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Purification of Roof-Harvested Rainwater using Progressive Freeze Concentration

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Rainwater can serve as a safe and sustainable source of drinking water. One common way to harvest rainwater is through runoff from the roofs of buildings. Depending on the cleanliness and material of the roofs, this roof-harvested rainwater can have very different water quality like colour, turbidity, pH and total dissolved solids (TDS) compared to the rainwater. Roof-harvested rainwater TDS was not correlated with antecedent dry days (ADD) and weakly negatively correlated with rainfall amount. This paper demonstrated the use of progressive freeze concentration (PFC) to purify rainwater through reduction of colour, turbidity and TDS. In lieu of a specialised PFC equipment, falling-film ice making equipment was used to purify rainwater. Falling-film equipment was chosen for PFC as supercooling that could adversely affect separation efficiency was not observed. Two parameters, circulation time and initial rainwater quality were investigated for their effects on yield and quality of purified rainwater quality (colour, turbidity and TDS). Preliminary results showed the reduction of colour to within drinking water standard and TDS by more than 93 %. Higher circulation time was shown to decrease yield but improve effective partition coefficient (K). Initial rainwater quality has negligible effect on yield and removal efficiency. In conclusion, PFC is a potential method to purify roof-harvested rainwater into drinking water.

1. Introduction

Rainwater can be purified in a variety of ways to be used for both potable and non-potable purposes around the world. One novel technique known as Progressive Freeze Concentration (PFC) has been demonstrated to purify rainwater. When rainwater is frozen slowly in PFC, pure ice crystal forms while rejecting dissolved contaminants in the unfrozen mother liquor. To proliferate the use of rainwater as a source of drinking water, several challenges like harvesting adequate raw rainwater, cost and reliability of the rainwater harvesting system must be overcome. While first flush system is an effective way to reduce contamination of roof-harvested rainwater by diverting the initial portion of rainwater runoff to drain, it is unclear how much rainwater should be wasted as the contamination level depends on roof cleanliness which in turns depends on antecedent dry days (ADD). The first objective is to create a model relating rainwater quality, ADD and rainfall amount by examining various factors affecting rainwater quality.

Supercooling is thought to contaminate the ice formed in previous PFC studies and additional steps were taken to prevent supercooling during PFC experiment. The precautionary steps could complicate and increase the cost of the PFC equipment setup. The second objective is to scrutinize the phenomenon of supercooling in falling-film PFC equipment by measuring temperature change throughout the PFC process.While PFC is effective in reducing dissolved contaminant in rainwater, it is unclear to whether the separation efficiency of PFC in rainwater is dependent on circulation time and initial rainwater quality like other PFC studies. The third objective is to explore the effects of PFC on rainwater purification, specifically circulation time and initial rainwater quality.

As roof-harvested rainwater comes from the roof, the cleanliness of the roof is of the utmost importance in ensuring minimal contamination to the rainwater. Roof surfaces can be contaminated by particulate matter from

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dry deposition, animal droppings and plant matter from nearby vegetations. According to Gikas and Tsihrintzis (2017), the longer the dry period before each rainfall event (ADD), the higher the amount of contaminants on the roof surface, leading to greater contamination of the rainwater runoffs. Conversely, a heavier rainfall event will result in a lower concentration of contaminants in rainwater (Maniquiz et al., 2010) due to the availability of more rainwater to dissolve a finite amount of contaminants on roof surface. While metallic roofs tend to leach metallic ion into roof-harvested rainwater due to the slightly acidic nature of rainwater, Mendez et al. (2011) showed that the concentration of zinc ion from galvanized metal roof-harvested rainwater was not higher compared to other types of roofs if the roof was in good condition.

Rainwater runoff quality can be mathematically approximated by assuming the amount of pollutant, which can be removed during a storm event, depends on rainfall duration and initial quantity of pollutant available (Eslamian, 2014). According to Robinson and Ward (2017), most of the dissolved material in rain came from the sea salts (Na^+, Cl^-, Mg^{2+} and K^+), the concentration of these ions decreases with increasing distance inland from the coast. On the other hand, Ca^{2+}, NH_4^+, SO_4^{2-}, HCO_3^- and NO_3^- ions came predominantly from terrestrial sources like dusts and anthropogenic emissions.

