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Freezing process – a new approach for nitrate removal from drinking water

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ABSTRACT

Nowadays a considerable number of people all around the world are affected by severe water scarcity. Nitrate and nitrite are widespread in the environment. Nitrate is the most common chemical contaminant in the world's groundwater aquifers. Conventional water treatment processes used at municipal water treatment plants such as coagulation, sedimentation, filtration, and chlorination, and even some of the most common tertiary processes like adsorption are not effective for nitrate removal. Among the novel techniques for nitrogen removal from aqueous solutions, the freezing-melting process is of the most convenient and effortless especially for a nonexpert person. The main objective of this study was to evaluate the performance of nitrate removal from aqueous solutions through freezing-melting process. Eight different nitrate concentrations ranged from 50 up to 250 mg/L were used in this study. Nitrate content of these solutions was reduced to its drinking water standard just by a single run or right after the first repetition of the process.

Keywords: Nitrate; Freezing process; Drinking water; Crystallization

1. Introduction

Nowadays a considerable number of people all around the world are affected by severe water scarcity. Today, 1.1 billion people live without clean drinking water. Water is scarce in numerous regions of the world and many suffer from perpetual shortages. According to estimates, the world will only have 60% of the water which needs by the year 2030 without significant global policy changes [1,2]. Purification and treatment of water is a fast-growing area of interest among the environmental health studies [3–13].

Nitrate and nitrite are widespread chemicals in the environment. They are naturally produced by the oxidation of nitrogen by microorganisms and, to a lesser extent, by lightning. The most common sources of these substances are human activities, including agricultural activities, wastewater treatment, and discharges from industrial processes, motor vehicles, and application of fertilizers [14]. Nitrate is the most common chemical contaminant in the world's groundwater aquifers [15].

A number of guidelines and technical reports [16] were reviewed and assessed all identified health risks associated with nitrate and nitrite in drinking water. These health risks include a wide range of adverse effects from methemoglobinemia in bottle-fed infants to incremental lifetime cancer risk for the entire society [14,16,17].

Nitrate is a stable and highly soluble ion with low potential for coprecipitation or adsorption. Thus, conventional treatment technologies cannot be used for its removal. Conventional water treatment processes used at municipal water treatment plants are coagulation, sedimentation, filtration, and chlorination, and even some of the most common tertiary processes like conventional adsorption are not effective and suitable for nitrate removal [14,16,18]. There are a number of studies reviewing various techniques for nitrate removal from drinking water in terms of their effectiveness, simplicity of operation, and costs [3,19]. Effective nitrate removal technologies to be used for municipal water supplies include ion exchange, biological denitrification, membrane bioreactors, reverse osmosis, chemical denitrification,

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and electrodialysis [3,14,16]. Some of these methods could be used for simultaneous removal of multiple pollutants from aqueous solutions as well [20–23]. Regarding lack of facilities available in rural and slum areas, most of these treatment methods are not applicable or they are too expensive to provide public potable water for considerably populated communities. However, some household water purification methods could be used as low-cost interventions to mitigate the water quality problems [24–27]. Among the novel techniques for nitrogen removal from aqueous solutions, the freezing–melting process is of the most convenient and effortless ones, especially for a nonexpert person [28]. Natural and manmade formed layers of ice could be removed from the enriched nitrate solution. This process is an old method of brine water desalination, and could be the easiest and the most accessible method to produce a more aesthetically acceptable and healthy potable water in rural and slum areas. This process could be done in two ways, with direct contact of the coolant and the solution (direct freezing) or with indirect contact of the coolant and the solution (indirect freezing). There are some excellent studies on application of this method both individually and in combination with other desalination methods [29–31].

The main objective of this study was to evaluate freezing–melting process performance for the removal of nitrate from aqueous solutions. Nitrate solutions used in this study involved eight different concentrations ranged from 50 up to 250 mg/L (50, 75, 100, 125, 150, 175, 200, and 250), chosen based on reports [4,14,16] on this pollutant in aquifers around the world.

