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Establishing the optimal condition for nutrient recovery from domestic wastewater using the freeze concentration method

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ABSTRACT

The freezing concentration method is one of the potential techniques for recovering nutrients from wastewater. In this study, the method of freeze concentration was studied to establish its optimal conditions in recovering nitrate-nitrogen and phosphate nutrients from domestic wastewater. Water in the form of an ice crystal block is produced and leaves behind a solution with a higher concentration. The effects of coolant temperature from −10 to −80 °C, freezing time from 1 to 8 h, and energy consumption on nutrient recovery were investigated. The optimal conditions were found at a coolant temperature of −20 °C, freezing time of 7 h, and energy consumption of 0.197 kWh/L that resulted in the highest nitrate-nitrogen and phosphate nutrient recovery values of 1.114 and 4.667, respectively, at the inlet of anaerobic digester 1; 1.325 and 4.975, respectively, at the outlet of anaerobic digester 1; 1.099 and 4.859, respectively, at the inlet of anaerobic digester 2; 1.132 and 4.755, respectively, at the outlet of anaerobic digester 2; and for gravel filter at the outlet the values were 1.111 and 4.861, respectively. The recovered nutrients can be used as biofertilizers.

Key words: biofertilizer, freeze concentration, resource recovery, wastewater

HIGHLIGHTS

- Effects of coolant temperature at −10, −20, −30, −40, −50, −60, −70, and −80 °C on the operation of the freezing concentration technique.
- Effects of freezing time at 1,2,3,4,5,6,7 and 8 h on the operation of the freezing concentration method.
- Low energy consumption on the operation of the freezing concentration technique.
- High amount of nitrate-nitrogen and phosphate nutrients recovered from domestic wastewater.

1. INTRODUCTION

Pollution of water sources with excessive nutrient loads, most commonly phosphate and nitrogen, is a major environmental problem faced by many African countries. The water pollution problem is often associated with the rampant discharge of untreated or partially treated domestic (urban) wastewater into the environment. Although domestic wastewater contains nutrients (i.e., nitrogen, phosphorus, and potassium) that can be used in agriculture, its recovery and reuse are still a challenge. Some of the potential methods for recovering nutrients from wastewater are not only costly but also introduce a second pollutant into the waste stream. Potential methods such as struvite precipitation, ion exchange, electrochemical, algae, and freeze concentration have all been researched, but each has its own setbacks and drawbacks (Mavhungu *et al.* 2021).

Struvite precipitation is a method of recovering nutrients such as phosphorus used in crop production as a fertilizer. However, this technique uses chemicals that add a second pollutant to the environment (Sena *et al.* 2021).

Algae have been used to produce biofertilizers since they grow very well in wastewater while absorbing nitrogen and phosphorus nutrients which are important for crop production (Huo *et al.* 2020). However, there are numerous challenges and technical flaws, including the appearance of a second pollutant in wastewater, the high price of algal compounds, algal pollution, low availability of algae, potential cyanobacteria threats to the environment, and high-water consumption as highlighted by Zou *et al.* (2020).

The ion exchange method is one of the methods that can remove and recover phosphorus to form fertilizer. However, the presence of competing anions such as sulfates in wastewater provides a major bottleneck in limiting the selection of phosphorus (Ownby *et al.* 2021).

However, freeze concentration is reported as a promising technique for recovering nutrients from wastewater. It is also a clean technology with no secondary pollutant production, no chemical addition, and low equipment erosion. The freezing concentration technique is a physical method where a solution is concentrated by freezing out water content in the form of ice crystals (Samsuri *et al.* 2015). The freezing method because of its low latent heat of fusion consumes less energy compared to the evaporation method (Moharramzadeh *et al.* 2021). A saturated liquid phase and a solid crystalline phase are generated as a result of the freezing concentration technique (Lu *et al.* 2017).

This technology has numerous advantages over other procedures, such as a high rate of recovery, simultaneously recovering of both water and valuable minerals, and the absence of any additional supplemental information (Lu *et al.* 2017).

The freezing concentration technique has been used in wastewater treatment for solutions where the solubility of the solute is substantially dependent on temperature (Ab Hamid & Jami 2019). Researchers have previously employed the freezing concentration technique to remediate wastewater from the pharmaceutical, chemical, fluoride removal, and chromium(VI) removal industries (Ab Hamid & Jami 2019). However, limited studies have been done on establishing the optimal operating conditions for the freeze concentration method to recover nitrate-nitrogen and phosphate nutrients from domestic wastewater processed in an anaerobic digester.