According to Grund (2015), when water exists in liquid phase below normal freezing point even when a more stable state exists, the water is supercooled. While supercooling of water is important in nature for certain species to survive the winter by preventing ice crystal formation inside their bodies that is lethal, it is undesirable in progressive freeze concentration (PFC) as it will lead to dendrites formation and inclusion of impurities in the ice crystal lattice that affect the purity of ice produced. Various methods have been proposed to prevent supercooling in PFC, e.g. use of ice-nucleating protein (Liu et al., 1997), a seed lining of pure ice crystal prior to starting experiments (Miyawaki et al., 1998) and holes on cooling surface (Liu et al., 1998).

Interestingly, no supercooling prevention method has been proposed for falling-film PFC equipment (Sánchez et al., 2010). This is probably due to supercooling not happening in the equipment due to the inherent turbulent nature of circulating flow. Nonetheless, Shirai et al. (1999) demonstrated the importance of seed ice in producing pure ice from a high COD solution. It is unclear whether supercooling occurred during the experiment as no temperature data were presented in the paper. Therefore, an experiment has been proposed to investigate the presence of supercooling in falling-film equipment.

Harvested rainwater usually requires additional purification steps if meant for human consumption. This is to prevent waterborne illnesses from microorganisms. According to Novak et al. (2014), harvested rainwater is purified in two steps (filtration and disinfection). While similar to the equipment used in conventional water treatment plants, the filters used in rainwater harvesting systems should be simple, inexpensive and capable of intermittent use. The function of filtration is to reduce the amount of total suspended solids (TSS) that can shield pathogens from subsequent disinfection and can be done using fine-mesh screen filters, bag filters, cartridge filters and membrane filters. Disinfection inactivates pathogenic microorganisms and can be performed by exposing the filtered rainwater with ultraviolet (UV) light, chlorine or ozone. Rashid et al. (2022) used PFC to reduce the turbidity in landfill leachate by up to 83 %. This suggests that TSS in rainwater could similarly be reduced using PFC as turbidity is often used as a proxy of TSS. TDS in rainwater can be reduced using PFC as shown by How et al. (2023).

In this study, TDS was used as a proxy to rainwater quality in lieu of a conventional laboratory analysis of constituents present in rainwater because of low cost, ease of use and repeatability of TDS measurement. While TDS can serve as an overall indicator of contamination in rainwater, more parameters should be measured to ensure that purified rainwater is safe for human consumption. This paper investigated the relationship between independent variables (initial rainwater quality, circulation time) and dependent variables (yield, effective partition coefficient (K), removal efficiency) and discussed the effect of PFC on rainwater TDS, pH, colour and turbidity. Fujioka et al. (2013) showed that K increased with increasing initial concentrations in 1.75 wt% to 3.5 wt% sodium chloride solution. Samsuri et al. (2015) showed that both K and yield followed a decreasing trend with increasing circulation time from 40 min to 80 min.

2. Materials and Methods

2.1 Factors Affecting Rainwater Quality

Rainwater samples and rainfall data were collected from October 2023 to May 2024. Roof-harvested rainwater was collected from the roof of N12, Universiti Teknologi Malaysia, Johor Bahru, Malaysia (1.5655, 103.6395) (Figure 1a). TDS of rainwater was measured using a portable TDS meter. Rainfall amount was measured using a rain gauge (Figure 1b) and compared with the data from Department of Irrigation and Drainage Malaysia (DID, 2024). A linear regression model and Pearson correlation coefficient (ρ), Eq(1) were used to determine the relationship between rainfall amount, ADD and rainwater TDS, where cov is covariance and σ is standard deviation (Steiner, 2008).

Figure 1: Actual site of (a) Rainwater harvesting system; (b) Rain gauge

$$\rho_{x,y} = \frac{cov(x,y)}{\sigma_x \sigma_y} \tag{1}$$

2.2 Supercooling in Falling-Film PFC

An ice making equipment HICON HZB-60FAB was used in this PFC experiment as falling-film equipment with conventional vapour-compression refrigeration cycle using R290 (propane) refrigerant to cool a stainless-steel cooling surface to below freezing point. Distilled water was fed to the equipment via an internal feed pump while a circulating pump was used to circulate the feed during PFC experiment. A calibrated temperature sensor was placed in the sump of the circulating solution and recorded every 30 s. The experiment was repeated using roof-harvested rainwater.

