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Review

A Review on Chlorination of Harvested Rainwater

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Abstract: The supply of safe drinking water to rural communities has always been challenging, unlike in most large cities where government authorities have constructed central water supply systems. In many rural areas, primary water sources such as surface water and groundwater are at risk of contamination with rapid agricultural and industrial growth and climate change-related issues. Rainwater harvesting is an ancient practice for rural communities, and the momentum around its use is continually growing in recent years. However, the lack of sustainable treatment facilities on a small scale encourages dwellers to consume harvested rainwater (HRW) without any treatment even though drinking untreated HRW may have multiple health impacts in many cases. There are several methods of treating HRW. While chlorination is extensively used to disinfect water in large volumes, e.g., central drinking water supply systems), it has not been widely adopted for treating water on a small scale. We present a scoping review to explore whether chlorination could be a viable option for disinfecting HRW at a domestic level. It is found that inadequate treatment prior to chlorine disinfection could produce chlorine disinfection byproducts (DBPs). Limited data on DBP concentrations in HRW are available to assess its health implications. Based on this review, it is argued that chlorination could be an option for treating HRW at a domestic level when limitations associated with this method (such as safe storage, appropriate sustainable technology, and lessening DBPs by lowering total organic carbon before chlorination through other treatment methods) are resolved.

Keywords: rainwater harvesting; water treatment; disinfection; chlorination; drinking water

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1. Introduction

Clean water resources are under stress globally to meet growing demand. The rapid industrial and urban growths often cause water pollution [1,2]. Many industries in developing countries discharge untreated wastewater into surface water bodies [3,4]. Many primary water sources such as groundwater and surface water bodies in both developing and developed countries are exposed to various industrial and agricultural pollutants [5,6]. As a result, people suffer from cancer and other diseases, which are caused by water pollution among other factors [7–9]. In many rural areas, groundwater is either unavailable or contaminated (such as arsenic and fluoride contamination). Currently, 783 million people do not have access to clean water, and 84% of them live in rural areas of developing countries. Many rural communities have been suffering from various acute waterborne diseases such as hepatitis, cholera, dysentery, giardiasis, diarrhoea, typhoid, and cryptosporidiosis [10–12]. The water treatment cost is relatively high, and many developing countries cannot afford to treat polluted water [13]. In many locations, rural and low-income communities are deprived of the benefit of modern water treatment technologies [14].

To meet clean water demand, harvested rainwater (HRW) is getting more attention in remote and some urban areas as rainwater is generally fresh in nature [15–19]. A question is often raised whether HRW needs disinfection before human consumption. For example, Chubbaka et al. [20] identified that some households in South Australia prefer drinking untreated HRW; however, some residents treat HRW using a filtration system.

Research suggests that in many parts of the world, the quality of untreated HRW does not comply with the World Health Organisation's (WHO) [20,21] drinking water guidelines. The reasons include (i) location of roof catchment [22], (ii) surrounding sources of pollutants [22], (iii) roof materials, (iv) air quality, (v) tank size, (vi) animal waste [23], and (vii) materials used in HRW storage and collection systems [24]. It indicates that at many instances HRW contains heavy metals [25], and microbiological contaminants. The consumption of untreated HRW for an extended period may have both immediate and long-term health consequences [21]. Therefore, HRW treatment at a small scale has become more relevant to the communities now than in the past.

Researchers indicate that the efficacy of water treatment methods may vary depending on the water source, environment, and catchment [26]. For example, Brown and Sobsey [27] conducted a test on the performance of ceramic filters between two types of water catchments, (a) rainwater and (b) surface water. They showed that rainwater has less turbidity compared to surface water. Lantagne et al. [28] proposed five well-known methods for household water treatment: (i) chlorination, (ii) filtration (bio-sand and ceramic), (iii) solar disinfection, (iv) combination of filtration and chlorination, and (v) combination of flocculation and chlorination. They noted that water quality improved significantly after treatment, reducing water-borne diseases. They also compared the performances of various filtration and disinfection methods as shown in Table 1.

Table 1. Performance analysis of various household water treatment methods [28–32].

Treatment Method	Can Remove Virus	Can Remove Bacteria	Can Remove Protozoa	Does the Treatment Method Has a Residual Effect	Cost of Treatment	Disinfection by Product Production	Reference
Disinfection by Chlorination	Yes—Medium	Yes—high	Yes—Low	Yes	US $0.09–0.37 per bottle of chlorine solution	Yes	[28]
Filtration by bio-sand	Not known	Yes—Medium to High	Yes—High	No	-	No	[28]
Filtration by ceramic	Not known	Yes—High	Yes—High	No	Water cost US $0.3–0.5 Filter cost US $2.5–4	No	[29,30]
Filtration and chlorination	Yes—Medium	Yes—High	Yes—High	Yes	US $0.09–0.37 per bottle of chlorine solution	Yes—Medium	[28]
Disinfection by solar	Yes—High	Yes—High	Yes—High	No	US $0, bottle cost not included	No	[28]
Flocculation and chlorination	Yes—High	Yes—High	Yes—High	Yes	US $0.07	Yes	[31]
Aeration + filtration + carbon filtration + ultra violet disinfection	Yes—High	Yes—High	Yes—High	Yes	AUD 4.59/m^3	No	[32]

Many non-governmentgovernment organisations (NGOs) have used hypochlorite solution for water disinfection at domestic level [32–34] because hypochlorite solution has additional advantages such as (i) lower cost, (ii) residual effect, and (iii) greater availability [34]. Among the advantages, the residual effect is one of the critical factors for hypo-chlorite solution, which can prevent stored water from potential recontamination. Ali et al. [35] noticed regrowth of coliform bacteria after treating water by coagulation. Therefore, it can be argued that there is a potential risk of recontamination of the disinfected rainwater while it is being stored. Hence, a hypochlorite solution could be an effective and preferred disinfectant for rainwater disinfection.

However, using the hypochlorite solution has some concerns that must be considered before recommending it for disinfecting HRW at the household level. Hypochlorite solution can produce disinfection byproducts (DBP) when it reacts with organic contents present in water [28,36]. HRW may contain organic matter, and hypochlorite solution can produce DBPs when applied to untreated rainwater [37]. DBP consumption through drinking water may trigger severe health issues like cancer [38]. Hence, both inadequate and excess chlorine dose in water treatment is undesirable.

Raising awareness on microbiological contaminants and chlorine DBPs in HRW is essential to people who consume rainwater. However, there is a lack of knowledge on the pros and cons of chlorination in the HRW. Therefore, this review paper focuses on:

a. Common organic pollutants in HRW;
b. DBPs formation in chlorinated water;
c. Health impacts of consuming water with DBPs and
d. Possible ways of minimising formation of DBPs in HRW.

2. Methodology

In carrying out this review, the following questions have been investigated:

1. Why does HRW need disinfection for drinking?
2. Can hypochlorite solution be considered for disinfecting HRW, and if so, what are its limitations?
3. How can DBPs be reduced after chlorination?
4. What is the possibility of formation of DBPs in HRW and what are the health repercussions if HRW is disinfected by hypochlorite solution?
5. What are the acceptable limits of the DBPs found in the HRW as per drinking water guidelines?
6. How can DBPs be reduced in HRW?

