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Assessment of Free Chlorine Residual and Microbiological Parameters in Supply Water of Centralized Supply Water Companies in Vietnam

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Abstract

In the study, the free chlorine residual and microbiological parameters in supply water of centralized supply water companies were assessed with a total of 1,612 samples in the period of 2020 and 2022. The results showed that 334 samples did not meet the permitted limit QCVN 01-1:2018/BYT for free chlorine residual (0.2-1.0 mg/L), of which, about 80% of sample have concentration less than the lower limit. The reason is due to the inaccurately used chloramine dose, mainly quantified without adjusting for actual output quality and distribution system. Some companies even periodically disinfect water 1-3 times/week. In many cases, the water samples at the companies' storage tanks have high free chlorine residual, about 2 times greater than upper limit (>2.0 mg/L). Regarding microbiological parameters, for total coliforms about 40-50% surveyed provinces have water samples that were non-compliance with the acceptable quality standard for QCVN 01-1:2018/BYT with total of 102, 57 and 105 samples. For those water samples, free chlorine residual found also lower than required limit (<0.06 mg/L). Apart from total coliforms and E. coli, Pseudomonas aeruginosa (Ps. aeruginosa) was also detected in water samples in some provinces and cities with levels exceeding the allowable standards from several times to dozens of times. In addition, the number of provinces/cities where Ps. aeruginosa was detected in supply water also increased over the years but insignificantly. Water samples that did not meet the standard are mostly from water supply companies with a capacity of less than 1,000 m^3/day. Correlation analysis by regression coefficient R^2 and Pearson correlation coefficient showed that there was a negative correlation between free chlorine residual and microbiological parameters (total coliforms, E.coli), but the correlation was low and non-linear.

Keywords: supply water, free chlorine residual, microbiological parameters, total coliforms, *E.coli, Ps. aeruginosa*

1. Introduction

The supply water demand for daily life and meals is increasing, especially in urban areas with high population density, urban expansion, and development of socio-economic activities. According to the Ministry of Construction, by 2022, Vietnam has 750 supply water companies in urban and rural areas with a total capacity of 12.6 million m^3/day. There was 94.2% of urban residents using clean water through a centralized water supply system that was an increase of 2.2% compared to 2021 [1]. At present urban water supply companies, raw water exploited from underground sources accounts for nearly 40% and surface water accounts for over 60%. Underground water extraction works are mainly on-site water extraction works, concentrated in urban areas or in neighbouring areas - densely populated places with many construction and production activities related to discharge sources, environment and surface water are polluted, so the input water of water supply companies is at risk of contamination and affects the output water quality. In 2014, the Ministry of Health assessed water quality of 166 samples taken from households of 06 districts in Hanoi city and showed that some indicators did not meet the allowable standards such as: permanganate index, free chlorine residual, ammonium, nitrite, aluminium, arsenic, total coliforms and *E.coli*.

In 2018, the Institute of Occupational and Environmental Health evaluated the quality of water supplied by 80 companies providing supply water for daily life and concluded that 45% of water samples did not meet the standards with free chlorine residual parameters, ammonium, permanganate index. The 2019 monitoring showed that 117/319 samples (36.7%) did not meet the standard also related to free chlorine residual, ammonium, permanganate index) [2].

Free chlorine residual depends on many factors including factors that directly affect the "CT value" (concentration of disinfectant and contact time) such

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as temperature and pH of the water [3]. In addition, there are many factors affecting the decomposition and free chlorine residual in the feed water, including hydraulic regime and characteristics of the water distribution system. In the study by Amrina *et al*. (2020) water distribution piping systems and the corresponding free chlorine residual levels were analysed and simulated in order to propose design and improve the performance of hydraulic systems, improve water quality and safety for users. The average decomposition rate constant of free chlorine residual was determined to be 0.00033 and the free chlorine residual concentration in the water distributed in the range of 0.1 to 0.2 mg/L [4].

In this study, the free chlorine residual and microbiological parameters (total coliforms and *E.coli*) in supply water of centralized supply water companies in provinces/cities in Vietnam will be investigated.

