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Influence of tap water stagnation on copper and iron concentration as a function of pH

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Abstract

The water at the consumer's tap may have a quality far removed from that obtained from the distribution service due to contamination by metallic trace elements. This is why this work is interested in studying the influence of the stagnation of tap water on the concentration of copper and iron. Thus, the levels of Cu and Fe in water with a composition similar to that of the public water supply of Niamey (Niger) in stagnation in four types of pipe were monitored. These are cast iron, galvanized steel, polyvinyl chloride (PVC) and polypropylene (PPR) pipes. The analyzes of the water samples were carried out by flame atomic absorption spectrometry. It appears from this study that all the pipes made up of these pipes and accessories release Cu and Fe into the water under the conditions of the experiment. Metal pipes, in particular the cast iron pipe, release these two metals much more than plastic pipes, in particular PVC. Thus, the new cast iron pipe releases exceptional maximum quantities of iron of the order of 1 to 1800 times the guideline value of 0.3 mg.L-1 after 4 hours to 90 days of water stagnation.

Keywords: Tap water; Trace Metals; Copper; Iron; Niamey

1. Introduction

Copper and iron are usually present in drinking water due to their leaching by the elements of the distribution network or plumbing [1]. These metals are frequently used in service lines and fittings, making their presence in drinking water more likely [2-5]. Since copper and iron concentrations at the consumer's tap can be significantly higher than those observed at the processing plant or in the distribution system, strategies to reduce copper exposure should focus on corrosion control in distribution systems and plumbing systems [6-8]. Monitoring for copper and iron should primarily be done where new plumbing exists or could exist and where water quality conditions are potentially corrosive [9]. [10] Have shown that the phenomenon of dissolution of water pipe leather is complex. Thus, the quality of the water, the oxidizing agents, the age of the material and the composition of the alloys have a significant impact on the evolution of the concentration of copper. This article studies the evolution of copper concentration and those of iron associated with water stagnation in different brand new pipes and accessories most commonly used in domestic installations at three pH values (pH 5.11; 7.04 and 9.11). These are polyvinyl chloride pipe, galvanized steel pipe, polypropylene pipe and cast iron pipe.

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2. Material and methods

2.1. Reagents and solutions

All reagents used are of analytical grade. The solutions are prepared in distilled water. The solutions used to power the pipes are prepared from the following salts: CaCO₃, KNO₃, NaCl and MgSO₄.7H₂O. The calcium carbonate (CaCO₃, 98%) comes from Micheltronche. Potassium nitrate (KNO₃ 99.5-100%) comes from Prolabo/France. Magnesium sulfate heptahydrate (MgSO₄.7H₂O \ge 99.5%) is from VWR Chemicals/France. Sodium Chloride (NaCl) is from Merck/Indonesia. Nitric acid HNO₃ 65% from VWR/France is used to acidify standard solutions and samples. The hydrochloric acid solution (prepared from 35% HCl, VWR/France) 0.1 M was used to bring the pH back to 7.04 and to 5.11 which was initially at 9.11. The 1000 mg.L⁻¹ standard solutions for copper, iron, zinc and lead AAS come from VWR/France.

The initial solution of pH 9.11 is prepared from the above salts. Then, the solutions with an average pH of 7.04 and that with a pH of 5.11 are obtained from the initial solution by lowering its pH using a dilute solution of hydrochloric acid. The concentrations of the chemical parameters of the pH 7.11 solution represent on average the concentrations of these same parameters (except Na+ and K+) in the public water supply of Niamey at the time of work.

After their preparation, the solutions are subjected to analyzes by atomic absorption with flame. Following his tests, no trace of Cu and Fe was detected in these solutions.

2.2. Experimental procedure

The assembly consists of three systems, made of four pipes each. The four pipes are (1) polyvinyl chloride (PVC), (2) galvanized steel, (3) Polypropylene (PPR) and (4) cast iron. The PVC pipe has a PVC elbow and tap. The PPR pipe has a elbow and a PPR valve. For the galvanized steel pipe and the cast iron pipe, each is equipped with a galvanized steel elbow and a brass tap.

The four pipes of system 1, 2 and 3 are supplied with the solution of pH 5.11; 7.04 and 9.11 respectively.

The physicochemical parameters of the water supplying these pipes, similar to those of tap water, are chosen to reproduce in the laboratory the similar facts inside the individual connection pipes. The variation in the quantity of copper and iron is studied according to the type of pipe, the pH, the variation in dissolved oxygen and the residence time of the water in the pipes.