2. Material and methods

2.1. Reagents

All chemicals were of analytical reagent grade. This stage was done based on the standard operating procedure for nitrate measurement [32]. Standard stock solutions of 1,000 mg/L of NO_3-NO_3 were prepared by dissolving 1.63 g of formerly dried (in an oven for 24 h at 105°C) potassium nitrate (KNO_3; Merck, Germany) in double-distilled water and diluting it to volume of 1,000 mL. Working solutions were prepared by diluting the nitrate stock solutions. Series of the samples and standard solutions were prepared by pipetting suitable volumes of nitrate solution (1,000, 500, and 250 mg/L) using a Scienceware® Roxy M™ Repeating Pipettor. Fresh solutions were prepared prior to each experiment. The solutions were prepared in 250 mL Erlenmeyer Flasks and subsequently poured into polypropylene beakers to be examined discretely.

2.2. Electrical conductivity and total dissolved solids measurement

All of the samples were examined for determination of electrical conductivity (EC) and total dissolved solids (TDS) in accordance with Standard Methods for the Examination of Water and Wastewater [33].

2.3. Crystallization

The same volume of the samples was poured into same identical flasks followed by putting them inside a Pars-PAMCO-FRZNF170 freezer with an energy consumption of 0.1 kW/h to be crystallized by indirect freezing. Samples were continuously controlled for the amount of the produced ice. By passing the retention times of 30 min up to 2 h, samples with different amounts of ice recovered from the freezer so that the layers and crystals of ice could be extracted from the solution. 10 mL of chilled (0°C) double-distilled water was used in order to drain the residual nitrate polluted water from the produced ice before letting them be melted at the room temperature in a capped jar to prevent dry deposition and adsorption of gases and particles.

2.4. EC and TDS remeasurement

Residual solution from all of the samples along with the water produced from melted ice were examined for determination of EC and TDS using methods ASTM D1125-14 and 2,540 C and in accordance with American Society for Testing and Materials Standard Methods [34] and Standard Methods for the Examination of Water and Wastewater [33].

2.5. Measurement of nitrate concentration

The concentration of nitrate in both melted ice and residual solution were examined in compliance with method 4500-NO_3 B. of Methods for the Examination of Water and Wastewater [33] by means of a DR 5000™ UV-Vis Spectrophotometer complying with the device manual [35]. All the measurements were repeated three times to ensure data validity.

The performance of the freezing process for reduction of nitrate and TDS concentration and EC value were expressed using the following equation:

$$\text{Removal }\% = \frac{P_1 - P_2}{P_1} \times 100 \tag{1}$$

where P_1: the initial value of the parameter; P_2: the final value of the parameter; and Removal %: removal percentage as a performance indicator.

3. Results and discussion

3.1. Reduction of nitrate concentration due to freezing

It is shown by the following figures that how freezing in different proportional volumes of freezing can separate ice crystals from the nitrate containing water and reduce the nitrate concentration in melted ice compared with the solution.

The initial nitrate concentrations were 50, 75, 100, 125, 150, 175, 200, and 250 mg/L. Figs. 1–8 show the effects of various proportional volumes of freezing (from 10% up to 50% of the initial solution volume) on the reduction of nitrate concentration from several solutions with different initial concentrations of it.

It was shown that for solutions with a nitrate concentration of 75, 100, and 125 mg/L, the removal performances were about 60% up to 70% when the freezing took place for 10% up to 15% of the total volume (shown by Figs. 1–4). However, an identical performance of nitrate removal (60%) for a solution

