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ORIGINAL RESEARCH PAPER

AN INDEXING APPROACH FOR THE ASSESSMENT OF HEAVY METALS IN DRINKING WATER PRODUCED BY MAURITANIAN WATER TREATMENT PLANT

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Abstract: In the present work, the assessment of drinking water quality was carried out through a monitoring of heavy metals in the treated and consumed waters in the city of Nouakchott (Mauritania). Monthly sampling was conducted for a period of 24 months between January 2012 and December 2013. Nine parameters were evaluated: *p*H, T (°C), Turbidity (NTU), Al, Fe, Cu, Mn, $Al_2(SO_4)_3$ and CaO. Indexing approaches have been applied by calculating the Heavy Metal Pollution Index (HPI) and Metal Index (MI) for the assessment of influence of heavy metals on the overall quality of water. The obtained results for heavy metals are in good agreement with World Health Organization (WHO) standards. Though the aluminum concentration remains in the limits set by WHO, yet it shows a major contribution in the indices. This has been verified by the statistical analysis which demonstrates fair correlations between aluminum, HPI (r = 0.9) and MI (r = 0.77). Aluminum showed the important influence of seasonal change in the year as well as the doses of reagents injected during the treatment process on the concentration of aluminum is detailed.

Keywords: *heavy metals, HPI, Mauritania, MI, Water treatment*

INTRODUCTION

Monitoring and study of heavy metals in drinking water present an immense importance for the protection of public health [1 – 4]. According to the United Nations [5], 3.5 million people die each year due to poor water supply conditions. Indeed, Africa is the driest continent after Australia. In an African country like Mauritania where water is increasingly scarce [6], monitoring and control of pollution remains a major challenge for the protection of public health. About 2150 Mauritanian which 1700 children under 5 years die each year due to the consumption of the water with poor quality [7]. Contamination of drinking water by heavy metals has always generated a grave consequences on the health of populations and their socio-economic life [8]. The metal trace elements play an essential role for human health; Copper, Iron and Zinc are necessary for life when they do not exceed the limits; On the other hand, the Lead and the Mercury are very dangerous for the living beings [9, 10]. In addition to their natural origins in water, heavy metals come mainly from anthropogenic activities such as, industrial, agricultural and domestic discharges [10, 11]. Different methods like adsorption, chemical precipitation, physical separation, ion exchange, membrane filtration, distillation and hybrid methods are applied for the removal of heavy metals [10]. The efficiency of the methods adopted during treatment plays a predominant role in reducing the level of these elements in order to achieve tolerable values in agreement with national/international standards, while also avoiding the production of corrosive water in order to eliminate the risk of corrosion of pipes which represent a potential source of heavy metals in drinking water [1, 12]. Different studies have been carried out throughout the world for the investigation of heavy metals in drinking water [8 – 15]. Several indexing methods have been developed to evaluate the influence of heavy metals on the overall quality of water. Among these methods, we cite some of them [16 – 20].

Nouakchott (capital of Mauritania) experienced a water shortage during the past years [21]. Although since 2011, the water treatment plants (project of Aftout Essahili) began the supply of drinking water from the Senegal River located at a distance of more than 170 km, with a production capacity of 220 000 m^3/day. Nevertheless, the quality of produced water is still an issue which needs to be addressed.

To the best of authors' knowledge, no studies have been reported on heavy metals in treated and consumed waters in the city of Nouakchott. The objective of this study is therefore the quantitative evaluation of the heavy metals in the waters produced by the Mauritanian water treatment plants. For this purpose, concentrations of aluminum, iron, manganese, and copper as well as certain physical parameters including, T (°C), *p*H and Turbidity will be investigated by following a monthly sampling carried out on water and recovered at the output of water treatment plants. In addition, data will be collected on chemical reagents (aluminum sulphate ($Al_2(SO_4)_3$) and lime (CaO) injected daily, during treatment. The influence of heavy metals on the overall quality of water and their additive effect will be assessed by applying an indexing approach by calculating the Heavy Metal Pollution Index (HPI) and the Metal Index (MI). In the end, multivariate statistical analysis will be employed on the obtained results and interpreted.

