

Reduction of lead concentrations in drinking water: evaluation of some industrial treatments

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1. Introduction

Since 1998, a new European drinking water legislation has been promulgated with new standard values for lead. Nowadays, this regulation requires that lead concentration must be lower than 25 µg/L. From 2013, the lead standard in drinking water will be lowered to 10 µg/L. Moreover, this legislation defines the control point as a water tap where regular consumption is achieved, which could be the tap in the kitchen in most cases. Thus, not only the sources of lead leaching in the public distribution network have to be eradicated, but solutions should also be available for in-house networks.

Already more than 20 years ago, some industrial treatments were proposed and evaluated for reducing lead leaching at consumer's tap. By modifying the water composition, a reduced lead leaching could be obtained in both public distribution and in-house networks. The main processes used to achieve reduced lead concentrations in drinking water were phosphate addition and decarbonation.

Within the context of the new European lead legislation, a new working group has been setup in CEOCOR Sector B. The task of this new working group will be to gather, summarize and analyse information about the most convenient industrial water treatment technologies that could provide lead concentrations lower than 25 µg/L at the consumer's tap. This article will review two water treatment technologies considered by the working group and already used: decarbonation and phosphate treatment. Benefits and drawbacks of both water treatment technologies will be presented and discussed, not only for lead dissolution but also for other aspects of drinking water quality such corrosion, bacterial growth, health and environmental impacts. Besides this general review, this presentation is also intended to promote discussion and exchange of experience with other CEOCOR members. This article will also present unanswered questions about these water treatment technologies and clarify some widespread and inaccurate ideas about lead corrosion. At the end, this work is expected to serve as guidance for water professionals not familiar with corrosion.

2. Plumbosolvency in water

The lead concentration in water has been explained by the dissolution of two different deposits present at the inner surface of lead pipes, for water pH ranging from 7 to 10: cerussite (PbCO_3) and hydroxycerussite ($\text{Pb}_3(\text{CO}_3)_2(\text{OH})_2$), both deposits arising from the presence of carbonate in natural waters (21). While cerussite is thermodynamically favoured and less soluble, analysis of real lead pipes have revealed that the more soluble hydroxycerussite is often found, either alone or in combination with cerussite (4). Thus, the solubility of lead will be controlled by the nature of the deposit, the most often encountered hydroxycerussite leading to an increased plumbosolvency (plumbosolvency represents the capacity of the water to bring lead into solution).

The solubility of lead is also influenced by many dissolved lead complexes represented in Figure 1, such as carbonate, hydroxy, chloride and sulphate ionic complexes (15). Among the

possible lead complexes, the ion pair $(\text{PbCO}_3)^{\circ}$ is certainly the dominant soluble complex over most of the pH range and alkalinity occurring in natural waters (21, 22). The presence of these complexing inorganic anions, together with some complexing organic matter, will promote lead dissolution from deposits.

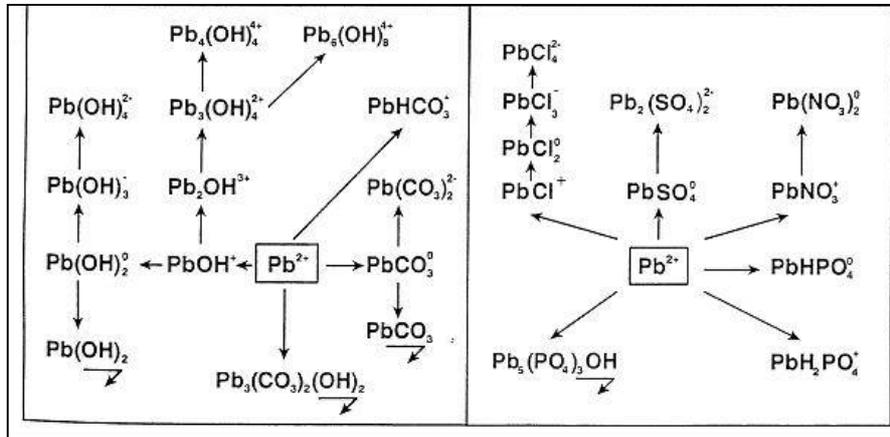


Figure 1: Lead complexes formed in solution (reproduced from Leroy, 1993).