Literature data were used to answer the above questions. Since there were six specific questions, the keywords that were used to identify relevant papers were categorised into six groups. Each group had subtopics to extract essential information from the existing literature. The keyword categories and subtopics are presented in Figure 1.

A literature search was conducted based on the selected keywords. There were 113 peer-reviewed journal articles, and 97 were selected to collect data for this study based on relevance. The sorting of journal articles followed the method explained by [22]. Three literature search engines were used for this research: (i) Google Scholar, (ii) UWS library and (iii) Scopus. These search engines also assisted in identifying various relevant regulatory organisations in different countries across the globe. They are (i) Water Environment Partnership in Asia (WEPA, Tokyo, Japan), (ii) New Zealand Ministry of Health (NZMH, Wellington, New Zealand), (iii) South African Department of Water Affairs and Forestry (DWAFSA, Pretoria, South Africa), (iv) National Public Health Service for Wales (Cardiff, Wales), (v) World Health Organization (WHO, Geneva, Switzerland), (vi) Bureau of Indian Standards (BIS, New Delhi, India), (vii) United States Environmental Protection Agency (USEPA, Washington, DC, USA), (viii) Council of European Union (Brussels, Belgium), (ix) Australian Government National Health and Medical Research Council (NHMRC, Canberra, Australia), (x) Canadian Federal Provincial Territorial Committee on Health and the Environment (Moncton NB, Canada), (xi) International Agency for Research on Cancer (IARC, Lyon, France), (xii) National Standard of the People's Republic of China, and (xiii) Japanese Ministry of Health, Labour and Welfare (Tokyo, Japan). The review also considered various guidelines for drinking water developed by relevant regulatory authorities. The result section analyses the obtained information, followed by a discussion section.

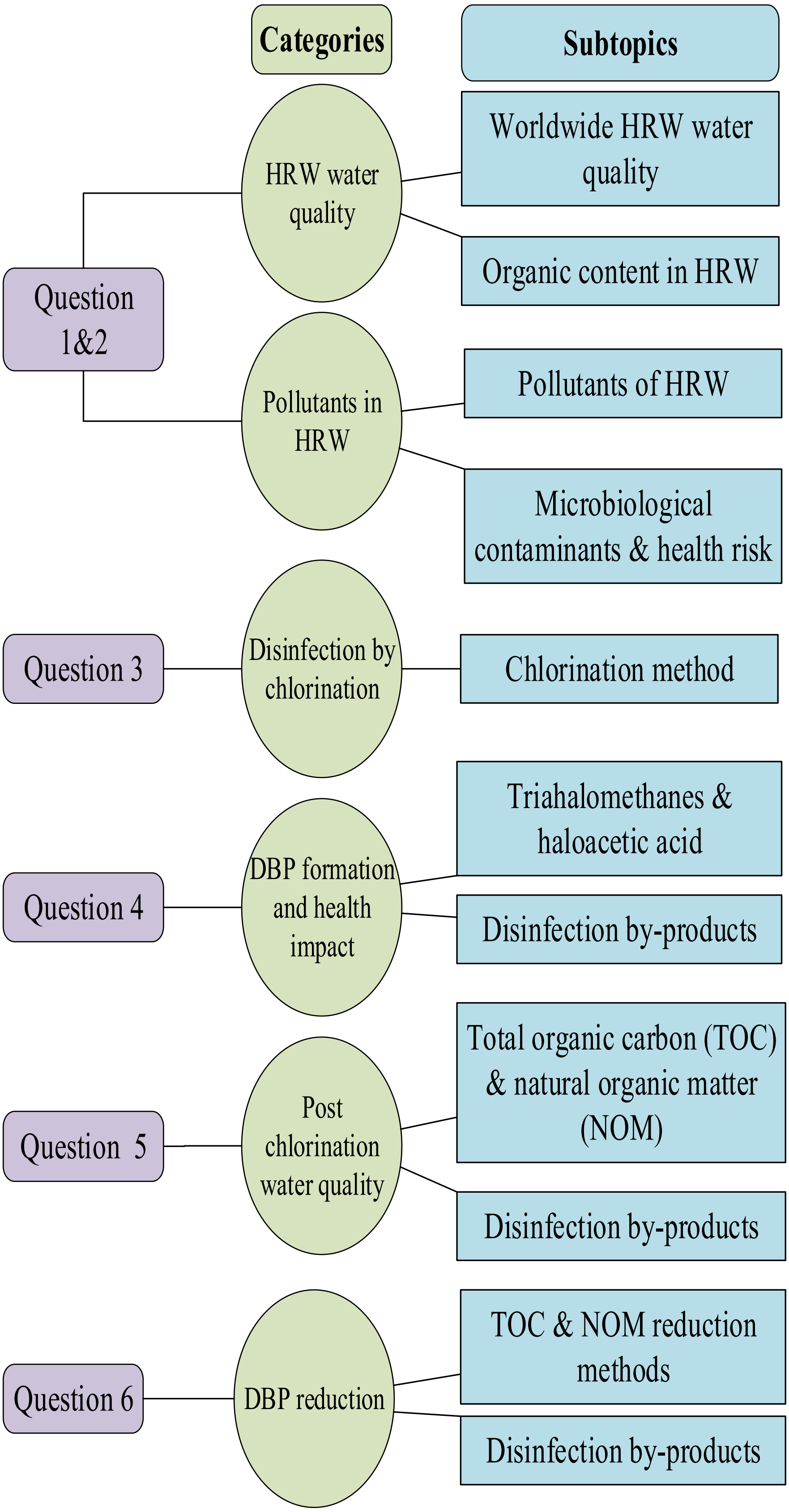

Figure 1. Keyword categories and subtopics to answer research questions.

3. Water Quality of Harvested Rainwater (HRW): Secondary Data Analysis of Microbiological Contaminants

Bacteria, viruses, and protozoa are the major microbiological pollutants in HRW [39]. The primary sources of microbial contaminants are the faeces of animals such as birds, rats, squirrels, and possums [23]. These unwanted animal excrements are washed out from the roof and accumulated in rainwater tanks during rainfall. Figure 2 shows various harmful microorganisms in rainwater tank collected from 15 countries. Among the microbiological pollutants, faecal and total coliform have been identified in higher concentrations than *E. coli* and Enterococci. Figure 2 demonstrates that faecal and total coliform were found excessive in rainwater worldwide in both developed and developing countries. This indicates that rainwater treatment is mainly ignored on a small scale, irrespective of the country's economic status.

