

## Article

# Onsite Chlorination of Greywater in a Vertical Flow Constructed Wetland—Significance of Trihalomethane Formation

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**Abstract:** To reduce public health hazards, greywater reuse may involve disinfection, which is often performed through chlorination. The formation of toxic disinfection by-products is a negative side-effect of chlorine's reaction with organic matter, of which trihalomethanes (THM) are one of the most dominant (though not most toxic) groups. Greywater treatment in vertical flow constructed wetlands leads to a decrease in dissolved organic matter. We hypothesized that these dissolved organic carbon (DOC) changes would be reflected in differences in THM formation. Greywater samples, at different treatment levels (i.e., decreasing organic matter content), were exposed to 5 mg/L of chlorine for 1 h. THM formation in raw greywater samples was significantly lower than in the more treated (recirculated) samples, despite their significantly higher DO concentrations. This trend was verified in six different systems. Furthermore, this was also shown when greywater was exposed to higher chlorine doses (25 and 50 mg/L). It is suggested that the increase in THM formation for longer recirculated water is the result of two factors: competition between a higher number of reactive sites in the raw water's organic matter, which leads to smaller THM yields, and changes in the abundance of THM formation moieties in the recirculated water's DOC. The latter was reflected in the SUVA increase in the treated water. Overall, THM formation, following treated greywater chlorination at the lower chlorine concentration studied, is not expected to pose an environmental health risk when the water is reused for irrigation.

**Keywords:** disinfection by-products; greywater; DOC; chlorination



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## 1. Introduction

Greywater (GW) is domestic wastewater, excluding the stream from the toilet and, usually, the one from the kitchen. Responsible on-site GW reuse may result in water and economic savings, as well as pollution reduction. For example, it was estimated that urban reuse of GW for toilet flushing alone may reduce approximately 30% of a building's water consumption [1]. It was also estimated that 30% savings in multi-story buildings in Israel may result in an annual overall reduction of 150 MCM of freshwater use. The latter is equal to the savings of one desalination plant, without all of the plant's environmental impacts (e.g., land use, electricity demand, brine treatment, and greenhouse-gas emissions) [1,2]. Yet, inappropriate reuse may compromise public health and the environment [3]. Thus, the potential greywater-associated risks should be identified and addressed to enhance safe reuse. Microbial hazards are a major source of concern when on-site GW reuse for irrigation is considered [4]. Indeed, with regard to the recent COVID-19 pandemic, it was just discovered that SARS-CoVs survive in domestic wastewater for several days and that conventional on-site wastewater treatment, such as wetlands, bioreactors, and even more advanced membrane bioreactors [5], might only partially remove them [6]. The authors postulated that either safe disposal or reuse depends on the efficacy of the final disinfection. Similar findings with the presence of other pathogens in GW (e.g., [7]) have motivated

many regulatory agencies to recommend application of water disinfection in order to meet irrigation standards and ensure safe reuse [8,9]. Many countries have standards for greywater reuse [10]. In Israel, for example, the standard for the *Escherichia coli* count set by the Ministry of Health for unlimited wastewater, including greywater reuse, is less than 1 CFU/100 mL, and chlorine disinfection is required [11]. In the UK, the standard allows no more than 250 CFU/100 mL for garden watering, which also implies the use of disinfection to meet the standards [12].

