# Influence of stagnation and pH of tap water on the concentration of metallic trace elements: case of lead and zinc

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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 focused on the study of the influence of the stagnation of tap water on the concentration of lead and zinc. Thus, the levels of lead and zinc 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 and accessories used release Pb and Zn into the water under the conditions of the experiment. Thus, the cast iron pipe releases exceptional maximum quantities of zinc of the order of 71 to 5000 times the guide value after 4 hours to 90 days of water stagnation. The PPR pipe equipped with a brand new PPR elbow and valve releases an exceptional maximum quantity of lead of the order of 100 to 9400 times the guide value after 4 hours to 60 days of stagnation of the lead. water. Dissolved oxygen evolves in a similar way in plastic pipes and in metal pipes.

Keywords: Tap water, metallic trace elements, lead, zinc, Niamey

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### I. Introduction

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The presence of metallic trace elements such as zinc and particularly lead in tap water is mainly attributable to the dissolution of the elements of distribution networks and domestic plumbing systems that contain these metals such as pipes, fittings, welds or service connections to homes [1]. Zinc is mainly used for protective coatings of metals against corrosion (electroplating, metallization, immersion treatment). It is also used in the composition of various alloys (brass, bronze, light alloys) [2].

The dissolution of zinc and lead can be caused by several factors, including the type of material used, the age of the pipes and fittings, the residence time of water in the pipes and the quality of the water such as pH and alkalinity [3]. Lead service lines have been shown to remain a high source of lead [4-6]. They can represent 50 to 75% of the total lead content at the tap after a prolonged period of stagnation [7]. Galvanized pipes release zinc (from the galvanized layer) and can also release cadmium and lead. Corrosion can be a particular problem where galvanized steel or iron piping comes into contact with dissimilar materials such as brass used in valves. In addition, electrolytic corrosion can occur when galvanized steel or iron pipes or fittings come into contact with copper pipes or brass fittings [8].

Many studies have identified several factors that can cause lead and zinc to be released in high concentrations to water long after the installation of lead-containing brass service lines, solders and fittings, including the characteristics water quality including temperature, pH, alkalinity, chloride content), stagnation time, flow rate, lead content and area of lead and brass fittings [9-13]. This work thus analyzes several factors that can lead to the dissolution of lead and zinc in tap water. These include the nature of the pipes, the duration of stagnation, the pH.

# II. Materials And Methods

# 2.1. Reagents and solutions

All the reagents used are of analytical quality and the solutions are prepared in distilled water. The solutions used to power the pipes are prepared from the following salts: CaCO<sub>3</sub>, KNO<sub>3</sub>, NaCl and MgSO<sub>4</sub>.7H<sub>2</sub>O. The calcium carbonate (CaCO<sub>3</sub>, 98%) comes from Micheltronche. Potassium nitrate (KNO<sub>3</sub> 99.5-100%) comes from Prolabo/France. Magnesium sulfate heptahydrate (MgSO<sub>4</sub>.7H<sub>2</sub>O  $\geq$  99.5%) is from VWR Chemicals/France. Sodium Chloride (NaCl) is from Merck/Indonesia. Nitric acid HNO<sub>3</sub> 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<sup>-1</sup> standard solutions of copper, iron, zinc and lead 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 in the public water supply of Niamey at the time of work.

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

# 2.2. Experimental Procedure

The assembly consists of three systems, consisting 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. This is what excludes the use of distilled water, because it is demineralized and has a more aggressive effect than tap water. On the other hand, tap water was not used in this experiment in order to control or easily understand the phenomenon of dissolution of metallic trace elements in the water having stayed in these types of pipe. Because tap water is much more complex than synthetic water.

# 2.3. Sampling and analysis

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. According to the experimental procedure, two hundred and four (204) samples were taken over the entire duration of the experiment and analyzed by flame atomic absorption spectrometry as described by [14]. Dissolved oxygen was measured at the sampling site. The concentration of Cu and Fe is determined in the laboratory 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 (1 day), 2 days, 3 days, 4 days, 5 days, 6 days, 7 days (1 week), 2 weeks, 3 weeks, 4 weeks (28 days), 60 days and 90 days. Seventeen (17) samples were taken from each pipe.