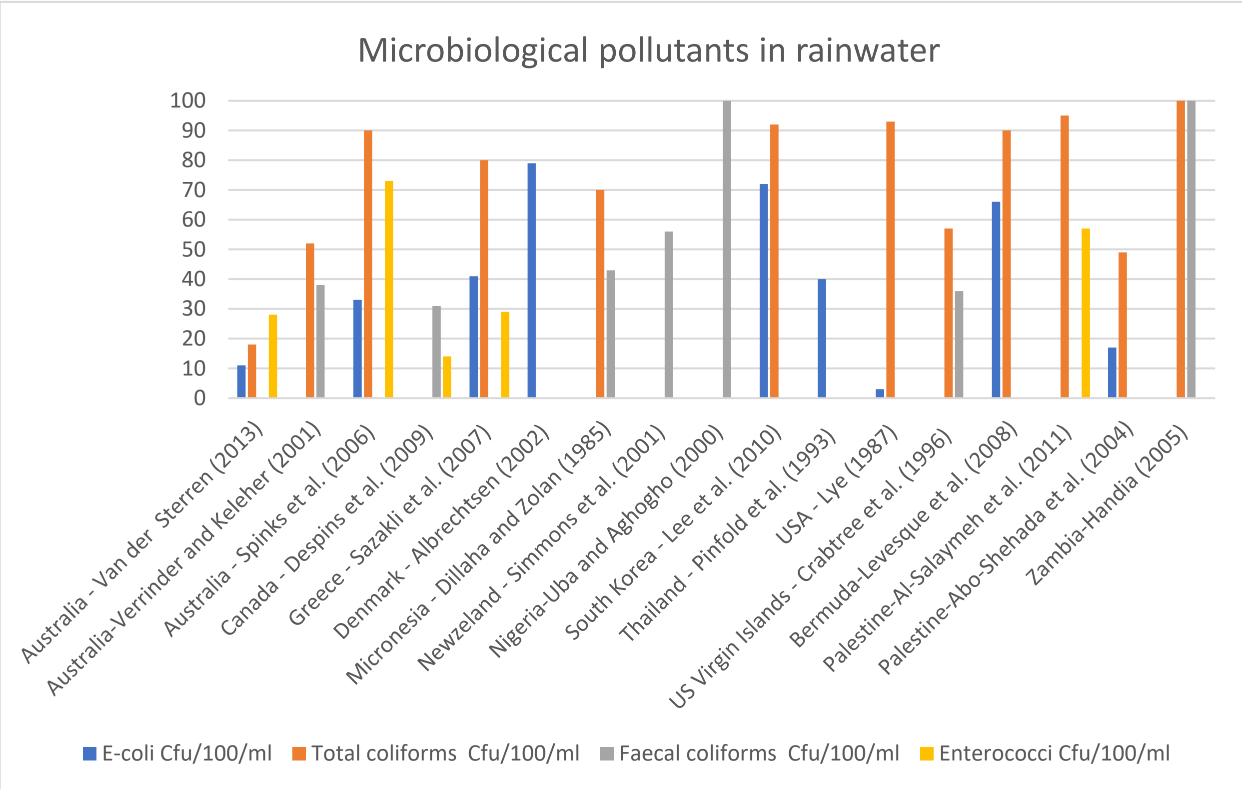

Figure 2. Microbiological pollutants in harvested rainwater (HRW) in various countries [40–56].

WHO drinking water guidelines [57] suggest that drinking water must be free from *E. coli*, total coliform, and thermotolerant coliform bacteria. However, Figure 2 demonstrates that rainwater contains a substantial amount of these microbiological pollutants [21]. They are responsible for waterborne diseases such as Diarrhoea, Cholera, Typhoid, and long-term gastrointestinal disorders [58]. It was further noticed that less awareness and poor preventive control spread these diseases quickly across the community [59]. Figure 3 further explains human health damage while consuming untreated HRW. Diarrhoea and vomiting are the most common symptoms when waterborne diseases infect the body.

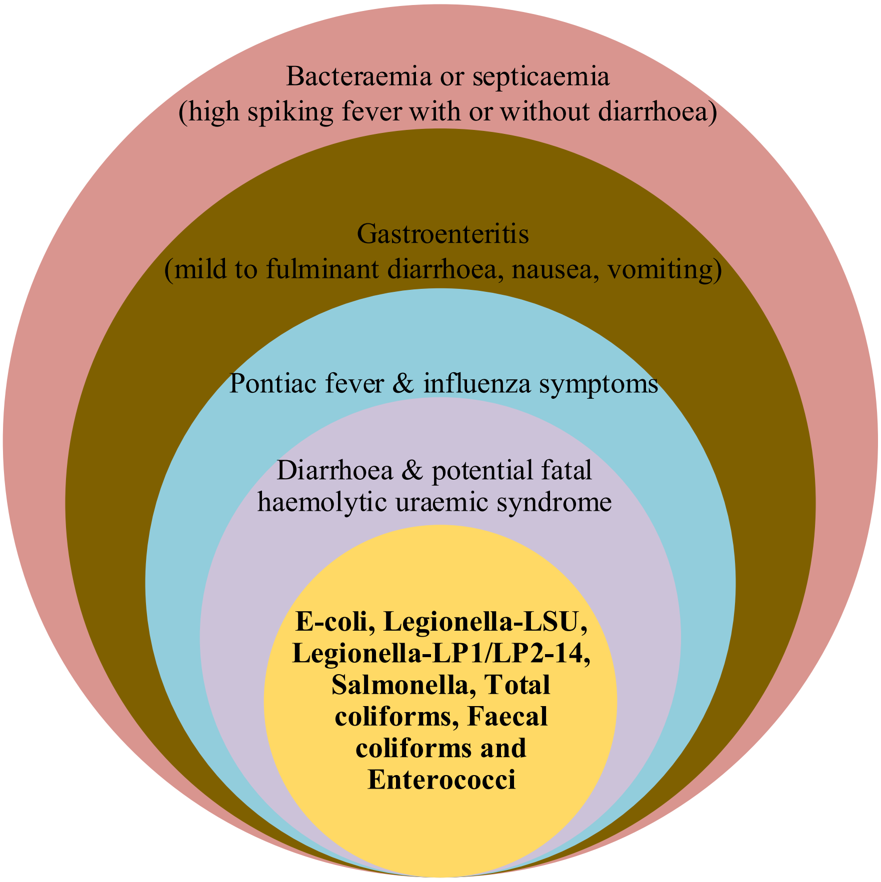

Figure 3. Microorganisms that are responsible for waterborne diseases.

Furthermore, remote communities are more vulnerable as they experience delays in having primary care services than large cities [60,61]. Therefore, the most viable solution to avert the spreading of disease in remote areas is to treat the rainwater and ensure water is clean and disinfected [59,62]. Therefore, it can be argued that untreated rainwater is not recommended to drink.

4. Opportunities for Hypochlorite Solution to Disinfect HRW

Among the disinfection methods, chlorination is the most common method in large-scale water treatment schemes [38,63–65]. It is also one of the most cost-effective water disinfection processes [66]. It is further suggested that free chlorine could be extensively efficacious against most waterborne pathogens and cause destruction of the cell DNA of these microorganisms, except for Cryptosporidium parvum oocysts and Mycobacteria species [66]. Consequently, this method could reduce waterborne diseases in many countries [32,47]. In addition, chlorine tablets and liquid pool chlorine are reasonably accessible and safe for transportation to the end-users for residential purposes [32].