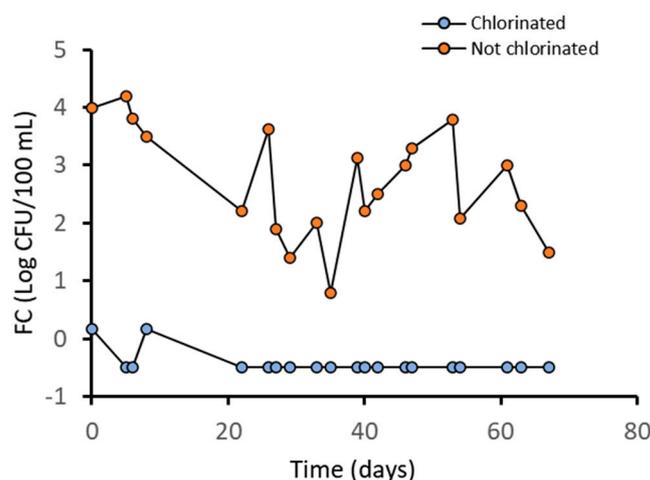
Chlorination is one of the most popular disinfection techniques for small on-site greywater treatment systems [4,13,14]. As a strong oxidizer, chlorine promotes the inactivation of, or lethality effects on, microorganisms, reducing pathogen hazards. Despite the clear positive aspects of chlorine application, chlorine can further react with organic matter and form toxic and potentially carcinogenic disinfection by-products (DBP) [15]. Of the disinfection by-products formed, trihalomethanes (THM) are one of the most abundant groups [16,17]. Although disinfection by-products of higher toxicities are known and increasingly documented, such as the chlorinated furanone “mutagen X” [18], most of them are not routinely monitored. Moreover, drinking water regulations [15], as well as swimming pool guidelines [19], do not take these into consideration. THM formation has been intensively studied in drinking water since the 1970s [15,17]. Studies on other water types, such as wastewater, have been increasingly undertaken as well (e.g., [20]), though they are less numerous for greywater. Although greywater is not used for drinking water, concerns were raised regarding the consumption of raw vegetables that are irrigated with greywater, as well as aerosol inhalation and dermal contact risks where greywater is reused [21,22]. Regardless, even if health risks may be considered low, understanding the extent of THM formation, especially as on-site greywater reuse practice is expected to expand [23], is of interest.

THM formation is positively correlated with chlorine dose and bromide concentrations [24,25]. The organic matter, on the other hand, plays a more complex role in THM formation. Organic matter in wastewater may be extremely heterogeneous in molecular size and structure, leading to differences in DBP formation, kinetics, and distribution. For example, resorcinol structures are fast-reacting THM precursors and present high yields, whereas phenolic compounds react more slowly, with lower THM yields [16,24,26]. Reviewing a large number of studies [27], it may be generally accepted that the hydrophobic organic fraction and the 1–10 kDa molecular weight fraction are of greater significance as THM precursors. Nevertheless, contradictory observations can be identified as well, stemming from differences, for example, in organic matter isolation procedures, chlorination methodologies, origins of dissolved organic matter (DOM), and the nomenclature used to describe DOM fractions [27]. This is becoming even more complex in the case of greywater, where not only natural organic matter is of concern, but also organic compounds in household products. Greywater was shown to present high oil and grease components, for example [28], and surfactants and detergents [29,30], which may react with chlorine. For instance, the widely used antibacterial soap component, triclosan, readily reacts with chlorine [31]. In addition, greywater quality was shown to change considerably over short periods of time [32], increasing the complexity of THM formation assessments.

Greywater treatment systems, as well as the treatment systems of other water types in constructed wetlands, differ in design, water source, water quality, and management. DBP precursors and THM formation should be strongly affected by these differences [33]. For example, no changes in DBP precursors were apparent throughout a small wetland area with short residence times in a study focusing on constructed wetlands that received agricultural return flows [33]; a decrease in THM formation was observed with increasing treatment time in a second study, focusing on a vertical and surface flow constructed wetland that received polluted river water [34]; and a THM increase was observed in wetland water that received secondary effluents, relative to THM in the inflow in a third study of a subsurface constructed wetland [35]. Clearly, DBP formation potential must be studied in terms of the system’s specific design and characteristics.

There is a growing global interest in greywater reuse, with developed countries such as Australia, the USA, and Japan leading the way [36].

The recirculating vertical flow constructed wetland (RVFCW) greywater system is economically sound, low-tech, and easily maintained [37,38]. In RVFCW systems, greywater is used for irrigation after it is circulated for several hours. While it is well documented that longer circulation is positive in the aspect of reducing greywater's biochemical oxygen demand (BOD), total suspended solids (TSS), and ammonium ( $\text{NH}_4^+$ ) [38], for example, the effects of circulation time and the consequent changes in water characteristics on THM formation are unknown. Our previous studies have indicated chlorine's efficiency in reducing pathogens and antibiotic resistance (Figure 1; [39,40]), and it is currently the most used disinfection technique for greywater [23]; thus, studying the magnitude of THM by-products is of interest. We hypothesized that changes in DOC concentrations in the treated greywater would result in differences in THM formation. This, however, was not yet studied for this system. The objective of this work was, therefore, to study the influence of greywater recirculation (treatment level) on THM formation in an on-site vertical flow recirculating constructed wetland. To this end, raw and treated greywater from six distinct vertical flow greywater systems were tested.