The lead solubility is mainly influenced by the pH and the inorganic carbonate concentration (22). The variation of lead solubility, at equilibrium, is represented in Figure 2 as a function of pH and total carbonate (related to alkalinity) and shows that:

- lead solubility increases when total carbonate increases, especially above pH 7.5, due to the formation of lead carbonate soluble complexes; systems with high total carbonate can dissolve as much lead as those with less total carbonate at lower pH values
- lead solubility is more sensitive to pH than to total carbonate
- dissolution of lead deposit is minimized at pH between 9 and 10, for a total carbonate between 20 and 60 mg CaCO_3/L (the minimum being at 30 mg CaCO_3/L for most pH's)

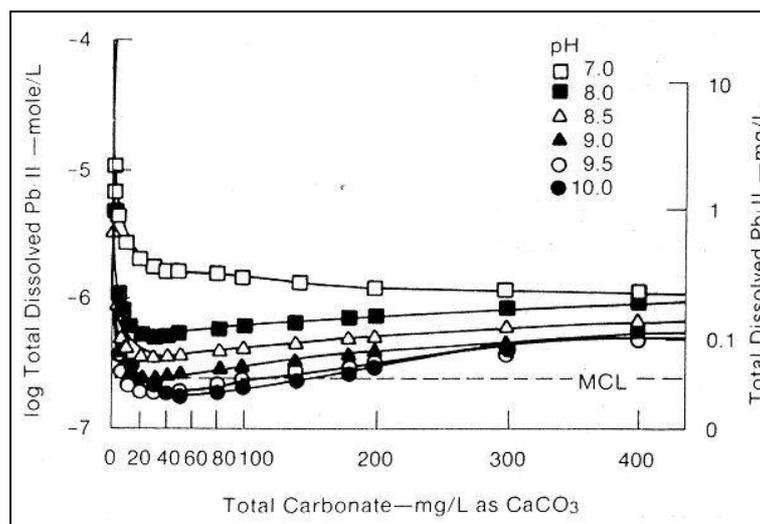


Figure 2: Lead solubility diagram as a function of pH and total carbonate, obtained from computational calculation at equilibrium conditions and 25 °C (reproduced from Schock, 1980).

A well-established idea is that reducing aggressivity of water will reduce its plumbosolvency. While this statement is verified when pH is increased, any increase in total carbonate made to reduce aggressivity might not necessarily diminish the lead solubility as soluble lead carbonate complex will improve it (24). Therefore, control strategies based on aggressivity correction might not always be successful. Moreover, the deposits controlling lead dissolution have been shown to be lead salts and not calcium carbonate salts (4, 21).

3. Decarbonation

Based on previous knowledge of lead deposits solubility, increasing the pH appears as the most efficient way to reduce plumbosolvency, especially for soft waters. For hard waters (calcium hardness > 250 mg CaCO₃/L), changing the pH will be rather difficult because of significant carbonate buffering effect and risk of calcium carbonate precipitation. To adjust the pH more easily, the total carbonate content must be reduced, which would contribute to decrease lead solubility too (15, 24). Changing both pH and total carbonate (or alkalinity) has been the underlining principle of the decarbonation process for lead control, as indicated with the arrow in Figure 3 (going from high to lower plumbosolvency water).

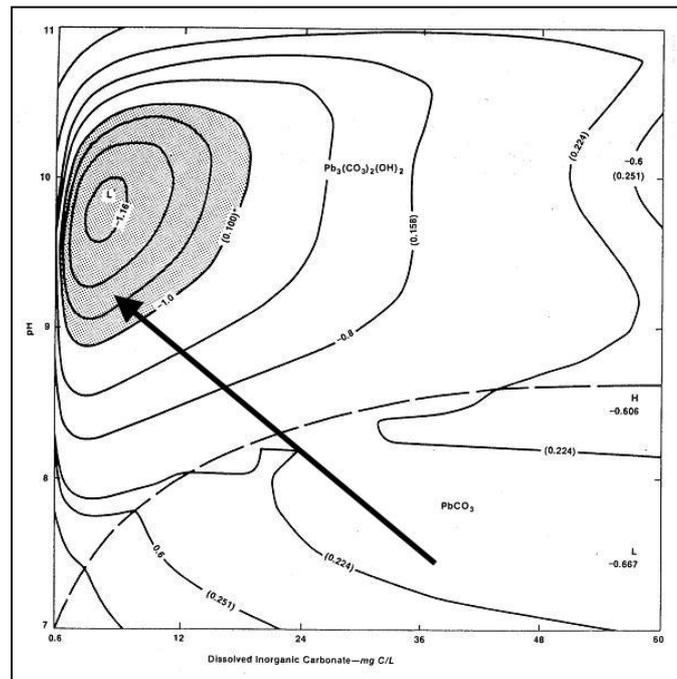
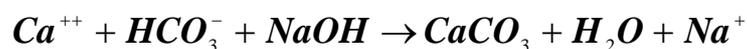


Figure 3: Lead solubility diagram as a function of pH and dissolved inorganic carbonate (equivalent to total carbonate), obtained from computational calculation at equilibrium conditions and 25 °C (reproduced from Schock, 1989).