The determination of Cu and Fe contents in the laboratory is carried out in the water contained in each pipe after the following stagnation times: 4 hours, 8 hours, 12 hours, 16 hours, 20 hours and 24 hours, 2 days, 3 days, 4 days, 5 days, 6 days, 7 days, 2 weeks, 3 weeks, 4 weeks, 60 days and 90 days. Seventeen (17) samples were taken from each of the 12 pipes.

2.3. Sampling

To monitor the concentration of Cu and Fe in the 4 types of pipes in the laboratory as a function of pH, new and sterile 60 mL polyethylene bottles were used to take the samples. Two hundred and four (204) samples were taken over the entire duration of the experiment. The water samples taken were analyzed by flame atomic absorption spectrometry as described by [11].

3. Results

3.1. Copper content

The presence of copper in tap water is mainly due to the leaching of components of distribution networks and plumbing systems that contain copper. Copper is widely used in drinking water applications, including residential plumbing pipes, fittings, gate valves and elbows [9].

3.1.1. Variation in copper content at pH 5

The study of the evolution of the concentration of copper in the water having remained in the pipes (used in domestic installations) was carried out in the laboratory under the finished conditions. Figure 1 illustrates the variation, as a function of time, at pH 5.11 of the concentration of copper in the water at the outlet of polyvinyl chloride (PVC), galvanized steel (TG), polypropylene (PPR) and cast iron. The initial point, stagnation time 0 hours, shows no trace of

copper according to the method of determination by atomic absorption with flame. This point represents the concentration of copper in the raw water that feeds the various pipes. Water stagnation after 4 hours showed an increase in copper concentration of 5.517 mg.L⁻¹ for the cast iron pipe, 4.155 mg.L⁻¹ for the in PPR, 0.656 mg.L⁻¹ for the galvanized steel pipe. The concentration of copper, very low, in the water from the PVC pipe represents only 0.001 mg.L⁻¹ during the same period and reaches the maximum of 0.023 mg.L⁻¹ after one day then goes down up to 0.000 mg.L⁻¹ after 90 days of water stagnation. These results show that the PVC pipe, elbow and gate valve contain very little or no copper following their exposure to acidic water of pH 5.11 for an extended period.

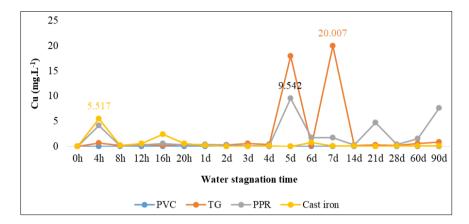


Figure 1 Evolution of copper concentration as a function of stagnation time of fresh water at pH 5.11 in different pipes and plumbing fixtures

The copper concentration decreases in the water from the TG after 8 hours of stagnation then continues to fluctuate to reach a high value after 5 days and a maximum value of 20.007 mg.L⁻¹ after 6 days before falling towards very low values up to 90 days. For the PPR pipe, the maximum concentration (9.542 mg.L⁻¹) is reached on the fifth day of water stagnation.

3.1.2. Variation in copper content at pH 7

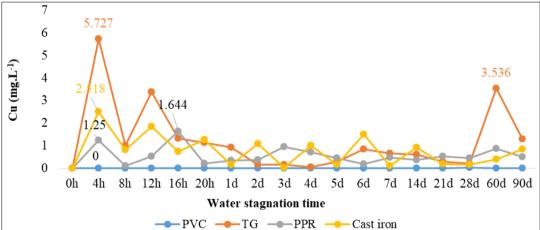


Figure 2 Evolution of the concentration of copper as a function of the stagnation time of fresh water at pH 7.04 in various pipes and plumbing accessories

The evolution of copper concentration as a function of water stagnation time was studied at pH 7.04. Figure 2 shows widely varying concentrations of copper in different pipes. The concentration of copper increases after 4 hours of water stagnation in three pipes. The copper concentrations are 5.727 mg.L⁻¹, 2.518 mg.L⁻¹, 1.250 mg.L⁻¹ and 0.000 mg.L⁻¹ respectively in the TG, cast iron, and PPR and PVC pipes. These concentrations fluctuate and gradually decrease during the stay of the water until the 90th day, but a rebound in the concentration of Cu is observed on the 60th day of stagnation of the water in the channel made by the galvanized steel pipe (TG). The formation of copper oxide and hydroxycarbonate could explain the gradual decrease in copper levels over a long period [12]. The maximum copper concentrations are reached after 4 hours in the TG pipe (5.727 mg.L⁻¹) and in the cast iron pipe (2.518 mg.L⁻¹) and after

16 hours in the PPR pipe. The concentration of Cu in the PVC pipe is very low; the maximum of 0.007 mg.L⁻¹ is reached after 8 hours of water stagnation.