MATERIALS AND METHODS

Samples collection was carried out at Beni Nadji water treatment plant on the right bank of the Senegal River (Figure 1). The plant has the capacity of 220 000 m^3/day. It provides water to more than 960000 inhabitants. The raw water is extracted from the Senegal River which is Mauritania's only permanent waterway, located between Mauritania, Senegal, Guinea, and Mali (1800 km). It is used for irrigation, navigation, hydro power generation, and drinking. The water treatment process consists essentially of preliminary chlorination ($Ca(ClO)_2$). The second stage is coagulation, which involves the addition of a chemical coagulant ($Al_2(SO_4)_3$). Then the flocculation combines small particles into larger ones which settle out of the water as sediment. The next stage is the filtration process, where the particles passing through the previous stages are removed. The filtered water is also disinfected by a final chlorination. The water is then stored and ready for distribution. Monthly sampling was conducted over a 24-month period, 2012-2013. The sampling and analysis protocols were carried out according to the standard methods recommended by AFNOR [22] and APHA [23], described by [24]. The samples were collected in polyethylene bottles. Temperature T (°C), *p*H, and Turbidity were measured in situ using a pHmeter (Hsen SION2) and Turbidimeter (Hach Range 2100 P). The other parameters were analyzed immediately in the laboratory; the concentrations of aluminum (Al), iron (Fe), manganese (Mn) and copper (Cu) was measured by an Atomic Absorption Spectrometer (AAS) Flame and Graphite Furnace (type PG 990). Doses of chemical reagents ($Al_2(SO_4)_3$ and CaO) injected, while treatment, were also recorded during the study period. All reagents used were of analytical grade.

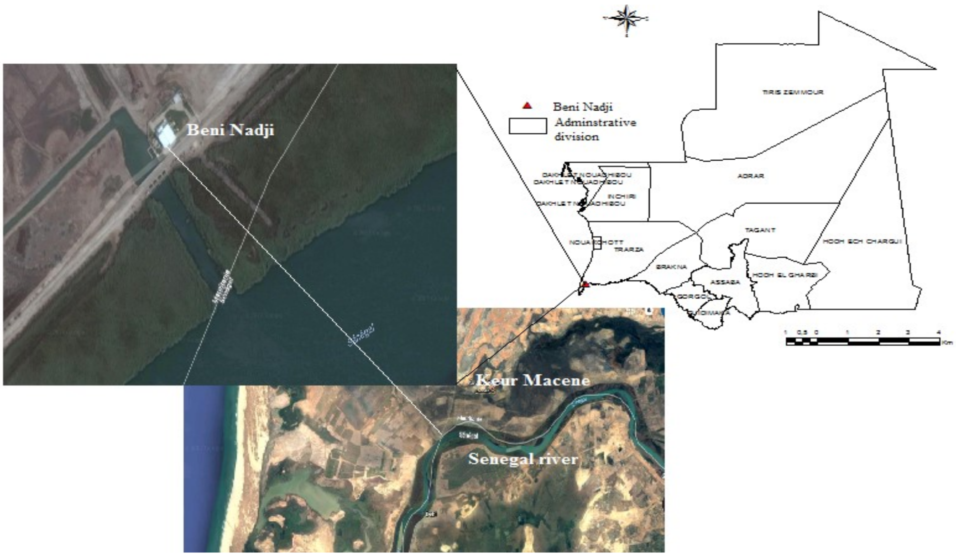

***Figure 1**. Sampling location in Water Treatment Plant, Beni Nadji south Mauritania*

Heavy Metal Pollution Index (HPI): The HPI is a metal evaluation method and an effective tool which makes it possible to demonstrate the influence of heavy metals on the overall water quality [18, 25]. This technique is based on the evaluation of the unit weight assigned to each selected parameter. The HPI is usually calculated according to the equation proposed by [16]. We considered the limit values set by [1] for calculating HPI.

$$HPI = \frac{\sum_{i=1}^{n}(Wi * Qi)}{\sum_{i=0}^{n} Wi} \quad (1)$$

Qi: Sub index of the ith parameter;

Wi: The unit weight of the ith parameter and n is the number of parameters considered.

