Comparison of water treatments used to reduce the aggressiveness of soft waters

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Abstract

Soft waters tend to reduce the life time of pipes from drinking water distribution systems and the induced corrosion is likely to deteriorate water quality at the consumer's tap. The new French legislation (decree 2001-1220 from 20/12/2001) effected by the end of 2003, recommends the supplied water not to be aggressive. Two types of treatments are commonly used to limit the corrosion effect of soft waters with low alkalinity: (1) the addition of phosphates corrosion inhibitors and (2) the neutralization or remineralisation of soft water. In order to provide guidelines for network operator to choose suitable treatment methods, a comparison must be made based on technical plan (reliability and effectiveness) and economical point (treatment cost and impact on pipes renewing). Within this study, a pilot experiment has been carried out by the end of 2003 to compare the impact of phosphate inhibitors with remineralisation on the degradation of pipes commonly used in drinking water distribution systems (cast iron, cement and steel) and household plumbing systems (copper). The impact of water treatments was studied by means of water analysis (pH, metal leaching, etc) and corrosion measurements (electrochemical probes, weight loss coupons). The results obtained over a 1-year period proved that remineralisation will reduce metal pipe corrosion and improve water quality of soft waters better than phosphate corrosion inhibitor.

Keywords

Pipe corrosion, remineralisation, phosphate inhibitors; drinking water distribution system, water quality

INTRODUCTION

The maintenance of distributed water quality up to the consumer's tap represents one of the main concerns of the network operator and sanitary authorities. It requires the knowledge of factors which deteriorate water quality causing internal corrosion of drinking water distribution systems.

In drinking water distribution systems, interactions between supplied water and pipes internal wall are important. They depend on both the nature of materials (cast iron, cement based material, steel) of the pipe and the physico-chemical characteristics (pH, conductivity, alkalinity, carbonate calcium saturation, etc) of the supplied water.

The current study was undertaken to examine at a pilot scale, the impact of two treatments used to limit the aggressiveness of soft waters. Indeed, soft waters tend to reduce the life time of the pipes in public and/or household plumbing distribution systems due to their aggressiveness towards calcium carbonate scale and to their corrosivity towards pipe walls. The induced corrosion has a high economic incidence for water utilities with the failure of the distribution system pipes such as water leakage, pipe breaks, overflows, clogging of pipes with corrosion products. One other effect of corrosion is consumers complaints, due to a degraded water quality (red water, bad taste and odour as well as eventually health problems depending on the pipe materials). Coloured water often results from improperly treated water that contains iron and / or from the dissolution of the corrosion scales in corroded iron pipes (Sarin *et al,* 2003). The overall consequences of pipes corrosion might be minimized by rehabilitations and more frequent renewing. However, such operations often represent an important cost issue, and preventive water treatments usually offer good economical alternatives.

In order to reduce the corrosivity of soft waters or low-alkalinity waters, two types of treatment are commonly used (1) the addition of phosphate corrosion inhibitors or (2) the neutralisation or remineralisation treatment which is a process correcting the mineral composition of the water.

Phosphate inhibitors are one of the most common methods used to prevent iron corrosion in drinking water distribution systems. Phosphates have been introduced in distribution systems since 1940 (Hoover and Rice, 1939). Phosphate inhibitors include phosphates and metaphosphates linear and cyclic polyphosphate, glassy polyphosphate, orthophosphate, blends of ortho and polyphosphate. The most common types of phosphate inhibitors used nowadays are polyphosphate (hexametaphosphate) or orthophosphate. Phosphate addition is a passivation approach to corrosion control. Several studies indicate the advantages of using polyphosphate inhibitors for preventing iron corrosion or controlling red water (see McNeill and Edwards, 2001; AWWARF, 1996). Several studies discussed the theory of polyphosphate corrosion prevention. Some researchers that claimed phosphates inhibitors had little effect on iron corrosion at low flow or stagnant conditions (McNeill and Edwards, 2000; Maddison and Gagnon, 1999, Rompre et al., 1999). Unlike polyphosphate, orthophosphate are added for scale formation but not for iron sequestration (Wagner, 1992; Benjamin et al., 1990).

The main purpose of the remineralisation is to obtain passivation of the pipe material by forming calcium hydroxide, calcium carbonate shell on its surface. This can be achieved by adding calcium hydroxide, calcium carbonate or sodium hydroxide to the water in order to reach an equilibrium pH, which is relatively high for soft water ($pH > 8$).

A 24-month pilot study was initiated to compare the effectiveness of these two treatments on four types of materials commonly used in public distribution systems (iron, scrapped and unscrapped old cast iron and cement ductile iron) and on one material only used in plumbing distribution systems (copper). The aim of this study is to better understand, on a long term basis, the interactions between water treatment and pipe corrosion. From these results, guidelines shall be proposed to help the operator to find the optimal strategy which would minimise the corrosion rates on iron, copper and cement-based materials in term of reliability and effectiveness.

After a brief review of the general experimental approach and a discussion of the quality control procedures of the pilot, 12-month results of the study will be presented. Finally, the implications of these results to better understand the impact of phosphate inhibitors towards remineralisation have been evaluated.

MATERIALS AND METHODS

Selection of treatments and pilot plant configuration

The pilot plant (Figure 1) is located at the reservoir of the city of Maisons-Laffitte (France). The pilot plant configuration was set up to compare five distinct water qualities, which characteristics are reported in table 1.

Table 1. Average water quality characteristics for the five units plant of the pilot from January, 2004 to March, 2005

Water quality parameter	Unit 1 : reference water	Unit 2 : soft water	Unit 3 : remineralisation of soft water	Unit 4 : soft water + polyphosphates	Unit 5 : soft water + orthophosphates
Temperature (°C)	19,2	19,4	18,2	18,8	19,0
pH	7,90	7,00	8,31	7,05	6,92
Conductivity (µS/cm)	295	102	268	100	105
Calcium (mg/l $CaCO_3$)	127	41	90	43	45
Alkalinity (mg/l $CaCO_3$	71	23	93	25	223
Hardness (mg/l $CaCO_3$)	101	35	104	35	35
Chloride (mg/l)	6,1	1,9	1,9	2,0	2,2
Sulfates(mg/l)	10,3	4,0	40,8	4,1	4,4
Saturation pH	7,95	8,87	8,01	8,83	8,84
Saturation index	-0,05	-1,8	0,32	-1,84	-1,83

The pilot plant included alternative water treatments strategies under consideration (neutralisation, addition of polyphosphates and orthophosphates) in order to minimize metal release. Four water qualities were therefore obtained by adequate treatment of the tap water form the public network, which served as reference water. Each water quality was then used to feed a pilot plant unit, composed of pipe loops with different materials (Figure 1).

The first unit (unit 1) was supplied with the water distributed by the drinking water system of Maisons-laffitte with no complementary treatment. This water was in equilibrium with a saturation index ranging from – 0, 2 to 0,08.

Unit 2 was fed with soft water, obtained form the reference water passing through a softener and a reverse osmosis unit. The final composition was as close as possible to natural soft water with low alkalinity (pH ≈ 7, hardness ≈ 40 mg $CaCO_3$/l and alkalinity ≈ 40 mg $CaCO_3$/l). Table 1 shows the expected characteristics of the soft water was obtained with a good accuracy. The soft water supplied the unit 2 and also constituted the aggressive and corrosive reference water for anti-corrosion tests of units 3, 4 and 5.