3.1.3. Variation in copper content at pH 9

Figure 3 shows the evolution of copper concentration as a function of water stagnation time at pH 9.11. After 4 hours of stagnation time, the concentrations of copper in stagnant water in PVC, TG, PPR and cast iron pipes are respectively 0.005 mg.L⁻¹; 1.006 mg.L⁻¹; 0.519 mg.L⁻¹ and 1.082 mg.L⁻¹. These very low concentrations fluctuate between 0 and 0.005 mg.L⁻¹ in the PVC pipe, between 0.052 and 1.442 mg.L⁻¹ in the TG pipe, between 0.038 and 1.786 mg.L⁻¹ in the in PPR and between 0.062 and 1.082 mg.L⁻¹ in the cast iron pipe from midnight to the 28th day of stay in water with a pH of 9.11. On the 60th day of water stagnation, an increase in the concentration of copper is observed in the water contained in the galvanized steel pipe (96.001 mg.L⁻¹) and the PPR pipe (46.406 mg.L⁻¹) before dropping after 90 days. These concentrations constitute the maximum concentrations reached in these pipes over the entire duration of the experiment.

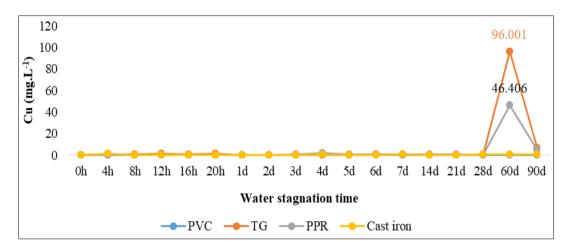
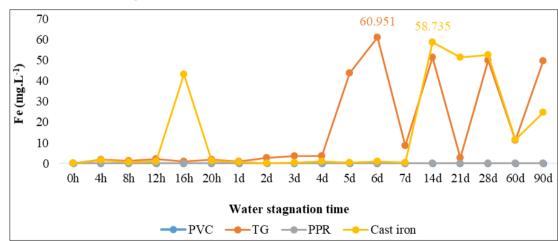
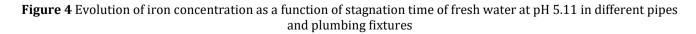


Figure 3 Evolution of copper concentration as a function of stagnation time of fresh water at pH 9.11 in various pipes and plumbing fixtures

3.2. Iron content



3.2.1. Variation in iron content at pH 5



The evolution of iron concentration as a function of water stagnation time was studied at pH 5.11 in PVC, TG, PPR and cast iron pipes (Figure 4). In raw water, the concentration of iron is 0 mg.L⁻¹. The contact of this water with these pipes after 4 hours of time leads to an increase of 0.059 mg.L⁻¹, 1.748 mg.L⁻¹, 0.13 mg.L⁻¹ and 0.679 mg.L⁻¹ of iron in PVC, TG, PPR and Cast Iron pipes respectively. These iron concentrations vary between 0 mg.L⁻¹ and 0.089 mg.L⁻¹ and from 0 to

0.151 mg.L⁻¹ throughout the stagnation period in PVC and PPR pipes; the maximum is reached after 1 d and 5 d respectively. For the water having stayed in the galvanized steel pipe, the concentration of iron increases slightly until the 4th day and reaches the maximum of 60.951 mg.L⁻¹ at the end of the 6th day then decreases with fluctuations until the 90th day. For water that has stagnated in the cast iron pipe, the concentration of iron also fluctuates slightly until 12 p.m., reaches a value of 43.037 mg.L⁻¹ after 4 p.m., returns to low values from 8 p.m. and reaches the maximum of 58.735 mg.L⁻¹ at the end of the 90th day.