2. Research Objects and Methods

2.1. Research Objects

The surveys were implemented during 2020-2022:

(i) in 2020: 540 samples (06 samples/company x 02 water supply companies x 45 provinces/citites);

(ii) in 2021: 512 samples (04 samples/company x 04 water supply companies x 32 provinces/cities);

(iii) in 2022: 5560 samples (04 samples/company x 05 water supply companies x 28 provinces/cities). Number of water supply companies with a capacity of greater and equal 1,000 m^3/day is 88 and that less than 1,000 m^3/day is 40.

2.2. Methods

Each water supplier took 04 water samples including: 01 sample at the storage tank after treatment at the water supplier before being put into the distribution pipeline network, 03 samples at the faucet used randomly on the distribution network (including water distribution vehicles such as tank trucks or water tankers) according to Circular 41/2018/TT-BYT). Supply water samples are taken and preserved according to TCVN 6663-5:2009 (ISO 5667-5:2009), Water quality - Sampling - Part 5: Guidelines for sampling drinking water from treatment companies and water distribution systems and TCVN 6663-3:2016 (ISO 5667-3:2012), Water quality - Sampling - Part 3: Preservation and treatment of water samples. Only in 2020, each water supplier took 02 more water samples at some areas having high density of population, such as schools, restaurants, hotels, v.v.

2.2.1 Determination of free chlorine residual

The free chlorine residual concentration is determined by (1) a color change to identify the presence of chlorine, (2) a measurement of the intensity of that color to determine how much free chlorine is present. Specifically, color wheel test kits are used in which a powder or tablet chemical DPD (N,N diethyl-p-phenylene diamine) was added into water sample to make the color change into pink in the presence of chlorine [5].

2.2.2. Determination of total coliforms, E.coli, Ps. aeruginosa:

Total coliforms and *E.coli* are analysed by TCVN 6187-1:2019 Water quality - Detection and enumeration of *Escherichia coli* and total coliforms bacteria - Part 1: Membrane filtration method.

Ps. aeruginosa was identified according to TCVN 8881:2011 (ISO 16266:2006): Water quality - Detection and enumeration *Ps. aeruginosa* - Membrane filtration method.

2.2.3. Statistical analysis

The relationship between the free chlorine residual and the pathogenic bacterial counts at different chlorine doses was carried out using regression coefficient analysis (R^2) that processed by SPSS version 16.0, computer application. The Pearson correlation coefficients were calculated using the following equation [6].

$$x = \frac{\sum_i (x_i - \bar{x})(y_i - \bar{y})}{\sqrt{\sum_i (x_i - \bar{x})^2} \sqrt{\sum_i (y_i - \bar{y})^2}} \quad (1)$$

3. Results and Discussion

3.1. Free Chlorine Residual Analysis

The main input water sources are surface water (about 69.5%) and groundwater (about 14.1%), Particularly, in companies with a capacity of greater and equal 1,000 m^3/day (74/102 companies used survey water, accounting for 72.5%) while this rate is 57.7% for companies with designed capacity of less than 1,000 m^3/day.

When using surface water, water supply companies will face difficulties because surface water is at risk of being polluted due to having to receive waste sources from agricultural, industrial, and domestic production activities. human pollution, especially high organic matter pollution, high turbidity/suspension, microbial contamination. In coastal areas there is also high chloride content.

For water disinfection: Common chemicals used to disinfect water companies in Vietnam are mainly chlorine or its compounds. For water companies inspected in 2021, chlorine liquid or gas or sodium hypochlorite (electrolyzed directly from salt in some companies) is added to the water before entering the treated water storage tanks.