As a result, the goal of this research was to find the optimal conditions for the freeze concentration method to recover nutrients from domestic wastewater processed in an anaerobic digester. The freezing concentration performance was evaluated using nitrate-nitrogen and phosphate nutrient recovery values. At various coolant temperatures, freezing time, and energy consumption, the performance of freezing concentration was examined. Changes in nutrient recovery were used to investigate the consequences of these various operating conditions. The recovered nutrients will be used as a fertilizer for crop production in agriculture.

2. MATERIALS AND METHODS

2.1. Wastewater samples

The feed water for the freezing concentration trials in this investigation was real domestic wastewater effluent from two anaerobic biodigester and gravel filter treatment stages put in a sequence located at The Nelson Mandela African Institution of Science and Technology in Arusha, Tanzania. The wastewater for the tests was collected from the inlet and outlet of each treatment plant as shown in Figure 1.

Domestic wastewater is a complicated mixture of accumulated chemicals and contaminants in water. Due to varying flow rates induced by water usage and precipitation, the quality and composition of the wastewater also vary on a regular basis.

2.2. Experimental setup

The apparatus setup for the freeze concentration process is shown in Figure 2. Thermal scientific freezer model numbers 713CD and 813CV were used for temperatures of −10 to −40 °C and −50 to −80 °C, respectively. This freezer provides power and control temperature. The average energy consumption of the coolant was 8.1 and 11.6 kWh/day for temperatures of −10 to −40 °C and −50 to −80 °C, respectively.

2.3. Experimental procedure

At the inlet and outlet of each anaerobic biodigester and a gravel filter treatment plant, wastewater samples were collected. Samples were filtered by using Cellulose Nitrate Filter, pore size of 0.45 μmm before being placed in the sample vessel during the experiment. Following the sample filtering, 400 mL of wastewater was added to the sample plastic vessel for the freezing concentration process. Vessels containing the sample solution were placed in the freezer and the operating parameters were adjusted as needed. The temperature of the sample was 25.43 °C ± 0.73 °C at the start of the freezing concentration procedure. The coolant temperature and time tested in this study are shown in Table 1. The sample vessel was removed after the freezing concentration procedure was finished under the specified operating conditions. The volume and concentration of the

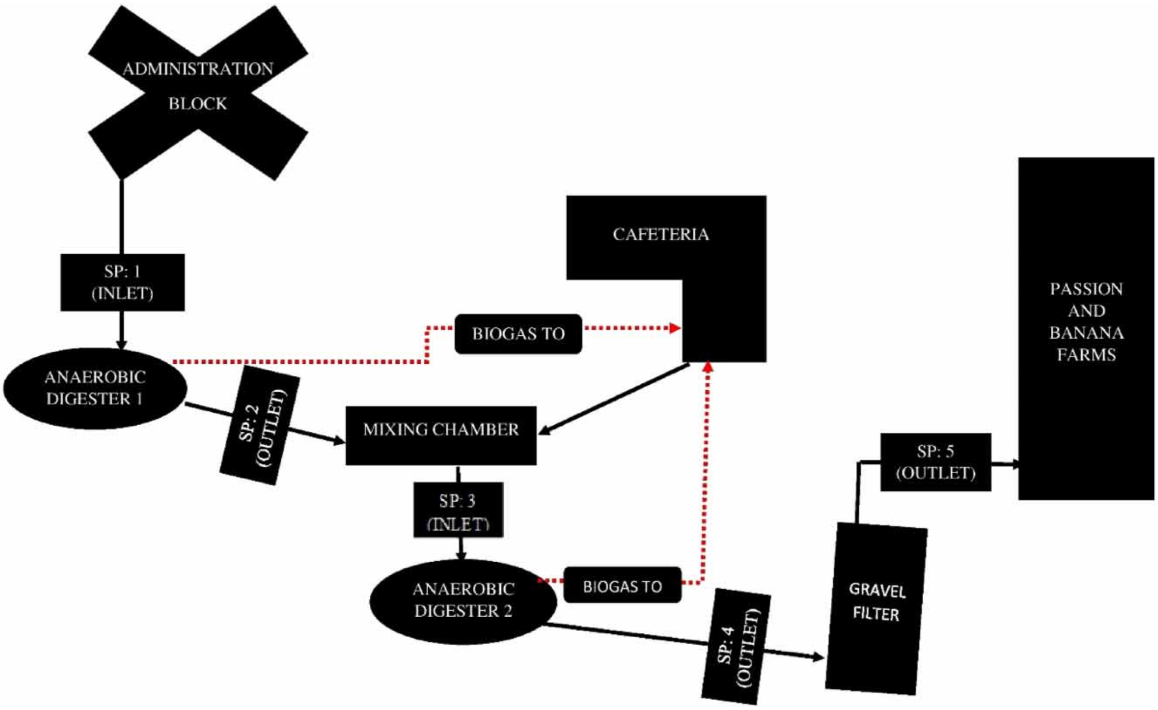

Figure 1 | Process flow of anaerobic biodigesters and gravel filter treatment stage.