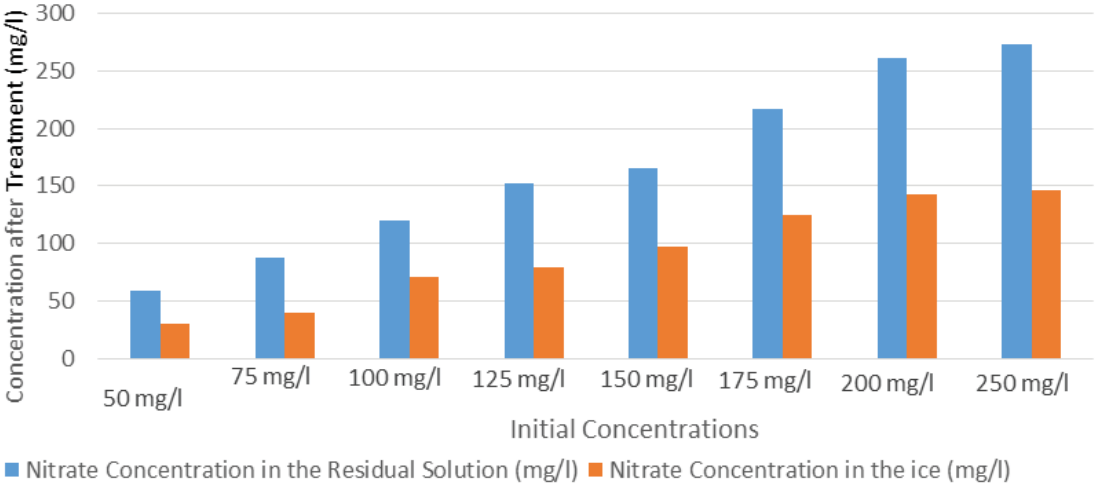


Fig. 1. Compression of nitrate concentration in solid and liquid phases.

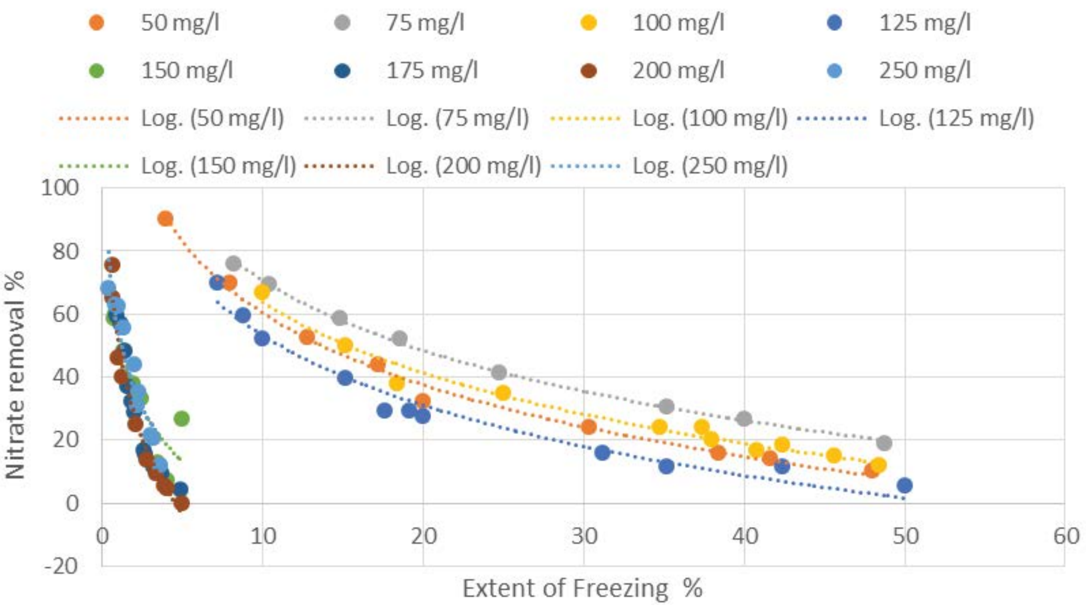
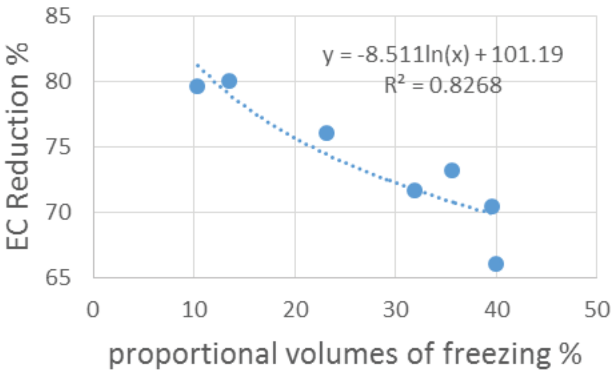


Fig. 2. The effect of proportional volumes of freezing on nitrate removal (The initial concentrations of 50 up to 250 mg/L).