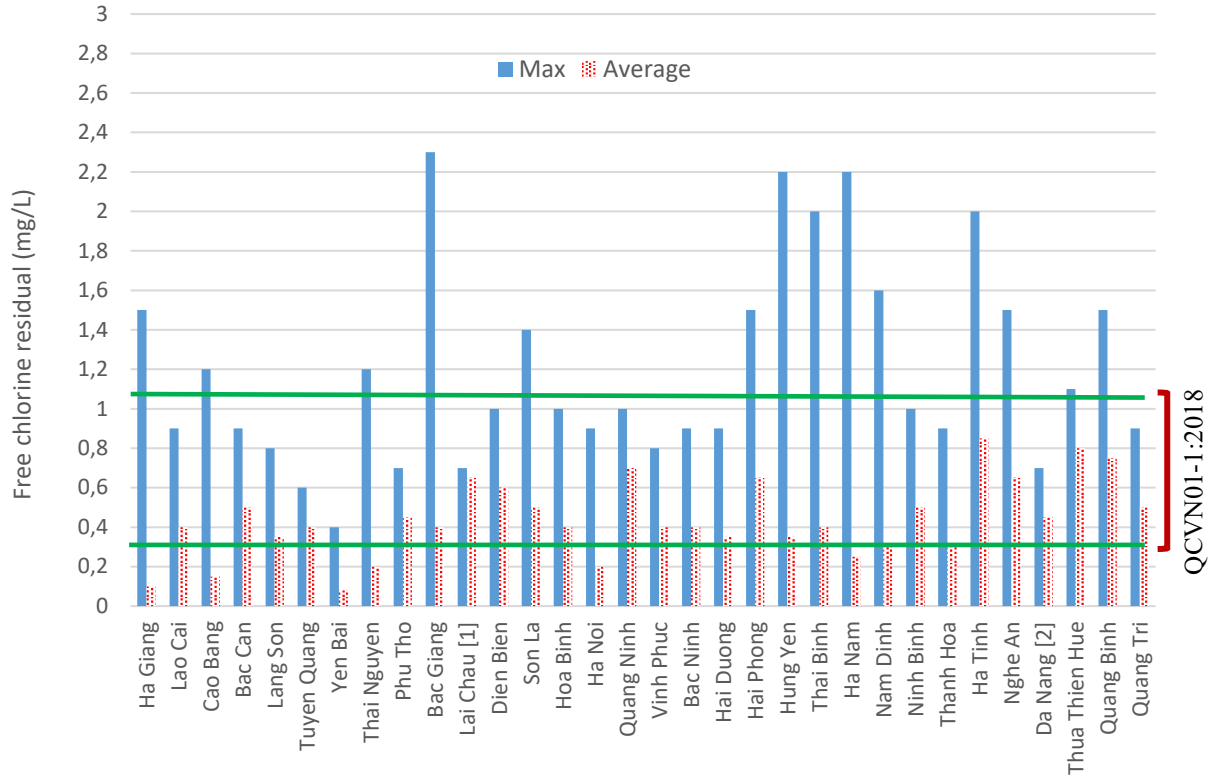

Fig 1. Results of analysis of free chlorine residual (max and average values) in provinces/cities during 2021 campaign

Table 1. Results of analysis of total coliforms and *E.coli* criteria of domestic water samples during 2020-2022

No.	Parameters QCVN 01-1:2018/BYT	No. of provinces with water samples did not meet the permitted standard						No. of water samples did not meet the permitted standard					
		2020		2021		2022		2020		2021		2022	
		N	%	N	%	N	%	n	%	n	%	n	%
1	Total coliforms	21	*46.7*	13	*40.6*	15	*53.4*	102	*18.9*	57	*11.1*	105	*18.8*
2	*E. coli*	3	*6.7*	5	*1.7*	5	*1.8*	7.0	*1.3*	20	*3.9*	50	*8.9*
3	Free chlorine residual	25	*55.6*	24	*75*	19	*67.9*	102	*18.9*	102	*19.9*	130	*23.2*

As can be seen in Fig. 1 and Table 1, in 2021 campaign 24/32 provinces and cities had at least 01 test water sample that did not meet the free chlorine residual parameter. For provinces that the average free chlorine residual content was lower than or equal to the allowable limit (0.2 mg/L) there are 9-14 water samples that did not meet QCVN 01-1:2018/BYT such as Yen Bai, Ha Giang, Cao Bang and Thai Nguyen, respectively, in the ranges of 0.06-0.4; 0.06-1.5; 0.06-1.2 and 0.1-1.2 mg/L). In addition, the percentage of water samples having average the free chlorine residual content non-compliance with the acceptable quality standard for QCVN 01-1:2018/BYT mostly were found in water supply companies with a capacity of less than 1,000 m^3/day. The results were similar in the study of Nguyen Thi Hai Ha *et al*., (2022). In the year 2020-2021, the analysis results of 345 water samples from centralized water supply companies with a capacity of greater than 1000 m^3/day and night show that 44 and 79 samples respectively do not meet the requirements of total coliforms and free chlorine residual criteria, respectively [7].