**Figure 1.** Fecal coliform count of treated, but not disinfected, greywater and after chlorination over time. Values were monitored for a single recirculating vertical flow constructed wetland over >60 days. Treated greywater flow through slow-release trichloroisocyanuric acid (TCCA) chlorine tablets (Kingnod Co. Ltd., Qingdao, China) containing 90% active chlorine. Tablets were placed in an irrigation filter casing, and water flow rate was 100 L/h, with an expected contact time of 36 s of chlorine disinfection.

## 2. Materials and Methods

### 2.1. Greywater and Its Treatment

This study was conducted by comparing six recirculating vertical flow constructed wetland (RVFCW) systems that have been treating greywater on-site for more than seven years each [41,42]. All six greywater systems are located in Midreshet Ben-Gurion in the Negev Desert, Israel. The collected raw water is considered light greywater, including sink, shower, and laundry effluent.

An RVFCW is composed of two containers (0.95 m wide  $\times$  0.95 m long  $\times$  0.55 m high; approximately 500 L each), with one stacked directly on top of the other. Raw GW flows through the upper container, which comprises soil planted with vegetation and a filter bed, to the lower container, which is located directly beneath and serves as a water reservoir. GW is continuously recirculated through the system at a given rate by a pump (a depiction of the RVFCW is provided in Figure S1). The system details are described by Gross et al. [37].

The greywater treatment follows a daily cycle to ensure that sufficient water quality is attained for unlimited garden irrigation [11]. Treated greywater is usable after a circulation period of at least 6 h. Previous studies have shown that this circulation time is required to ensure adequate water quality [41]. Once 70% of the treated water is used, irrigation ceases, and a new batch of raw greywater is pumped into the RVFCW. The system stops receiving raw greywater either when the basin becomes full or at the point of 6 h before the next irrigation cycle. This treatment cycle continues in this way indefinitely.

## 2.2. Experimental Setup

Two sampling campaigns were carried out. In the first, raw greywater was grab sampled in all systems, as well as water after 6 h of circulation. Water samples were analyzed on-site for electrical conductivity (EC), dissolved oxygen (DO), pH, and temperature (MultiLine 3420, WTW, Oberbayern, Germany), as well as for free chlorine (C201 chlorine colorimeter, Eutech). Samples were kept on ice for no longer than 6 h and transferred to the lab where they were kept refrigerated until further analysis.

In the lab, samples of raw and treated greywater from each household were dosed with sodium hypochlorite (Sigma-Aldrich, Darmstadt, Germany) to a calculated initial concentration of 5 mg/L of free chlorine. This experimental setup was intended to mimic a realistic field chlorination scenario, in which a constant dose of chlorine is often applied, regardless of water quality. The experiment was performed in duplicates in 40 mL glass vials sealed with Teflon-lined septa. Samples were analyzed after incubation of 1 h at room temperature. The reaction was quenched with ammonium chloride and analyzed for THM concentrations. DOC was measured (in duplicates) after filtration (0.45 µm) in a TOC/TN Jena analyzer 2100S (Analytik Jena AG, Jena, Germany). TSS were analyzed by the gravimetric method (APHA, 2005). Treatments were performed in duplicates.

A second sampling campaign was aimed at obtaining a clearer insight into the influence of greywater circulation time on THM formation. For this, system S5, which presented the greatest difference in DOC during the first sampling event (Table 1), was grab sampled in finer intervals of 0.0, 0.5, 1.0, 3.0, and 6.0 h after feeding the system with raw greywater. Samples followed the same experimental setup and analytical procedure as above. Additionally, raw and 6 h circulated water samples were also dosed with 25 and 50 mg/L of free chlorine (in addition to 5 mg/L free chlorine) and analyzed for THM and residual chlorine after 1 h of incubation. Chlorine was analyzed by an ExStik Chlorine Meter (Extech Instruments, Waltham, MA, USA), which is unaffected by turbidity.