Despite having some advantages, chlorination is not the preferred option in the residential or small-scale context [67,68]. Alekal et al. [67] and Burch and Thomas [29] identified three primary reasons which deter dwellers from implementing chlorination in their houses for water disinfection: (1) difficulties in mixing chemicals, (2) lack of convenience in storing chemicals, and (3) poor taste and odour issues because of incorrect dosing.

5. Chlorination for Disinfection

Various chlorine compounds are used for disinfection, such as chlorine gas, sodium-hypochlorite, chlorine tablets, calcium hypochlorite, bleaching powder, and chlorine dioxide [69,70]. The most common ones are (a) chlorine gas, (b) liquid pool chlorine (sodium hypochlorite, NaOCL) solution, and (c) calcium hypochlorite $Ca(OCl)_2$ as solid [71]. When a chlorine-contained disinfectant is administered in water, it transforms into hypo-chlorous acid (HOCl) and hypochlorite ion (OCl^-). Free available chlorine (FAC) is the combination of HOCl and OCl^- in water [70,71]. The associated chemical reactions are shown in Equations (1)–(4):

$$Cl_2 + H_2O \rightarrow HOCl + H^+ + Cl^- \quad (1)$$

$$HOCl \leftrightarrows H^+ + OCl^- \quad (2)$$

Similarly, when NaOCL and $Ca(OCl)_2$ are applied, they also form HOCl:

$$NaOCL + H_2O \rightarrow HOCl + NaOH \quad (3)$$

$$Ca(OCl)_2 + H_2O \rightarrow HOCl + Ca(OH)_2 \quad (4)$$

Wei-Ling and Jensen [70] suggested total residual chlorine combines FAC and chloramine concentrations in water. FAC predominantly reacts with ammonia in water and forms mono-chloramine first and then dichloramine [70]. Also, FAC reacts with multiple substances available in water and influences chlorine demand [70].

Amy et al. [71] suggested hypochlorite ion is an active oxidising substance. Hypochlorite ion originated from the reversible ionisation process of (HOCl) as shown in Equation (2). However, HOCl performs better than OCl^- against germs [71]. On the contrary, the unwanted microorganisms regrow in treated water after free chlorine fades out [72,73]. Amy et al. [63] stated hypochlorous acid is more stable if pH remains under 7.5; otherwise, it turns to OCl^-. Amy et al. [63] further suggested that chlorination's breaking point occurs when the total residual concentration remains unchanged.

Wei-Ling and Jensen [70] and Alim et al. [72] conducted tests on chlorine decay in laboratories, and they added chlorine and hypochlorite to water sample, respectively. They identified that total residual chlorine (TRC) dropped initially and increased later when chlorination proceeded further. When the chlorine dose exceeds the TRC, it indicates the breakpoint. Alim et al. [72] suggested that Sydney Water Corporation's standard is to maintain chlorine levels between 1 and 3 mg/L. According to WHO [73] guidelines, chlorine contact time (CT) should be held at 15 mg. min/L for proper disinfection. That means the free chlorine residual must be kept more than 0.5 mg/L in the water for 30 min.

6. Impurities Impacting Chlorination

HRW contains precursors, as listed in Table 2. The highest-concentration precursors are ammonia, total organic carbon (TOC), nitrate, and nitrite. These precursors react with chlorine in water [70] and increase chlorine demand [71]. Organic precursors such as natural organic material (NOM) react with free chlorine and produce DBPs [71]. Table 3 shows that a filter can reduce precursors from HRW; for example, TOC concentration declines by 22%, other contaminants such as nitrite by 0.2%, ammonia by 1.8%, and nitrate by 4.5%.

Table 2. Chemicals that react with and consume chlorine in water [49,54,72,74,75].

Country	NH_3 (mg/L)	NO_2 (mg/L)	NO_3 (mg/L)	Reference
Australia	0.35 ± 0.01	0.006 ± 0.001	0.196 ± 0.02	[72]
China	0.01			[74]
Jordan	0.06		1.56	[75]
South Korea	0.02		2.2	[49]
Palestine	1.4		4.2	[54]

Table 3. Chemicals reduction [64].

Parameter	Before Filtration	After Filtration	Contamination Reduction	TAP Water	Australian Standard
NH_3	0.35	0.332	1.80%	0.013	0.2
NO_2	0.006	0	0.60%	0	3
NO_3	0.196	0.151	4.50%	0.026	50
TOC	0.8874	0.667	22.00%	0	

7. Water Quality Meta-Analysis after Chlorination

Amy et al. [71] considered NOMs organic and Bromite ions nonorganic precursors. NOM concentration in water is quantified by TOC [71]. They further suggested that precursors react with disinfectants and form DBPs. Researchers have identified almost 1000 chlorination DBPs in water [76]. Table 4 shows the most common organic byproducts, which belong to three major groups, Trihalomethans (THMs), haloacetonitriles (HAAs), and halonitromethanes (HANs) [71–77]. Amy et al. [71] opined that the quantity of DBPs produced from THMs, HAAs, and HANs groups is much higher than other precursors.

Table 4. Lists of the most common DBPs. This table needs formatting.

THMs	HANs	HNMs
Chloroform Dibromochloromethane (DBCM) Bromodichloromethane (BDCM) Bromoform	Monochloroacetic (MCAA), Monobromoacetic (MBAA), Dichloroacetic (DCAA), Trichloroacetic (TCAA), Bromochloroacetic (BCAA), Dibromoacetic (DBAA), Bromodichloroacetic (BDCAA), Dibromochloroacetic (DBCAA), Tribromoacetic acid (TBAA).	Trichloroacetonitrile (TCAN), Dichloroacetonitirile (DCAN), Bromochloroacetonitrile (BCAN), Bichloroacetonitirile (DBAN).

Stefán et al. [78] conducted multiple studies on chlorination impacts on raw water at 12 drinking water treatment plants (DWTP) in Hungary. They applied chlorine gas at five plants and sodium hypochlorite at seven plants as disinfectants up to the breakpoint. Table 5 compiles the relevant data from the studies and compares how DBPs change with high and low pH, temperature, precursors, and disinfectants in water. Table 5 reveals that at a minimum temperature of 13.1 °C and Ph of 7.5, bromide ion, TOC, and bromate concentrations have been found at 0.07, 1.9, and 0.01, respectively. In that condition, the applied chlorine gas concentration was 19 mg/L, and the hypochlorite concentration was 16. After chlorination, these chemicals reacted with disinfectants and formed DBPs such as THMs, HAAs, and HANs, and the DBP concentrations were 0.014 µg/L, 0.01 µg/L and 0.001µg/ L, respectively. The study also showed that the concentration of unreacted chlorine-containing disinfectants such as chlorate, free chlorine, and combined are insignificant. On the other hand, when chlorine gas (24 mg/L) and liquid sodium hypochlorite (33 mg/L) were applied, bromide ion, TOC, and bromate concentrations increased, 0.14 µg/L, 0.13 µg/L, and 0.02 µg/L, in water, respectively. The pH and temperature in water were recorded at 8.5 and 54.2 °C. THMs, HAAs, and HANs concentrations reached 0.14 µg/L, 0.13 µg/L, and 0.02 µg/ L, respectively.