The permitted minimum level of free chlorine residual in some countries are higher than that in Vietnam. In the study of Perez *et al* (2022) [8], the concentration of active chlorine in the supply water distribution network was measured and estimated according to the model to optimize the dosage used and reduce the risk of forming by-products (chlorinated hydrocarbon). The results showed that the free chlorine residual concentration ranges from 0.4 to

1.2 mg/L (permitted level from 0.4-1.0 mg/L). The higher minimum level of free chlorine residual in the feed water may cause the risk of forming by-products such as trihalomethanes (THMs) when water transported in the distribution network are significantly dependent on temperature. To reduce the risk of THMs by-products some water supply companies have used a combination of ozone and chlorine [9].

In the study of NTP Thao *et al*. (2020) [8] the correlation between free chlorine residual content and *E. coli* and total coliforms parameters of drinking and supply water sources (end of water supply network) in districts of Ho Chi Minh city were determined and found the average free chlorine residual content was within the allowable range of 0.3-0.5 mg/L (QCVN 01-1:2018/BYT). However, the average concentration of *E. coli* and total coliforms of the analysed water samples all exceeded the allowable limits of less than 1 and less than 3 CFU/100 mL, respectively [10].

According to the results of the assessment of free chlorine residual content in the field, it is found that many cases of water samples in the companies' reservoir have high free chlorine residual content (greater than 2.0 mg/L) but samples taken on the system distribution with a significantly reduced free chlorine residual content (less than 0.1 mg/L). The reason might be the consumption of free chlorine by organic matters and ammonium during the distribution process. However, the correlation between organic matters and free chlorine residual is weak as only several water samples in 1 or 2 water companies had high concentration of permanganate and low concentration of free chlorine residual at the same sampling points. There is no correlation between free chlorine residual and ammonium. Another possibility is that there might be leaks along the distribution system, especially the tertiary ones, due to the old and degraded pipes or effects by construction work. In contrast, water samples of some water supply companies, especially in the northern mountainous areas such as Cao Bang, Ha Giang, Yen Bai, Tuyen Quang, Thai Nguyen, etc., have low free chlorine residual content less than 0.1 mg/L, including the water sample at the companies' storage tanks.

3.2. Total Coliforms and E.coli Analysis

Total coliforms and *E.coli* parameters are the two main microbial indicators that show whether the water is treated up to the standards or re-contaminated with microorganisms during the distribution process

The results showed that in 2020, 2021 and 2022 102/540, 57/512 and 105/560 supply water samples, respectively did not meet permitted standard on total coliforms and these numbers for *E.coli* were 7/540, 20/512 and 50/560. The water samples that did not meet these two microbiological criteria were all taken from the distribution system of water supply companies' capacity less than 1,000 m^3/day, which means treated water was re-contaminated with microorganisms during distribution. One of the main reasons is that the residual free chlorine residual content in the clean water is not enough to maintain the effectiveness of water disinfection during the distribution process. The results of the assessment of free chlorine residual above also showed similarity, when the water samples of 8/13 provinces in 2021 campaign did not meet these two criteria and was lower than the minimum allowable limit. In addition, as discussed above, the old and low quality of water pipelines could lead to leaks and the reduction of free chlorine and intrusion of contaminants from the outside could happen. Therefore, supply water taken on the distribution system was contaminated with total coliforms and *E.coli* partly because of that. Once, microorganisms present, it could create biofilms inside the pipelines and the disinfection effectiveness of free chlorine could not remain. Therefore, although some water samples have high free chlorine residual content, even higher than the maximum allowable limit, total coliforms or *E.coli* were still found. The relationship between total coliforms, *E. coli* and free chlorine residual in supply water samples by regions (Table 2).