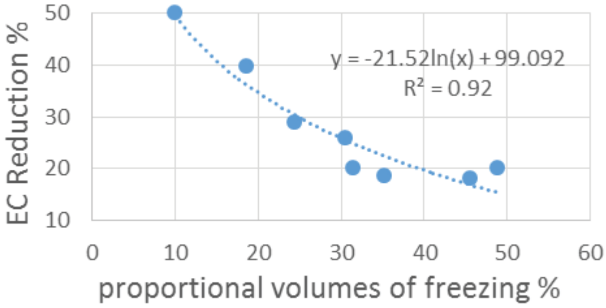


Fig. 3. The effect of proportional volumes of freezing on EC reduction for an initial EC of 250 μmho/cm.

Fig. 4. The effect of proportional volumes of freezing on EC reduction for an initial EC of 500 μmho/cm.

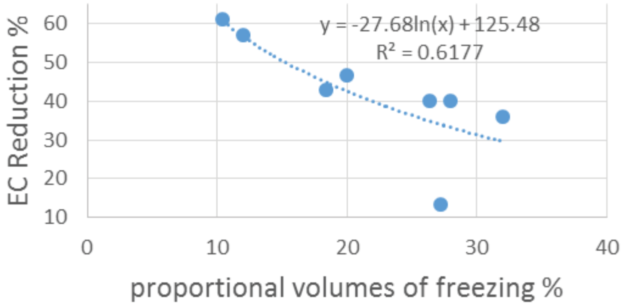


Fig. 5. The effect of proportional volumes of freezing on EC reduction for an initial EC of 750 µmho/cm.

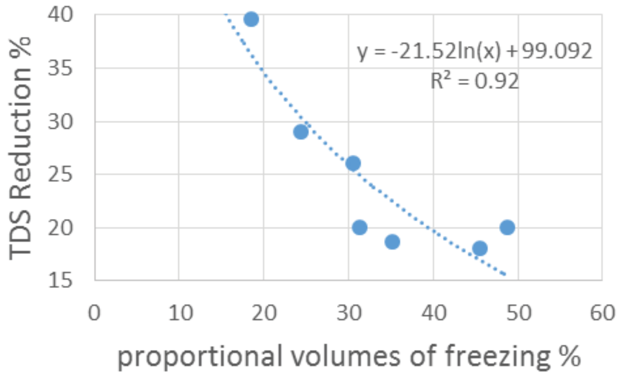


Fig. 6. The effect of proportional volumes of freezing on TDS reduction for an initial concentration of 250 mg/L.

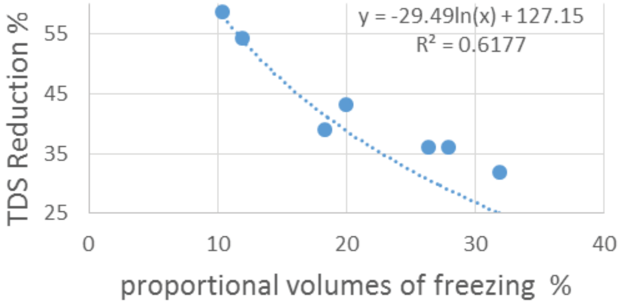


Fig. 7. The effect of proportional volumes of freezing on TDS reduction for an initial concentration of 352 mg/L.

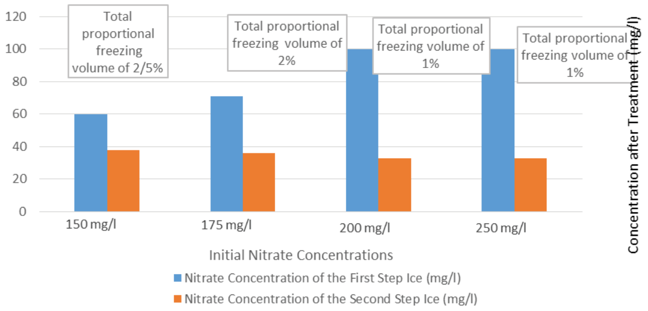


Fig. 8. Nitrate concentration in the first and second stage's ice (with a 10% proportional volume of the initial freezing).

with greater nitrate concentration (i.e., 150, 175, 200, and 250 mg/L) was not enough to meet the standard requirements. Hence the freezing and melting process implemented in two stages to pass the standard level (20, 35, 35, and 38 mg/L of nitrate, respectively, against the maximum contaminant level of 50 mg/L). It was also found that nitrate concentration in liquid phase always tends to be more than the frozen part.