Unit 3 supported the remineralisation approach. This approach involves the formation of $CaCO_3$ scale in order to create a protective scale to block the contact between the electrolyte and the anodes and cathodes. The process used during this study is the soft water (same as unit 2) passing through a column of calcareous alga (NeutralG$^{®}$). The reactions involved are as follows:

[1] $CaCO_3 + CO_2 + H_2O \rightarrow Ca(HCO_3)_2$
[2] $MgCO_3 + CO_2 + H_2O \rightarrow Mg(HCO_3)_2$

An addition of sodium hydroxide was performed at the exit of the column in order to have a pH close to 8,3. This value permit to obtain a saturation index ranging between 0,3 and 0,4 in order to accelerate the formation of a ferricalcite scale (Boireau, 2000).

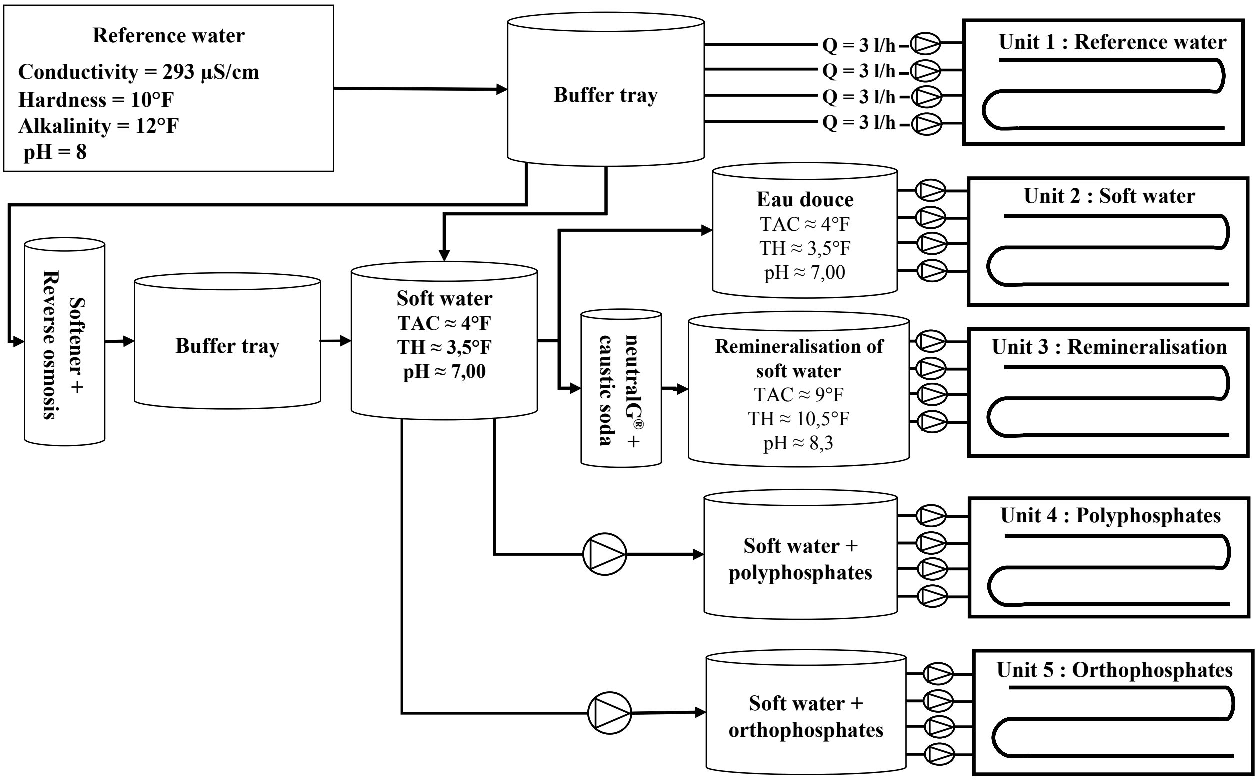


Figure 1. Pilot plant schematic diagram of the pilot set-up

Unit 4 and Unit 5 were fed with phosphate treated soft (same as Unit 2) water. Based on the characteristics of the soft water, two inhibitors were selected by the Aquarex –Arcie Company: Hydrex 3140 (Hexametaphosphate = polyphosphate) and Hydrex 3420 (orthophosphate). They proceed to be the most efficient to reduce the corrosivity of the soft water. The physico-chemical characteristics of the water produced in each of these units were closed to those obtained for unit 2. It involves the description of only soft water characteristics in Table 2.

The treatment rate was fixed and stabilized in each unit plant at 3 mgPO$_4$/l. Static inline mixers were located downstream of the injection ports to ensure fast and reliable mixing. As shows in Figure 2, orthophosphate and polyphosphate doses were maintained at the desired concentration during the first 12 months of the study.

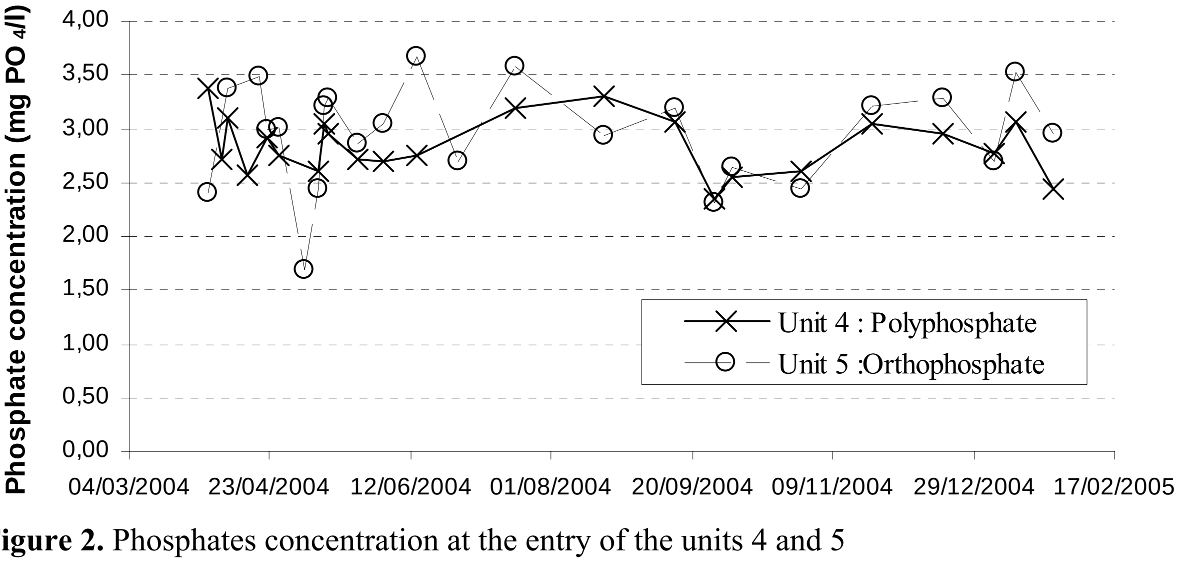


Figure 2. Phosphates concentration at the entry of the units 4 and 5

Pilot pipe-loop construction

Each of the five units (Fig. 3) consisted of five independent flow-through PVC loops. Each loop, excepted copper loops, offers six removable 30 cm long pipe sections of 60 mm inner diameter, connected with quick fit coupling, so that each pipe section could be easily sampled for further analysis. These inserts were machined to fit the assemblies and to minimize flow distortions. The inserts were coated with epoxy on the outer surface and edges to limit corrosion to interior surfaces. Pipe section samples were made up of different type of materials:

- loop 1 = Portland cemented ductile iron pipe samples
- loop 2 = steel pipe samples
- loop 3 = scrapped old unlined cast iron samples
- loop 4 = old unlined cast iron samples
- loop 5 = only with copper : copper loops were made of a single 6 m long line of 12 mm diameter copper tube.