Table 2. Comparison of total coliforms, *E. coli* and free chlorine residual in supply water samples by regions

Parameters		Total coliforms	*E. coli*	Free chlorine residual
		CFU/100mL		mg/L
Northern midland and mountainous region (15 provinces)	Min	ND	ND	ND
	Max	**23**	**17**	**2.3**
	Aver	ND	ND	0.4
Red river delta region (10 provinces)	Min	ND	ND	ND
	Max	**18**	**16**	**2.2**
	Aver	ND	ND	0.4
Central region (including North Central region) (7 provinces)	Min	ND	ND	ND
	Max	**23**		**2.0**
	Aver	ND		0.6
Total of 32 investigated Provinces/ cities	Min	ND	ND	ND
	Max	**23**	**17**	**2.3**
	Aver	ND	ND	ND

Note: *ND is Non Detection*
LOA of Total coliforms: less than 3.0 CFU/100mL
LOA of E. coli: less than 1.0 CFU/100mL
LOA of Free chlorine residual: less than 0.06 mg/L

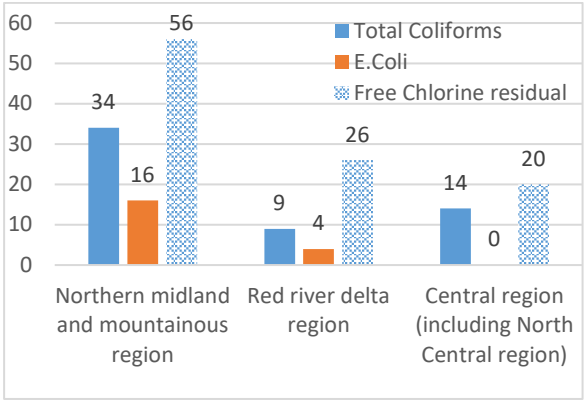

Fig. 2. Number of samples do not meet the allowable standards by regions in 2021 campaign

Table 2 shows that all 3 regions, values of free chlorine residual have the same maximum value range and about 2 times higher than the allowable standards (QCVN 01-1:2018/BYT). For total coliforms and *E.coli*, the maximum number greater than the allowed upper limit 6-8 and 16-17 times, respectively. Regarding to the maximum value of free chlorine residual, total coliforms and *E coli*, it should be noted that these are the range of values for all analysed water samples. In fact, supply water samples with high free chlorine residual will normally have very low or undetectable total coliforms and *E.coli* parameters except for a few special cases. In addition, in the Northern Midlands and Mountains, there are high number of water samples that do not meet the water quality criteria, especially for microbiological indicators (total coliforms, *E. coli*, free chlorine residual (Fig. 2).

Apart from *E.coli* and total coliforms, *Ps. Aeruginosa* and *Staphylococcus aureus* are also 02 microbial indicators for clean water used for domestic purposes specified in QCVN 01-1:2018/BYT.

As can be seen in Table 3, similar to total coliforms and *E.coli*, in 2021 campaign, out of 11 provinces where *Ps. aeruginosa* were detected in water samples, up to 6 provinces had low free chlorine residual content (< 0.06 mg/L) or even not disinfected water (Lai Chau, Da Nang). There were 2 samples that *Staphylococcus Aureus* (in Tuyen Quang) was detected also having low free chlorine residual content. However, there are still water samples in 5 provinces/cities showing results for detecting *Ps. aeruginosa* but the free chlorine residual content reaches QCVN 01-1:2018/BYT or higher than the maximum permitted limit. Since *Ps. aeruginosa* is an easy microorganism to survive in the environment (can survive up to 6 months in an environment without nutrients and without oxygen), therefore, along with the existence of total coliforms, microorganisms can be grown into biofilms in water distribution pipes and are a favourable environment for microorganisms to exist and grow. This can be a reasonable explanation for the presence of microbial parameters in the finished water despite the high free chlorine residual content.