**Table 1.** Average ± standard deviation of raw and treated greywater quality from the six recirculating vertical flow constructed wetlands. TSS, BOD, and fecal coliform results are based on eight sampling events, twice per season.

System	pH Raw GW	EC Raw GW (µs/cm)	DO Raw GW (mg/L)	D° C (mg/L)		TSS (mg/L)		BOD <sub>5</sub> (mg/L)		Fecal Coliforms (Log CFU/mL)	
				Raw GW	Treated	Raw GW	Treated	Raw GW	Treated	Raw GW	Treated
S1	7.6	555	0.84	10.8 ± 0.8	6.3 ± 0.7	115 ± 18	2.0 ± 2.2	221 ± 71	5.9 ± 8.9	4.1 ± 3.0	3.1 ± 3.1
S2	7.3	391	0.33	9.5 ± 1.0	6.7 ± 0.7	140 ± 85	17.6 ± 11.3	121 ± 20	5.0 ± 2.8	7.1 ± 6.8	6.6 ± 3.0
S3	7.2	357	0.06	7.2 ± 0.8	6.0 ± 0.7	260 ± 74	1.9 ± 1.6	92 ± 69	2.8 ± 2.5	3.3 ± 2.1	0.3 ± 0.1
S4	7.4	760	0.70	40.5 ± 3.1	12.6 ± 0.5	155 ± 73	2.1 ± 1.6	190 ± 13	2.6 ± 2.1	2.4 ± 2.2	0.4 ± 0.7
S5	6.8	663	0.50	130.9 ± 1.9	19.9 ± 0.2	365 ± 113	17.8 ± 11.3	208 ± 44	5.7 ± 3.0	4.5 ± 4.1	1.7 ± 0.7
S6	9.0	884	0.95	54.8 ± 1.8	9.4 ± 0.2	152 ± 22	8.1 ± 8.3	146 ± 51	4.9 ± 1.4	3.0 ± 3.2	0.2 ± 0.1

Finally, changes in specific UVA absorbance were separately monitored in system S1, to evaluate changes in organic matter characteristics [43].

## 2.3. Analytical Methods

THM samples were analyzed by a GC/MS (Trace 1310 GC coupled to an ISQ LT quadrupole MS, Thermo Fisher Scientific, Dreieich, Germany). Samples (10 mL in 20 mL glass vials) were analyzed in headspace injections following 20 min of agitation at 65 °C. Compound

separation was achieved using a Restek Rxi-5Sil column (30 m × 0.25 mm × 0.25 μm) under a temperature program of 25 °C (hold 7 min), followed by a 15 °C /min ramp to 170 °C, followed by a 20 °C/min ramp to 190 °C (hold 2 min). Peaks were detected using the selected ion mode (SIM) with selected m/z of 47, 83, 85 (chloroform); 83, 85, 129 (dichlorobromomethane); 79, 127, 129 (chlorodibromomethane); and 91, 173, 252 (bromoform). Concentrations were quantified against an external calibration curve.

UV–visible absorbance measurements were performed on a Tecan Spark instrument. A 96-well quartz microplate was used for simultaneous duplicate measurements of the samples. SUVA values were determined by dividing the UV absorbance measured at a wavelength of 254 nm by the DOC concentration, and are reported as liter per milligram carbon per meter.

### 3. Results

#### 3.1. Influence of Greywater Treatment on THM Formation

Water samples from the six systems differed significantly in their DOC, varying from 7.2 to 130.9 mg/L. Dissolved oxygen in the raw water was lower than 1.0 mg/L in all systems, the pH varied from 6.8 to 7.6 in five of the systems, and a sixth exceptional system presented a pH of 9.0 (Table 1). The raw water in the systems originated from different households, thus reflecting differences in domestic water usages. Large variation in raw greywater quality is typical for the systems tested [32].

