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Factors influencing lead and iron release from some Egyptian drinking water pipes☆

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ABSTRACT

The major objective of this study is to assess the effect of stagnation time, pipe age, pipes material and water quality parameters such as pH, alkalinity and chloride to sulfate mass ratio on lead and iron release from different types of water pipes used in Egypt namely polyvinyl chloride (PVC), polypropylene (PP) and galvanized iron (GI), by using fill and dump method.

Low pH increased lead and iron release from pipes. Lead and iron release decreased as pH and alkalinity increased. Lead and iron release increased with increasing chloride to sulfate mass ratio in all pipes.

EDTA was used as an example of natural organic matter which may be influence metals release. It is found that lead and iron release increased then this release decreased with time.

In general, GI pipes showed to be the most effected by water quality parameters tested and the highest iron release. PVC pipes are the most lead releasing pipes while PP pipes are the least releasing.

1. Introduction

The toxicity of lead remains a matter of public health concern even today, due to the pervasiveness of lead in the environment and the awareness about its toxic effects [1]. Exposure to lead contaminated tap water is a persistent problem and is a significant health risk. Once ingested through the gastrointestinal track, lead accumulates in vital organs and bones, and finally causes a number of diseases ranging from anemia to nervous system degeneration [2]. The World Health Organization (WHO) recommended guidelines of 10 μg/l for lead in drinking water and the same level is adopted by the Egyptian drinking water standards [3].

The major source of lead in drinking water was identified to be plumbing materials. The dissolution of lead in water is called plumbosolvency [4]. Lead pipes, lead-based solder, brass fittings and plumbing fixtures such as pipe's jointing faucets are known to be dominant lead sources in public water supply systems [5–7]. Lead pipes were replaced with other types of pipes such as polymer materials like polyvinyl chloride (PVC), polyethylene (PE) and polypropylene (PP). They currently make up about 54% of the all pipes installed worldwide and PVC makes up 62% of this demand [8]. PVC polymer is mixed with a number of additives including stabilizers in order to provide the range of properties needed in the final products. Stabilizers are often composed of salts of metals like lead, and cadmium [9]. The maximum allowable level of lead extraction is 5.0 μg/l for products of PVC that come in contact with drinking water according to the new National Sanitation Foundation (NSF) standard number 61 [10]. Unlike PVC, other plastics including polyethylene and polypropylene do not require metallic heat stabilizers. In addition galvanized iron (GI) pipes can release significant amounts of lead into standing water, as the zinc coating contains about 1% lead impurities [7].

The corrosivity of water is influenced by a number of factors including pH, alkalinity, chloride and sulfate. In general soft acidic waters are more corrosive to lead and iron than hard waters [7,11].

The main objective of this work is to assess the effect of stagnation time, pipe age, pipes material and water quality parameters such as pH, alkalinity and chloride to sulfate mass ratio on lead and iron release.

2. Materials and methods

2.1. Test conditions

Tap water from surface water sources was used in the experiment and its quality characteristics are listed in Table 1.

The pipes were exposed to six conditions:

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Table 1
Water quality characteristics of tested tap water

Parameter	Value
pH	7.5
Total hardness (mg/l) as $CaCO_3$	160
Calcium hardness (mg/l) as $CaCO_3$	80
Total alkalinity (mg/l)	124
Chloride (mg/l)	30
Sulfate (mg/l)	36
Lead (Pb) (mg/l)	0.01
Iron (Fe) (mg/l)	0.03

(1) Tap water without further treatment (untreated) to show the effect of stagnation time and pipe age on lead and iron release (control pipe).
The tested tap water used was modified in other pipes as below giving different conditions to test.
(2) Tap water with two different pHs (gives two conditions), low pH (pH 6), adjusted with 1 M HNO_3, and high pH (pH 8), adjusted with 1 M NaOH.
(3) Tap water with high alkalinity (250 mg/l), adjusted with $NaHCO_3$.
(4) Tap water with high (Cl/SO_4) mass ratio (mass ratio = 2), adjusted with NaCl.
(5) Tap water with organic chelating agent (ethylene diamine tetracetic acid (EDTA)) solution with different concentrations (0.5, 1, 2, 5, 10 mg/l of EDTA using a stock solution 1 M EDTA) were exposed to 5 months aged pipes and compared with the untreated pipe. Exposure lasted for 7 weeks .All chemicals were analytical grade from Merck.