The sub index Qi is given by the following formula:

$$Qi = \sum_{i=1}^{n} \left(\frac{Mi - Ii}{Si - Ii}\right) * 100 \quad (2)$$

Mi: Monitored value of heavy metal of the ith parameter;
Si: Standard value of ith parameter;
Ii : Ideal value of the ith parameter.
The unit Weight (Wi) is obtained by equation (3):

$$Wi = \frac{1}{Si} \quad (3)$$

The critical value of the metal pollution index is 100.
Metal Index (MI): In order to calculate this index, the model proposed by [20] is used. The MI is an assessment method that provides an overview of the overall water quality based on the additive effect of heavy metals [26, 27]. It is given by the following equation:

$$MI = \sum_{i=1}^{n} \left(\frac{Ci}{(MAC)i}\right) \quad (4)$$

Ci: is the concentration of each metal;
(MAC)i: is Maximum Allowable Concentration.

If the MI is higher than unity, the water cannot be used for human consumption.
Statistical analysis: Statistical methods are commonly used in the interpretation of water quality data [26 – 29]. In this study, statistical analysis was performed with software Origine 8.5 and SPSS 20. The correlation matrix was used to explain correlations between variables studied as well as the sources of the various metallic elements.

RESULTS AND DISCUSSION

The results obtained on the physical parameters are presented in Table 1. These results show that the treated water is slightly basic with annual average *p*H values for 2012 and 2013 respectively 7.79 ± 0.177 and 7.83 ± 0.10. Average water temperatures range between 24.90 ± 2.85 and 25.68 ± 2.48 °C. Turbidity of the water vary within the ranges 0.30 - 1.25 and 0.25 - 0.59 NTU. During the period of this study, the physical parameters demonstrated the results in accordance with the standards set by the [1]. These results are in good agreement with those obtained by [30].
The overall assessment of heavy metals is presented in Table 1. The lowest concentrations were recorded for manganese (Mn) with mean values of 0.006 ± 0.004 and 0.002 ± 0.002 mg·L^{-1}. These values correspond to the WHO standard (0.4 mg·L^{-1}). Mean iron (Fe) concentrations are lower than the WHO standard (0.3 mg·L^{-1}), oscillating between 0.023 ± 0.016 and 0.017 ± 0.01 mg·L^{-1}. The copper (Cu) level is

very low compared to the WHO standard (1 mg·L^{-1}) with mean values of 0.028 ± 0.017 and 0.021 ± 0.01 mg·L^{-1}.

Table 1. *Descriptive statistics of the physical parameters and metal concentrations*

Parameter	Unit	Si*	2012				2013			
			Min	Average	Max	SD	Min	Average	Max	SD
*p*H	-	6-8.5	7.54	7.79	8.12	0.177	7.69	7.83	7.99	0.1
Temperature	°C	25	20.57	25.68	29	2.482	20.7	24.9	28.32	2.85
Turbidity	NTU	5	0.3	0.68	1.25	0.354	0.25	0.44	0.59	0.09
Al	mg·L^{-1}	0,2	0.04	0.06	0.08	0.014	0.055	0.07	0.1	0.02
Fe	mg·L^{-1}	0.3	0	0.023	0.056	0.016	0	0.017	0.04	0.01
Mn	mg·L^{-1}	0,4	0.0008	0.006	0.014	0.004	0	0.002	0.006	0.002
Cu	mg·L^{-1}	2	0	0.028	0.06	0.017	0	0.021	0.05	0.01

*S*i*: Standard value of *i*th parameter

The obtained results for manganese, iron and copper are generally low compared to those obtained by [31, 32], for the assessment of heavy metals in raw water before treatments. Coagulation, flocculation and sedimentation allows the elimination of heavy metals [33]. These results show the efficiency of the processes applied in treatment plant to reduce the level of metallic trace elements which, according to [32], are highly correlated with the suspended matter. Aluminum (Al) is the most predominant element compared to other heavy metals, with concentrations ranging from 0.06 ± 0.014 to 0.07 ± 0.02 mg·L^{-1}, with maximum values of 0.08 mg·L^{-1} in 2012 and 0.10 mg·L^{-1} in 2013. The contents of aluminum are below the standard set by the WHO (0.2 mg·L^{-1}). Aluminum sulphate ($Al_2(SO_4)_3$) added during coagulation represents the main source of increase in the level of aluminum in drinking water [34, 35].