As a whole, the pilot is composed of twenty five loops (5 loops with cement pipe samples, 5 loops with steel pipe samples, 5 loops with old unlined cast-iron pipe, 5 loops with scrapped old cast-iron and 5 loops in copper), figuring five independent units fed by five water qualities.

Each cast-iron loop was constructed with samples from old, unlined cast-iron pipe removed from the Syndicat des Eaux d'Ile de France (SEDIF), water distribution system in the city of Saint-Denis. This pipe was 90-100 years old and had an internal diameter of 60 mm (2,4 in.). The internal surface of the pipe was only slightly turbaculated. In order to study the impact of rehabilitation on the water quality and iron release, half of the old cast-iron pipes collected was scraped and installed on a distinct loop in each treatment unit. Detailed information on the quality of water that passed through this pipe over the years of its use was not available. Concerning loops made with few cemented ductile iron and loops made with steel pipes, brand new pipes were used for the study.

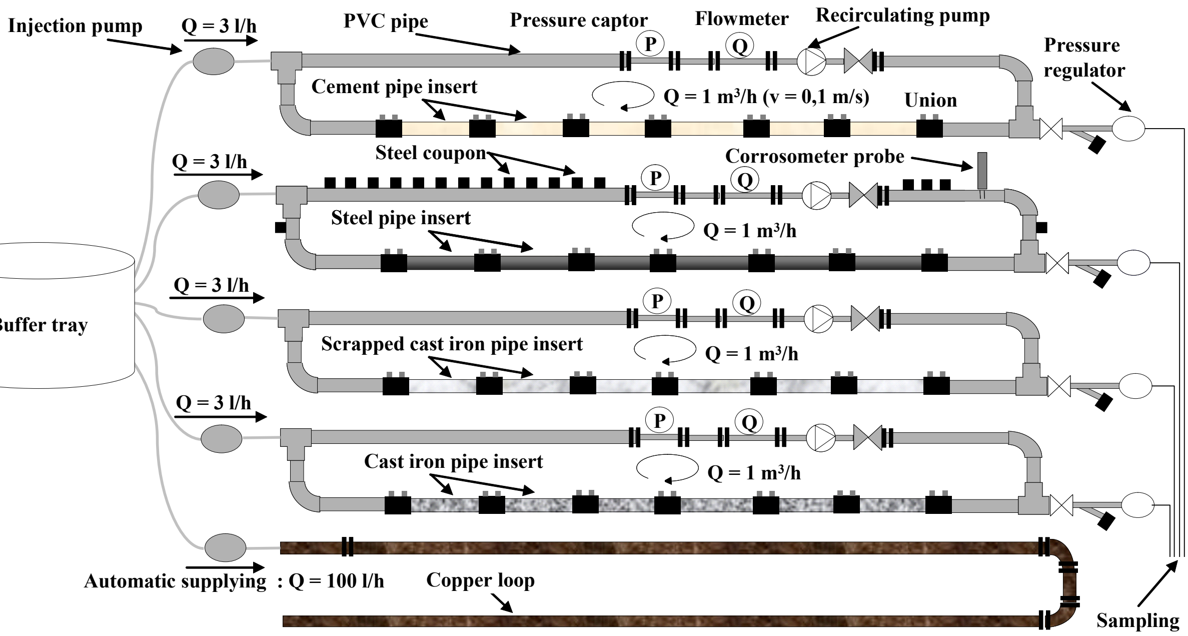


Figure 3. Schematic diagram of a treatment unit

Operational procedures: case of loops with cement, steel and cast iron

The inlet and outlet flow of these loops is about 3 l/h. The initial flow rate through each loop was 1 m^3/h which resulted in a velocity of 1 m/s. and a pressure of 3 bars. The volume of each loop being 18 L, the residence time was about 6 hours. These hydraulics characteristics are close to those which can be met in a real distribution system. They are based on hydraulic modelling on real drinking water distribution systems in France (Jaeger *et al.*, 2002; Jaeger *et al.*, 2003)

The corrosion impact of the different water qualities on materials was assessed through a weekly sampling and analysis in the influent of each of the five treatment units and the effluent of each loop. Sampling were carried out in order to monitor pH, alkalinity, temperature, turbidity, conductivity, hardness, calcium carbonate, total iron in the loops constituted with iron or cast iron and aluminium for cement pipe loops and the phosphate rate for the involved loops. The aluminium and total iron measurements were made twice a week during the study using an atomic absorption spectrophotometer (PERKIN ELMER Optima 3300 DV) in accordance with Standard Methods (NF EN ISO 11885).

Steel corrosion coupon

To each of the five loops installed by using six steel pipe inserts, 18 weight loss corrosion coupons made of mild steel were also installed. These coupons were developed by Anjou Recherche to obtain a reliable tool to measure the corrosion in distribution system. The corrosion coupons are made of a steel washer on a Teflon^{TM} support (Fig. 4.a). These coupons were inserted flush to the pipe wall via a 20/27 fitting (Fig. 4.b). The purpose of the use of such coupons was to evaluate the corrosion in distribution systems which can be subsequently performed at different levels:

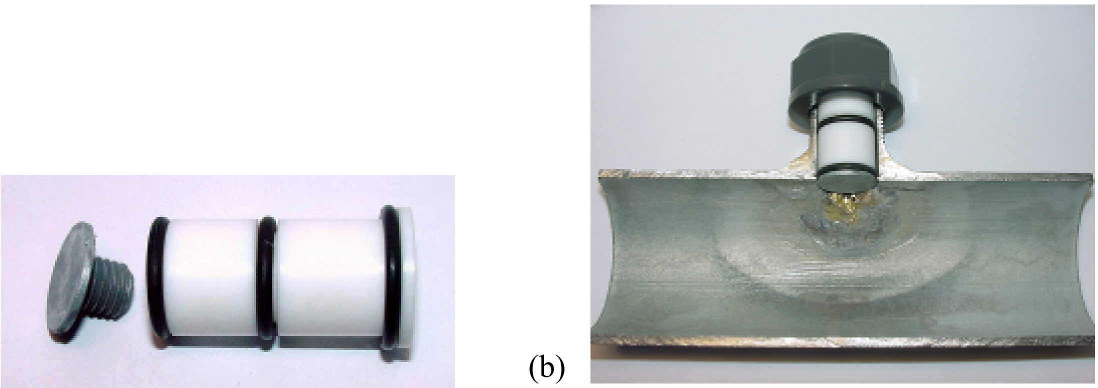
- The easier exploitation consists in a visual observation of the coupon aspect to evaluate the corrosion scale, the presence or lack of corrosion-inhibition product, the eventual presence of pitting. This inspection is totally non-destructive. After the examination, the coupon can be replaced in the pipe for further investigation.
- A second degree of exploitation consists on performing weight loss measurements in order to evaluate the quantity of oxidized metal. Such a measure requires a suitable preparation of the coupon in order to remove scrap the adherent scale according to NF ISO 8407. The corrosion rate can be calculated by the following equation :

  [3] $$v_{corr} = \frac{M_i - M_f}{m_v \times S \times t} \times 365 \times 10^4$$

  where v_{corr} = average corrosion rate (µm/year); M_i = initial mass of the coupon (g); M_f = final mass of the coupon (g); m_v = volumetric mass of the metal (g/cm^3: 7,8 g/cm^3 for steel); S = exposed coupon area (cm^2) and t = exposition time of the coupon (day)
- These coupons can also be used for a more complete corrosion characterization as well: surface analysis, nature of both the corrosion scale or corrosion-inhibition product, biofilm analysis

Within the frame of this pilot study, the above two first evaluation levels were considered. The steel coupons weight loss rates were expressed in terms of equivalent rates of penetration in micrometers per year. At 3-months intervals for the first year and 6-months for the second year, three iron sampling were carried out by removing from each iron loop and subsequently replaced with a new set of the same type. Thus, the final set will include coupons exposed for

3, 6, 9, 12, 18 and 24, providing replicates of the 6, 12, 15, 18, 21 exposures although for different times of the year.