Table 5. Raw water quality and DBP formation after chlorination (disinfect chemicals Cl_2 and NAOCl) [78].

Physical and Chemical Properties	pH	Temp	Raw Water mg/L					
			Br^- mg/L	TOC. mg/L	Bromate mg/L	$NH4^+$ mg/L	NaOCL mg/L	CL_2 gas mg/L
Maximum	8.1	54.2	0.34	11	0.34	4.5	33	24
Minimum	7.5	13.1	0.07	1.9	0.01	0.76	19	16
DBP mg/L								
Physical and Chemical Properties	THMs µg/L	HAAs µg/L	HANs µg/L	Chlorite ion, mg/L	Chlorate ion, mg/L	Free chlorine, mg/L	Combine chlorine, mg/L	
Maximum	0.14	0.13	0.02	<0	0.002	0.001	0.002	
Minimum	0.01	0.01	<0		<0	<0	0	

Table 6 shows that HRW samples from various countries contain precursor that can form DBP if the disinfectant is applied. For example, in the coastal mega-cities of southern China, the TOC concentration from HRW was recorded 1.03 to 4.23 mg/L, Ontario (Canada) 0.53 to 5.7 mg/L, Alaska (USA) 0.53 to 5.7 mg/L, Italy (University of Salerno, Fisciano, Italy) 5.1 to 7.1 mg/L, Pakistan (Karachi) 3.31 to 7.9 mg/L, and Australia (Werrington) 0.89 to 0.9 mg/L.

Table 6. Disinfection byproduct precursor found in HRW in different countries [43,72,79–82].

Parameter	Southern China	Ontario/ Canada	Alaska/ USA	Fisciano SA/Italy	Karachi/ Pakistan	Werrington/ Australia
TOC mg/L Reference	1.03–4.21 [79]	1.8–8.5 [43]	0.53–5.7 [80]	6.61–5.19 [81]	12 [82]	0.89 ± 0.05 [72]

8. Disinfection Byproduct and Health Concerns

In 1900, cholera spread was ended in the Western world by applying chlorination in the municipality-supplied water. However, in 1974 some researchers discovered that NOM reacted with chlorine and formed trihalomethanes, and the concentration reached 160 µg/L [83,84]. It has been proven that DBPs affect the human body when exposed to public water supplies and swimming pools [36,85]. The health risk of DBPs is high because of their carcinogenic nature [38].

Chlorination is the most common disinfection method globally. Researchers have identified 600 to 700 chlorinated DBPs in drinking water [36,76]. Besides the chlorinated DBPs, other disinfectants (such as hydrogen peroxide, ozone, and chloramines) produce DBPs when they react with NOM in water [35]. Hence, trihalomethanes (THMs) and haloacetic concentrations are much higher than other DBPs in drinking water [36]. THM has four species that may cause cancer [86]. These are TTHMs, chloroform, bromodichloromethane (BDCM), chlorodibromomethane (CDBM), and bromoform. Mishaqa et al. [86] also suggested that THMs can enter into the human body via three routes. They are oral ingestion, inhalation, and dermal absorption. Table 7 shows that by inhalation 97.86% BDCM, 97.66% CDBM, 81.46% Bromoform, and 68.95% TTHMs can enter a human body. On the other hand, chloroform can enter a human body at 55.23% through dermal absorption.

Table 7. Percentage of THMs exposure route to human body [79].

CDBP Contaminants	Oral Ingestion	In halation	Dermal Absorption
Chloroform	37.97	6.8	55.23
BDCM	0.78	97.86	1.36
CDBM	0.93	97.66	1.41
Bromoform	16.6	81.46	2.14
TTHMs	2.1	68.95	27.94

WHO published a guideline [85] in 1993, which outlined the allowable limits of the DBPs in drinking water. Since then, epidemiologists have been researching the link between THM4 resulting from chlorinated water and various health occurrences (such as cancers, miscarriages, and premature baby birth) [87]. Li and Mitch [87] further identified that THM4 concentration greater than 50 micrograms per litre increased bladder cancer chances. On the contrary, no conclusive evidence was found regarding DBP's adverse impact on women and children [36]. Table 8 demonstrates the acceptable concentration of common DBPs specified by various countries and WHO. It is distinctly noted that, for an unknown reason, not all countries are concerned about all variances of DBPs. That is why they did not set any limit on some DBPs. However, WHO has identified the DBPs and their permissible limits vary extensively. If HRW is chlorinated, the DBPs level should be checked occasionally to meet the standard mentioned in Figure 4.

Table 8. HRW quality analysis on pre and post-filtration, and compared with tap water and Australian standard.

Sources of Water	pH	Electrical Conductivity	Dissolved Oxygen	NH_3	NO_2	NO_3	TOC	Turbidity
Australian Standard	6.5–8.5	200–800		0.2	3	50		5
Sydney Water supplied water quality	6.73 ± 0.2	247 ± 2	7.5 ± 1	0.013 ± 0.01	0 ± 0.01	0.026 ± 0.02	0.23 ± 0.05	0 ± 0.5
HRW (after filtration)	6.59 ± 0.2	71 ± 2	9 ± 1	0.332 ± 0.01	0 ± 0.001	0.151 ± 0.02	0.667 ± 0.05	2 ± 0.5
HRW(before filtration)	6 ± 0.2	21.54 ± 2	9.5 ± 1	0.35 ± 0.01	0.006 ± 0.001	0.196 ± 0.02	0.887 ± 0.05	2 ± 0.5

THM

DBPs	WHO[1]	US[2]	UK[3]	Canada[4]	EU[5]	South Africa[6]	Japan[7]	China[8]	Australia[9]	New Zealand[10]	India[11]
Chloroform	0.3	0.08	-	-	-	0.3	0.06	0.06	0.4	0.2	
BDCM	0.06	0.08				0.06	0.03	0.06		0.06	0.06
DBCM	0.1	0.08				0.1	0.1	0.1	0.15		
Bromo form	0.1	0.08				0.1					
TTHM		0.08				0.1	0.1		0.25		

HAA

DBPs	WHO	US	UK	Canada	EU	South Africa	Japan	China	Australia	Newzealand	India
Total HAAs	-	0.06	-	0.08	0.06	-	-	-	-	-	
Mono chloroacetic acid	0.02	0.06					0.02		0.15	0.02	
Dichloroacetic acid	0.05	0.06					0.03	0.05	0.1	0.05	
Tri chloro acetic acid	0.2	0.06					0.03	0.1	0.1	0.2	
Di bromoacetic acid											
Bromochloro acetic acid											

HAN

DBPs	WHO	US	UK	Canada	EU	South Africa	Japan	China	Australia	Newzealand	India
Di chloroaceonitrile	0.02						0.04			0.02	
Trichloro acetonitrile											
Di bromoacetonitrile	0.07									0.08	

Figure 4. Disinfection byproduct (DBP) limit across the world [1] [38], [2] [74], [4] [88], [3] [89], [5] [90], [6] [91], [7] [92], [8] [93], [9] [94], [10] [95], [11] [96].