The data obtained for the heavy metals (Mn, Fe, Cu and Al) allowed the estimation of the Heavy Metal Pollution Index (HPI) based on equation (1), proposed by [16], and the standards of [1]. Annual changes in HPI are presented in Table 2. Indeed, the obtained values are below the critical threshold (<100). The HPI indicates maximum values in August 2012 and in February 2013, while minimum values are recorded in November (Figure 2). The HPI values show a low level during the months following the rainy season (October, November and December). This season is characterized by a raw water highly charged with suspended matter [36].

Table 2. *Values of HPI and MI determined during the study period*

	2012				2013			
	Min	Average	Max	SD	Min	Average	Max	SD
HPI	12.12	18.11	23.11	3.44	13.78	18.87	26.64	3.80
MI	0.31	0.45	0.60	0.08	0.34	0.46	0.65	0.08

The application of equation (4), proposed by [20], on the concentration of heavy metals (Mn, Fe, Cu and Al) made it possible to obtain the Metal Index (MI) Table 2. The monthly variation (Figure 3) shows that the maximum values are recorded in the months of August 2012 and February 2013. The MI remains below the limit (<1). The months of October, November and December are also characterized by low values of MI. In general, HPI and MI tend to increase in 2013 compared to 2012.

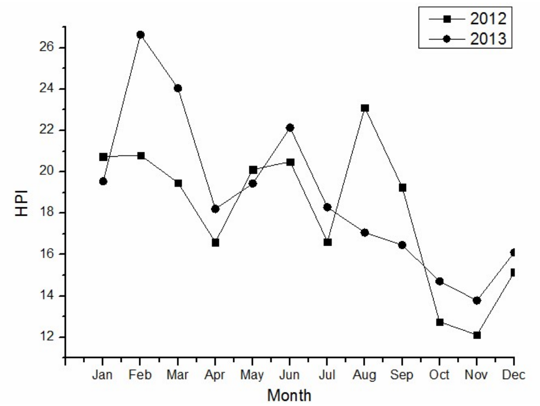

***Figure 2.** Variation of the monthly values of HPI determined for treated water*

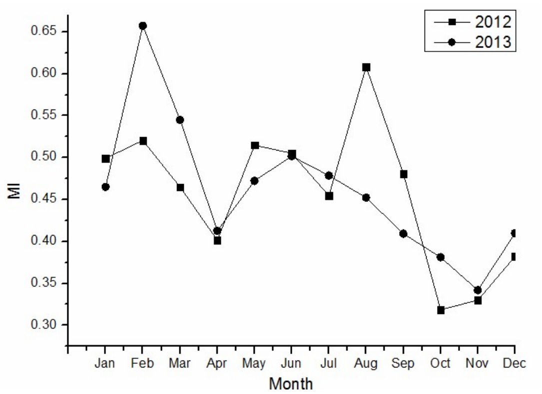

***Figure 3.** Variation of the monthly values of MI determined for treated water*

The statistical analysis made it possible to establish the Pearson correlation coefficients between different variables (physical parameter, heavy metals and chemical reagents). The results show a perfect positive correlation between the HPI and MI (r = 0.968). Aluminum shows a positive correlation with HPI (r = 0.908) and MI (r = 0.777). With the exception of copper, which is negatively correlated with HPI (r = -0.6) and aluminum (r = -0.718); other metals show low correlations with the indices. The *p*H shows a positive correlation with aluminum (r = 0.525), while temperature has a negative correlation with aluminum (r = -0.506) and *p*H (r = -0.507). Turbidity indicates negative correlations with HPI (r = -0.648), MI (r = -0.587), aluminum (r = -0.655), and positive correlation with copper (r = 0.512). Aluminum sulfate is negatively correlated with HPI (r = -0.610) and aluminum (r = -0.799). The negative correlation between *p*H and aluminum sulphate (r = -0.617) is explained by the effect of the latter on acidification of water. Lime shows negative correlations with HPI (r = -0.701), MI (r = -0.577) and aluminum (r = -0.793), and positive correlations with copper (r = 0.709), turbidity (r = 0.529) and aluminum sulphate (r = 0.824). The Turbidity is generally weakly correlated with the physical parameters: *p*H (0.064), and temperature (0.168). The origin of turbidity is commonly associated with the presence of suspended matter which is correlated with the trace metals parameters. These correlation analyses show, in general, that aluminum has a major contribution in HPI and MI. Other metals have only little contributions. The origin of aluminum in treated water is largely related to the use of $Al_2(SO_4)_3$ during coagulation [35, 37]. According to [24], a water treatment plant, in good condition, should produce water with an aluminum concentration less than 50 $\mu g \cdot l^{-1}$. The *p*H, temperature and turbidity of water represent factors determining the solubility of residual aluminum concentration (particulate aluminum, organic monomeric aluminum, inorganic monomeric aluminum) [34, 37]. Figure 4 shows that the maximum values of the aluminum are recorded during the January-June period in which the turbidity and temperature values are low compared to those recorded during the other periods of the year (rainy season). This is in good agreement with correlation analyses (Table 3) and it shows that the aluminum concentration rate decreases during the rainy season and increases during the dry period.