(a) (b)

Figure 4. Steel corrosion coupon and Teflon^{TM} support (a) and steel corrosion coupon inserted in pipe (b)

Mild steel corrosion probes (corrosometer)

Corrosion probes were also used. They consisted in a measurements device (type Corrater 9030+^{TM} from Rohrback Cosasco Systems) linked to a probe with two mild steel electrodes. This device proposes to determine the polarization resistance of the corrosion process. The resistance is used to assess the intensity of the corrosion current (i_{corr}), by using Stern and Geary equation [Eq. 4]. The corrosion rate is then calculated from the corrosion current deriving from the Faraday law.

[4] $$R_p = \frac{1}{2{,}3.i_{corr}} \frac{b_a.b_c}{b_a + b_c} = \frac{B}{i_{corr}}$$

where b_a and b_c which are the Tafel coefficients: they are considered constant and ranging from 40 mV to 150 mV respectively. The value of B factor can be estimated around 20 mV.

In the framework of the pilot study, mild steel probes were installed on each of the five steel-loops. The corrosion rate was measured in continuous.

Operational procedures: case study using copper loops

Currently, copper pipes are used in most of household plumbing systems. In order to quantify the impacts of these treatments on the copper release, copper pipe have been installed on the pilot.

As a simulation of the behaviour of household plumbing systems, the copper loop has a different pattern in comparison with the others. An electronic automate has been installed in order to simulate household using copper. This protocol is based on research work led by European laboratories and coordinated by the CRECEP (Centre de Recherche et de Contrôle des Eaux de Paris) (European Commission, 2000; DIN, 1999). The following schedule was used:

7:30 – 9:00 : valve open, water flowing for 1 h 30
9:00 – 11:30 : valve closed, no flow for 2 h 30
11:30 – 12:30 : valve open, water flowing for 1 h
12:30 – 17:00 : valve closed, no flow for 4 h 30

17:00 – 19:30 : valve open, water flowing for 2 h 30
19:30 – 21:30 : valve closed, no flow for 3 h
21:30 – 22:30 : valve open, water flowing for 1 h

Thus, water flowed for 6 hours each day. The initial flow rate through each copper loop was 100 l/h which resulted in a consumption of 600 l/day which is representative for about four households. The copper measurements in the outlet water were made weekly during the study on a PERKIN ELMER Optima 3300 DV atomic absorption spectrophotometer in accordance with Standard Methods (NF EN ISO 11885). Three samples were collected weekly in order to monitor copper release after three different stagnation times (0, 30 minutes and 9 hours).

RESULTS AND DISCUSSION

Effect of water treatments on copper release

The main objective of this phase was to examine the effect of different water treatment strategies on copper-releases rates. Copper release was monitored in the effluent of all the 5-loops with copper. For copper release, the results of loops supplied with the reference water were not considered because the goal of this study is to compare the impact of remineralisation and addition of phosphate inhibitors on the soft water. The measured copper level in the effluent of copper pipes for three stagnation times (0, 30 minutes and 9 hours) are represented graphically in Figures 5, 6 and 7.

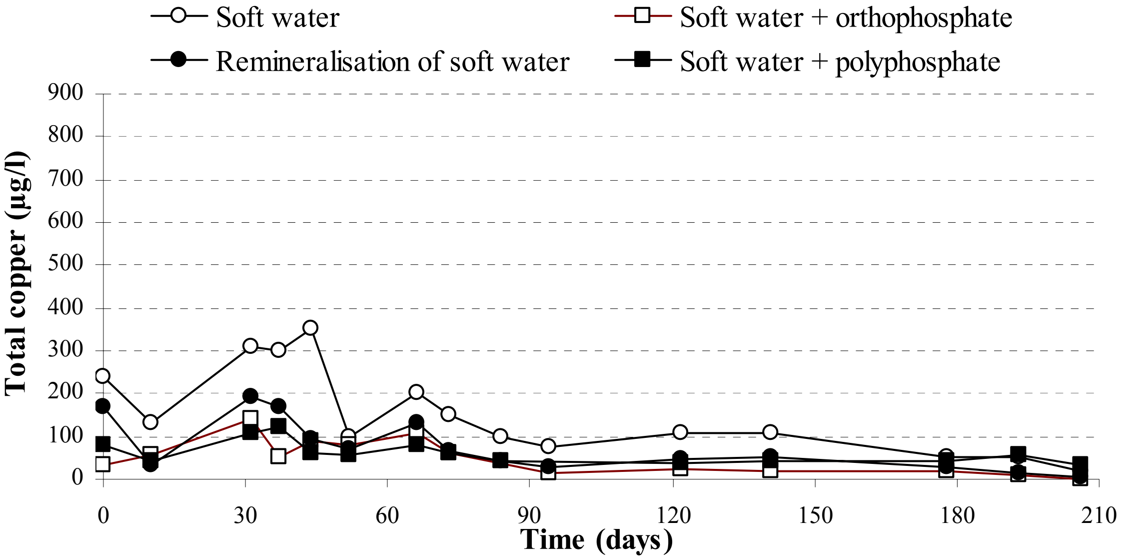


Figure 5. Copper levels in brand new pipes after no stagnation

Without stagnation, the lowest copper levels were from orthophosphate treated loop, followed by remineralisation and polyphosphate loops. Figure 5 shows that all the treated loops had copper levels above the European legislation standard of 200 µg/l. The highest copper concentrations were measured outlet the pipe supplied with the soft water. For the soft water, the highest copper concentration occurred in the form of pikes (e.g. > 200 µg/l during the first month of the study). However, there was a decreasing trend in copper levels with time in the soft water. Whatever the type of treatment, this no-stagnation study shows an average improvement of at least 60% for the copper release reduction. This improvement is based on the following equation:

$$[5] \quad \%\ \text{improvement} = \frac{(\text{Total copper release with treatment} - \text{Total copper release without treatment}}{\text{Total copper release without treatment}} \times 100$$