After the raw water was recirculated in the systems for 6 h, the greywater DOC decreased to varying extents, with the most significant decrease in raw water with high DOC levels (Table 1). A similar trend was shown for TSS. This trend is related to the microbial consumption of organic matter and is in accordance with previous studies of the systems in focus (e.g., [32,41]).

Both raw greywater and 6 h recirculated greywater samples were spiked with 5 mg/L chlorine and incubated for 1 h. Total THM formation after 1 h of incubation did not exceed 34.3 μg/L, for either the raw or the recirculated greywater (Figure 2). This concentration range is lower than the maximal concentration limit for drinking water (the US EPA: 80 μg/L [44], the WHO: 100 μg/L [45]), as well as guidelines for swimming pools [19], and does not pose a health risk when used for irrigation.

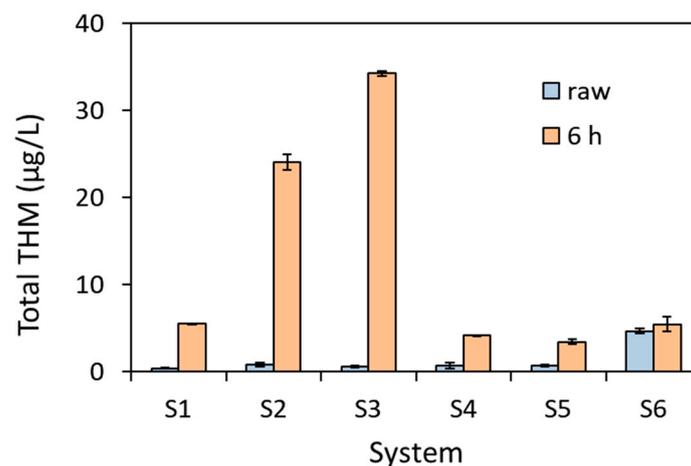
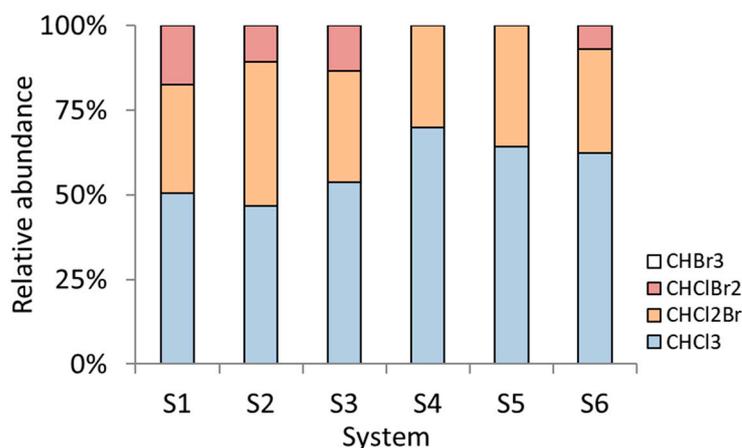


Figure 2. Total trihalomethane concentrations in raw water and water treated for 6 h.

Great differences in the THM concentrations between the raw and the recirculated water were observed in all systems, except S6. THM formation was significantly lower in the raw greywater with concentrations in the single μg/L range (with the exception of system S6), in comparison to the recirculated water with a maximal concentration of 34.3 μg/L. This implies that changes in water characteristics during treatment promote THM formation. Of the THM species formed, chloroform was the most abundant (usual abundance

of  $\geq 50\%$ ), whereas bromoform was not detected in any of the samples (Figure 3). The distribution of chlorinated versus brominated species is related to bromide concentrations in the greywater [16]. Thus, this distribution is related to the water source, rather than to the treatment process. It may be expected that for bromide-rich raw greywater, more brominated species will be formed [16,46].