2.3 Rainwater Purification Using PFC

An ice making equipment HICON HZB-60FAB was used in this PFC experiment. Two sets of experiments (20 min and 26 min circulation time) were conducted for each batch of rainwater. TDS, pH, colour, turbidity and volume of raw rainwater, purified rainwater and purge (unfrozen, concentrated portion of the feed rainwater) were measured. TDS and pH were measured using portable TDS meter and pH meter respectively. Colour and turbidity were measured using Macherey-Nagel PF-12Plus photometer. Effects of circulation time were analysed using paired-sample t-test while effects of initial rainwater quality were analysed using linear regression. Yield, K and removal efficiency were calculated using the following formulae Eq(2), Eq(3) and Eq(4).

$$Yield\ \left(\frac{ml}{h}\right) = \frac{Volume\ of\ purified\ rainwater\ (ml)}{Time\ taken\ (h)} \tag{2}$$

$$K = \frac{TDS\ of\ purified\ rainwater\ (ppm)}{TDS\ of\ purge\ (ppm)} \tag{3}$$

$$Removal\ efficiency\ (\%) = \frac{(TDS\ of\ feed - TDS\ of\ purified\ rainwater)}{TDS\ of\ feed} \times 100\ \% \tag{4}$$

3. Results and Discussion

3.1 Factors Affecting Rainwater Quality

Excerpt of rainfall data from October 2023 to May 2024 was tabulated in Table 1. Most rainfall events had ADD which were quite short with a median of 2 days and mean of 2.9 days. This is not surprising, considering Malaysia is a tropical country. The median and mean of the rainfall amount were 17.4 mm and 23.6 mm. All rainwater samples had TDS less than 50 ppm, which was below the TDS of local tap water supply. TDS was found to be not correlated (ρ = 0.076) with ADD (Figure 2a) and weakly negatively correlated (ρ = -0.320) with rainfall amount (Figure 2b). However, the correlations were not strong and other more important parameters are directly involved in the rainwater quality, e.g. type of roof surface, location, duration of the rain and level of anthropogenic greenhouse gas emissions.

Table 1: Excerpt of raw rainfall data

Date	From	To	Manual rainfall (mm)	DID rainfall (mm)	Average rainfall (mm)	TDS (ppm)	ADD
30/12/23	0630H	2000H	1.7	5	3.4	27	2
31/12/23	1630H	1700H	2.1	2	2.1	29	1
1/1/24	1230H	2300H	7.2	55	31.1	35	1
3/1/24	1530H	1600H	1.6	3	2.3	18	2
4/1/24	0330H	2200H	40.8	48	44.4	14	1

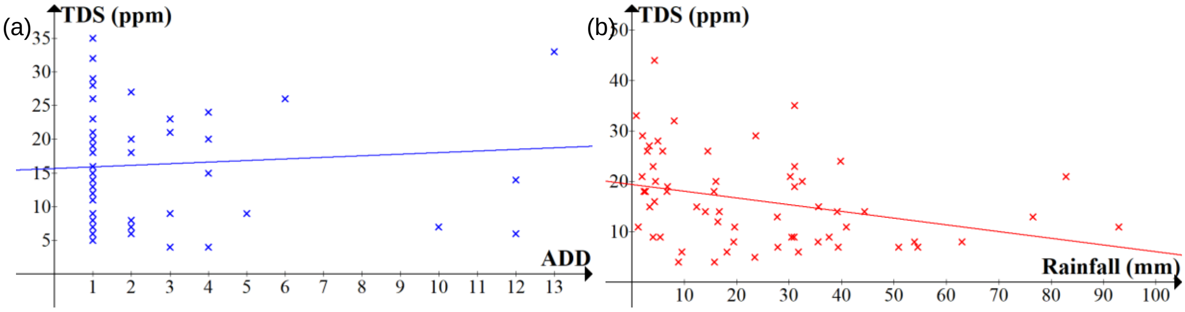

Figure 2: Graph of (a) TDS of rainwater against antecedent dry days (ADD); (b) TDS of rainwater against rainfall amount

3.2 Supercooling in Falling-Film PFC

Ice formed on the stainless steel cooling surface (Figure 3) as the temperature of circulating solution reached the freezing point.

Figure 3: Ice formation in falling-film PFC experimental setup

No supercooling was observed in both distilled water and rainwater during cooling and formation of ice as the temperature of the circulating solution never dropped below the freezing point prior to ice formation as shown in Figure 4a. Supercooling did not happen in a falling-film PFC setup as compared to other setup was probably due to the different way fluid was circulated in falling-film. Fluid was pumped to the top of the cooling surface and gravity flowed to the bottom, during which the fluid became turbulent and aerated. The air bubbles formed served as ice-nucleating sites and inhibited supercooling. In contrast, conventional PFC setup employing stirrers cannot generate enough agitation to promote heterogeneous nucleation as in falling-film setup.