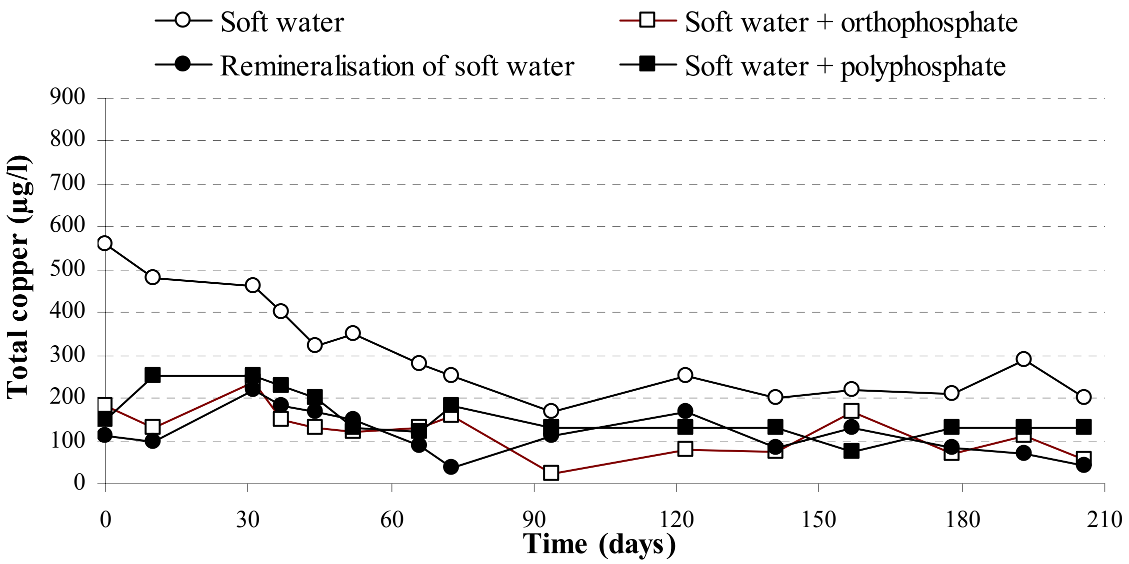


Figure 6. Copper levels in brand new pipes after 30-minutes stagnation

For 30-minutes stagnation, the highest copper levels were measured for the soft water loop. As shown in Figure 5, a decreasing trend in copper level in the soft water was observed, but this trend was less pronounced. Whatever the treatment, the average improvement of copper release is similar. So, average copper levels in treated loops were 50% lower than those measured outlet the soft water loop.

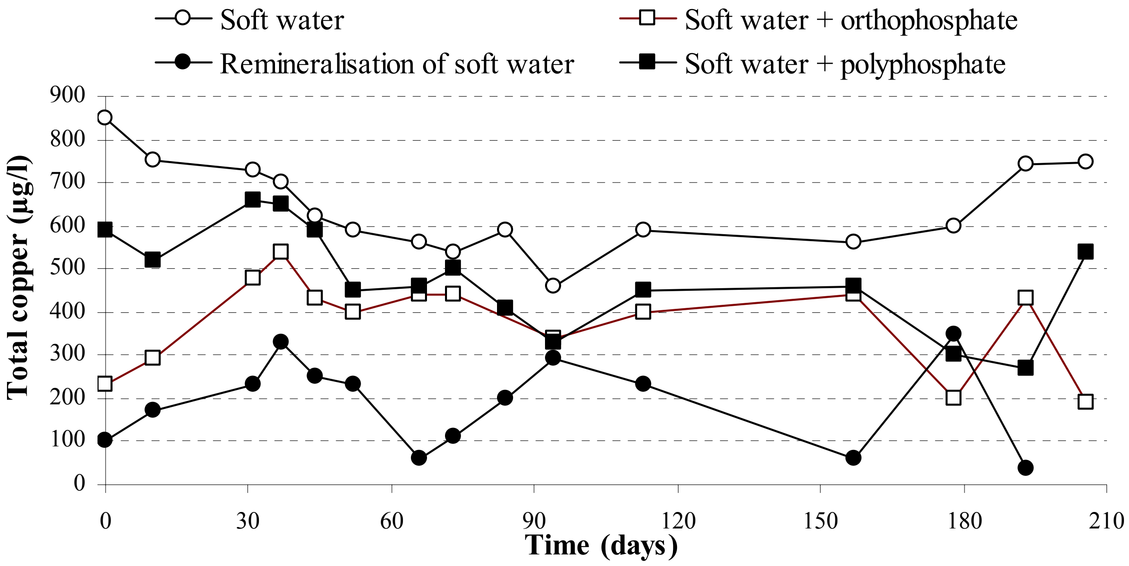


Figure 7. Copper levels in brand new pipes after 9-hours stagnation

For 9-hours stagnation, high copper levels were encountered particularly for soft water. Contrary to previous observations, there was not a decreasing trend in copper levels with time in soft water loop. The lowest copper levels were from remineralisation treated loop, followed by phosphate inhibitors loops. Average copper levels in remineralisation loop were 50 %

lower than for orthophosphate loop, the next lowest loop. Overall, the impact of phosphate inhibitors on copper release reduction is less observable than in figures 5 and 6.

In summary, for all stagnation times, the lowest copper levels were measured outlet the remineralisation loop. Nevertheless, phosphate-based inhibitor treatments appeared to provide a significant degree of beneficial protection to the copper pipe over that from soft water. Overall, the inhibitors protection tends to decrease when the stagnation time increases. The difference between the soft water and the treated loops is a combination of less aggressive environment (higher pH and alkalinity for remineralisation and addition of phosphates). Outlet the copper loop treated with the remineralisation, this treatment seems to facilitate the formation of a protection scale (CuO, $Cu(OH)_2CO_3$) at the surface of the copper pipe. Nevertheless, this study underlines the beneficial effect of phosphate-based inhibitors addition in copper release reduction for water whose characteristics are pH ≈ 7,0 and alkalinity ≈ 40 mg/l as $CaCO_3$. Overall, orthophosphate had lower copper release than did polyphosphate but this performance was a slightly significant for the three stagnation time studied. Dosing of 3 mg PO_4/l orthophosphate led to reductions in copper release from 70 % for no-stagnation time to 40 % for 9-hours stagnation time. For polyphosphate, a dosing of 3 mg PO_4/l led to reductions in copper release from 60 % for no-stagnation time to 30 % for 9-hours stagnation time. The chemical explanation of phosphate impact on copper release reduction is not well known. Maybe, phosphates complex copper at the surface of the pipe to form a film or in contrary, phosphates contribute to accelerate the formation of Cu_2O on the copper pipe wall. In summary, for brand new copper pipes supplied with a soft water treated or not, copper release decreased in the following order:

Remineralisation treatment > orthophosphate > polyphosphate > no treatment

Effect of Portland cemented ductile iron pipes on water quality

Some research have revealed that the degradation of cement based materials involves the dissolution of calcium hydroxide and an increase of the water's original pH which can be accompanied by aluminium dissolution (Kristiansen *et al,* 1978; Vik and Weidborg, 1991; Conroy and Oliphant, 1991).

The water quality monitoring performed on cement loops don't take into evidence important difference between the water qualities (Table 2 gives the pH level measured after a contact time of 6 hours and Table 3 gives the aluminium concentration).

Table2. pH levels before and after a contact time of 6 hours in cement loops

Time (months)	Soft water		Remineralisation of soft water		Soft water + orthophosphate		Soft water + polyphosphate	
	Inlet	Outlet	Inlet	Outlet	Inlet	Outlet	Inlet	Outlet
0	7,01	7,40	8,25	8,20	6,95	7,40	6,97	7,54
0,25	7,04	7,42	8,18	8,20	7,00	7,42	7,01	7,59
0,5	7,02	7,46	8,54	8,50	7,00	7,46	7,00	7,63
0,75	6,99	7,57	8,41	8,40	6,95	7,57	6,96	7,55
1	7,01	7,62	8,30	8,3	6,98	7,62	6,96	7,44
2	6,99	7,75	8,35	8,35	6,95	7,75	6,98	7,37
3	7,03	7,80	8,20	8,20	7,01	7,80	7,02	7,34
6	6,96	7,72	8,41	8,41	6,98	7,72	6,98	7,33
9	7,05	7,61	8,30	8,30	7,03	7,19	7,05	7,19
12	6,98	7,5	8,30	8,30	6,94	7,19	6,94	7,18

The table results show that an increase of pH and aluminium can be observed outlet the cement loop supplied with both soft water and phosphates inhibitors. However, the impact of phosphates is less important than soft water on these decreases. The higher values of pH and aluminium concentrations were measured after 3-months operation. After this period, these values decrease but there is always a difference of 0,3 pH unit between the inlet and the outlet of each cement loop. Finally, phosphates have no impact to really limit the $Ca(OH)_2$ leaching. Concerning aluminium, no release was observed for phosphates treatment after 6 months contrary to soft water where the aluminium release tends to disappear after only 12 months. On the other hand, the remineralisation enables to limit the degradation of the cement lining that's why no increase of pH or aluminium concentration was measured.