Table 3. The values of total coliforms and *Ps. Aeruginosa* identified in supply water samples in 2021 campaign

No.	Provinces/ cities	Total coliforms (CFU/100mL)			*Ps. aeruginosa* (CFU/100mL)		
		Min	Max	Mean	Min	Max	Mean
1.	Ha Giang	ND	**16**	ND	ND	ND	ND
2.	Lao Cai	ND	**9**	ND	ND	**16**	ND
3.	Cao Bang	ND	**18**	**5**	ND	**21**	ND
4.	Tuyen Quang	ND	**23**	ND	ND	ND	ND
5.	Yen Bai	ND	**23**	ND	ND	**37**	ND
6.	Thai Nguyen	ND	**18**	ND	ND	**18**	ND
7.	Phu Tho	ND	**18**	ND	ND	ND	ND
8.	Lai Chau	ND	**23**	**16**	0	**9**	ND
9.	Ha Noi	ND	0	ND	ND	**9**	ND
10.	Vinh Phuc	ND	**18**	ND	ND	**18**	ND
11.	Hung Yen	ND	**18**	ND	ND	**21**	ND
12.	Ha Nam	ND	**6**	ND	ND	ND	ND
13.	Nam Dinh	ND	**18**	ND	ND	ND	ND
14.	Thanh Hoa	ND	ND	ND	ND	**18**	ND
15.	Da Nang	ND	**18**	3	ND	ND	ND
16.	Thua Thien Hue	ND	ND	ND	ND	**15**	ND
17.	Quang Binh	ND	ND	ND	ND	**21**	ND
18.	Quang Tri	ND	**23**	ND	ND	**48**	ND
QCVN 01-1:2018/BYT		**<3**			**<1**		

Note:
LOA of Total coliforms: less than 3.0 CFU/100mL
LOA of Ps. aeruginosa: less than 1.0 CFU/100mL

Compared with 2020 campaign, the level of total coliforms infection and *Ps. aeruginosa* tended to increase both in the number of provinces with samples exceeding the allowed regulations and the level of excess although not significant. Number of provinces and cities with samples exceeding the standard. For total coliforms is 14/32 and 21/45; *Ps. aeruginosa* is 12/32 and 15/45 respectively in 2021 and 2020 (at maximum value).

3.3. The Correlation Between Free Chlorine Residual Content and Total Coliforms, E. coli

The Fig. 3 showed the correlation between free chlorine residual and microbiological parameters in supply water of centralized supply water companies in Vietnam in the period of 2020-2022. To calculate the correlation the outliers have been already removed/taken out. These outliers include water samples with a satisfactory free chlorine residual content but having substandard microbiological parameters. There are other reasons may affect the content of microbiological parameters. The correlation between free chlorine residual and total coliforms was weak negative correlation, strongest one; between free chlorine residual and *E. coli* was found very weak negative correlation. The results of the Pearson correlation coefficient showed that there was a much larger negative correlation between total coliforms and *E.coli*. The Pearson correlation coefficients of both are very low, -0.282 and -0.091, respectively. However, these found higher than the Pearson correlation coefficients of correlation coefficients and total coliforms, *E.coli* in rural areas of Poldasht, Iran [11].

The impact level of free chlorine residual on total coliforms found greater than that on *E. coli.* The correlation between free chlorine residual and both parameters are negative which means that the decrease of free chlorine residual content can lead to the increase of presence of total coliforms and *E.coli*. However, free chlorine residual has little effect on microorganisms.

All taken water samples having the concentration of free chlorine residual that meet the permitted limit do not impact the variation of microbiological parameters. In addition, most of water samples that have influence on the change of microorganism content have free chlorine residual content being lower than 0.2 mg/L. A greater removal efficiency of both microbiological parameters was achieved at higher residual chlorine concentration. This may be due to the high residual chlorine content causing inactivation in the bacterial cells. At the same time, there are also many hypotheses that substances with strong oxidizing capacity such as residual chlorine have a higher oxidizing capacity to inactivate bacteria [12].

In 2022 campaign, the similar results with previous campaigns have been obtained according to the supply water quality analysis in 28 provinces/cities. As can be seen from Fig. 3, the relationship of free chlorine residual and total coliforms and *E.coli* were observed non-linear with very low regression coefficients. The results found were different from some other studies with higher R^2 values [13, 14].