# 3.1. Lead content

# III. Results And Discussion

# 3.1.1. Variation in lead content at pH 5

Figure 1 shows a rise in lead concentration after the first 4 hours of stagnant water at pH 5.11 in all pipes. The maximum concentration of  $6.522 \text{ mg.L}^{-1}$  is reached in the case of the cast iron pipe. This concentration gradually decreases during the stay of the water in this pipe. It becomes very low (0.202 mg.L<sup>-1</sup>) after two days of water stagnation and fluctuates around this value apart from a rebound on the 6th day. For the galvanized steel pipe, the concentration of Pb is 2.430 mg.L<sup>-1</sup> after 4 hours of water stagnation at pH 5.11. The lead concentration fluctuates around this value then increases slightly to reach a peak of 2.935 on the 7th day of water stagnation before dropping to 0.268 mg.L<sup>-1</sup> at the end of the 14th day and fluctuates around this value until on the 90th day stagnation. The stagnant water in the PPR pipeline has a Pb content of 0.795 mg.L<sup>-1</sup> after 4 hours of stagnant water with a pH of 5.11. This concentration decreases slightly to reach a maximum value of 2.685 mg.L<sup>-1</sup> after 90 days. The stagnant water in the PVC pipe contains a Pb concentration of 0.140 mg.L<sup>-1</sup> reached after 4 hours of stagnation. It gradually increases to reach the maximum of 0.331 mg.L<sup>-1</sup> after 14 days of stagnation.



Figure 1: Evolution of lead concentration as a function of stagnation time of fresh water at pH 5.11 in various pipes and plumbing accessories.

Thus, the exposure of these different pipes to fresh water of pH 5.11 showed a leaching of the quantity of lead in this order Pb (cast iron) > Pb (TG) > Pb (PPR) > Pb (PVC) in the after 4 hours with these plumbing materials.

# 3.1.2. Variation in lead content at pH 7

The evolution of the concentration of lead in water of pH 7 in stagnation in different pipes shows a sudden increase in the concentration of lead of 6.168 mg.L<sup>-1</sup>, 5.881 mg.L<sup>-1</sup> and 4.351 mg.L<sup>-1</sup> after 4 hours in galvanized steel, cast iron and polypropylene pipes respectively (Figure 2). These are the maximum concentrations reached. They gradually decrease during water stagnation. This decrease would be due to the formation of sparingly soluble lead hydroxycarbonate [15]. However, some rebounds in concentrations are observed at 12 noon for the TG and the cast iron pipe, at 4 p.m. and after 3 days of stagnation for the PPR pipe. The Pb concentration increases gradually in the water remaining in the PVC pipe to reach the maximum concentration of 0.251 mg.L<sup>-1</sup> after 3 days of stagnation. This concentration fluctuates a little below the maximum value until the end of the experiment. Thus, the concentrations of lead in water of pH 7.04 in stagnation in different pipes ranks according to this order after the first four hours of contact: Pb (TG) > Pb (Cast iron) > Pb (PPR) > Pb (PVC). This order is reversed for Pb (TG) and Pb (Cast iron) in the case where the pH of the water is 5.11. A similar evolution of the Pb concentration in softened water was observed during a study carried out by [16].