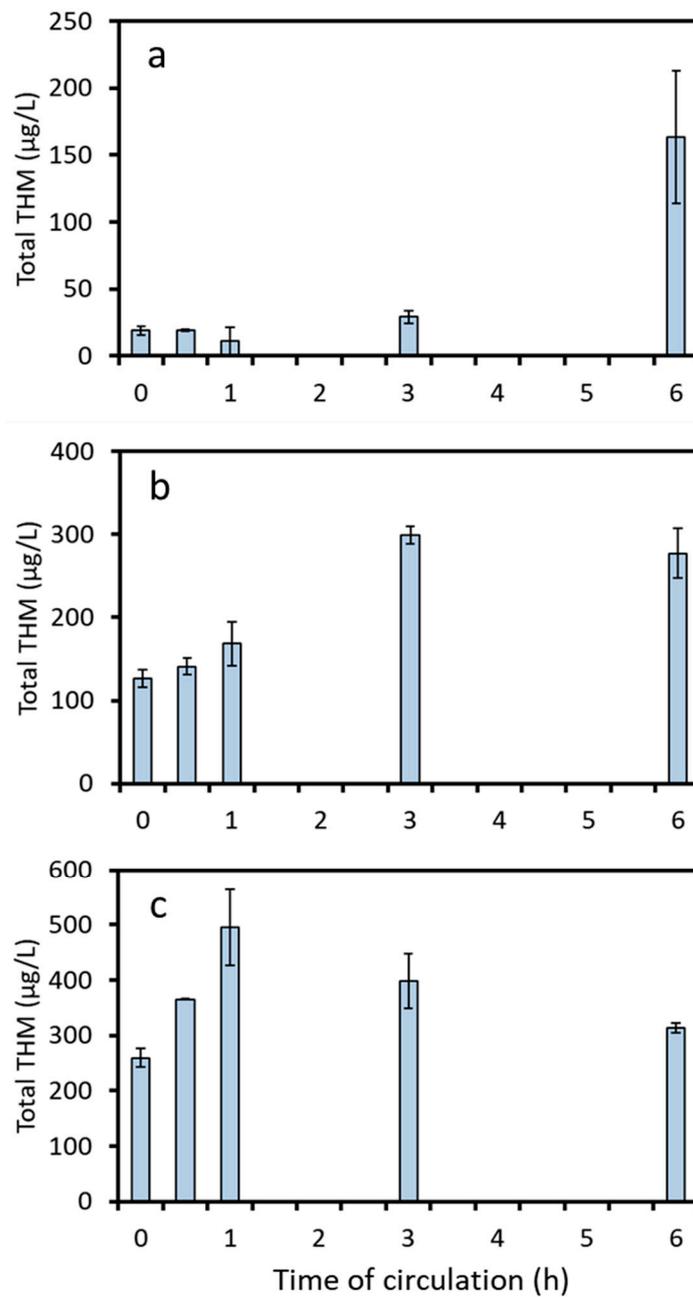


**Figure 3.** The relative abundance of the different THM species ( $\text{CHCl}_3$ ,  $\text{CHCl}_2\text{Br}$ ,  $\text{CHClBr}_2$ ,  $\text{CHBr}_3$ ) in 6 h recirculated water after incubation with 5 mg/L chlorine for 1 h.

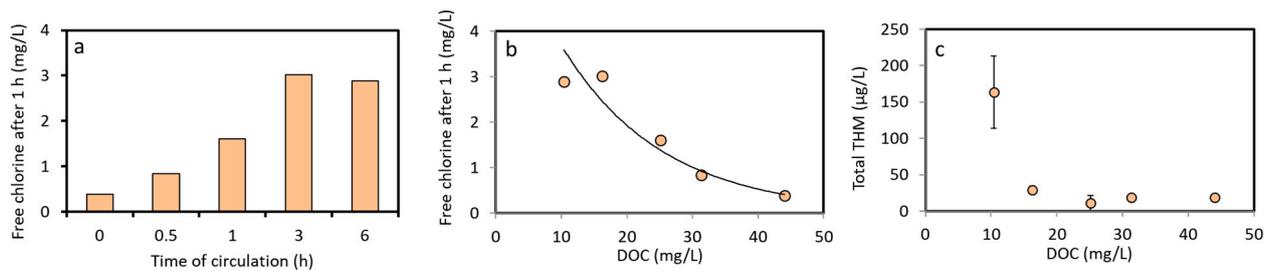
### 3.2. Influence of Organic Matter on Residual Free Chlorine and THM Formation