3.1 Decarbonation processes

The conventional decarbonation process relies on the precipitation of calcium carbonate when raising the pH. This can be achieved in a fluidized bed reactor by adding caustic soda or lime to a suspension of fine heavy sand, which will serve as nucleation centre. For caustic soda, the stoichiometry of the reaction is given by:



For lime, the stoichiometry is the following:



After reduction of calcium and carbonate concentrations, the pH is easily adjusted above 8. For the caustic soda decarbonation, calcium and carbonate content (or alkalinity) will be reduced similarly while for the lime process, the carbonate content will be reduced twice more than calcium (on a molar basis). This implies that for removing carbonate, the lime process might be a better choice, but more sludge will be produced than with the caustic soda process. On the other end, the use of caustic soda is easier on industrial scale since it is a liquid ready to use while the lime must be prepared on site by dissolution into water and will be the source of many technical problems due to scaling. But, it should be remembered that the sodium concentration will increase in treated water when using caustic soda.

The fluidized bed reactor is well suited for large water volume production. For small production units, this process might be too heavy to handle: regular technical problems with pipes and pumping systems need a lot of maintenance. The fluidized bed reactor needs a height of more than 10 m to insure a high flow velocity (80-100 m/h) and an adequate contact time (5-10 min). High flow velocity is needed to fluidise sand into the reactor.

Industrial experience from the Netherlands shows that the plumbosolvency of water can be reduced by 50% after decarbonation with fluidized bed reactors (Figure 4).

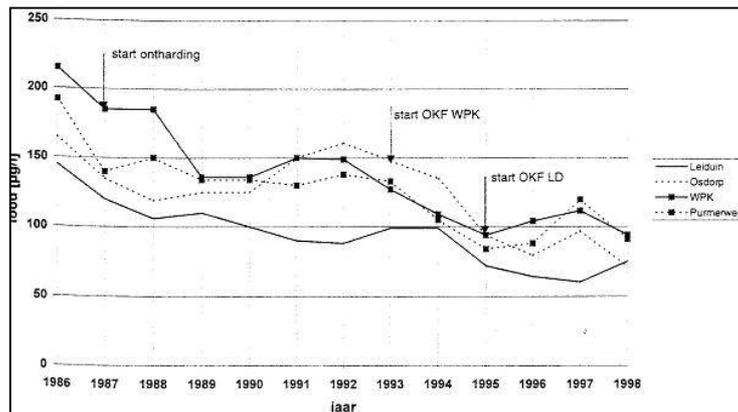
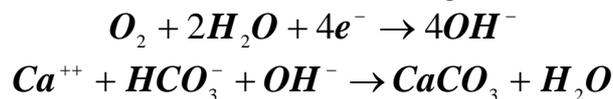


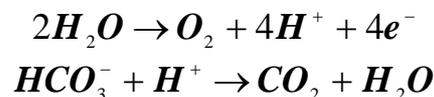
Figure 4: Evolution of the plumbosolvency of decarbonated water distributed in Amsterdam (The Netherlands), since decarbonation was started in 1987, measured from pipe rig tests after 16 hours stagnation (reproduced from Zillig et al., 2000).

An industrial electrochemical version of the precipitation process has been developed (8). By reducing oxygen present in water into hydroxide anion, calcium carbonate will precipitate at the cathode and fall at the bottom of the reactor, according to the following stoichiometry:



The reduction of water will also generate hydrogen gas at the cathode.

At the anode, oxidation of water will occur and due to the overall stoichiometry, treated water will become acidic:



Thus, the total carbonate concentration will decrease by calcium carbonate precipitation and by the formation of carbon dioxide, the latter being removed by air stripping. With this process, pure calcium carbonate is obtained while for the fluidized bed reactor decarbonation, the core of the calcium carbonate particle is a sand particle. Nevertheless, the electro-decarbonation is limited by the size of the reactor and can only be applied for smaller production units (the maximal flow treated with one electro-decarbonation unit is 36 m³/h, with a 30 min contact time).