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

### 3.1.3. Variation in lead content at pH 9

The variation of the Pb concentration was studied at pH 9.11. Figure 3 illustrates the results obtained. The lead concentration reaches the maximum peak of 4.022 mg.L<sup>-1</sup> in the water that has remained in the cast iron pipe after 4 hours and then decreases according to the stagnation time. This maximum concentration is lower than that found at pH 5.11 (6.522 mg.L<sup>-1</sup>) and at pH 7.04 (5.881 mg.L<sup>-1</sup>) in the same pipe (cast iron) after the same duration . In the case of the PPR pipe, the lead concentration after 4 hours is 5.043 mg.L<sup>-1</sup>. This concentration increases slightly during stagnation to reach a peak of 16.444 mg.L<sup>-1</sup> on the 4th day of stagnation before falling back to a low value of 2.502 mg.L<sup>-1</sup> after the 28th day. Then, this value rises to reach 94.449 mg.L<sup>-1</sup> on the 60th day of stagnation. In the TG, lead increases in water to reach 1.495 mg.L<sup>-1</sup> then decreases slightly until the 28th day before rising to a maximum concentration of 6.730 mg.L<sup>-1</sup> after 60 days of stagnation. This concentration decreases between 60 and 90 days of water stagnation. In the stagnant water in the PVC pipe the Pb increases after 20 hours of stagnation to reach 0.276 mg.L<sup>-1</sup>. A plateau is observed around this value until the 60th day, then the Pb increases slightly to 0.282 mg.L<sup>-1</sup>, the maximum concentration reached on the 90th day.



Figure 3: Evolution of lead concentration as a function of stagnation time of fresh water at pH 9.11 in various pipes and plumbing accessories.

The concentration of lead in the water after 4 hours in different pipes is then classified in this order (Cu) PPR > (Cu) Cast iron > (Cu) TG > (Cu) PVC. The order of peak concentrations reached is as follows: Cu (in PPR pipe at day 60) > Cu (in TG at day 60) > Cu (in Cast iron pipe at 4 hour) > Cu (in pipe in PVC on the 90th day).

# 3.2. Zinc content

# 3.2.1. Variation in zinc content at pH 5

The study of the evolution of the concentration of zinc in stagnant water in the pipes used in domestic installations was carried out in the laboratory under the finished experimental conditions. Figure 4 presents the variation, as a function of time, at pH 5.11 of the concentration of zinc in the water at the outlet of the polyvinyl chloride (PVC), galvanized steel (TG), polypropylene (PPR) and cast iron. The cast iron pipe channel fitted with a galvanized steel elbow and a brass tap has the highest concentration of zinc until the 60th day of water stagnation. The lowest concentration of 30.959 mg.L<sup>-1</sup> is released into the water just 4 hours of water stagnation and the maximum concentration of 255.764 mg.L<sup>-1</sup> of zinc is reached on the 6th day. The TG pipe fitted with a galvanized steel elbow and a brass tap showed a zinc concentration of 22.469 mg.L<sup>-1</sup> in water at pH 5.11 in stagnation after 4 hours of stagnation. This concentration increases slightly during the stagnation period. It reaches the maximum value of 162.903 mg.L<sup>-1</sup> after 90 days of water stay in contact with this piping.



Figure 4: Evolution of the concentration of zinc as a function of the stagnation time of fresh water at pH 5.11 in various pipes and plumbing accessories.

In the PPR pipe, the water has a concentration of  $3.927 \text{ mg.L}^{-1}$  after 4 hours of contact. The zinc concentration fluctuates slightly below this value for the remainder of the experiment. However, peaks with significant concentrations were observed respectively on the 5th day (12.489 mg.L<sup>-1</sup>) and the 90th day (13.851 mg.l<sup>-1</sup>) of water stagnation. For the PVC pipe, stagnant water has a concentration of 0.031 mg.L<sup>-1</sup> at the 4th hour of contact. The Zn concentration fluctuates around this value until 90 days of stagnation. But peaks with higher values were observed on the 1st day (0.783 mg.L<sup>-1</sup>) and on the 28th day (0.949 mg.L<sup>-1</sup>), which is the maximum zinc concentration obtained.

### 3.2.2. Variation in zinc content at pH 7

The evolution of the concentration of zinc in water of pH 7.04 in stagnation in different pipes is presented in Figure 5. These results show that the water at the outlet of the cast iron pipe has higher concentrations of Zn than in the other pipes during the 90 days of stagnation. These results would be related to the nature of this material. It could contain easily leachable zinc and form less insoluble products. The concentrations found gradually increase from 27.990 mg.L<sup>-1</sup> (at the 4th hour) to the maximum concentration of 875.082 mg.L<sup>-1</sup> (at the 90th day) during the stagnation. Galvanized steel pipe comes second only to cast iron in terms of releasing zinc into water at pH 7.04. Zn concentrations vary continuously from 26.113 mg.L<sup>-1</sup> after 4 hours to the maximum value of 598.111 mg.L<sup>-1</sup> after 90 days of stagnation.