Table 3. *Pearson correlation coefficient (r) between parameters, indices and chemical reagents*

	HPI	MI	Al	Fe	Mn	Cu	*p*H	T	Tur	$Al_2(SO_4)_3$	CaO
HPI	1.000										
MI	.968	1.000									
Al	.908	.777	1.000								
Fe	.153	.381	-.270	1.000							
Mn	.074	.147	-.005	.042	1.000						
Cu	-.600	-.446	-.718	.294	.092	1.000					
***p*H**	.381	.268	.525	-.351	-.119	-.322	1.000				
T	-.466	-.410	-.506	.112	.080	.228	-.507	1.000			
Tur	-.648	-.587	-.655	.068	-.103	.512	.064	.168	1.000		
$Al_2(SO_4)_3$	-.610	-.448	-.799	.476	.068	.609	-.617	.521	.443	1.000	
CaO	-.701	-.577	-.793	.247	.063	.709	-.332	.296	.529	.824	1.000

The raw water of Senegal River is highly charged in suspended matter during the rainy season [30], and high doses of aluminum sulphate ($Al_2(SO_4)_3$) are required during coagulation. Figure 5 shows the correlation between aluminum sulphate and aluminum (r = -0.799), confirming that high doses of $Al_2(SO_4)_3$ are not the cause of increase in the level of residual Aluminum in the treated water.

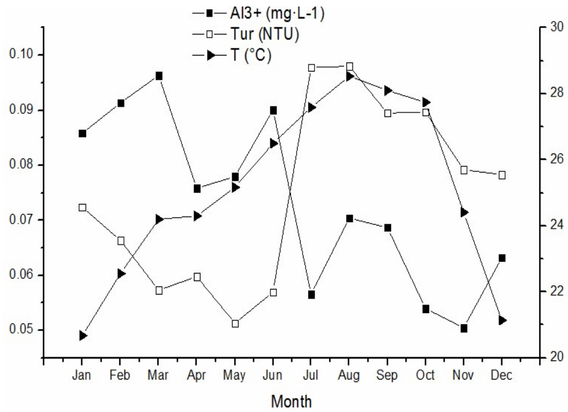

Figure 4. *Variation of the monthly values of the Al, Turbidity and T(°C)*

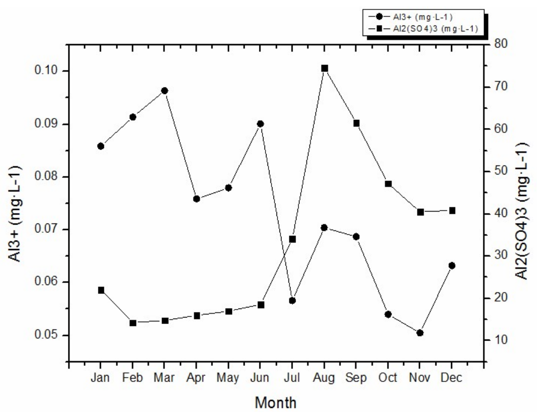

Figure 5. *Variation of the monthly values of the Al and $Al_2(SO_4)_3$*

The addition of $Al_2(SO_4)_3$ is accompanied by a drop in *p*H and loss of alkalinity according to the hydrolysis equation [38]:

$$Al_2(SO_4)_3 \cdot 18\ H_2O + 3\ Ca(HCO_3)_2 \rightarrow 18\ H_2O + 3\ CaSO_4 + 2\ Al(OH)_3 + 6\ CO_2$$