Figure 2 | Experimental setup for freezing concentration.

Table 1 | Values of coolant temperature and freezing time tested

Coolant temperature (°C)	−10	−20	−30	−40	−50	−60	−70	−80
Freezing time (h)	1–8	1–7	1–5.5	1–4.5	1–4.25	1–4	1–3.75	1–3.5

concentrate (unfrozen liquid), and melt solution (solution from melted ice) were measured and recorded by using standard techniques for the examination of water and wastewater (APHA 2012). The value of nutrient recovery efficiency was used to determine the efficiency of the freezing concentration process (Ab Hamid & Jami 2019).

2.4. Analytical procedure

Using graduated cylinders (500 mL), the volumes of the initial input water sample (collected water samples before freezing method), unfrozen liquid (samples of concentrate formed after freezing method), and melting ice samples (samples of melted ice formed after freezing method) were collected and measured. Standard techniques for the examination of water and wastewater were used to measure nitrate-nitrogen (NO_3-N) and phosphate (PO_4^{3-}) nutrients in liquid and frozen liquid (APHA 2012).

The analysis of nitrate-nitrogen (NO_3-N) concentration was measured by using DR 900 UV-Visible spectrophotometer method number 355 HR. Phosphate (PO_4^{3-}) was measured by using the same spectrophotometer method 490 HR.

2.5. Nutrient recovery efficiency

Nutrient recovery is the ratio of nutrient amount in the unfrozen liquid to that in the original solution (Ab Hamid & Jami 2019). To get the nutrient recovery, Equation (1) was used.

$$\text{Nutrient recovery} = \frac{C_L}{C_O} \tag{1}$$

where C_L is the concentration of nutrients in the concentrated solution (mg/L), while C_O is the concentration in the original solution (mg/L). A higher value of nutrient recovery gives a good efficiency of the freezing concentration technique.

2.6. Energy consumption

The energy used in each freeze concentration stage was calculated taking into account the pre-cooling of the reactors and the experimental time of the freezing process. The power measured with an energy meter (ZHURUI TEC-PR10, CH) was averaged to determine the energy usage.

3. RESULTS AND DISCUSSION

By examining nutrient recovery, the coolant temperature effect, time and energy consumption on the freezing concentration process were explored. Freeze concentration efficiency in the system was given by measuring variations in the value of nutrient recovery from the wastewater. Ice crystals were seen growing on the vessel once the freezing concentration process was completed under the specified operating conditions.

3.1. Effect of coolant temperature

The effect of freezing temperature on the operation of a freezing concentration system was investigated using nutrient recovery values as a metric. Figure 3 illustrates the relationship between the temperature of coolant and the average value of nutrient recovery at different points of treatment plants. It can be observed that nutrient recovery value was increased with decreasing coolant temperature from −10 to −20 °C and the maximum nutrient recovery was reached at a coolant temperature of −20 °C for samples collected from the inlet and outlet of both anaerobic digesters and gravel filter treatment plants. This recommended range gives a result that agrees with Samsuri *et al.* (2015) and Amran & Jusoh (2016), who both found that lower freezing temperatures will result in a higher nutrient recovery value. As the freezer temperature reduced, the difference in temperature between the freezer and wastewater became high, implying an increase in the rate of heat transfer which leads to a better efficiency of the freeze concentration method (Ab Hamid & Jami 2019).

However, when the freezer temperature was reduced further from −30 to −80 °C, the trend shifted, and the nutrient recovery value declined marginally. This suggests that the process' efficiency has decreased. The discrepancy could be due to a supercooling effect, which occurs when the freezing temperature is at its lowest (Ab Hamid & Jami 2019). The lower nutrient recovery value was caused by the supercooling phenomenon, which reduced the effectiveness of the freezing concentration process. The supercooling effect speeds up the creation of an ice crystal layer, resulting in more solute inclusion on the side of the ice.