The PPR pipe and accessories used released a zinc concentration of 1.097 mg.L<sup>-1</sup> in the 4th hour and 2.802 mg.L<sup>-1</sup>, maximum concentration, after 90 days. In the PVC pipe, the amount of zinc released is much lower than in the other pipes. The Zn concentration varies from 0.030 mg.L<sup>-1</sup> after 4 hours of contact and fluctuates around this value until the 90th day of water stagnation. But some increases in concentration were observed on the 1st day (1.180 mg.L<sup>-1</sup>), the 5th day (0.406 mg.L<sup>-1</sup>) and the 28th day (1.338 mg.L<sup>-1</sup>) which is the maximum concentration.



Figure 5: Evolution of zinc concentration as a function of stagnation time of fresh water at pH 7.04 in various pipes and plumbing accessories.

# 3.2.3. Variation in zinc content at pH 9

The change in zinc concentration in stagnant water in the four types of pipes was evaluated as a function of time at pH 9.11. The results in Figure 6 show that cast iron pipe releases significantly higher amounts of zinc than galvanized steel, PPR and PVC pipe. These amounts are also higher in the same pipe at pH 5.11 and at pH 7.04. The concentration of leached zinc varies from 28.888 to 3370.882 mg.L<sup>-1</sup>, maximum value after 90 days of stagnation. It generally increases during water stagnation. The concentration of zinc is 71.041 mg.L<sup>-1</sup> from 4 hours of contact time with water, it reaches 289.002 mg.L<sup>-1</sup> after 20 hours of contact. Exceptional increases in concentration are also observed on the 4th day (692,200 mg.L<sup>-1</sup>) and the 6th day (1612,847 mg.L<sup>-1</sup>).



Figure 6: Evolution of zinc concentration as a function of stagnation time of fresh water at pH 9.11 in various pipes and plumbing fixtures.

The concentration of zinc in the TG reaches  $11.326 \text{ mg.L}^{-1}$  at pH 9.11. It fluctuates around this value until 28 days of stagnation. But high concentrations were observed at the 12th hour (49.865 mg.L<sup>-1</sup>) and the first week (33.450 mg.L<sup>-1</sup>) of water stagnation. The Zn concentration reaches the maximum peak of 201.975 mg.L<sup>-1</sup> from the 28th to the 60th day of water stay. Thus, the TG, although it contains a layer of zinc deposited on the steel, does not release zinc in as large a quantity as the cast iron pipe. This suggests that the zinc present in the TG is not easily leachable than that present in the cast iron pipe.

The PPR pipe also releases zinc in contact with water of pH 9.11, but at very low concentrations at the start of stagnation. After 4 hours, the concentration of Zn is 0.976 mg.L<sup>-1</sup>, it fluctuates around this value until 28 days of stagnation. Then, it increases to reach the maximum value of 58.864 mg.L<sup>-1</sup> after 60 days and drops to 46.020 mg.L<sup>-1</sup> on the 90th day. The zinc in the PPR pipe could mainly come from the PPR gate valve because it has a metal part that contains zinc. The results obtained show that the leaching of large quantities of Zn requires a long period of stagnation of water with a pH of 9.11. PVC pipe also releases zinc in quantities similar to those released during early stagnation in PPR pipe. These concentrations fluctuate around 0.725 mg.L<sup>-1</sup> obtained after 4 hours until the end of the experiment. The maximum zinc concentration of 0.806 mg.L<sup>-1</sup> is obtained after 12 hours of water residence at pH 9.11.