3.2 Effect on the health

From a health perspective, mainly the calcium removal from the decarbonation process should be evaluated. The impact of total hardness on health has been widely studied, but definitive conclusions cannot be drawn for calcium (13, 26). Several studies have highlighted the contribution of water total hardness for reducing cardiovascular diseases, hard water being more beneficial than soft water. According to the latest WHO reports (9), sufficient evidence is now available to confirm that an increased risk for cardiovascular diseases, and especially for sudden cardiac death, is essentially related to low concentration of magnesium. As water decarbonation does not affect significantly the magnesium concentration, no adverse health effect is to be expected for cardiovascular diseases.

On the other end, lower concentration of calcium may be associated with a higher risk of fracture in children, certain neurodegenerative diseases, pre-term birth and low weight at birth and some types of cancer (13). In fact, consumption of moderately hard water is believed to provide an important incremental percentage of the calcium and magnesium daily intake (5-20%) since modern diet may not be an adequate source of minerals.

Of course, indirect beneficial impact on health can be expected from reduced corrosion of other metals. Moreover, higher concentrations of calcium have an antitoxic activity, preventing the absorption of some toxic elements, such as lead, from the intestine into the blood (13).

These health considerations lead WHO to recommend (unofficial) avoiding calcium hardness below 50 mg CaCO₃/L or 0.5 mmol/L (13).

3.3 Effect on corrosion of other metals

As decarbonation will transform any aggressive water into a calcifying one after pH adjustment, cement corrosion should be strongly reduced for originally aggressive waters. For galvanised steel and copper pipes, increasing pH and decreasing alkalinity have been shown to be beneficial for reducing metal dissolution (7, 23). In fact, similarly to lead, several carbonate and hydrogen carbonate complexes are responsible for the increased copper and zinc dissolution from deposits at lower pH. Industrial water decarbonation in The Netherlands has been successfully used to reduce copper leaching from drinking water distribution pipes to the environment (28).

For iron corrosion, several investigations have highlighted the important role of siderite (FeCO₃) as an intermediate in the formation of strong protective layers (2, 19, 25). According to the siderite model, more resistant deposits are formed when the water has a higher buffer capacity (higher alkalinity or total carbonate) and a pH closer to neutrality, since the deposits contain more Fe²⁺. When the buffer capacity is decreased and the pH closer to 8, the iron deposits were shown to be less resistant, due to the more rapid oxidation of Fe²⁺ to Fe³⁺, and any calcium carbonate precipitation in these conditions would form nonprotective CaCO₃ amorphous deposits. Thus, according to this model, the lower buffer capacity and the increased pH obtained after any decarbonation process would lead to an increased iron corrosion rate. Later, Berghult et al. noticed that since iron corrosion can be decreased by high concentrations of carbon dioxide and copper corrosion is facilitated by the same

parameter, the two materials need very different water qualities if corrosion is to be minimised (2).

Other investigations (11, 16) mentioned that supersaturation of the water with calcium carbonate would efficiently protect iron from corrosion, due to calcium carbonate deposition on the surface of the pipe. Nevertheless, the reduced corrosion from supersaturation of calcium carbonate was mainly reported for soft waters (11, 16).

On the other end, higher concentrations of chloride and/or sulphate are well-known to increase iron corrosion rate. This effect has been expressed quantitatively by a ratio, known as the Larson index (17). Since carbonate concentration will decrease while chloride and sulphate concentrations will stay unchanged after decarbonation, the Larson index will inevitably increase and this may lead to accelerated iron corrosion.

3.4 Effect on disinfection

As the pH is adjusted above 8 at the end of the decarbonation process, the chlorine disinfection will be less efficient. In fact, it is well-known that the active disinfectant is hypochlorous acid (HClO) while its anion (ClO^-) is much less efficient. From the acidity constant of hypochlorous acid, it can be predicted that the efficiency will be 75 % lower at pH 8.5 compared to pH 7.5 (at 20°C, Figure 5). It implies that more chlorine must be dosed to keep the disinfection efficiency at higher pH.