3.2.2. Variation in iron content at pH 7

Figure 5 shows the evolution of iron concentration as a function of water stagnation time at pH 7.04. The concentration of iron released in the water from the PVC pipe and the PPR pipe is very low during the period of 0 h - 90 days of stagnation. It varies respectively in these pipes from 0 to 0.241 mg.L⁻¹ and from 0 to 0.183 mg.L⁻¹. The iron concentration of 3.463 mg.L⁻¹ is reached after 4 hours of residence of water at pH 7.04 in the TG. The iron concentration fluctuates below this value during the 90 days of the experiment, but a peak is observed on the 6th day when the iron concentration reaches the maximum value of 47.625 mg.L⁻¹. The very low concentration of iron at the start of water stagnation in the cast iron pipe for up to 5 days, reaches a high value of 43.409 mg.L⁻¹ on the 6th day, then fluctuates around similar values until the 60th day. It reaches the maximum of 189.878 mg.L⁻¹ after 90 days.

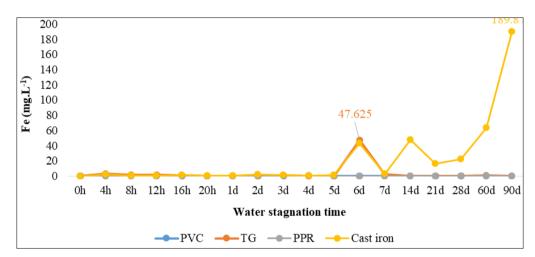


Figure 5 Evolution of iron concentration as a function of stagnation time of fresh water at pH 7.04 in different pipes and plumbing fixtures

3.2.3. Variation in iron content at pH 9

The follow-up of the evolution of the concentration of iron in the water staying in different pipes was also carried out at pH 9.11. Figure 6 illustrates the results obtained.

The concentration of iron in the PVC pipe is very low throughout the duration of the study, the maximum concentration of 0.419 mg.L⁻¹ is reached after 1 day. The concentration of iron in the water in the PPR pipe was also low throughout the duration of the study, the maximum concentration of 2.242 mg.L⁻¹ being reached after 60 days. In the standing water in the galvanized steel pipe, the concentration of iron fluctuates between 0 and 1.805 mg.L⁻¹ from 0 to 28 days. A maximum value of 55.639 mg.L⁻¹ is reached after 60 days of stagnation then drops to 28.912 mg.L⁻¹ on the 90th day. For the cast iron pipe, the monitoring of variation in iron concentration showed a fluctuation of 0 to 2.473 mg.L⁻¹ between 0 -12 h then a rise in concentration of 63.681 mg.L⁻¹ (at 4 p.m.) and 79.225 mg.L⁻¹ (at 8 p.m.) before falling to values of the order of 2 mg.L⁻¹ between 1-3 days. Subsequently, the concentration of iron varies between 1.651 and 45.598 mg.L⁻¹ from 4 to 14th day of stagnation before rising gradually to reach an extreme value of 554.011 mg.L⁻¹ after 90 days.

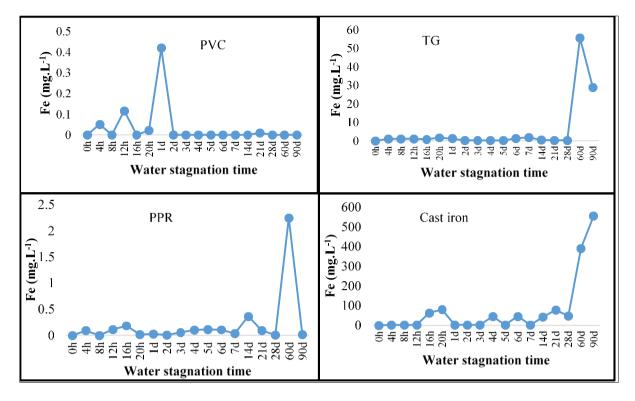


Figure 6 Evolution of iron concentration as a function of stagnation time of fresh water at pH 9.11 in different pipes and plumbing fixtures

4. Discussion

The results obtained show that the leaching level of copper in a pipeline consisting of the pipe, the elbow and the PPR gate valve when exposed to acidic water of pH 5.11. Indeed, the PPR pipe is a tube formed essentially of polypropylene, the copper leaching could come mainly from the gate valve because it has a metal part inside it. Water of similar composition could then lead to the dissolution of the cooker of the order of 4.155 mg.L⁻¹ after 4 hours of contact time and a maximum value of the order of 9.542 mg.L⁻¹ after 5 days of stagnation in a newly installed identical pipe. The concentration of copper in the cast iron pipe reaches the maximum of 5.517 mg.L⁻¹ after a stagnation of 4 hours then decreases and remains very low, apart from a rise to 2.368 mg.L⁻¹ on the 6th day, until the 90th day of stagnation.