### 3.3. Evolution of dissolved oxygen concentration

Figures 7, 8 and 9 show the evolution of the dissolved oxygen concentration in the different pipes respectively at pH 5.11; 7.04 and 9.11. The analysis of these results shows that at each pH considered, the dissolved oxygen evolves in a similar way in the PVC pipe and in the PPR pipe. This similarity is linked to their nature because they are all made of plastic materials and contain very few metallic elements (apart from the PPR gate valve). On the other hand, the dissolved oxygen concentration evolves in the same way during water stagnation in cast iron and galvanized steel pipes. This resemblance highlights the similar characteristics of these pipes because they are made of metal alloys composed mainly of iron with more or less the presence of zinc, copper, lead, nickel and also carbon. These three figures show that the concentration of dissolved oxygen is lower in stagnant water in pipes of a metallic nature (TG and cast iron) compared to the concentration of dissolved oxygen observed would be linked to its consumption during water stagnation, because dissolved oxygen is involved in the corrosion of metal tubes and pipes.



Figure 7: Evolution of dissolved oxygen concentration in water held in different pipes at pH 5.11.

Figure 7 above shows that the concentration of dissolved oxygen in the PVC pipe at pH 5.11 is higher compared to that in the PPR pipe. Indeed, the dissolved oxygen involved in the corrosion of the pipes is very little used in the case of the PVC pipe because this pipe contains only low concentrations of metallic trace elements, lower than those found in the case of the PPR pipe. However, at pH 7.04 (Figure 8) and at pH 9.11 (Figure 9) the dissolved oxygen concentration evolves during the stay of the water in the PVC and PPR pipes with similar values.

In a slightly acidic medium with a pH of 5.11, Figure 7 illustrates lower dissolved  $O_2$  concentrations at the start of water stagnation (4 - 8 h) in cast iron and galvanized steel pipes. This justifies the release of high concentrations of zinc, iron and lead in stagnant water in these pipes at the start of stagnation because  $O_2$  is used for the oxidation of metals in the pipes.



Figure 8: Evolution of dissolved oxygen concentration in water held in different pipes at pH 7.04.

In a neutral medium of pH 7.04, Figure 8 shows low dissolved  $O_2$  concentrations at the start of water stagnation (4 - 8 h) in cast iron and galvanized steel pipes. But these concentrations are higher than those found at pH 5.11. They reach lower values after a long period (60 - 90 days) of water stagnation in the pipes. This means that dissolved oxygen is moderately used at the start of water stagnation at pH 7.04 but heavily used during prolonged contact with metal piping. This is closely related to the quantities of metals leached into the

water. Thus, the more dissolved oxygen is used, the more its residual concentration in the pipe decreases and the more the concentration of metals leached in the water increases.

Figure 9 shows the evolution of the  $O_2$  concentration at pH 9.11 similar to that observed at pH 7.04, especially at the start of stagnation. But, after 90 days of water stagnation, the  $O_2$  concentration is marked by an intense decrease (0.13 mg.L<sup>-1</sup>) especially within the cast iron pipe marking its high consumption. This result is also consistent with an exceptional rise in Zn (3370.882 mg.L<sup>-1</sup>) concentrations in stagnant water within this pipe over an extended period.



Figure 9: Evolution of dissolved oxygen concentration in water held in different pipes at pH 9.11.

### IV. Conclusion

The evolution of Pb and Zn levels as a function of water residence time in polyvinyl chloride, galvanized steel, polypropylene and cast iron pipe in the laboratory was studied at pH 5.11, 7.04 and 9.11. It appears from this study that all new pipes made of these pipes and accessories release lead and zinc into the water under the defined conditions. Metal pipes (particularly the cast iron pipe) release much more zinc than plastic pipes (particularly PVC). Thus, the cast iron pipe releases exceptional maximum quantities of zinc of the order of 71 to 5000 times the guide value after 4 hours to 90 days of water stagnation. The PPR pipe equipped with a brand new PPR elbow and tap releases an exceptional maximum quantity of lead of the order of 100 to 9400 times the guide value after 4 hours to 60 days of stagnation of lead. water. Dissolved oxygen evolves in a similar way in plastic pipes for the three pH values studied. This evolution is also similar in metal pipes for the three pH values considered. The similarity of evolution observed is related to the nature of these pipes; the more dissolved oxygen is involved in oxidation-reduction reactions, the lower its remaining concentration in stagnant water.

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