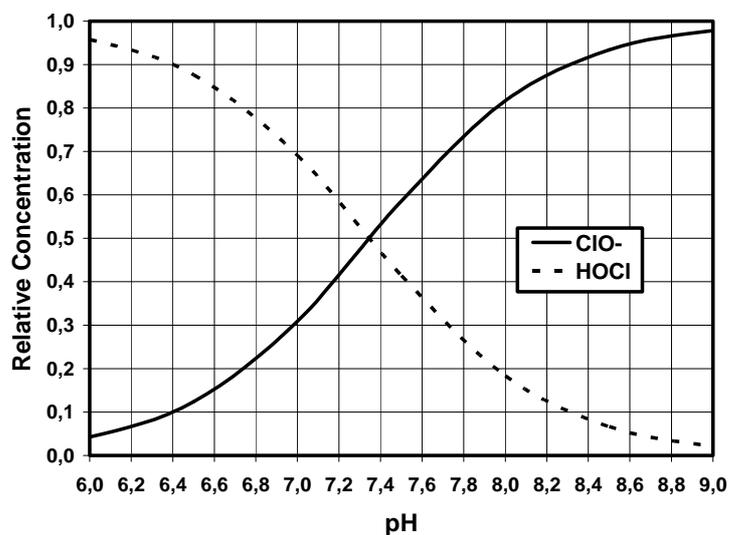


Figure 5: Relative concentrations of hypochlorous acid (HClO) and hypochlorite anion (ClO^-) as a function of pH, when considering the acid dissociation constant at 20 °C for pure water.

For ozone, a pH above 8 is unfavourable for disinfection since both ozone self-decomposition and bromate formation will increase. When ozone is used, the ozonation should be realized at lower pH, thus before water decarbonation. Moreover, carbonate slows down ozone decomposition through radical trapping by hydrogen carbonate anion, thus improving micro-organism disinfection.

Chlorine dioxide and chloramines should be recommended in case of water decarbonation as their disinfection efficiency does not vary with the pH. But these disinfectants suffer from other drawbacks, which have limited their use in Europe (27).

3.5 Effect on bacterial growth

As bacterial growth in water is essentially resulting from biofilm development on the inner surface of pipes, controlling biofilm will reduce bacteria in water. If we consider that biofilm will mainly develop in crevices and holes (cracks, deposits, tubercles), reduced corrosion of

either cement or metals should disfavour biofilm growth and thus reviviscence of bacteria in drinking water, and reduce also microbial influenced corrosion. This was shown for iron pipes, where reducing corrosion rate and tuberculation improved the disinfection efficiency of chlorine (14). Unfortunately, no experimental work has tackled the impact of decarbonation on biofilm growth.

3.6 Environmental impact

The waste from the decarbonation process has a weak environmental impact since the calcium carbonate produced is a harmless compound. The produced sludge can be used in cement works or as nutrient additive in the chicken industry. With the pure calcium carbonate generated from the electrochemical process, other applications can be considered such as fillers for paper production, possibly creating added value. Anyway, as sludge will be generated in large amount whatever the decarbonation process, disposal remains the main problem for all decarbonation processes.

4. Orthophosphate treatment

The orthophosphate treatment relies simply on the addition of orthophosphate (either as phosphoric acid or as an orthophosphate, hydrogen phosphate or dihydrogen phosphate inorganic salt) to drinking water. Orthophosphate anion will decrease dissolved lead concentration because lead phosphate deposits have a lower solubility than lead carbonate deposits (21, 24). Under drinking water conditions, evidence points out that the most likely phosphate deposit should be hydroxypyromorphite ($\text{Pb}_5(\text{PO}_4)_3\text{OH}$), but other deposits have been considered (21, 24).

The orthophosphate treatment for lowering plumbosolvency is quite efficient for soft waters (total alkalinity < 100 mg CaCO_3/L), even at pH as low as 6.5, with a dose of 0.5 mg P/L (24). This can be illustrated by Figure 6, where the variation of lead solubility in the presence of orthophosphate, at equilibrium, is represented as a function of pH and dissolved inorganic carbonate (or DIC, which is similar to total carbonate and related to alkalinity). From this figure, the following comments can be made:

- lead solubility increases when DIC increases due to the formation of lead carbonate soluble complexes
- lead solubility is sensitive to DIC and rather insensitive to pH from 7 to 8, implying that pH should not be adjusted in this range when injecting orthophosphate
- dissolution of lead deposit is minimized at about pH 7.5, whatever the dissolved inorganic carbon