9. DBP Reduction by Pretreatment before Chlorination of Water

Several researchers identified three elements such as NOM [87], TOC [78], and DOC [97,98] as the precursors that form DBPs in chlorination. In addition, temperature and free chlorine have shown strong relationships in growing chlorinated DBPs [78]. These DBPs had been produced mainly at the chlorine breakpoint stage. However, many water filtration treatment plants use excessive sodium hypochlorite [78] for proper disinfection. Since hypochlorite ions reacted with water's impurities, excess hypochlorite reduced some solids dissolved in water [78]. However, this strategy is likely to form DBPs. On the other hand, many researchers have considered coagulation and filtration as effective pre-treatment processes before disinfection [97–100]. For lessening DBPs in chlorinated water, coagulation/flocculation and filtration are effective options [101–103], as discussed below.

9.1. Coagulation

Coagulation is one of the effective ways to remove NOM from water [102]. Randdtke [102] suggested that organic substances are removed through coagulation by three main mechanisms: (i) colloid destabilization, (ii) precipitation, and (iii) coprecipitation. Iron salts (ferric and ferrous chloride) and alum are used as coagulants in many water treatment plants. These coagulants help NOM to precipitate quicker and reduce the solid load on filters. However, coagulation is impacted by various factors such as coagulation conditions, characteristics of NOM, nature, and concentrations of inorganic compounds, and the design and operation of a treatment plant [103]. Uyak and Toroz [98] stated that ferric chloride removed more DOC than alum. They further suggested that optimum coagulation can be achieved at pH 5.25 for ferric chloride and 5.50 for alum. For more NOM and TOC reduction, additional treatment is required (such as filtration) [98].

9.2. Filtration

A filter separates suspended solids from a mixture of solid and liquid when this mixture passes through the filter media [104]. They can remove suspended solid particles, DOC/NOM, and TOC from raw water. Various media are used for filtration, such as granular activated carbon filters [33], sand filters [105], cloth filters [106,107] and ceramic filters [26].

Jun et al. [105] found that the depth of a slow sand filter significantly impacts the removal of DOC from raw water. Researchers identified that the ability to remove solid particles varies with the types of filter media, their depth, and filter run time [105]. Figure 5 shows DOC removal efficiency by filters at different slow sand filter depths.

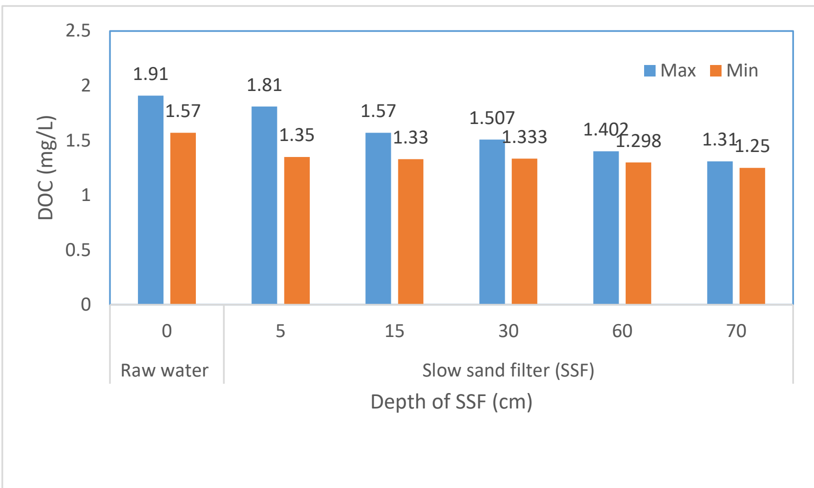

Figure 5. Comparison of dissolved organic carbon (DOC) in raw water and filtered water at various depth of slow sand filter (SSF) [105].

On the other hand, Zeng et al. [100] suggested that flow direction and filter media affect DOC removal. In an up-flow roughing filter with ceramic media, DOC removal efficiency is 8% [100], and in a down-flow roughing filter with a slow sand filter it is 3% [108].

The granular activated carbon (GAC) filter's biodegradation also plays an essential role in improving TOC removal efficiency [109]. In GAC filters, DOC removal efficiency is initially 77–81%, and after 200 days, it is 13% [110]. Furthermore, Van der Aa et al. [110] suggested that biodegradation occurs due to the presence of NOM. They further identified pre-oxidation by converting ozone DOC into assimilation organic carbon (AOC). As a result, AOC removal efficiency improved to 70%.

10. Suitability of HRW Treatment on a Small Scale

The argument regarding the feasibility of HRW treatment has appeared to ensure a clean water supply to remote communities [111]. Gomez and Teixera [111] suggested that HRW treatment is technically and economically feasible. Alim et al. [72] conducted an experiment where they installed a rainwater tank fitted with a dual media filter at the University of Western Sydney, Werrington in Australia. They compared HRW quality before and after filtration. Their findings are summarised in Table 8. They identified that most contaminants were reduced after filtration, except turbidity. Though the turbidity of filtered HRW was found to be four times higher than the supply water, their filtered HRW complied with the local drinking water guideline. However, as the faecal coliform, E-coli, and enterococci were not tested, it is unsure whether the filtered HRW complied with the Australian Drinking water guidelines under disinfection requirements.

On the contrary, in countries where centralised water is less expensive and the water sources have sufficient capacity, such as the Netherlands, HRW is not considered a cost-effective option for potable usage [112]. On the other hand, from 2008 onwards, chlorination was no longer applied to water for disinfection in the Netherlands. Instead, disinfection is conducted by a UV (ultraviolet) system, hydrogen peroxide (H_2O_2), or a combination of UV and H_2O_2 [113].

On the other hand, in Australia, Senevirathna et al. [33] built an affordable treatment system that can treat HRW (500 L/day). It has an aerator, an adsorption unit, a sedimentation unit, and a UV disinfection unit [33]. The whole package is powered by solar power. But for 24/7 treatment, the system needs 24 h uninterrupted power supply, which solar panels cannot provide.

As for applying chlorination on a small scale, Pickering et al. [34] conducted studies in Dhaka, Bangladesh. They chlorinated water in two ways, (i) by an automated chlorinator and (ii) by chlorine tablets. The automated chlorinator was connected to a hand pump. However, the dosing rate was not consistent with the design expectation. They did not arrange any treatment before chlorination. The study found some concerns about bad taste and odour in the chlorinated water; however, this study had two positive outcomes: (i) micro-biological contaminants dropped in the treated drinking water, and (ii) it presented a further opportunity for automatic chlorination in the domestic context [34].

On the other hand, Neto et al. [114] engaged a slow sand filter and chlorine tablet to treat HRW. The disinfectant was chlorine tablets (Genco, composed of trichloroisocyanuric acid with 90% active chlorine). In the study, they noticed free chlorine (FRC) varied with the outflow in the chlorinator; 81% recorded FRC values were between 0 to 2 mg/L and 23% below 0.5 mg/L. The treated water was used mainly for non-ptoable purposes such as toilet flushing and cooling system and the maximum efficiency of total coliform and E.coli were 4 and 3 logarithmic units, respectively. Their study proved that (i) HRW could save a significant amount of potable water for nonpotable purposes, and (ii) proper treatment can protect human health from unexpected contaminations.