2.2. Pipe rig experiments

Polyvinyl chloride, polypropylene and galvanized iron pipes were used in the experiment and each pipe is 1 m (39.3699 in.) long and 0.01905 m (0.75 in.) diameter with a volume of approximately 300 ml. Pipe rigs were conducted to illustrate changes in lead and iron release in response to water quality changes and aging.

Bran new pipes were used without additional treatment. All pipes were initially rinsed three times with distilled water then with the tested water. The same rinsing is done for aged pipes used in EDTA experiment. Ends of the pipes were plugged with stoppers from the same kind of material as pipes. The pipes were filled with the appropriate water quality, then laid side by side and maintained in a horizontal position at all times. Pipes were emptied and then refilled with fresh water every Sunday, Tuesday, Thursday of every week resulting in a regular 2, 2 and 3 days (48, 48 and 72 h) weekly stagnation periods. To collect samples, the pipe was gently inverted three times, one stopper carefully removed from the pipe, and the water poured out into the sample container. Within 10 s a volume of the appropriate fresh solution was returned to the pipe.

The pipes were aged by regular water change for the duration of this study which lasted for 5 months (20 weeks) .All pipes were exposed at each condition yielding 18 pipe experiments for each metal (six water qualities × three types of pipes) and duplicate samples were collected from duplicate pipes.

2.3. Chemical analysis and instrumentation

- Influent and effluent water samples were monitored for pH, alkalinity, total hardness, chloride, sulfate and conductivity. Measurement was carried out according to "Standard Methods" [12].
- Total Lead and iron were analyzed using Atomic Absorption spectrometer Varian Spectra AA (220) by using flame and graphite furnace (GTA 110) techniques .For each series of measurements absorption calibration curve was constructed composed of a blank and three or more standards. External reference standards from Merck, Germany, and quality control sample from U.S. EPA were used to confirm the instrument metal concentration reading.

2.4. Statistical analysis

Statistical analysis of the data is carried out using statistic package for social science (SPSS) software, version 14.

3. Results and discussion

3.1. Effect of water and pipe age

The 72-h stagnation time showed higher lead and iron release than 48-h of stagnation. This increase was certified in many results such that established by Lytle and Schock [13] who found that metal levels increased exponentially with respect to stagnation time, particularly for stagnation times in the 20–24 h range. Lead and iron increase as pipe age increases.

Lead release from PVC pipes showed to be the highest of all pipes. Results clearly demonstrate the steady increase in lead concentration released from the PVC pipes with respect to time. Concentrations were higher than the lead standard in drinking water adopted in Egypt. A study carried out by Al-Malack [14] showed by using a circulatory method that lead concentration migrated from PVC pipes into the circulated water after 10 h of exposure reached a value of 0.43 mg/l and by the end of the experiment (48 h), it increased to 0.78 mg/l.

The results showed that lead concentrations were ranged between 0.09 and 0.13 mg/l with mean concentrations of 0.095 and 0.12 mg/l in 2 and 20 weeks, respectively aged pipes (Fig. 1). As shown in Fig. 2, lead release from PP pipes was the least of all pipes and mean concentrations of lead were 0.03 and 0.04 mg/l, while lead mean concentrations were 0.053 and 0.064 mg/l in GI pipes for 2 and 20 weeks aged pipes after 72 h of stagnation as shown in Fig. 3.

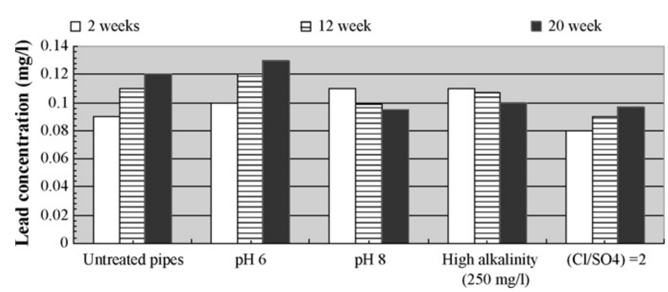

Fig. 1. Mean concentrations of lead release from PVC pipes (72 h stagnation).