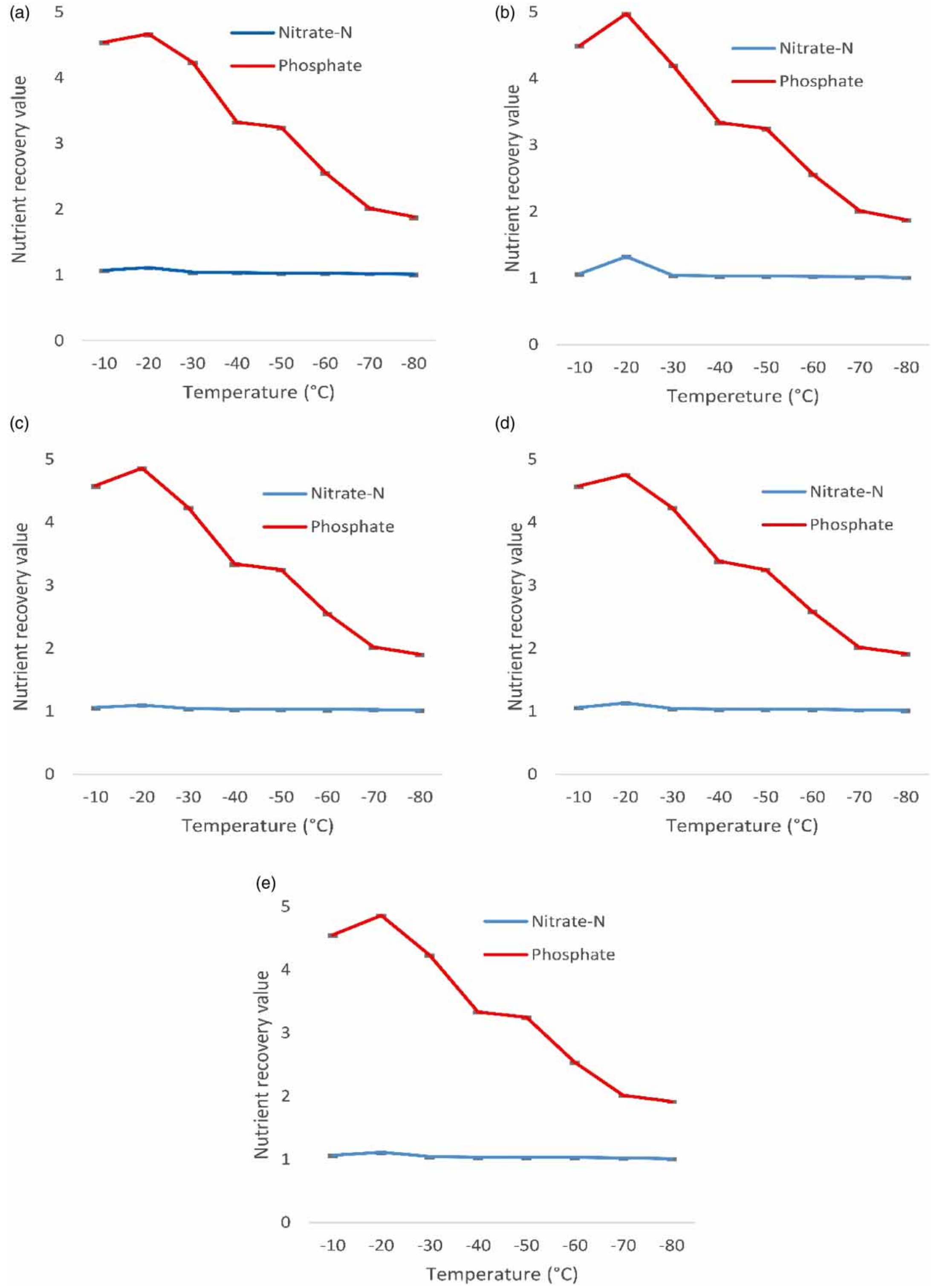

Figure 3 | Effect of the coolant temperature on nitrate-nitrogen and phosphate nutrient recovery at (a) anaerobic digester 1 inlet (SP1); (b) anaerobic digester 1 outlet (SP2); (c) anaerobic digester 2 inlet (SP3); (d) anaerobic digester 2 outlet (SP4)l; and (e) gravel filter outlet (SP5).

Furthermore, depending on the findings of Ab Hamid & Jami (2019) minimum temperatures gave a higher growth rate of ice crystals. It is also important to note that **a** freezing temperature of −20 °C was chosen as the optimal freezing temperature for this system for both sample points because of its better nutrient recovery value.

Nitrate-nitrogen gave lower nutrient recovery values compared to phosphate. The observed disparities in nutrient recovery of nitrate-nitrogen and phosphate by freezing concentration could be due to the morphology of the ice created, which was a multi-crystalline dendritic ice structure that held more nitrate-nitrogen than phosphate. Also, it could possibly be due to molecular weight and size discrepancies between nitrate-nitrogen and phosphate, nutrients of large molecular weight are more easily recovered compared to nutrients of small molecular weight (Gu 2016).

Since it gives a freezing rate, the freezing temperature is important in controlling the process (Melak *et al.* 2016). Examination of the effect of freezing temperature is required to give the ideal temperature for the method. The temperature difference between the freezing temperature, the surface temperature, and the temperature of the wastewater causes the transfer of heat between the coolant and sample solutions across the vessel wall surface during the freezing concentration process (Ab Hamid & Jami 2019). The temperature difference between the coolant and wastewater solution is directly related to the heat transfer rate (Samsuri *et al.* 2015). The heat transfer rate increases as the freezing temperature is lowered. As a result, a lower coolant temperature is better, as this improves the transfer of heat between the coolant and wastewater. A lower temperature is obtained at the surface when there is a lower freezing temperature providing an acceptable initial supercooling environment for ice formation.