11. Discussion

11.1. Challenges with HRW Disinfection and How Hypochlorite Can Resolve It

From a heavy metal contamination point of view, HRW is generally safe, even though some exceptions are explicit [22]. In contrast, microbiological contaminations are very common in HRW. Figure 3 shows that HRW contains unacceptable concentrations of microbial impurities and can cause harm to public health. Hence, it is detrimental to health to drink untreated HRW. Regrettably, HRW treatment from the storage tanks at the domestic level has failed to obtain much attention. The possible reasons are:

1. Mains water is available to a large number of consumers who live in cities and they have little concern to treat HRW.
2. As many HRW consumers are from impoverished areas, the treatment is not affordable to them.
3. People suffering from water scarcity and contamination accept untreated HRW as they may not have any other options.
4. Some consumers put taste priority over health impacts caused by germs. In addition, some people have little awareness and education about water contaminants and their health impacts.
5. The poor investment return.
6. Lack of data on organic content in HRW to develop design criteria.

Many researchers have proposed low-cost and straightforward solutions for disinfection, such as heating water in bottles with solar power (known as thermal pasteurization), treating water in bottles with ultraviolet (UV) rays (known as UV method), and diluting sodium hypochlorite solution or tablet in water (known as chlorination method). Those low-cost solutions showed improvement against spreading waterborne diseases. However, these methods have not received substantial attention from dwellers for two reasons. Firstly, the inconvenience and inconsistent efficacy of producing clean and disinfected drinking water in all weather conditions [22]. For example, solar pasteurization can be applied during summertime when intense sunlight is available, UV needs an uninterrupted power supply, and to use hypochlorite solution or tablets; convenient automatic systems are unavailable. Secondly, proposed solutions show insufficient evidence of protecting treated water against recontamination when it is stored for extended periods. For example, after UV, solar, or thermal disinfection, germs may regrow in the reserved water if the water remains in the storage tank longer [115].

To encounter recontamination, Latif et al. [22] argued that hypochlorite solution has residual effects that can disinfect water irrespective of weather conditions and deter microorganisms from growing for a certain period after disinfection. Moreover, hypochlorite solution, especially sodium hypochlorite is the most desirable due to its low cost and availability among other chemical disinfectants (such as hydrogen peroxide, ozone, and hypochlorite solution) [115].

11.2. Potential Challenges with HRW Disinfection by Hypochlorite and Possible Solution

Section 4 demonstrates that when hypochlorite is injected, HOCl (hypochlorous acid) and OCl- (hypochlorite) are formed, known as free chlorine. Free chlorine not only disinfects but also reacts with organic contents in water and forms DBPs. Researchers identified that OCL^- produces more DBPs than HOCL, and HOCL changes to OCL^- through ionisation when pH is maintained over 7.5 in chlorinated water [71]. It can be suggested that the direct application of hypochlorite solution will likely form DBPs in HRW in the locations where pH is high such as 8.31 in Kefalonia Island (Greece) [44], and 8.2 in Hebron (Palestine) [54]. Therefore, high pH in HRW needs pH treatment before using hypochlorite solution and consumption as potable water.

On the other hand, consumers' satisfaction regarding the taste of disinfected water by sodium-hypochlorite was not very optimistic. In a survey by Pickering et al. [34], the treatment was limited to chlorination only, and no pre-treatment was performed to lessen solid contents in water. After injecting sodium hypochlorite by an automatic disinfection system, the germs were reduced significantly. The survey indicated that the taste of chlorinated water was one of the significant reasons many householders disliked their technology for automatic chlorination. Though exact causes were not determined, it can be argued that chlorine DBPs could be one of the reasons. According to Jamal et al. [115], the TDS of the supplied water in Dhaka city is high for various reasons, which is between 151 to 407 mg/L. On the contrary, it has also been noted that Neto et al. [114] used chlorine tablets in disinfecting HRW for nonpotable purposes and did not test DBP formation in chlorine-treated water. Hence, conducting a further survey on hypochlorite-treated HRW like Pickering et al. [34] and measuring chlorine DBPs will be prudent.

DBP concentration could differ in the chlorinated water at various locations at the same chlorine dose rate because of the quality variation of HRW [44]. Table 5 suggests that DBPs were formed when raw water was chlorinated until it reached the breaking point. Furthermore, Table 5 shows that temperature, pH, and DBP precursors (Br^-, TOC, $NH4^4$) directly influence DBP concentration in water. Likewise, Tables 3 and 7 indicate that rainwater contains TOC. Mendez et al. [116] and Abbasi and Abbasi [117] identified that TOC concentrations and pH in HRW differ with countries. The possible reasons for these variations in HRW quality include (a) variation of roof materials, (b) surrounding environment, and (c) differences among water catchments.

11.3. Analysis of DBPs Formation in HRW by Comparison with Other Raw Water and WHO-Acceptable Limits

Stefan et al. [78] suggested (Table 6) that sodium hypochlorite was added to raw water, and DBPs were produced, which exceeded WHO accepted limits. With a similar quality of HRW, DBPs can grow, and their concentrations are higher than acceptable limits if an equivalent amount of hypochlorite solution is injected into raw water. Therefore, it is conceptually undeniable that if chlorination is used in untreated HRW DBPs will be formed. However, to prove this argument, not enough supporting data are available. The possible main reason is chlorination in HRW is not widely applied.

The WHO and the environmental protection authorities in multiple countries have outlined the acceptable limits of DBPs in the drinking water guideline due to growing concern about the health impact of daily intake of DBPs through chlorinated drinking water. However, Figure 4 indicates the inconsistency in acceptable limits of DBPs in drinking water among the environmental protection authorities of the respective countries. For example, in the USA [88] and Canada [118], the THM's acceptable limit is 0.08 microgram/L, the lowest compared to WHO and other countries. Some countries (such as the UK, China, Japan, South Africa, Australia, New Zealand and India) have not considered limiting HAAs and HANs substances. On the other hand, the USA and WHO have identified HAA and HANs as toxic products and set their acceptable limits in potable water.

11.4. DBP Reduction through Pre-Treatment

Section 9 demonstrates that pre-treatment methods can reduce the DBPs concentration by minimising TOC before disinfection, which has been noticed in large drinking water treatment plants. Similarly, HRW pretreatment and chlorination are technically feasible for small-scale drinking. Evidence has been found in some studies where the ultraviolet (UV) process [33] and filtration [72] have treated HRW, and chlorination has disinfected stored water on a small scale [34]. Generally, HRW does not contain too much TOC and DOC, and HRW needs filtration and other techniques to reduce TOC and DOC contents which will lessen DBPs if chlorination is adopted. Apart from DBP formation in drinking water, the following challenges are still present in applying chlorination at the domestic scale [67]:

1. Storing sodium hypochlorite and mixing it safely before dosing;
2. Reduction of odour and improving taste and
3. Removing Cryptosporidium parvum oocysts and Giardia lamblia cysts.