It is often stated that orthophosphate treatment is only appropriate for soft waters. While orthophosphate treatment is surely less efficient for hard waters than for soft waters, it has nevertheless been used successfully for lowering plumbosolvency of hard waters (or high total alkalinity waters). Due to its simplicity, the orthophosphate treatment has even been recommended as the favoured method for lowering plumbosolvency of hard waters in the UK (5, 21). But, in hard waters, some concern can be raised about precipitation of a calcium orthophosphate product into the water phase, especially when the water is warmed (6). Experience in natural waters has shown that a large degree of supersaturation of calcium phosphate solid can occur without precipitation up to concentrations of orthophosphate greater than 2 mg P/L (21). For calcium hardness higher than 300 mg CaCO₃/L, it was concluded from field trials that a blend of orthophosphate and polyphosphate would be recommended to avoid calcium phosphate precipitation, this blend acting as both a corrosion inhibitor and a calcium anti-scaling agent (6).

As the phosphate deposits form slowly on the inner surface of lead pipes, especially for old pipes, the orthophosphate treatment should start with a higher dose (1 mg P/L) than the optimal one (generally 0.5 mg P/L), to speed up the phosphate deposit formation. Nevertheless, some reports indicate that, even then, a year or more might be needed to reach the full efficiency of the orthophosphate treatment (5), while other state that full performance can be achieved after a few months (3). After stable conditions have been obtained, the orthophosphate dose can be decreased to 0.5 mg P/L without adverse effects on lead solubility (3), provided that an adequate orthophosphate concentration reaches the extremities of the distribution network. Then, lead solubility at the consumer's tap can be reduced from 65% to 90% of its original concentration when applied in a distribution network (3). It should also be taken into account that other metallic pipes will adsorb phosphate at their inner surface. Polyphosphates have often been reported to lower plumbosolvency of drinking water. However, some studies showed unambiguously that polyphosphates were often not only ineffective in reducing lead levels, but they could actually increase dissolved lead concentrations by complexation and solubilization of protective deposits (12, 15, 24). Anyway, some success in reducing plumbosolvency with polyphosphates has been reported (6) but could be ascribed to the presence of orthophosphate, either present in the polyphosphate blend or from the natural hydrolysis of polyphosphate in water. Thus, conclusive experimental and field evidence does not support the use of polyphosphates for lowering plumbosolvency (24).

4.1 Effect on the health

The addition of phosphorus should not lead to adverse effects for human health. On the other end, indirect beneficial effects on health can be expected when considering the reduced corrosion of other metallic pipes in the network.

4.2 Effect on corrosion of other metals

Beneficial effects for iron and zinc corrosion have been reported (6, 24, 28), while less effect is to be expected for copper. Nevertheless, the reduction of copper solubility has been reported to be as high as 90% for trials in a distribution network (3). Similarly to lead, phosphate deposits with lower solubility than the usual deposits will be formed and decrease the concentration of dissolved metals.

4.3 Effect on disinfection

No adverse effect on disinfection efficiency is to be expected for chlorine because the pH should remain unchanged. For chlorine dioxide and chloramines, disinfection efficiency

should also be unaffected. On the contrary, phosphate could improve the disinfectant stability, thus disinfection efficiency, by reducing the concentration of dissolved metallic cations and stabilizing corrosion deposits, both known for contributing to the disinfectant demand of drinking water. But, improvement of disinfectant stability under phosphate treatment has not yet been reported.

4.4 Effect on bacterial growth

Some studies have reported that phosphorus can be a limiting factor for bacterial growth in water. Thus, adding phosphorus to waters where it is a limiting factor can promote bacterial growth. But, the phosphorus has been shown to be a limiting factor for bacterial growth only at concentrations below 10 µg P/L (18, 20). Above this concentration, little if any effect on bacterial growth was observed. As for most drinking waters through Europe this limiting phosphorus level is already reached in raw water and will be preserved after treatment, adding orthophosphate to drinking water should not be expected to promote bacterial growth, excepted for some very pristine European waters.

When looking at the biofilm growth, some contradictory reports have been published. According to Gaujous et al. (10), increased biofilm formation (about 1 Log unit) was observed both with laboratory experiments and within a drinking water distribution network, during the summer. But, for iron corroded pipes, Appenzeller et al. (1) showed that phosphate did not promote biofilm growth, and could even lead to a drastic drop of bacterial production through release of iron oxide deposits, which promote bacterial growth. In fact, most studies show that the bacteriological quality of water treated with orthophosphate will not change (3).