3.3 Rainwater Purification Using PFC

Summary of the experimental data was shown in Table 2. In general, PFC decreased the TDS and pH of rainwater by preventing the inclusion of dissolved, charged species in the ice crystal formation. While no significant effect was observed for colour and turbidity, purge showed increased colour, implying the dissolved organic matters in rainwater were concentrated in the unfrozen fraction, achieving rainwater purification. Initial rainwater quality (TDS) has no significant effect on yield, K and removal efficiency. This is probably due to the very low concentration of contaminants in rainwater.

Table 2: Summary of experimental data

Parameter	TDS (ppm)	pH	Colour	Turbidity (NTU)	Volume (ml)	Yield (ml/h)	K	RE (%)
Feed	14	6.76	< 25	2				
Purified rainwater (20 min)	< 1	6.19	< 25	1	1,590	970	< 0.024	> 93
Purified rainwater (26 min)	< 1	6.12	< 25	2	1,830	840	< 0.021	> 93
Purge (20 min)	42	7.12	28	2	890			
Purge (26 min)	48	7.13	32	2	850			

Purge TDS is positively and linearly correlated with feed rainwater TDS with R^2 = 0.9523 (Figure 4b). Circulation time has no significant effect on colour and turbidity.

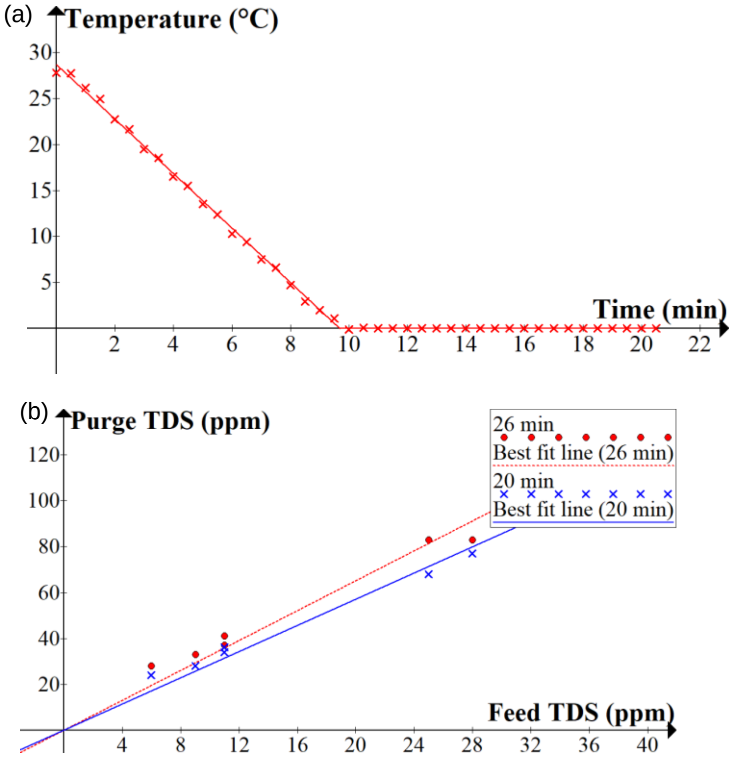

Figure 4: Graph of (a) temperature of distilled water *against* time during cooling; (b) purge TDS against feed TDS

Due to the detection limit (< 1 ppm) of the TDS meter, TDS of purified rainwater in both 20 min and 26 min cannot be measured accurately. Therefore, TDS removal efficiencies of both circulation time were assumed to be the same. Yield is 13.0 % lower and K is 13.2 % lower in 26 min circulation time compared to 20 min circulation time. This means that longer circulation time can improve purity of the ice product at the expense of yield. Optimum circulation time should be determined on a case-by-case basis as it depends on the experimental setup and feed solution.

4. Conclusions

Roof-harvested rainwater TDS was not correlated with antecedent dry days (ADD) and weakly negatively correlated with rainfall amount. The heavier the rainfall event, the higher the quality of the roof-harvested rainwater from that rainfall event. No supercooling was observed in the falling-film PFC equipment, which simplified operation. At very low concentration, TDS did not affect separation efficiency of PFC. Longer circulation time improved separation efficiency at the expense of the yield of ice. PFC is a potentially viable and feasible technique to purify rainwater into drinking water. Further research is necessary to determine the extent of PFC rainwater purification on other contaminants.

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