When salt water freezes, ice crystals are essentially salt-free. Pure water crystallization takes place prior to crystallization of dissolved salts, however, as the freezing extends, salts crystallization is also becoming more and more promoted as the result of solubility reduction due to the thermodynamic limitations of dissolution. Therefore, it is noteworthy to mention the importance of time and temperature control in order to minimize salts crystallization and maximizing pure water production. In other words, ice production is a function of temperature and freezing time. By advancing the elapsed time and/or reduction of freezing temperature, more amount of ice could be produced, however, this rise in amount is in coincidence with a fall in quality.

Nitrate concentration reduction (as the percent of the initial concentration) was plotted against the proportional volumes of freezing. Different percentages of nitrate removal were plotted against the associated proportional volumes of freezing for all the initial concentrations of nitrate and at last the resulted trend line for all these charts seemed to be similar to each other. It is worth mentioning that for the initial nitrate concentrations of greater than 125 mg/L, the freezing and melting process should be done again to pass the standard requirements and therefore results from the first and the second step are also compared with each other in the figures. For all the double-staged freezing and melting experiments, the extent of the first stage freezing was about 10%, water from this stage melted ice undertook a wide range of proportional volumes of freezing to scrutinize the effect of different levels of freezing on nitrate removal rate. There was some negative coefficient logarithmic relationship between removals and the proportional volumes of freezing. This fact supports the previous paragraph's statements.

EC and TDS are also plotted against the proportional volumes of freezing and the resulted charts for these two parameters were identical for each initial concentration. Reduction of EC and TDS because of water purification due to freezing had a logarithmic relationship with the proportional volumes of freezing, however, this relationship was not like the nitrate removal ones. This inconsistency was interpreted as a result of the uneven tendency of the ions to be crystallized and to be diffused in the solid phase and also as a result of differences in electroconductive characteristics of different ions.

Fig. 1 compares nitrate concentration of the residual solution and the solid phase for different initial nitrate concentrations following the first stage of the freezing–melting process.

Fig. 2 illustrates that how variations of freezing amount affect nitrate removal. For the initial nitrate concentrations of 50 up to 250 mg/L when the extent of freezing is less than 30%, nitrate concentration could be reduced more than 60%.

Fig. 3 illustrates that how variations of freezing amount affect EC reduction for an aqueous solution with an initial EC of 250 µmho/cm. This plot shows the reverse relationship between these two variables.

Fig. 4 illustrates that how variations of freezing amount affect EC reduction for an aqueous solution with an initial EC of 500 µmho/cm. This plot shows the reverse relationship between these two variables.

Fig. 5 illustrates that how variations of freezing amount affect EC reduction for an aqueous solution with an initial EC of 750 µmho/cm. This plot shows the reverse relationship between these two variables.

Fig. 6 illustrates that how variations of freezing amount affect TDS reduction for an aqueous solution with an initial TDS of 250 mg/L. This plot shows the reverse relationship between these two variables.

Fig. 7 illustrates that how variations of freezing amount affect TDS reduction for an aqueous solution with an initial TDS of 352 mg/L. This plot shows the reverse relationship between these two variables.

Fig. 8 illustrates the additive effects of each freezing step (first and second) on nitrate concentrations for several initial concentrations. The first steps proportional volume of freezing was 10%.

Different normality tests (Anderson–Darling, Lilliefors, Jarque–Bera, and Shapiro–Wilk) were used to determine the appropriate correlation method. Since the data did not follow a normal distribution, Spearman correlation method has been chosen. The two-sample Kolmogorov–Smirnov test is also used to make an illustrative comparison between the variables' distribution. Identical distribution trends were inferred as a result of variables' correspondence. The following charts and tables exhibit results of these statistical analyses.

Table 1 summarizes the *p*-values for different normality tests. Since the *p*-values are lesser than $\alpha = 0.05$, except for Jarque–Bera and Lilliefors *p*-values for nitrate reduction, therefore the null hypothesis (sample is derived from a population with normal distribution) could be rejected.