4.5 Environmental impact

The use of orthophosphate in drinking water network will inevitably bring phosphorus to waste waters, which will be discharged into surface water or treated in waste water treatment plant. Concerning waste water treatment plants, as phosphorus concentrations are in the range of 10 mg P/L, about 1 mg P/L more (worst case) would represent a 10% phosphorus concentration increase (6) and thus should not be a problem, especially when a tertiary treatment is running. For surface water, direct discharge of 1 mg P/L would certainly add some pressure to the fight against eutrophication. As most European cities should have waste water treatment plants in the near future, the use of orthophosphate might no more be considered as an environmental problem.

5. Water quality objectives

In the Netherlands, the legal objective is a calcium hardness between 1 mmol/L and 2.5 mmol/L (corresponding to a calcium hardness of 100 mg CaCO₃/L and 250 mg CaCO₃/L). The water industry developed fluidized bed decarbonation processes to reach a calcium hardness of 1.5 mmol/L. Such a decarbonation strategy has proven useful for limiting both lead and copper dissolution and some water companies have even considered lowering the calcium hardness to 1 mmol/L (28). While carbonate concentration should be considered for controlling lead solubility, setting a goal for calcium hardness will of course reduce the carbonate content of the water.

In France, the first objective is to reach equilibrium pH of the distributed water. Next, to satisfy the 25 µg/L lead guideline, both decarbonation and phosphate treatments are considered for a total alkalinity higher than 80 mg CaCO₃/L. Both decarbonation and phosphate treatment must be applied when the total alkalinity is higher than 300 mg CaCO₃/L. For total alkalinities in the range of 80 mg CaCO₃/L and 300 mg CaCO₃/L, phosphate treatment must be considered while decarbonation is recommended but not mandatory. It must be

mentioned that phosphate treatment will only be applied if the water equilibrium pH is lower than 7.5. No official information is given about the alkalinity target that should be reached after decarbonation, but studies performed by CRECEP showed that a total alkalinity of 80 mg CaCO₃/L, together with a calcium hardness of 80 mg CaCO₃/L and a pH between 8 and 8.5 would reduce significantly corrosion of most metals found in distribution systems (17). In Sweden, the water quality goal, implemented under the name 'hardness increase', was to reach a total alkalinity of 1 mmol/L (50 mg CaCO₃/L) with a calcium hardness between 0.5 mmol/L and 1.5 mmol/L. This strategy was mainly developed to reduce copper corrosion.

6. Conclusions

Both decarbonation and phosphate treatment will reduce the water plumbosolvency. Thus, these industrial treatments could bring a solution for both public distribution and in-house networks. Today, it is recognised that both treatments can reduce the lead concentration below 25 µg/L, with the phosphate treatment reducing more efficiently the plumbosolvency. Even if phosphate treatment was shown to reduce the lead concentration below 10 µg/L, satisfying the 10 µg/L guideline is not considered as achievable on a long term with any of these treatments.

When compared to each other, the decarbonation treatment could present two drawbacks: first, chlorine disinfection efficiency will be reduced, and next, some detrimental impact on iron corrosion might be expected. Concerning iron pipes, no definitive conclusions can be drawn whether decarbonation would increase or decrease corrosion. As a matter of fact, iron corrosion studies should be first conducted before implementing any decarbonation process. The phosphate treatment may suffer some drawback when the water is warmed: precipitation of calcium salts might be an important point to consider when applying phosphate, even if the available reports are optimistic on this topic.

For both treatments, no detrimental health effect is to be expected, and microbial growth may be reduced in both cases, excepted if iron corrosion would increase when applying decarbonation. Most European water quality objectives satisfy the WHO's unofficial recommendation for minimum calcium concentration (50 mg CaCO₃/L). But, the optimal carbonate content needed to minimize plumbosolvency (30 mg CaCO₃/L) should not constitute a quality objective as the calcium concentration would then certainly lie under the WHO's recommendation.

From an industrial point of view, adding phosphate to water is by far more easily and more quickly realized than decarbonation, and should certainly be the less expensive option. But, when considering psychology, the phosphate treatment will appear as less environment-friendly due to bad renown of phosphorus.

7. References

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8. Conversion table

	meq/L	mmol/L	mg/L	mg CaCO ₃ /L	°F
HCO ₃ ⁻	1	1	61	50	5
Ca ²⁺	1	0.5	20	50	5