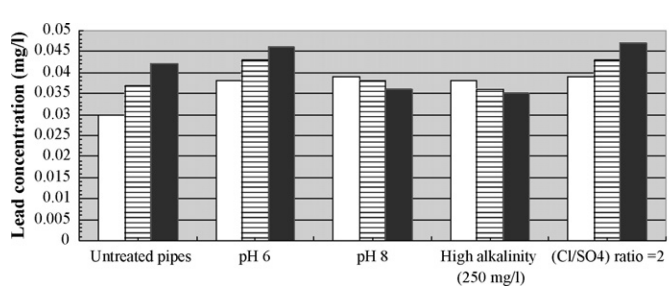

Fig. 2. Mean concentrations of lead release from PP pipes (72 h stagnation).

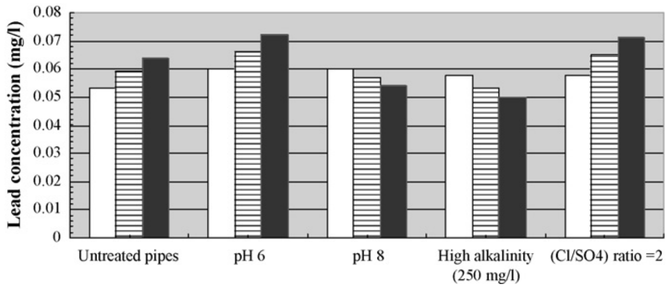

Fig. 3. Mean concentrations of lead release from GI pipes (72 h stagnation).

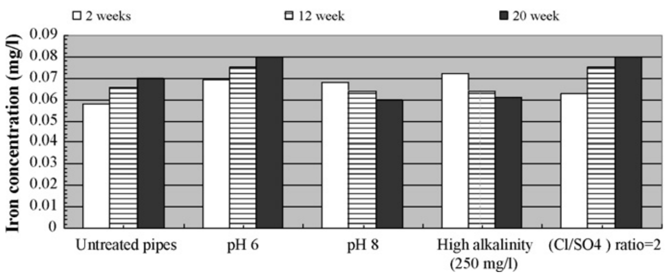

Fig. 4. Mean concentrations of iron release from PVC pipes (72 h stagnation).

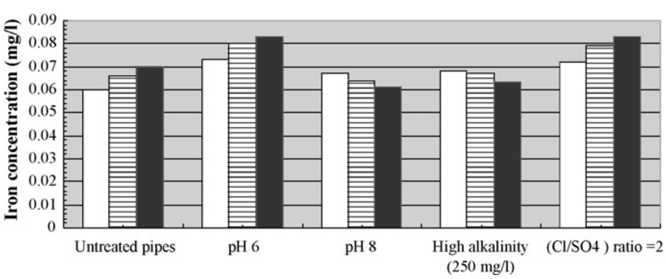

Fig. 5. Mean concentrations of iron release from PP pipes (72 h stagnation).

The mean concentrations of iron released from PVC pipes were 0.058 and 0.07 mg/l in 2 and 20 weeks aged pipes, respectively for 72 h of stagnation. Results are shown in Fig. 4. Iron released from PP pipes showed to be similar to that for PVC pipes and its mean concentrations were 0.06 and 0.07 mg/l as shown in Fig. 5 for the same pipe age and stagnation time. Iron release showed to be the highest from GI pipes. Fig. 6 shows mean concentrations of iron in GI pipe which were 0.7 and 1.44 mg/l in 2 and 20 weeks, respectively aged pipes after 72 h of stagnation. These concentrations are higher than Egyptian standard of iron which is 0.3 mg/l.

The levels of lead and iron released from the three pipes types indicate that each pipe shows a significant variation ($P < 0.05$) for

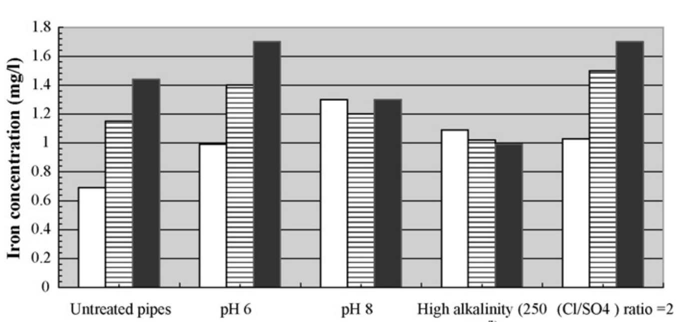

Fig. 6. Mean concentrations of iron release from GI pipes (72 h stagnation).