Probability–Probability (P-P) plots were also used to demonstrate these findings by plotting the normal cumulative distribution against the empirical cumulative distribution.

Fig. 9 compares EC reduction distribution with an absolute normal distribution by plotting the empirical cumulative distribution against the normal cumulative distribution. As the points in EC reduction P-P plot don't lie on the absolute normality line $y = x$, then this distribution is not a normal distribution.

Fig. 10 compares TDS reduction distribution with an absolute normal distribution by plotting the empirical cumulative distribution against the normal cumulative distribution. As the points in TDS reduction P–P plot don't lie on the absolute normality line $y = x$, then this distribution is not a normal distribution.

Table 1
Summary of normality test results

Variable/test	Shapiro–Wilk	Anderson–Darling	Lilliefors	Jarque–Bera
EC reduction	0.000	<0.0001	0.000	0.028
TDS reduction	0.000	<0.0001	0.002	0.027
Nitrate reduction	0.005	0.003	0.087	0.088

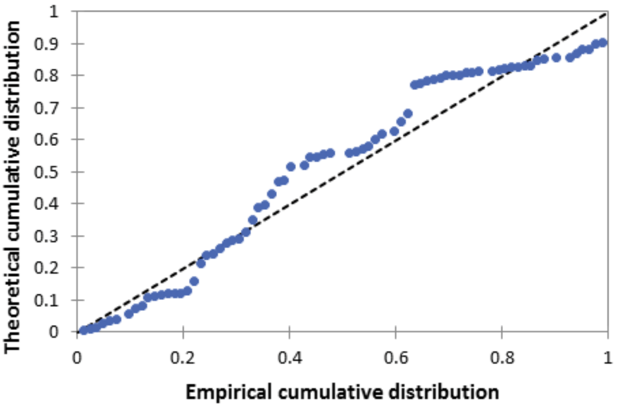


Fig. 9. Normal P-P plot for EC reduction.

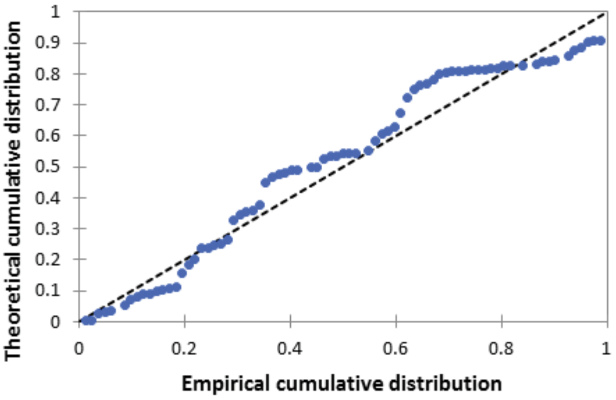


Fig. 10. Normal P-P plot for TDS reduction.

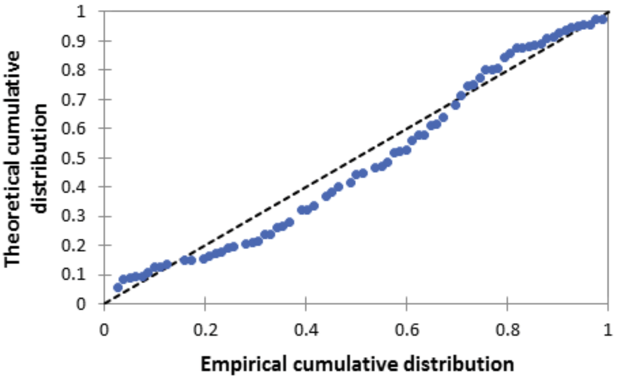


Fig. 11. Normal P-P plot for nitrate reduction.

Fig. 11 compares nitrate removal distribution with an absolute normal distribution by plotting the empirical cumulative distribution against the normal cumulative distribution. As the points in TDS reduction P–P plot don't lie on the absolute normality line $y = x$, then this distribution is not a normal distribution.

The two-sample Kolmogorov–Smirnov test is used for investigation of the goodness of fit between different variables. With regard to small but contrasting degrees of freedom (*D*) values, it was revealed that there were strong relationships between the variables, however, these strengths were not identical. Two samples comparison of distributions were also used in order to illustrate these relationships.