A detailed study of the recirculation time's influence on THM formation was undertaken in system S5, which was sampled in different time-steps over 6 h of recirculation. Water samples that were recirculated in the system for increasing durations (0.5, 1, 3 and 6 h) were spiked with 5 mg/L of free chlorine, and incubated for 1 h, following the same procedure as in the first experiment. The results were consistent with the general increasing trend in THM formation for longer recirculated water (Figure 4a), as shown in the first experiment. THM concentrations of  $>150 \mu\text{g/L}$  were recorded, which were higher than those observed in the first experiment (up to  $\approx 35 \mu\text{g/L}$ ), highlighting the temporal variability in the quality of the raw water feeding the system. For higher chlorine doses—25 and 50 mg/L—maximal THM concentrations were observed for the water that was circulated in the system for shorter periods. In the case of 25 mg/L free chlorine, the maximal THM concentration was recorded for water that was circulated for 3 h within the system, and in the case of 50 mg/L free chlorine, for water that was circulated for 1 h within the system.

The chlorine-spiked water samples were further monitored for residual free chlorine, showing a general increase in residual free chlorine for longer treated water (Figure 5). The free chlorine consumed during 1 h of incubation was shown to be correlated to DOC (Figure 5). The higher the DOC, the lower the free chlorine concentration after 1 h of incubation. This observation implies a higher number of reactive sites that are present with increasing organic loads, which serve as chlorine sinks. Nevertheless, the increase in chlorine consumption was not reflected in a parallel increase in THM formation. On the contrary, the highest THM formation was shown for the lowest DOC concentration (Figure 5).



**Figure 4.** Total THM after 1 h of incubation in system S5's recirculated water with (a) 5, (b) 25, and (c) 50 mg/L chlorine.



**Figure 5.** Residual free chlorine after 1 h of incubation (a) and as a function of DOC (b), and total THM formed (c) after 1 h of incubation as a function of DOC. Results for S5 water following differing circulation times.

#### 4. Discussion

As organic matter concentration decreases, chlorine competes on a smaller number of reactive sites, leading to higher THM yields (chlorine recovery in THM species increased from  $\approx 0.3\%$  to  $5.3\%$ ; Supplementary Material, Table S1). Nevertheless, it should be noted that not only the absolute amount of organic carbon drops with recirculation time, but the relative abundance of moieties that favor THM formation may change as well. The relative abundance of these moieties can be related to two sources: (1) It is expected that following microbial activity, labile molecules will be favorably consumed, increasing the relative abundance of more complex recalcitrant compounds. In agreement with this general expectation, Park et al. [47] showed an increase in aromaticity throughout a constructed wetland receiving secondary treated wastewater (shown for a free-water surface flow constructed wetland with a retention time of seven days). (2) Vegetation can serve as an important additional source of wetland organic matter, as in the RVFCW systems studied here. This source can alter the DOC characteristics. Leaf- and root-derived organic matter were shown to present higher aromaticity and to increase the THM formation potential [48]. Indeed, as demonstrated for subsurface wetland flow, it was observed that wetland-derived organic matter has higher aromaticity and is less biodegradable, resulting in an increase in chlorine reactivity [35]. An increase in aromaticity is also expected in the current systems, as reflected by the increase in SUVA during treatment in system S1 (Table 2). While raw greywater presented a relatively low SUVA value, this rapidly increased for recirculated water.

**Table 2.** SUVA values in system S1 during recirculation.

Recirculation Time (h)	DOC (mg/L)	SUVA (L/mg·m)
Raw	9.61	1.08 ± 0.04
0.5	5.56	2.09 ± 0.06
1.0	5.45	1.78 ± 0.12
1.5	4.97	2.91 ± 1.25
6.0	4.06	2.63 ± 0.15

Correlating THM formation with a single water parameter may seem overly simplistic. Previous attempts to construct predictive models for THM formation potential in water are summarized in the literature (e.g., [25,49,50]). Such models involve the fitting of THM formation to multiple parameters, often including both DOC and DOC characteristics, as well as factors such as pH,  $\text{Br}^-$ , chlorine dose, and more. This leads to complex, multi-parameter equations. In the current study, the dominance of bulk DOC versus the DOC characteristics can be tested for different recirculation times.