Table 2
Levels of lead and iron released from the three types of untreated pipes

	Mean	S.E.	*F*-ratio	*P*-value	LSD
Lead					
GI pipe (40)	0.062	0.001	726.8	<0.0001	(PP, PVC)
PP pipe (40)	0.040	0.001			(PVC, GI)
PVC pipe (40)	0.113	0.002			(PP, GI)
Iron					
GI pipe (40)	1.379	0.069	356.2	<0.0001	(PP, PVC)
PP pipe (40)	0.067	0.001			(GI)
PVC pipe (40)	0.068	0.001			(GI)

LSD: least significant difference

lead and iron release. Statistical analysis of lead and iron release as a function of pipe material as shown in Table 2 indicates that lead release is different from one pipe to another, while iron release had no difference in PVC and PP pipes and only differ between both and GI pipes. This result is obtained for all conditions which reveals the similar iron release from PVC and PP pipes.

Total alkalinity, hardness and chloride measured in pipes effluent showed no major changes. Final pH after stagnation increased in GI pipes to 8, while it reached to 7.6 (as control pipes) in plastic pipes.

3.2. *Effect of pH*

3.2.1. *Low pH (pH 6)*

Results showed that lead and iron increases with pH decrease comparing to control pipes. This could be explained by the fact that the solubility of lead is governed by the formation of lead carbonates as pipe deposits, below pH of 8 there is substantial decrease in the equilibrium carbonate concentration and the primary form of lead in water at low pH is predominantly Pb^{2+}, and less abundant inorganic forms include $Pb(SO_4)_2{}^{2-}$, $PbCO_3$ and $Pb_3(OH)_2(CO_3)_2$. Thus, plumbosolvency tends to be at a maximum in waters with a low pH [15].

Lead concentrations were ranged between 0.11 and 0.15 mg/l with mean concentrations of 0.1 and 0.13 mg/l in 2 and 20 weeks, respectively aged PVC pipes after 72 h of stagnation as shown in Fig. 1. These results were similar to that obtained by Al-Malack [14] who studied lead migration to the water at pH value of 5 and found to be 1 mg/l after 48 h of exposure. Burn and Sullivan [16] also reported that acidic conditions enhance extraction of lead from PVC pipes. Lead mean concentrations in 2 and 20 weeks, respectively aged pipes after 72 h of stagnation were 0.038 and 0.046 mg/l in PP pipes as shown in Fig. 2, while concentrations were 0.06 and 0.07 mg/l, respectively in GI pipes as presented in Fig. 3. Lead release in pipes associated with an increase of 8 and 5% in PVC and PP pipes respectively, while in GI pipes lead release increased by 12.5% at the end of experiment comparing with control pipes. This reveals that GI pipes are the highest affected for lead release by low pH.

Iron released from PVC as shown in Fig. 4 had levels between 0.068 and 0.098 mg/l in 72 h of stagnation. Iron released from PP pipes was shown in Fig. 5. Mean concentrations of iron in GI pipes were shown in Fig. 6. Iron levels, were usually reported to be increase with decreasing pH [17]. Increases of iron release were comparable in pipes and PVC had the least increase by 14.5% while iron increased by 18% in both PP and GI pipes at the end of the experiment.

As shown in Table 3, the mean concentrations of lead and iron release from pipes at pH 6 show a significant variation ($P < 0.05$) for lead and iron release from all pipes.

Water quality parameters measured in pipes effluent showed a very slight change in plastic pipes, in GI pipes final pH

Table 3
Effect of pH 6 on lead and iron release from the three types of pipes

	Control pipes		Pipes of pH 6		Independent *t*-test	
	Mean	S.E.	Mean	S.E.	*t*-test	*P*-value
Lead						
GI pipes	0.0618	0.0011	0.0701	0.0011	5.168	<0.0001
PP pipes	0.0401	0.0009	0.0457	0.0008	4.588	<0.0001
PVC pipes	0.1132	0.0020	0.1277	0.0021	5.066	<0.0001
Iron						
GI pipes	1.3788	0.0695	1.6453	0.0676	2.750	<0.01
PP pipes	0.0668	0.0007	0.0807	0.0009	12.028	<0.0001
PVC pipes	0.0685	0.0011	0.0807	0.0014	7.027	<0.0001

Table 5
Effect of high alkalinity on lead and iron release from the three types of pipes