3.2. Effect of freezing time

In this analysis, 7 h of freezing time at −20 °C coolant temperature was enough to give the highest performance of the process with the highest nutrient recovery value. At this condition, the average values of nutrient recovery of nitrate-nitrogen and phosphate were 1.114 and 4.667, respectively, at the inlet of anaerobic digester 1; 1.325 and 4.975, respectively, at the outlet of anaerobic digester 1; 1.099 and 4.859, respectively, at the inlet of anaerobic digester 2; 1.132 and 4.755, respectively, at the outlet of anaerobic digester 2; and for gravel filter at the outlet the values where 1.111 and 4.861, respectively.

However, at −30, −40, −50, −60, −70, and −80 °C the maximum freezing time was reached at 5.5, 4.5, 4.25, 4, 3.75, and 3.5 h, respectively, because of supercooling effect as described by Ab Hamid & Jami (2019). This result is in agreement with the findings of Moussaoui *et al.* (2021), Safiei *et al.* (2019) and Azman *et al.* (2018).

In general, a longer freezing duration could increase the freezing concentration process's efficiency (Amran & Jusoh 2016; Safiei *et al.* 2017). With the presence of a dendrite structure, the amount of crystallinity of the generated ice layer was still low when the freezing process began. By extending the freezing period, the ice layer thickens, leaving the unfrozen fluid in a state close to saturation (Yang *et al.* 2017). Figures 4 and 5 show the trends of nutrient recovery at different freezing times from the samples collected at different treatment plants. From these figures, it can be observed that nutrient recovery value for phosphate and nitrate-nitrogen increases as the freezing time increases.

3.3. Analysis of energy consumption

One of the major problems in several industries is to reduce energy consumption and operational costs (Ghannadzadeh & Sadeqzadeh 2016). For innovative resource recovery techniques to be adopted and established in the agricultural sector, it is essential to evaluate their economic implications. As a result, the present study examined the energy consumption of the entire process as an extra criterion to establish optimal conditions for the freezing concentration process to recover nutrients from domestic wastewater. Heat transfer and energy utilized per unit volume of the wastewater sample processed were used to analyze energy consumption to recover nutrients. However, the estimated energy efficiency is likely to be on the higher side given that freezer geometry was not considered in the calculation. A comparison of energy consumption and coolant temperature is shown in Table 2.

When compared to membrane-based technologies, which are one of the most widely used to concentrate wastewater and recover particular components, the freezing concentration process uses the same amount of energy during operation, if not less. Comparing freeze concentration to heating and evaporation procedures, there is great potential for energy savings (Uald-Lamkaddam *et al.* 2021). The amount of energy used depends on the technology being used, the feed solution being used, the ambient temperature, the desired recovery rate, and the electricity cost.

In comparison to the energy consumption of the most effective evaporation systems, Pazmiño *et al.* (2017) claimed energy savings of up to 30% while using a freezing concentration process, treating sucrose solutions, and combining them with the falling film technique. Mtombeni *et al.* (2013) further claimed that its use of the