Aluminum is insoluble at *p*H between 6.5 and 8.5 and highly soluble at an acid (*p*H < 6) or basic *p*H (*p*H > 8.5) [24, 34]. As a result, the residual aluminum concentration is related to the contents of HCO_3^- ions and to the formation of $Al(OH)_3$ according to the hydrolysis equation:

$$[Al(H_2O)_6] + H_2O \rightarrow [Al(H_2O)_5OH]^{2+} + H_3O^+$$

$$[Al(H_2O)_5OH]^{2+} + H_2O \rightarrow [Al(H_2O)_4(OH)_2]^+ + H_3O^+$$

$$[Al(H_2O)_4(OH)_2]^+ + H_2O \rightarrow [Al(OH)_3 \cdot 3(H_2O)]^+ + H_3O^+$$

The H_3O^+ ions formed during the hydrolysis of aluminum ions must be removed by adjusting the *p*H so as to facilitate the formation of $Al(OH)_3$:

$$H_3O^+ + HCO_3^- \rightarrow H_2CO_3 + H_2O$$

Adjustment of *p*H is achieved by adding lime (CaO) [38], which is injected in parallel with $Al_2(SO_4)_3$ in order to avoid the drop in *p*H, thus favoring the precipitation of aluminum in the form of $Al(OH)_3$. This will reduce the level of residual aluminum [33]. Figures 6 and 7 as well as the correlation between CaO and aluminum (r = -0.793) indicating that high doses of CaO cause a decrease in residual aluminum. Maximum values of residual aluminum are observed during the dry season when low doses of CaO are injected Figure 6. The major contribution of aluminum in the HPI and MI indices is strongly influenced by the seasonal variation as well as the doses of CaO injected during the treatment.

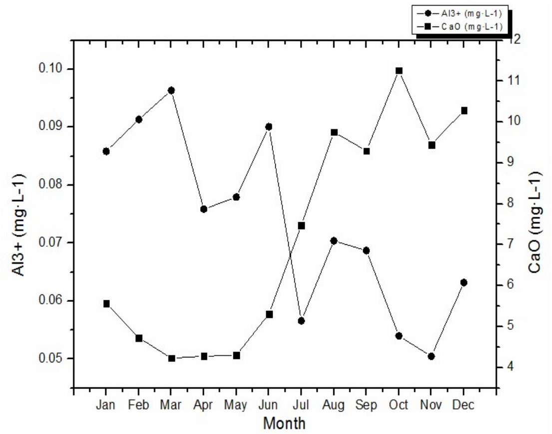

Figure 6. *Variation of the monthly values of the Al and CaO*

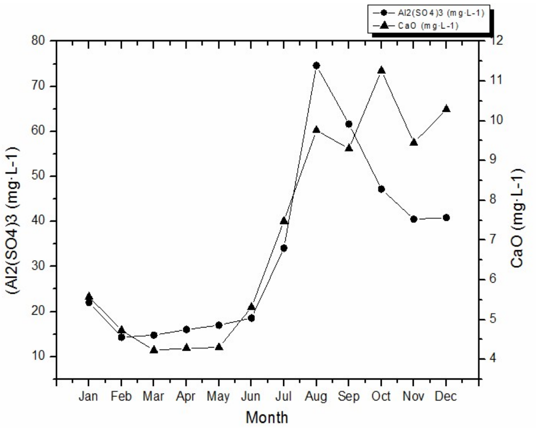

Figure 7. *Variation of the monthly values of the $Al_2(SO_4)_3$ and CaO*

CONCLUSION

In this study, our objective has been to quantitatively evaluate the heavy metals in the treated and consumed waters in the city of Nouakchott. The obtained results show that although the heavy metal levels fluctuate with the seasons of the year, they always remain below the standards set by World Health Organization. The metal concentrations show this order of predominance: Al > Fe > Cu > Mn. The evaluation of the influence of heavy metals on the overall quality of water and their additive effect were carried out using an indexing approach. HPI and MI indices remain below their critical thresholds. Aluminum demonstrates an important contribution in the indices. Aluminum concentration has been found to strongly relate the seasonal changes and is also influenced by the doses of chemical reagents injected during treatment. In the prospects, it would be particularly interesting to study the behavior of aluminum during the transport of water in the distribution network, with possibilities of decreasing the concentration due to precipitation of $Al(OH)_3$ in the pipes and reservoirs.

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