	Control pipes		High alkaline pipes		Independent *t*-test	
	Mean	S.E.	Mean	S.E.	*t*-test	*P*-value
Lead						
GI pipe	0.0618	0.0011	0.0495	0.0007	9.2550	<0.0001
PP pipe	0.0401	0.0009	0.0353	0.0003	5.0210	<0.0001
PVC pipe	0.1132	0.0020	0.0964	0.0013	7.1920	<0.0001
Iron						
GI pipe	1.3788	0.0695	0.9372	0.0110	6.2780	<0.0001
PP pipe	0.0668	0.0007	0.0633	0.0006	3.7550	<0.0001
PVC pipe	0.0685	0.0011	0.0594	0.0010	6.2650	<0.0001

increased to pH 8 while it showed no change in PVC and PP pipes.

3.2.2. High pH (pH 8)

Results showed that, lead and iron decreases with pH increases. The mean concentrations of lead release from PVC pipes were shown in Fig. 1. Lead levels had a mean concentration of 0.11 mg/l in 2 weeks aged pipes then reduced to 0.09 in 20 mg/l weeks aged pipes for 72 h of stagnation with a reduction in release by 20% comparing with control pipe.

Lead in PP pipes reduced by 14.3% and the mean concentration was 0.036 mg/l in 20 weeks aged pipes as shown in Fig. 2. Lead released in GI pipes as shown in Fig. 3 had mean concentrations of 0.06 and 0.05 mg/l in 2 and 20 weeks aged pipes.

Iron release showed to be similar in PVC and PP pipes with mean concentrations of 0.06 mg/l for both in 20 weeks aged pipes and iron release reduced by 14 and 13% in PVC and PP pipes, respectively comparing to control pipes as shown in Figs. 4 and 5. Iron release decreased in GI pipes by 9.7% comparing to control pipes with a mean concentration of 1.3 mg/l as shown in Fig. 6. Previous studies indicated that iron levels were usually reported to decrease with increasing pH [17,18].

A significant variation ($P < 0.05$) of lead and iron release as a function of pipe age is obtained from the statistical analysis in all pipes as shown in Table 4.

Final pH studied in pipes effluent showed no change in PVC and PP pipes but increased to 8.5 in GI pipes. Total alkalinity in samples effluent of GI pipes increased from 124 to 180 mg/l as $CaCO_3$ in this condition only with formation of carbonate alkalinity.

3.3. Effect of high alkalinity

Alkalinity is known to greatly influence the corrosivity of water always correlated with lower lead release [19].

In the study lead release decreased at high alkalinity. The mean concentrations of lead released were 0.11 and 0.1 mg/l in PVC pipes while concentrations were 0.038 and 0.035 mg/l in PP pipes for 2 and 20 weeks aged pipes after 72 h of stagnation as shown in Figs. 1 and 2. Concentrations of lead release from GI pipes had mean concentrations of 0.058 and 0.05 mg/l for the same pips age and stagnation time as shown in Fig. 3.

Table 4
Effect of pH 8 on lead and iron release from the three types of pipes

	Control pipe		Pipes of pH 8		Independent *t*-test	
	Mean	S.E.	Mean	S.E.	*t*-test	*P*-value
Lead						
GI pipes	0.0618	0.0011	0.0543	0.0006	6.008	<0.0001
PP pipes	0.0401	0.0009	0.0354	0.0004	4.757	<0.0001
PVC pipes	0.1132	0.0020	0.0931	0.0011	8.961	<0.0001
Iron						
GI pipes	1.3788	0.0695	1.1570	0.0109	3.154	<0.005
PP pipes	0.0668	0.0007	0.0619	0.0005	5.512	<0.0001
PVC pipes	0.0685	0.0011	0.0598	0.0008	6.511	<0.0001

The results show that lead release increased in the first few weeks of exposure then decreased as pipes aged. This may be explained by that alkalinity affects on the formation of lead carbonate on the pipe surface. When $[PbCO_3]$ (cerussite) is stable, increasing alkalinity reduces lead solubility; when $[Pb_3(CO_3)_2(OH)_2]$ (hydrocerussite) is stable, increasing alkalinity increases lead solubility [20].