The concentration of copper in the water after a first contact for 4 h (and also at 16 h) in different pipes then ranks in this order (Cu) Cast Iron > (Cu) PPR > (Cu) TG > (Cu) PVC. But, the trend is no longer the same on the 5th and 7th day of stagnation where it looks like this: (Cu) TG > (Cu) PPR > (Cu) Cast Iron > (Cu) PVC. From 21st to 90th day of stagnation, in other words for prolonged contact, Cu in PPR pipeline water has the highest concentration ahead of Cu (TG), Cu (Cast iron) and Cu (PVC) in this order. These trends show the degree of Cu leaching and the phenomena that facilitate its dissolution. These phenomena involve prolonged contact with acidic water of pH 5.11, the interaction between copper and associated materials.

The copper concentrations in the water having stagnated in the various pipes and accessories at pH 7.04, apart from the PVC pipe, over the entire duration of the experiment are generally average. Thus, the maximum concentrations of Cu reached are lower compared to the case where the pH of the water is 5.11. This phenomenon would be linked to the neutrality of the pH because the water in contact with the pipe materials being less aggressive than acidic water with a pH close to 5.

In practice, where the pH of drinking water is around 7, similar amounts of copper can be released by new installation of residential plumbing or after an extension of the drinking water distribution system where pipes and fittings (elbows, counters, faucet, etc.) are new. In this case, the guide value of 2 mg.L⁻¹ of Cu can then be exceeded. In addition, a decrease in the pH of the water can lead to very high concentrations of copper in the water coming out of the tap.

These results illustrate the intrinsic characteristics of water at pH 9.11 that has stagnated in these different pipes. Can then be cited, the leaching of very small quantities of copper over a long period of water stagnation. These results could

be explained by the formation of protective layers such as malachite, $Cu_2 (OH)_2CO_3$), copper (I) oxide (cuprite, Cu_2O) or copper (II) oxide (tenorite, CuO) on the walls of the pipes. This reduces, and in extreme cases prevents, water contact with the pipe material. The appearance of very high amounts of copper on the 60th day of water stagnation in galvanized steel (TG) and polypropylene (PPR) pipes would be linked to the detachment or displacement of these protective layers. This exposes the pipes to water and causes the copper to dissolve. This situation is sometimes associated, in practice, with the phenomena of localized corrosion which occurs when the pH is higher than 8 [13]. In addition, the solubility and physical properties of the encrusting solids inside the pipe containing the copper such as copper hydroxide, copper carbonate and copper oxide are critical to the release of copper into the water. They are also a means of predicting the concentration of copper in water [9].

The results obtained also show that the TG and cast iron pipes are those which release considerable quantities of iron because they are essentially made up of iron. The cast iron pipe released a large amount of iron (43.037 mg.L^{-1}) after 16 h while it would take 5 days of stagnation for the TG to release the similar amount (43.707 mg.L^{-1}).

The results show that the concentration of iron in water of pH 5.11 and in water of pH 7.04 in stagnation in the TG reaches the maximum value after 6 days. However, the maximum iron concentration at pH 5.11 of 60.951 mg.L⁻¹ is greater than 47.625 mg.L⁻¹ at pH 7.11. This is therefore due to the power of acidic water to dissolve the substances with which it comes into contact. For Cast Iron pipe, at pH 5.11, iron is released in large quantities during the early periods (at the 16th hour and the range of 7 to 60th day) of stagnation while at pH 7.04 the leaching of large amounts begins on the 6th day so that, subsequently, higher amounts of Fe are released into the water after 60 days.

5. Conclusion

The evolution of copper and iron content as a function of water stagnation time in polyvinyl chloride, galvanized steel, polypropylene and cast-iron pipes was studied in the laboratory at pH 5.11, 7, 04 and 9.11. It appears from this study that all the pipes made up of these pipes and accessories release Cu and Fe into the water under the defined conditions. Metal pipes, in particular the cast iron pipe, release these two metals much more in front of plastic pipes, in particular PVC. Thus, the new cast iron pipe releases exceptional maximum quantities of iron of the order of 1 to 1800 times the WHO guideline value of 0.3 mg. L⁻¹ after 4 hours to 90 days of iron the water stagnation.

Compliance with ethical standards

Acknowledgments

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Disclosure of conflict of interest

The authors stated that they have no competing interests.

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