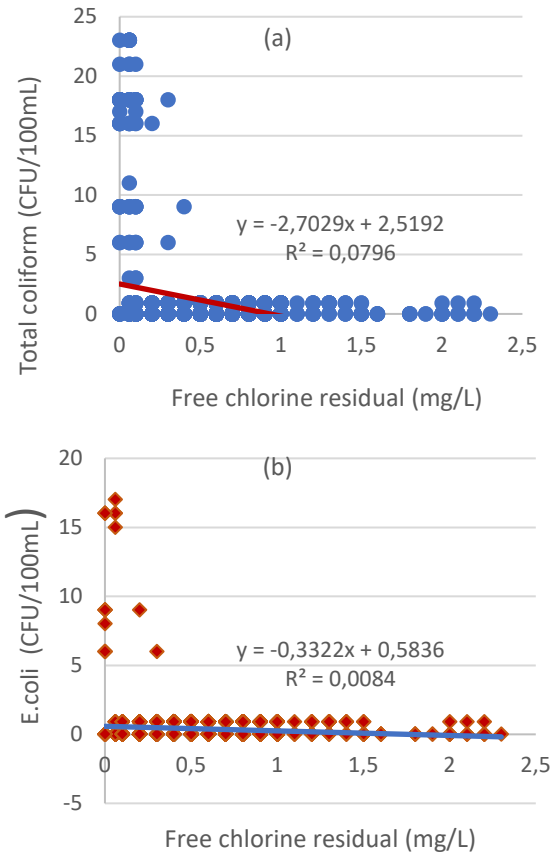

Fig. 3. The relationship between residual chlorine content and total coliforms (a) and *E. coli* (b) of 2020-2022 campaigns

In some previous studies the correlation between free chlorine residual and diseases related to microbiological contamination was found stronger. The correlation between water quality focusing on free residual chlorine (FRC), turbidity (T), total coliforms (TC), *E. coli* (EC) parameters and the status of several related diseases such as acute diarrheal disease (ADD) and hepatitis A in Brazil were assessed during 2012 and 2017. Results of evaluating correlations (r, $p \leq 0.2$) between the different variables that had satisfactory linear regression values ($p \leq 0.2$) in relative complaints index and non-compliance index for free residual chlorine obtained r=0.57737 with R^2=0.3334 [15]. Total of 180 water samples in the water supply network of Azogues city, Ecuador during the 6 months of the Covid 19 epidemics were analysed and evaluated the rate of free chlorine residual decomposition in the network using EPANET hydraulic models and measured data. The findings showed that the free chlorine residual decomposition

rate was 3.7 /day and the reaction rate constant on the water distribution pipe wall was 0.066 m/day. In addition, the reaction rate constant is strongly positively correlated with flow velocity and not much correlated with the pipeline diameter [16].

Analysis of 422 drinking water samples randomly taken at the tap in Kisii, Kenya showed that 31.2% of the samples had free chlorine residual content higher than 0.2 mg/L, however 39.3 and 17.5% of the samples were contaminated with total coliforms, and *E. coli*, respectively. The study also showed a strong correlation between bacterial infection and temperature, free chlorine residual in water samples [17]. A study of the status of opportunistic pathogens (OPs) and health risks associated with four water companies and distribution systems in western China found a negative correlation between OPs and free chlorine residual [18].

In further study, the impact of the presence of ammonium and some other components in water supply on the chlorination process and the formation of some by-products that pose health risks will be evaluated. In addition, the influence of some other factors like the water distribution system, pipe line length, pipe line status, pH and temperature on free chlorine residual also needs to be considered and investigated.

4. Conclusion

The results of assessment of free chlorine residual of 1612 water samples of water supply companies nationwide in 3 campaigns in 2020, 2021 and 2022 showed that about 20% of water samples did not meet both free chlorine residual and total coliforms criteria, except for 2021, only 10% of samples did not meet the requirements for total coliforms parameters. There is no difference between water supply companies in different regions, however, most of the water samples that fail are from companies with a capacity of less than 1000 m^3/day. There was no significant difference between regions in terms of free chlorine residual levels and total coliforms and *E.coli* contamination in the analysed supply water samples.

The evaluation of the correlation between free chlorine residual and the microbiological criteria of total coliforms and *E.coli* showed that free chlorine residual was negatively correlated with both parameters but not linearly with low regression coefficient R^2. Calculation of Pearson coefficient shows low and negative correlation. However, the correlation of free chlorine residual with total coliforms is much higher than that of *E.coli*, -0,282 and -0,091, respectively.

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