Many studies showed that lower iron concentrations in distribution systems have been associated with higher alkalinities [18,19]. In the study iron release decreased with high alkalinity. Fig. 4 shows the mean concentrations of iron released from PVC pipes which were 0.07 and 0.06 mg/l in 2 and 20 weeks aged pipes. The mean concentrations of iron released from PP pipes were 0.068 and 0.065 mg/l, while iron released from GI iron had mean concentrations of 1.1 and 0.99 mg/l in 2 and 20 weeks aged pipes, respectively for 72 h stagnation as shown in Figs. 5 and 6.

Table 5 indicates the significant variation ($P < 0.05$) of lead and iron release from all pipes.

Final pH studied in pipes effluent showed no change in PVC and PP pipes but increased to 9.5 in GI pipes.

3.4. Effect of high (Cl/SO_4) ratio

A review of lead levels reported by 365 water utilities, following the implementation of the U.S. EPA Lead and Copper Rule, carried out by Edwards et al. [21] revealed that higher $Cl^-:SO_4^{--}$ ratios were associated with higher 90th-percentile lead levels at the consumer's tap. The study showed that 100% of the utilities that delivered drinking water with $Cl^-:SO_4^{--}$ ratio below 0.58 met the U.S. EPA's action level for lead of 0.015 mg/l. However, only 36% of the utilities that delivered drinking water with $Cl^-:SO_4^{--}$ ratio higher than 0.58 met the U.S. EPA's action level for lead of 0.015 mg/l.

From the results, increasing (Cl/SO_4) mass ratio to 2 corresponded to increasing lead levels in PP and GI pipes. This may be due to that chloride is an aggressive anion which inhibits the formation of a protective film and could complex lead and iron ions to form more soluble compounds resulting in a higher lead and iron concentration in tap water [22]. The mean concentrations of lead released were 0.04 and 0.047 mg/l from PP pipes and 0.058 and 0.07 mg/l from GI pipes in 2 and 20 weeks aged pipes, respectively for 72 h stagnation as shown in Figs. 2 and 3.

In PVC pipes lead increased as chloride concentration increased but these levels showed to be less than released in control pipes and the mean concentrations were 0.08 and 0.1 mg/l in 2 and 20 weeks aged pipes for 72 h of stagnation as shown in Fig. 1. The effect of

Table 6
Effect of high Cl/SO_4 ratio on lead and iron release from the three types of pipes

	Control pipe		High Cl/SO_4 ratio pipes		Independent *t*-test	
	Mean	S.E.	Mean	S.E.	*t*-test	*P*-value
Lead						
GI pipe	0.0618	0.0011	0.0688	0.0012	4.314	<0.0001
PP pipe	0.0401	0.0009	0.0458	0.0009	4.63	<0.0001
PVC pipe	0.1132	0.0020	0.1004	0.0020	4.537	<0.0001
Iron						
GI pipe	1.3788	0.0695	1.6605	0.0728	2.799	<0.01
PP pipe	0.0668	0.0007	0.0802	0.0010	10.969	<0.0001
PVC pipe	0.0685	0.0011	0.0788	0.0014	5.769	<0.0001

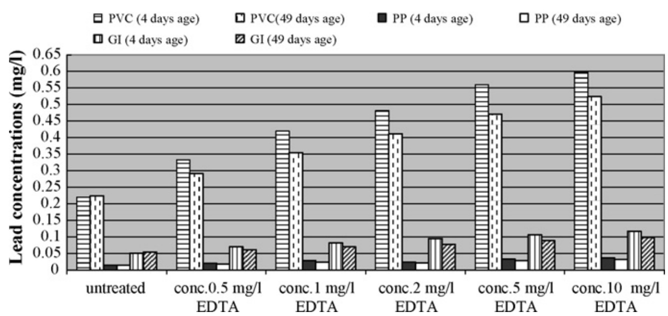

Fig. 7. Effect of EDTA on lead release from pipes at two different ages.

chloride on lead release from PVC pipes was similar to that of Koh et al. [23]. They stated that lead migration is enhanced by the presence of low concentrations of anions such as Cl^- and SO_4^{--}.

Fig. 4 shows the mean concentrations of iron in PVC pipes which were 0.06 and 0.08 mg/l, while as shown in Fig. 5, the mean concentrations were 0.07 and 0.08 mg/l in PP pipes for 2 and 20 weeks aged pipes, respectively after 72 h of stagnation. Fig. 6 shows iron mean concentrations in GI pipes which were 1 and 1.7 mg/l for the same pipes age and stagnation time. Table 6 indicates the significant variation ($P < 0.001$) of lead and iron release from all pipes.