Table3 Aluminium concentrations before and after a contact time of 6 hours in cement loops

Time (months)	Soft water		Remineralisation of soft water		Soft water + orthophosphate		Soft water + polyphosphate	
	Inlet	Outlet	Inlet	Outlet	Inlet	Outlet	Inlet	Outlet
0	< 0,02	0,03	< 0,02	0,02	< 0,02	0,02	< 0,02	0,02
0,25	< 0,02	0,04	< 0,02	0,02	< 0,02	0,04	< 0,02	0,04
0,5	< 0,02	0,05	< 0,02	0,02	< 0,02	0,05	< 0,02	0,04
0,75	< 0,02	0,05	< 0,02	0,03	< 0,02	0,05	< 0,02	0,04
1	< 0,02	0,06	< 0,02	0,03	< 0,02	0,05	< 0,02	0,07
2	< 0,02	0,06	< 0,02	0,03	< 0,02	0,06	< 0,02	0,05
3	< 0,02	0,05	< 0,02	0,03	< 0,02	0,06	< 0,02	0,04
6	< 0,02	0,04	< 0,02	< 0,02	< 0,02	0,03	< 0,02	0,03
9	< 0,02	0,03	< 0,02	< 0,02	< 0,02	< 0,02	< 0,02	< 0,02
12	< 0,02	< 0,02	< 0,02	< 0,02	< 0,02	< 0,02	< 0,02	< 0,02

Iron release studies

The goal of this phase was to examine the effect of different water treatment strategies on iron-releases rates. Iron release was monitored in all the 15-loops with metallic section (cast iron and steel). For iron release, the results of loops supplied with the reference water were not considered in order to only compare the impact of remineralisation and addition of phosphate inhibitors on the soft water.

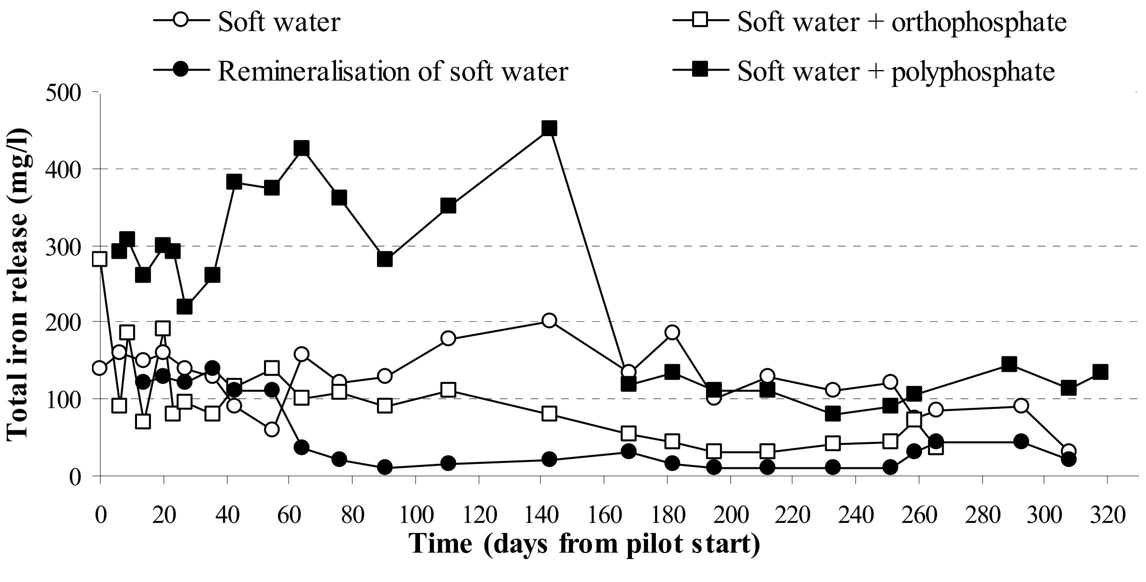


Figure 8. Iron release observed in loops with old unlined cast iron pipes

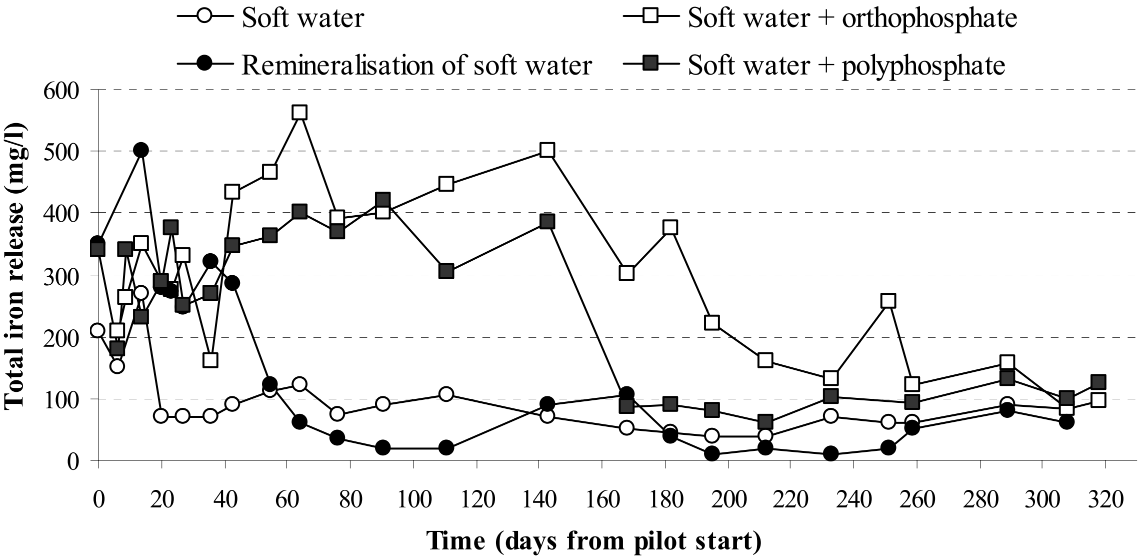


Figure 9. Iron release observed in loops with scrapped old unlined cast iron pipes

Effect of phosphate inhibitors

Many studies have demonstrated that phosphates can be effective in decreasing lead and cooper corrosion. Nevertheless, they can be quite detrimental to iron corrosion (MacNeill et al, 2000). If we assume all of the experimental parameters (two inhibitors types, three pipes-loops types), there were six different combinations of conditions. For five of these conditions, addition of either orthophosphate or polyphosphate increased the iron release in comparison with the results obtained for soft water (Figures 8 – 10). There was only one condition were the addition of orthophosphate reduced iron release (case of old unlined cast iron pipe-loop). For this condition, orthophosphate enables an average iron reduction of 60 % in total iron release.

The scraping has a significant effect on iron release during the first two months of the study, the iron release in the effluent of old unlined cast-iron pipes-loops (Figure 8) is lower than iron release in the effluent of scraped old unlined cast iron (Figures 9). The same trend was observed with iron pipes (Figure 10) which present like scrapped cast-iron pipe, a metallic surface uncovered.