The influence of organic matter and its characteristics is further magnified when increasing the chlorine dose in the same water samples. When the chlorine dose was increased in the 1-h incubation experiments from 5 to 25, and further to 50 mg/L, not only did the maximal THM concentrations increase, but the peak THM concentration was also observed in samples that underwent a shorter recirculation time (Figure 4). When spiked with 5 mg/L of chlorine, a maximal THM concentration of 163  $\mu\text{g/L}$  was obtained for water that underwent 6 h of recirculation. A maximal THM concentration of 496  $\mu\text{g/L}$  was detected for water that underwent 1 h of recirculation and was spiked with 50 mg/L chlorine. These results suggest that the higher the chlorine dose, the lower the competition between DOC reactive sites that serve as chlorine sinks and the THM-yielding reactive sites.

It may be questioned whether the increased THM levels observed in the water recirculated for longer times may also be related to changes in inorganic nitrogen species. Previous works on the systems tested here have shown that reduced nitrogen species dominate in raw wastewater ( $\text{NH}_3$  and  $\text{NH}_4^+$ ), and oxidized species dominate in recirculated water ( $\text{NO}_2^-$  and  $\text{NO}_3^-$ ) [38]. For greywater, the oxidized species dominated in both the raw and recirculated water [37].  $\text{NH}_3$  is known to play an important role in chlorine reactions, forming chloramines and decreasing the formation of THM. However, chlorine reactivity

with  $\text{NH}_4^+$  was reported as negligible in well-controlled studies [16,51]. Some field studies contradict the proposed negligible role of  $\text{NH}_4^+$  in THM formation (e.g., [52,53]), yet it is unclear whether ammonium was the sole factor leading to the contradictory results. Under the pH range of the studied systems (with the exception of system S6),  $\text{NH}_3$  levels are expected to be negligible and, therefore, are not expected to lead to an increasing trend in THM formation with prolonged recirculation.

Oxidized inorganic nitrogen species may also participate in chlorine chemistry. A main player is  $\text{NO}_2^-$ , which may be oxidized by HOCl to form  $\text{ClNO}_2$  [16]. This reaction directs a decrease in THM formation in recirculated water. This is opposed to the trend observed in this study. Therefore, it is postulated that the inorganic nitrogen species in these recirculated water samples cannot explain the increasing trend of THM formation with circulation time.

## 5. Conclusions

Chlorination of greywater prior to irrigation use is an accepted practice. This study shows that when chlorinated with the same amounts, THM formation in raw greywater is significantly lower than in treated greywater. While we do not encourage the use of raw greywater for household irrigation, it is often allowed by different countries, such as Australia and the US [10]. There is an increase in THM formation in recirculated (better treated) greywater, which is inversely correlated with DOC, TSS, and chlorine consumption. It is suggested that this trend is the result of both competition between a higher number of reactive sites, leading to smaller THM yields, and changes in the DOC characteristics towards a higher abundance of the moieties that support THM formation. Lastly, it was found that for the chlorine dose used here, THM concentrations may exceed drinking water regulations (e.g., the EPA [44] and the WHO [45]) and swimming water guidelines [19]. As contact with this type of water is not expected to be high, such as for drinking or swimming, this dose does not pose any health risk, and the water should not be restricted for irrigation. Further work is needed in order to define organic matter characteristics during treatment.

**Supplementary Materials:** The following are available online at <https://www.mdpi.com/article/10.3390/w13070903/s1>, Figure S1: A recirculating vertical flow constructed wetland system, Table S1: Chlorine yield as THM for system S5 water that was incubated with 5 mg/L chlorine for 1 h.

**Author Contributions:** Conceptualization, A.B., H.S., and A.G.; methodology, A.B., H.S., and A.G.; validation, A.B., H.S., and A.G.; investigation, A.B., H.S., A.G.K. and A.G.; resources, A.B., H.S., and A.G.; data curation, A.B., H.S., and A.G.; writing—original draft preparation, A.B.; writing—review and editing, A.B., H.S., A.G.K. and A.G.; visualization, A.B., H.S., and A.G. All authors have read and agreed to the published version of the manuscript.

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**Conflicts of Interest:** The authors declare no conflict of interest.

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