Water parameters studied in pipes effluent showed no change. Final pH in GI pipes increased to 8.5.

3.5. *Effect of ethylene diamine tetracetic acid (EDTA)*

The results indicated that lead and iron release initially increased in pipes then decreased as pipes aged. PVC released the highest lead concentrations while GI released the highest iron concentrations. Figs. 7 and 8 show the effect of EDTA on lead and iron release from pipes.

The main and combined effects of pipe type, EDTA and pipe age had been analyzed statistically and they were found to have a significant variation on lead and iron release as shown in Tables 7 and 8.

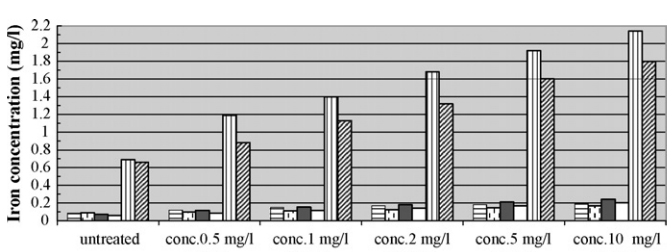

Fig. 8. Effect of EDTA on iron release from pipes at two different ages.

Table 7
Statistical analysis of the combined and the main effects of the different pipe types, EDTA concentrations and pipe age on lead release from the different pipe types

		Simple factorial	
		F	Sig.
Main effects	(Combined)	2116.17	<0.0001
	Pipe types	9869.54	<0.0001
	EDTA concentration	291.67	<0.0001
	Pipe age	19.55	<0.0001
2-Way interactions	(Combined)	46.96	<0.0001
	Pipe types × EDTA concentration	176.34	<0.0001
	Pipe types × pipe age	8.87	<0.0001
	EDTA concentration × pipe age	1.05	NS

Table 8
Statistical analysis of the combined and the main effects of the different pipe types, EDTA concentrations and pipe age on iron release from the different pipe types

		Simple factorial	
		F	Sig.
Main effects	(Combined)	2114.7	<0.0001
	Pipe types	9749.5	<0.0001
	EDTA concentration	386.3	<0.0001
	Pipe age	25.7	<0.0001
2-Way interactions	(Combined)	61.0	<0.0001
	Pipe types × EDTA concentration	229.3	<0.0001
	Pipe types × pipe age	13.4	<0.0001
	EDTA concentration × pipe age	0.7	NS

Table 9
The effects of interactions between pipe type, water quality parameter and pipes age on lead and iron release

	Lead		Iron	
	F-ratio	*P*-value	*F*-ratio	*P*-value
Pipe type	8815.00	<0.0001	5764.30	<0.0001
Water quality parameter	62.798	<0.0001	17.800	<0.0001
Pipe age	111.64	<0.0001	12.603	<0.0001
Pipe type × water quality parameter	33.393	<0.0001	14.881	<0.0001
Pipe type × pipe age	10.255	<0.0001	10.006	<0.0001
Water quality parameter × pipe age	0.035	NS	0.004	NS
Pipe type × water quality parameter × pipe age	0.022	NS	0.005	NS

3.6. *Statistical analysis of the influence of tested factors on metals release*

Statistical analysis for the effects of main and combined interaction between the factors namely (water quality parameter, pipe type and pipe age) was carried out. As shown in Table 9 each of pipe type, water quality parameter and pipe age mainly had a highly significant variation ($P < 0.0001$) for metals release. The combined and interaction of both water quality parameter, pipe type and pipe age gave the same significant variation for lead and iron release.

4. Conclusion

The study reveals a preliminary work on Egyptian drinking water pipes to highlight the lead and iron release from pipes. It can be concluded that

- PVC pipes showed the highest lead release.
- Polypropylene pipes showed the least levels of lead and iron release.
- Lead and iron release increases from all pipes at low pH.

- High alkalinity and pH decrease lead and iron release as pipes aged.
- High Cl/SO_4 mass ratio affects variably on lead release depending on the pipe material while increases iron release from all pipes.
- EDTA increased lead and iron release in all pipes which decreased with time.

A future work is needed to study metals scales and release in the existed distribution system pipes using mechanical testing, X-ray diffractometry and scanning electron microscopy.

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