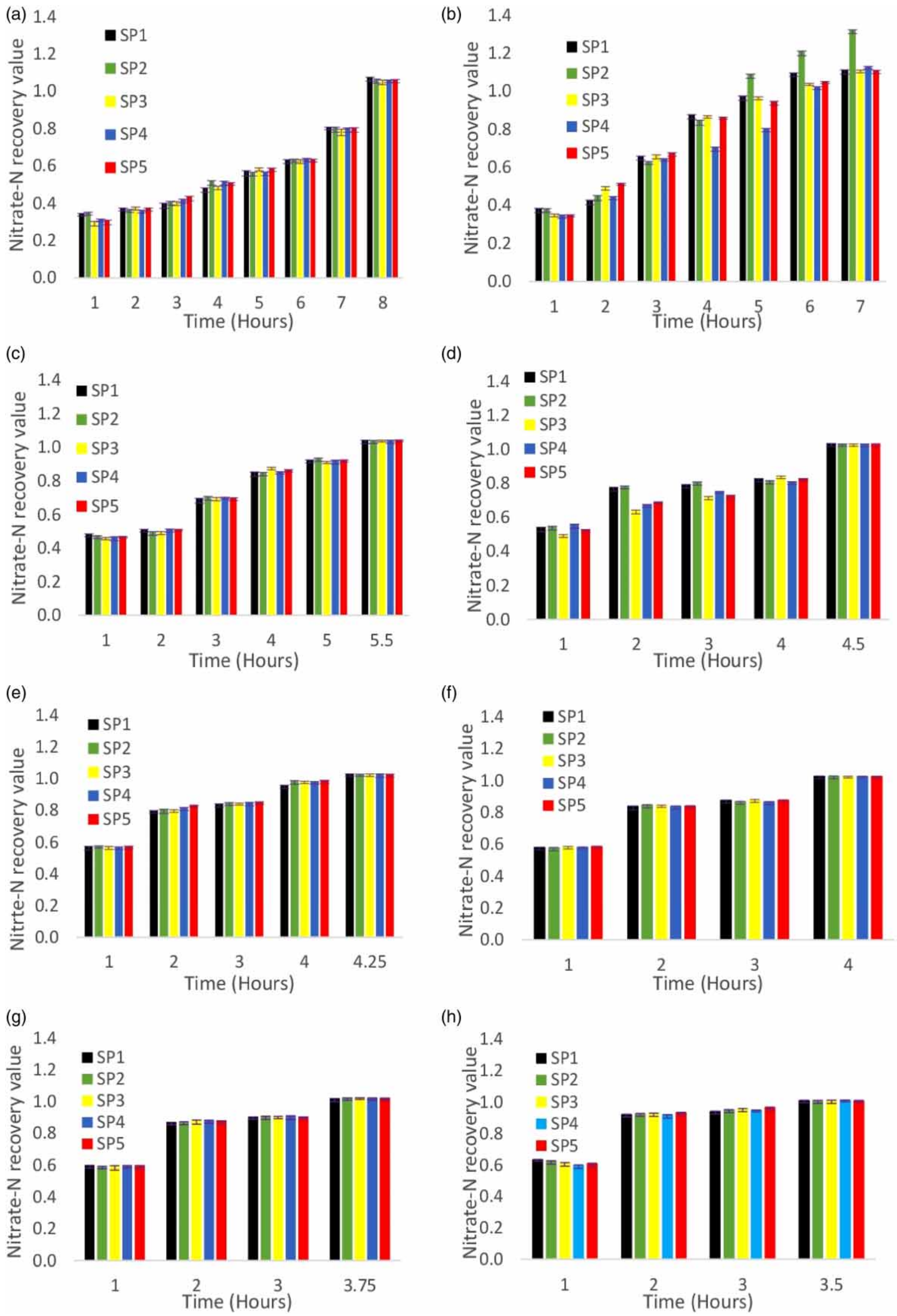

Figure 4 | Nitrate-nitrogen nutrient recovery value at different freezing times at a coolant temperature of (a) −10 °C; (b) −20 °C; (c) −30 °C; (d) −40 °C; (e) −50 °C; (f) −60 °C; (g) −70 °C; and (h) −80 °C.

freezing desalination technique to remove salts from wastewater resulted in the lowest energy consumption (0.39 kWh). This study's findings indicate that employing a freezing concentration system, 1 L of domestic wastewater will require 0.197 kWh energy to recover maximum nutrients. However, one of the most important considerations for separation and concentration technologies is the capital and operating expenses (Uald-Lamkaddam *et al.* 2021). While other separation techniques like ammonia stripping, thermal treatment, ion exchange, and