Table 2 summarizes degrees of freedom (*D*), asymptotic *p*-value, and predefined significance level (alpha) for the two-sample Kolmogorov–Smirnov test between TDS and EC reduction. This considerable *p*-value as well as the small degrees of freedom are showing that there were some relationships between the variables.

Table 3 summarizes degrees of freedom (*D*), asymptotic *p*-value, and predefined significance level (alpha) for the two-sample Kolmogorov–Smirnov test between TDS reduction and nitrate removal. This inconsiderable *p*-value is showing that there were not strong relationships between the variables. On another hand the small degrees of freedom shows that these variables variations were comparable.

Table 4 summarizes degrees of freedom (*D*), asymptotic *p*-value, and predefined significance level (alpha) for the two-sample Kolmogorov–Smirnov test between EC reduction and nitrate removal. This inconsiderable *p*-value is showing that there were not strong relationships between the variables. On another hand the small degrees of freedom shows that these variables variations were comparable.

Fig. 12 compares cumulative distributions of TDS and EC reductions. This plot shows that these two distributions goodness of fit is approximately high.

Fig. 13 compares cumulative distributions of nitrate removal and TDS reduction. This plot shows that these two distributions goodness of fit is low, however, their variations were comparable.

Fig. 14 compares cumulative distributions of nitrate removal and EC reduction. This plot shows that these two distributions goodness of fit is low, however, their variations were comparable.

Table 2
Two-sample Kolmogorov–Smirnov test for EC and TDS reduction

D	0.159
Asymptotic *p*-value	0.254
Alpha	0.05

Table 3
Two-sample Kolmogorov–Smirnov test for nitrate and TDS reduction

D	0.439
Asymptotic *p*-value	<0.0001
Alpha	0.05

Table 4
Two-sample Kolmogorov–Smirnov test for nitrate and EC reduction

D	0.476
Asymptotic *p*-value	<0.0001
Alpha	0.05

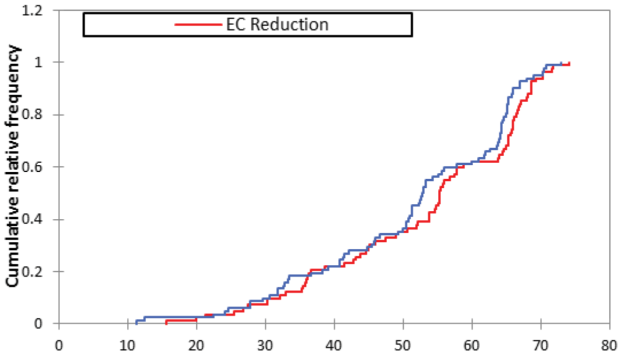


Fig. 12. Comparison of distributions (EC reduction/TDS reduction).

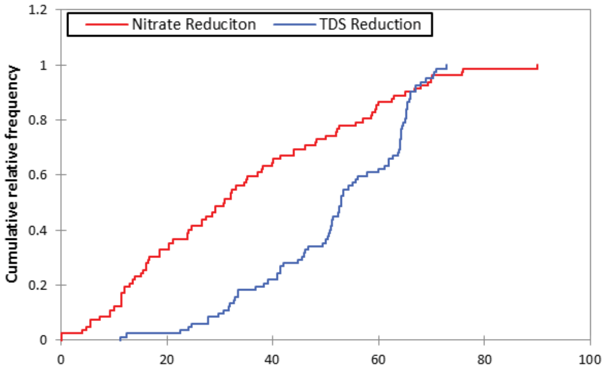


Fig. 13. Comparison of distributions (nitrate reduction/TDS reduction).