A possible solution for challenge 1 is to design and build a treatment system with automatic filtration, coagulation, and chlorination. A filtration system can address challenge 2. Ultraviolet (UV) disinfection can fix challenge 3. Senevirathna et al. [33] built a system that can disinfect Cryptosporidium parvum oocysts and Giardia lamblia cysts from water by UV. In addition, they also applied a granular activated carbon filter for filtration and aeration to improve the water taste and disinfection performance, respectively.

In contrast, according to Table 1 UV system cannot protect water from recontamination if it is stored for an extended period since UV does not have residual protection. Therefore, all three challenges can be overcome if a water treatment system has a pretreatment filter, UV, and a chlorination arrangement for disinfection. If pathogens like Cryptosporidium parvum oocysts and Giardia lamblia cysts are not significantly present in HRW, filtration, and chlorination can be a good combination. Another advantage of filtering water before chlorination is that fewer DBPs will be developed.

To consider building a HRW treatment system in a household context, proper guidance on the treatment of HRW is unavoidable. The possible reason for not outlining a sustainable HRW treatment method and guidelines is minimal research on quantifying the organic content in HRW worldwide. The same logic applies for not developing reliable and sustainable pretreatment systems for HRW. Nonetheless, it is likely to form DBPs in HRW if chlorination is used without minimizing the organic materials through treatment. Further studies are required on chlorine DBPs in HRW.

12. Further Research Opportunities

12.1. Insufficient Data on TOC from HRW

As TOC reacts with hypochlorite solution and produces DBPs, further data on organic contents in HRW are required to develop sustainable domestic treatment systems. On the other hand, many research studies have developed significant databases on TOC and DBP quantities in different drinking water sources compared to HRW. Therefore, collecting more data on TOC concentration in HRW will be prudent. It can add three benefits. Firstly, it can contribute to designing an appropriate filter to capture the expected quantity of TOC. Secondly, the optimum chlorine dose can be determined in the designing phase, resulting in fewer chlorine DBPs. Thirdly, chlorine demand can be calculated beforehand as it is essential to establish the correct disinfectant dose rate.

12.2. Develop an Independent Sustainable Automated Treatment Method

Many ways have been developed to treat HRW. An automatic treatment system for treating HRW can attract many dwellers who prefer rainwater over municipal water. However, the lack of convenience and automation in the current domestic treatment tools are the leading causes of getting little attention from most of the residents in urban and remote locations. On the contrary, the economic payback of the domestic HRW treatment system is also a critical factor influencing the implementation of HRW treatment facilities.

12.3. Insufficient Data on Chlorine DBPs in HRW

As chlorine is not widely used for treating HRW, not enough data are available to explore the impact of chlorination on HRW. Sometimes, liquid sodium hypochlorite or chlorine tablets were applied as low-cost disinfectants, but chlorine DBPs were not measured.

12.4. Outlining a Good HRW Treatment Strategy

It is desirable to develop a robust HRW treatment strategy based on multiple performance tests on various pre-treatment, and disinfection methods and their combinations. Once a suitable method of disinfecting HRW is found for a given community, a policy should be developed for its broader and safer implementation.

Not much data are available for treating HRW with multiple treatment methods. Functional materials and weather conditions in a given location can dictate selecting appropriate pre-treatment and disinfection methods. A single treatment method may not produce drinking water from HRW and comply with the WHO guidelines, such as, during the winter season, solar disinfection may not perform adequately. UV disinfection cannot function in highly remote areas when a conventional power supply is unavailable or partially available.

On the other hand, with proper design and instrumentation, disinfection by hypochlorite solution can be achieved at most locations, even though some germs can survive against it. Therefore, disinfection by only hypochlorite solution may not be able to provide complete quality treatment. Hence, combining other pre-treatment and disinfection concepts can improve outcomes. Thus, the treatment process flow chart, suggested in Figure 6 could be a sustainable domestic HRW treatment strategy.

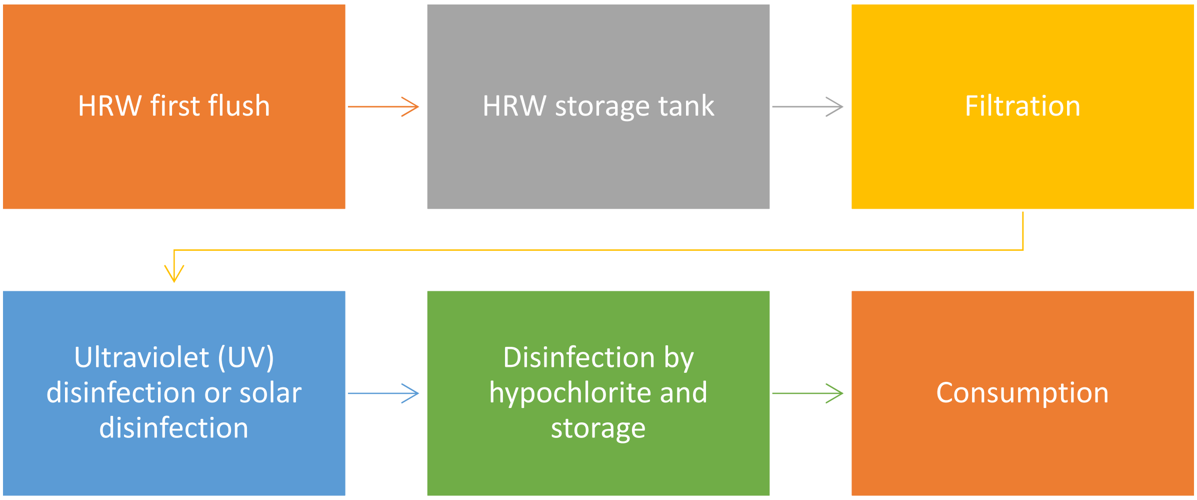

Figure 6. HRW treatment strategy.

If chlorination is recommended to disinfect HRW in developing countries, simplified water quality testing facilities should be grown to check for DBPs and other water quality parameters. In developed countries, water quality testing facilities are more accessible to the public than in developing countries.

13. Conclusions

Residual effect, cost, and availability have made hypochlorite solutions popular for large-scale water treatment facilities. It is not, however, widely used to disinfect HRW for small volumes. There is a lack of research on disinfecting HRW mainly for two reasons, (i) people believe rainwater is clean and safe to drink, and (ii) with low demand, HRW treatment fails to achieve attention from investors.

As chlorine is not widely used for treating HRW, not enough data are available to explore the impact of chlorination on HRW. HRW should be subjected to adequate pretreatment to reduce organic contents before chlorination so that the levels of DBPs are reduced.

The use of hypochlorite solution for HRW treatment within a domestic context may not be the only cost-effective method. Hypochlorite solution combined with other treatment methods may give better outcomes for treating HRW. In some situations, at remote locations, the use of hypochlorite solution might be the best solution, such as, in relatively less sunny areas, poor remote communities, and countries that are struggling to build infrastructure to provide clean water to all their citizens.

The use of hypochlorite solutions for HRW treatment requires more attention and financial investments from industrial and financial organisations. The invested funds can sponsor research to resolve the outstanding issues that have not been explored yet on HRW treatment by hypochlorite solution at the household level.

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