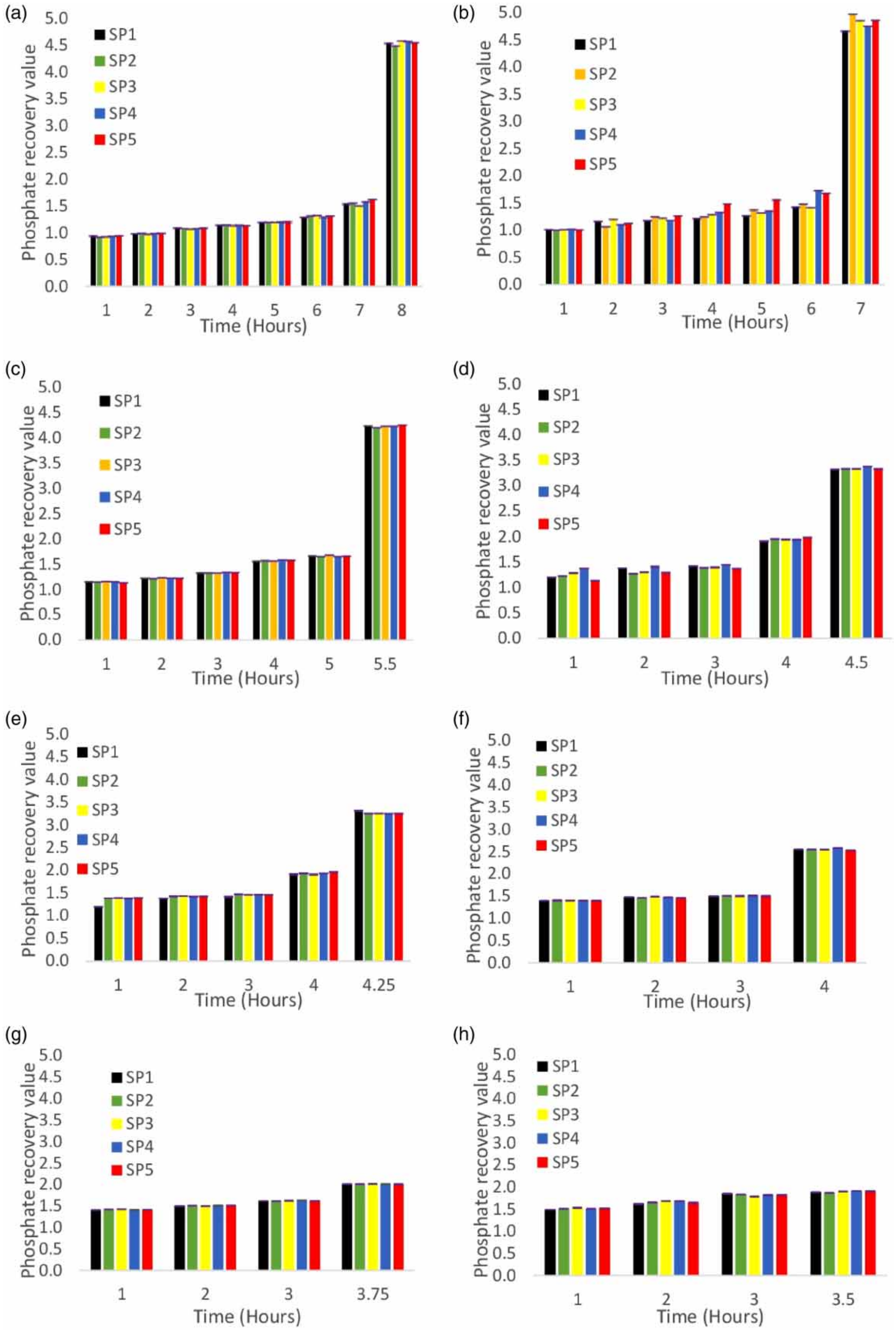

Figure 5 | Phosphate nutrient recovery value at different freezing times at a coolant temperature of (a) −10 °C; (b) −20 °C; (c) −30 °C; (d) −40 °C; (e) −50 °C; (f) −60 °C; (g) −70 °C; and (h) −80 °C.

Table 2 | Energy consumption at different coolant temperatures

Temperature (°C)	−10	−20	−30	−40	−50	−60	−70	−80
Energy consumption (kWh/L)	0.225	0.197	0.155	0.127	0.171	0.161	0.151	0.141
Standard deviation	0.009	0.001	0.007	0.007	0.003	0.006	0.002	0.008

adsorption may need more energy input and be influenced by factors such as pH and aeration, freezing concentration technology is recognized as an environmentally friendly separation process with simple operation, low energy consumption, and a high rejection rate (Shi *et al.* 2018).

4. CONCLUSION

The possibility of using a freezing concentration method to recover nutrients from domestic wastewater was proven in this study. This method, which is based on the freezing concentration mechanism, has been used in the separation of liquid and solid, e.g., in food and pharmaceutical industries and it has also been effectively used for seawater desalination. Nutrient recovery values were used to assess the effects of freezing temperature, cooling time, and energy consumption on the effectiveness of the freezing concentration process. It's critical to run this process to get the optimal nutrient recovery value. The freezing temperature of −20 °C and freezing time of 7 h gave the highest nutrient recovery value during the process. The amount of energy consumed by the coolant at this particular condition was 0.197 kWh/L. In general, the results show the possibility of the freezing concentration method and give conditions that can be used to recover nutrients from domestic wastewater. However, further analysis is needed on the application of this method in recovering heavy metals, emerging compounds, and other organic compounds from domestic wastewater, which could not be checked.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

REFERENCES

AB Hamid, F. H. & Jami, S. N. 2019 Progressive freeze concentration for wastewater treatment from food industry. Key Engineering Materials. Trans Tech Publ, 55–64.

Amran, N. A. & Jusoh, M. 2016 Effect of coolant temperature and circulation flowrate on the performance of a vertical finned crystallizer. *Procedia Engineering* **148**, 1408–1415.

APHA 2012 *Standard Methods for the Examination of Water and Wastewater*. American Public Health Association, Washington.

Azman, N., Samsuri, S. & Jusoh, M. 2018 Effect of freezing time and shaking speed on the performance of progressive freeze concentration via vertical finned crystallizer. *International Journal of Automotive and Mechanical Engineering* **15**, 5356–5356.