A general observation that can be made from the iron release data is that the addition of orthophosphate or polyphosphate inhibitors never had a beneficial effect on iron by-products release. It either increased iron release or had no effect compared to a pipe supplied with soft water. Nevertheless, orthophosphate has a slight effect on old unlined cast-iron where the average iron levels is lower than for untreated loop. For all other conditions, the addition of phosphate increased total iron concentration in comparison of iron levels measured.

The influent residual of 3 mg PO_4/l was reduced to 1-2 mg PO_4/l in the effluent from the loops with unlined either no-scraped or cast iron or iron pipes. The phosphate was either adsorbed on the corroded wall of the iron pipe or may have precipitated as iron phosphate compounds in the corrosion scales. This decrease was the most important in the effluent of iron pipes loops with a phosphate concentration which was 75% lower than the influent.

According to few studies, orthophosphate form, when corrosion scale are present at metallic pipe surface, protective “plugs” in pores in the surface layer or in the shell-like layer, and thus reduce porosity and the rate at which ferrous ions diffuse out (Sarin et al, 2002). Thus, orthophosphate can increase the scale impermeability and decrease iron release (Vik et al, 1996). These results can explain the low iron release in the effluent of the old unlined cast iron pipe-loop.

The observation of water outlet the metallic loop treated with phosphates didn't show any phenomena of red waters in spite of the high iron levels measured. This observation can be explained by the complexing of ferrous ions which involves the removing of red waters (reducing of Fe(III) to Fe(II)) but the increase of total iron levels.

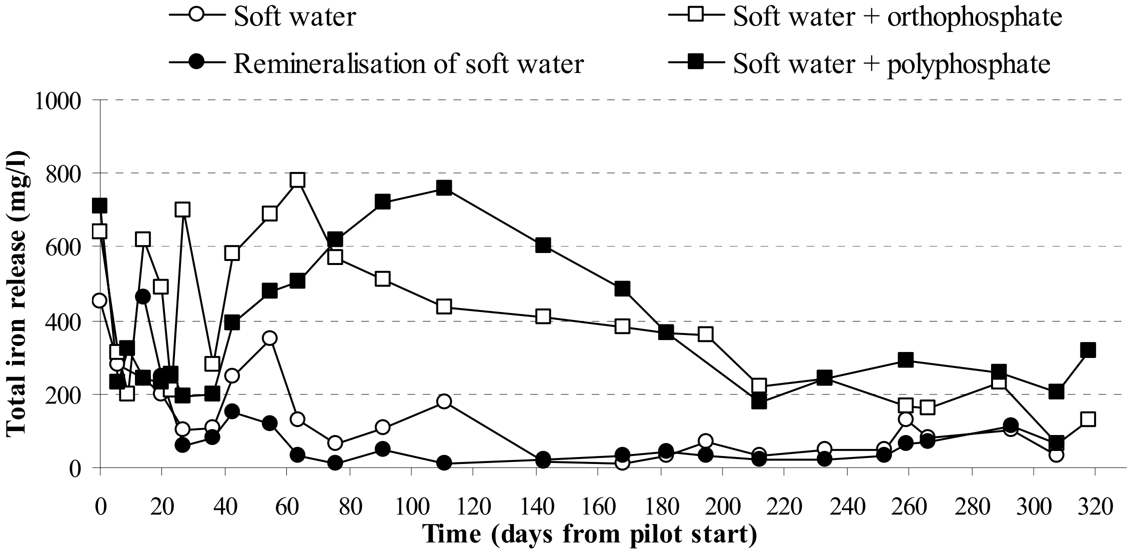


Figure 10. Iron release observed in loops with brand new iron pipes during the study

Effect of remineralisation

In response to the remineralisation, the iron levels are lower than 0,05 mg/l and that, whatever the type of analysed pipes. As shown in Figures 8, 9 and 10, the remineralisation reduced iron release over a period of 2 months. This period can probably be attributable to the implementation of a steady-state at the surface of the pipe material. Indeed, at the start of the study, pH and alkalinity slightly fluctuated so the formation of a protective scale has taken some time. Changes in alkalinity from 35 – 40 mg/ to 80 – 90 as CaCO3 showed that higher alkalinity resulted in lower iron release (Sarin *et al*, 2002). Calcite (CaCO3) plays a role in reducing iron release in old unlined cast iron pipes by reducing the porosity of the scale. It seems to be possible that a wide variety of conditions can exist locally; calcium carbonate deposits may form and block some pores inside corrosion scales. A comparison of iron release behaviour for phosphate inhibitors and remineralisation show that iron release was reduced to lower levels for remineralisation. Contrary to phosphate inhibitors, the remineralisation enables the formation of iron corrosion scale on the pipe wall of uncovered pipe wall (scrapped cast-iron and iron).

Steel coupon

The iron corrosion rates in the pilot study are illustrated by weight loss measurements in Figure 11. The highest corrosion rates were found for the soft water and soft water treated with the two corrosion inhibitors. The lowest corrosion rate was observed for the re-mineralised soft water.

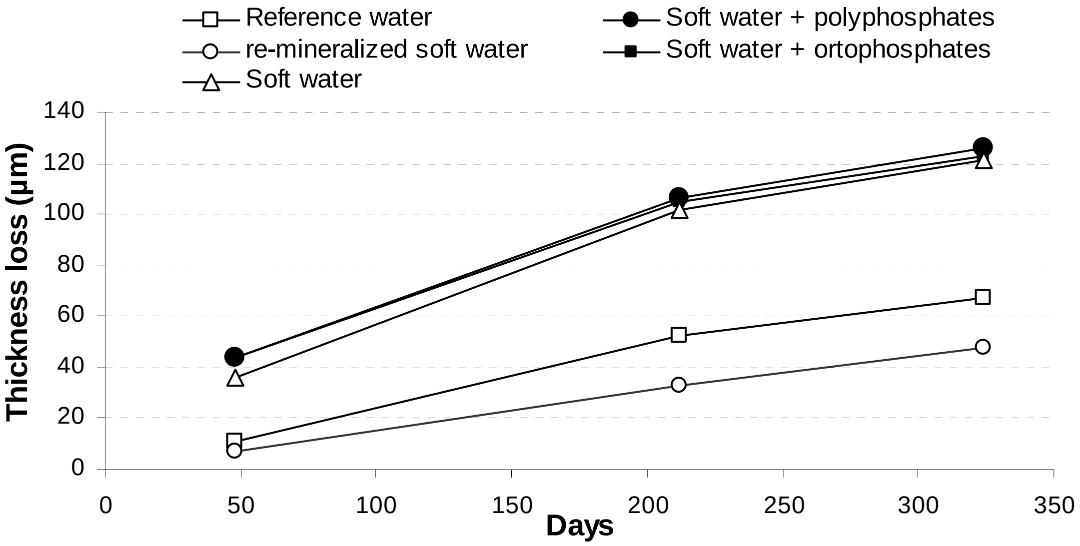


Figure 11. Iron corrosion measured with coupons (all points are averages of three samples)

For the remineralisation assumes to give a protective layer mainly with calcium carbonate, the corrosion rates decreased after a short period, while no similar decrease was observed when a water quality assumed to be protected by a passivation scale such as phosphate inhibitor. The initial corrosion during the first nine months of the study was higher for the soft water treated with corrosion inhibitors than for the soft water. In fact, the protective layer starts to be established rarely at short term with inhibitor treatments (Hem *et al*, 2001). Based on the two last coupon samples (i.e. between 6 month and 9 month in Figure 11), corrosion rate of steel was calculated for each water quality (Table 4).