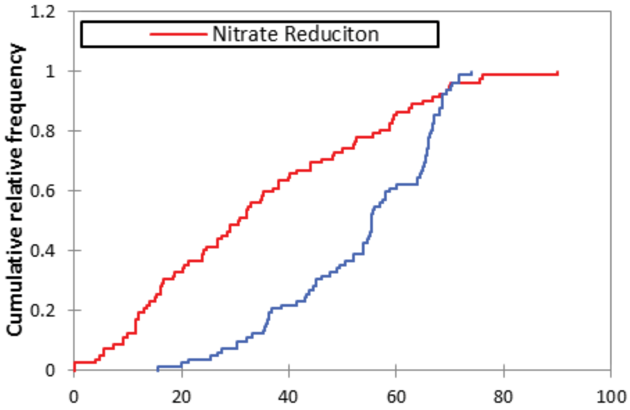


Fig. 14. Comparison of distributions (nitrate reduction/EC reduction).

Spearman correlation method is used to analyze inter-relationships of the variables. These values could vary between –1 and +1 in respect to the direction of correlation (reverse or direct), while a value of "0" reveals that there was no linear correlation.

Table 5 summarizes Spearman correlation coefficients among nitrate removal, EC reduction, and TDS reduction. Since these values are close to +1, it could be considered that there were some strong inter-relationships between these variables.

Table 5
Correlation matrix (Spearman)

Variables	EC reduction	TDS reduction	Nitrate reduction
EC reduction	1	0.933	0.782
TDS reduction	0.933	1	0.705
Nitrate reduction	0.782	0.705	1

Table 6 summarizes Spearman's correlation test's *p*-values for the aforementioned variables. Since these values are around 0, it could be considered that Spearman's correlation test's results have a high probability of trueness or goodness of fit.

Table 7 summarizes Spearman's correlation test's coefficients of determination (R^2) for the aforementioned variables. Since these values are considered, it could be considered that Spearman's correlation test results have a high probability of trueness or goodness of fit.

The results show that there is a strong correlation between EC and TDS. This relationship was much stronger than those belonging to TDS/nitrate and EC/nitrate. This inconsistency interpreted as a result of the uneven tendency of the ions being crystallized and diffused in the solid phase and also as a result of differences in electroconductive characteristics of different ions. Since double-distilled water used to prepare the solutions could not include a significant amount of impurities, therefore, the potassium ions are the probable cause of this contrariety.

These results are in agreement with correlative desalination literature, as a reverse relationship between the proportional volumes of freezing and the efficiency of desalination process was observed [6,36,37]. This process requires minimum capital costs with regard to other removal and treatment processes. It also does not add any other substances to water unlike ion-exchange or biological methods, moreover, there were no resin capacity reduction or increased chlorine consumption. This method is a quick and simple technique in comparison with biological methods [38] and does not require any high-priced nitrate selective resins or the use of graphene nano sheets which should be regenerated frequently [12,39]. This process, unlike catalytic reduction process, does not require high capital costs or expertized professionals for operation [11,40]. It is noteworthy that all the processes have their own advantages and disadvantages, therefore, some should select the best available technology based on practical, technical, and local limitations, and economic conditions. In some parts of Iran, nitrate is relatively high in groundwater sources and therefore freezing process is recommended for nitrate removal [41].

Table 6
Correlation test *p*-values

Variables	EC reduction	TDS reduction	Nitrate reduction
EC reduction	0	0.000	0.000
TDS reduction	<0.0001	0	<0.0001
Nitrate reduction	<0.0001	<0.0001	0

Table 7
Correlation test coefficients of determination (Spearman)

Variables	EC reduction	TDS reduction	Nitrate reduction
EC reduction	1	0.871	0.611
TDS reduction	0.871	1	0.497
Nitrate reduction	0.611	0.497	1

4. Conclusion

This manuscript was aimed to evaluate freezing and melting process performance for production of high-quality water. Results from this study confirmed that the process has the potential of nitrate removal from drinking water. In spite of small water production, this process is of interest with regard to its low capital expenditure (as nowadays there is a refrigerator in almost every home) and simplicity which makes it really convenient and applicable for a nonexpert person living in a developing country in order to produce a convenient supply of healthy potable water specially for infants. It can be concluded that freezing–melting process reduces the nitrate concentration to less than its standard level (50 mg/L as NO_3) from drinking water with nitrate concentrations of equal or less than 125 mg/L right after a single-stage freeze and melt. However, for more concentrated solutions, a repetition of the process is required. This method has its limitations especially for solutions with high nitrate concentrations and needs more improvements.

Acknowledgments

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