Ghannadzadeh, A. & Sadeqzadeh, M. 2016 Exergy analysis as a scoping tool for cleaner production of chemicals: a case study of an ethylene production process. *Journal of Cleaner Production* **129**, 508–520.

Gu, X. 2016 *Nutrients Recovery From Municipal Wastewater Effluent Using Electrochemical and Freeze Concentration Approaches*.

Huo, S., Liu, J., Addy, M., Chen, P., Necas, D., Cheng, P., Li, K., Chai, H., Liu, Y. & Ruan, R. 2020 The influence of microalgae on vegetable production and nutrient removal in greenhouse hydroponics. *Journal of Cleaner Production* **243**, 118563.

Lu, H., Wang, J., Wang, T., Wang, N., Bao, Y. & Hao, H. 2017 Crystallization techniques in wastewater treatment: an overview of applications. *Chemosphere* **173**, 474–484.

Mavhungu, A., Foteinis, S., Mbaya, R., Masindi, V., Kortidis, I., Mpenyana-Monyatsi, L. & Chatzisymeon, E. 2021 Environmental sustainability of municipal wastewater treatment through struvite precipitation: influence of operational parameters. *Journal of Cleaner Production* **285**, 124856.

Melak, F., Du Laing, G., Ambelu, A. & Alemayehu, E. 2016 Application of freeze desalination for chromium (VI) removal from water. *Desalination* **377**, 23–27.

Moharramzadeh, S., Ong, S. K., Alleman, J. & Cetin, K. S. 2021 Parametric study of the progressive freeze concentration for desalination. *Desalination* **510**, 115077.

Moussaoui, C., Blanco, M., Muñoz, I. D. B., Raventós, M. & Hernández, E. 2021 An approach to the optimization of the progressive freeze concentration of sucrose solutions in an agitated vessel. *Separation Science and Technology* **56**, 746–756.

Mtombeni, T., Maree, J., Zvinowanda, C., Asante, J., Oosthuizen, F. & Louw, W. 2013 Evaluation of the performance of a new freeze desalination technology. *International Journal of Environmental Science and Technology* **10**, 545–550.

Ownby, M., Desrosiers, D.-A. & Vaneeckhaute, C. 2021 Phosphorus removal and recovery from wastewater via hybrid ion exchange nanotechnology: a study on sustainable regeneration chemistries. *npj Clean Water* **4**, 1–8.

Pazmiño, N. V., Raventós Santamaria, M., Hernández Yáñez, E., Gulfo, R., Robles, C., Moreno, F. & Ruiz, Y. 2017 Continuous system of freeze concentration of sucrose solutions: process parameters and energy consumption. *Journal of Food Technology and Preservation* **1**, 1–5.

Safiei, N. Z., Ngadi, N., Johari, A., Zakaria, Z. Y. & Jusoh, M. 2017 Grape juice concentration by progressive freeze concentrator sequence system. *Journal of Food Processing and Preservation* **41**, e12910.

Safiei, N. Z., Danuri, N. F. N., Rosly, M. K. & Shaharuddin, S. 2019 Optimization of fractional freezing process for orange juice concentration. *Materials Today: Proceedings* **19**, 1591–1598.

Samsuri, S., Amran, N. A. & Jusoh, M. 2015 Spiral finned crystallizer for progressive freeze concentration process. *Chemical Engineering Research and Design* **104**, 280–286.

Sena, M., Seib, M., Noguera, D. R. & Hicks, A. 2021 Environmental impacts of phosphorus recovery through struvite precipitation in wastewater treatment. *Journal of Cleaner Production* **280**, 124222.

Shi, L., Simplicio, W. S., Wu, G., Hu, Z., Hu, H. & Zhan, X. 2018 Nutrient recovery from digestate of anaerobic digestion of livestock manure: a review. *Current Pollution Reports* **4**, 74–83.

Uald-Lamkaddam, I., Dadrasnia, A., Llenas, L., Ponsá, S., Colón, J., Vega, E. & Mora, M. 2021 Application of freeze concentration technologies to valorize nutrient-rich effluents generated from the anaerobic digestion of agro-industrial wastes. *Sustainability* **13**, 13769.

Yang, Y., Lu, Y., Guo, J. & Zhang, X. 2017 Application of freeze concentration for fluoride removal from water solution. *Journal of Water Process Engineering* **19**, 260–266.

Zou, Y., Zeng, Q., Li, H., Liu, H. & Lu, Q. 2020 Emerging technologies of algae-based wastewater remediation for bio-fertilizer production: a promising pathway to the sustainable agriculture. *Journal of Chemical Technology & Biotechnology* **96**, 551–563.

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