Table 4. Summary of iron corrosion rate measured with coupons during the last three months of the pilot study

Water quality	Average corrosion rate with coupons (µm/y)	Average corrosion rate with probe (µm/y)
Reference water	48 ± 4	35
Soft water	65 ± 6	51
Re-mineralized soft water	48 ± 5	32
Soft water + orthophosphate	59 ± 2	-
Soft water + polyphosphate	64 ± 2	48

As shown in Table 4, the corrosion rates were highest for iron exposed to the soft water and soft water treated with polyphosphate. The re-mineralised treatment is close to the reference water which tends to assume that the remineralisation treatment performed well. This water treatment enables to reduce the iron corrosion by 25%. The treatment with polyphosphate seems to have no effect on corrosion inhibition. Increased corrosion often occurs because of the complexing and sequestering properties of polyphosphate leading to the formation of less protective layers. Contrary to polyphosphate, orthophosphate reduced the iron corrosion by 10%. This inhibiting effect can be due to the corrosion products which have formed insoluble phosphates and have improved the protective scale quality by increasing its impermeability and adherence (AWWARF, 1996).

Mild Steel corrosion probes

The corrosion rates measured with probes during the first ten months of the pilot study are shown in Figure 12. For all the water qualities, a fast decrease of the corrosion rate was observed during the period which follows the immersion. This phenomenon is due to the higher reactivity of the metallic surface uncovered. This phenomenon decreases during the formation of corrosion scale more or less protective. After one month, the corrosion rate of both re-mineralized and reference water tends to stabilise in contrary to soft water and soft water treated with phosphate inhibitor which waited the height month to be stabilised. The Table 4 addresses the corrosion rates of this plateau for each water quality.

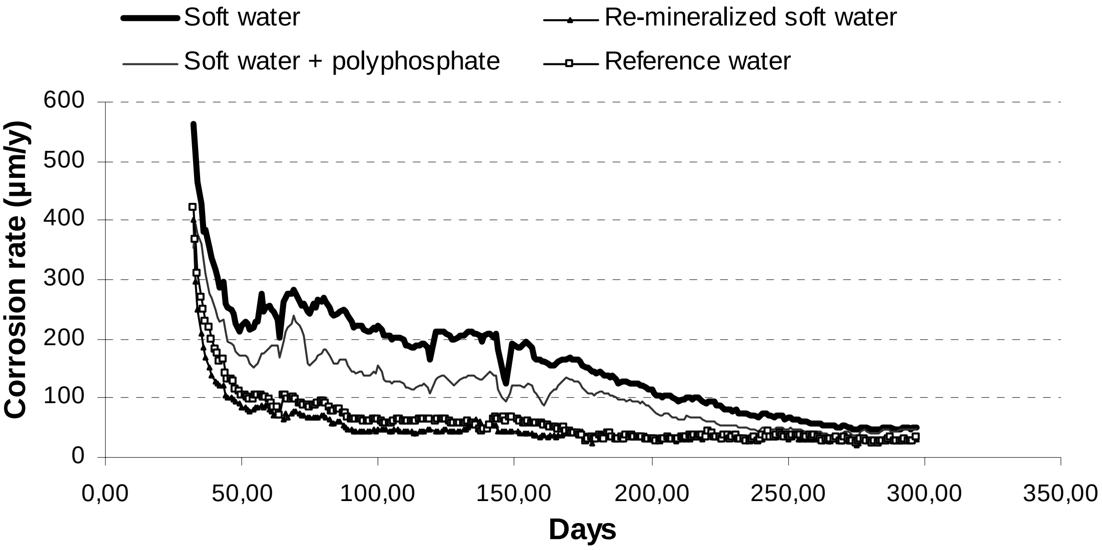


Figure 12. Iron corrosion rates measured with probes

The results of the probes confirm the results from the coupon tests, showing that the remineralisation of the soft water gives lower iron corrosion than soft water treated with polyphosphate inhibitor. The lowest corrosion rates have been observed for each corrosion tool with the reference water and the re-mineralised soft water. These two corrosion tools enable to distinguish without difficulty the situation of important corrosion (soft water and soft water treated with polyphosphate) and situations with moderated or low corrosion (re-mineralised water and reference water).

SUMMARY AND CONCLUSIONS

The main objective of this study was to study the impact of different water treatment such as phosphate-based inhibitors and remineralisation (pH and alkalinity adjustment) to limit the aggressiveness of soft water. This study was performed with a pilot pipe-loop system in order to compare the effectiveness of these treatments on few materials used in both drinking water distribution systems (cast iron, scrapped-cast iron and steel) and household plumbing systems (copper). Several means were used to carry out this comparison in order to have as many measures as possible to help the network operator to choose the suitable treatment. In this study, the main purpose was not to speculate on the processes and physicochemicals reactions involved in the pilot survey.

The results of the copper release studies enable to provide some information on the copper levels likely to be found as function as the type of the water quality. This monitoring of copper leaching was performed for three stagnation times in copper pipe. Phosphates inhibitors and the remineralisation appear to provide some significant degree of protection over that from the soft water. Generally speaking, the remineralisation seems to be the best treatment to limit the copper concentration at the consumer's tap, notably after long stagnation time.

The analytical survey performed on different materials representatives of those used in drinking water distribution systems underlines the beneficial impact of the remineralisation to reduce the iron release met in the soft water. Unlike to the remineralisation, the action of phosphate inhibitors was rarely significant for controlling iron release. Overall, these inhibitors tend to increase iron concentration. Compared to polyphosphate, orthophosphate treatment has a better effect to limit iron release. Nevertheless, this beneficial potential can be only observed on old unlined cast iron pipes due to the increasing of the scale impermeability leading to an iron release decreasing. Face to unprotected metallic pipes such as scrapped cast iron and brand new steel pipes installed in the pilot, orthophosphates and polyphosphates didn't appear to provide any significant effect to reduce iron release over that obtained from the soft water.

The corrosion probe experiment and the steel coupon experiment gave similar results and showed the remineralisation of soft water reduced corrosion rate below of this of the soft water. This water treatment tends to reduce the iron corrosion by 25 %.Contrary to remineralisation, orthophosphate reduced the corrosion rate by 10 % notably during the last three months of the study. This inhibiting effect can be due to the corrosion products. The inhibiting effect from orthophosphates can be due to the corrosion products which have formed insoluble phosphates and have improved the protective scale quality by increasing its impermeability and adherence Unlike these two treatments, polyphosphates tend to increase scale thickness and overall weight loss. In fact, polyphosphate can reduce red water phenomenon due to its complexing and sequestrating properties. These properties have no beneficial impact to reduce corrosion rate because they lead to the formation of less protective layers on the surface of pipes. Overall polyphosphate tend also to increase total iron levels at the consumer's tap due to the complexation of ferrous iron (Fe^{2+}).

This study provides some good information for water utilities supplied with corrosives soft water to better manage their network and improve the water quality at the consumer's tap. The remineralisation treatment is reliable on all types of materials (scrapped or not). This treatment protects the whole type of pipes. This study also enabled to study two different tools used to evaluate and follow the corrosion rate in a distribution system (coupons and electrochemical probes). The corrosion probes data corroborated the steel coupons data. They notably permitted to clearly distinguish corrosives or no corrosives water. For utilities, probes are relatively expensive but coupons are cheap. They can be easily installed in both a distribution system and a household plumbing system and visual observations can be also performed to help network managers. Moreover, these coupons can be used for biofilm analysis. Finally, these two tools constitute interesting devices to help water utilities to better survey and manage their drinking water distribution system.

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