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Effect of Different Disinfectants on Grey Water Quality during Storage

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ABSTRACT

Grey water offers potential to save large amount of potable water resources. However, grey water can only be stored for up to 24 hours in most parts of the world because of the fast deterioration in quality due to bacterial growth. Several physical, biological and chemical treatment methods or combination of them can be employed to safely increase the retention time. In this study, grey water sample collected from a typical urban house in Western Australia was used to investigate the effectiveness of physical, chemical and combination of them to treat for storing longer time. For chemical treatment, chlorine, chloramine and hydrogen peroxide were assessed whereas for physical treatment, grey water samples were filtered through 11μm filter paper to remove large suspended solids. Chloramine was found to be effective for storing longer time without combination of physical treatment. It was noted that organic matter present in grey water induces a greater chlorine demand and prevent proper disinfection. Chlorine was found to be highly reactive with impurities found in grey water, so removal of these impurities is essential through filtration. Hydrogen peroxide was ineffective at low doses and was the weakest of the three disinfectants.

Keywords: Disinfectants, Grey water, Reuse, Storage time, Total coliform

1.0 INTRODUCTION

Grey water recycling is becoming an increasingly common practice in Australia, as households look to save large amounts of potable water resources. The amount of grey water produced in a typical household was assessed by Water Corporation, Western Australia. It reported that 90 kL/yr of grey water (from the bathroom and the laundry) produced out of a total use of 282 kL/yr from an average household in Perth (Water Corporation, 2008). Similarly, outdoor use of water, mainly watering lawn, was found to be 83.98 L/person/day, the largest consumer (34 %) in a single household. The analysis shows that reusing grey water for irrigation purposes has the potential to save up to 40 % of potable water resources in Western Australia.

Despite advantages over reducing the use of fresh water resources, long time storage of grey water has been challenging. Untreated grey water can only be stored for a maximum of 24 hours. After 24 hours of storage, many parameters reach levels which are unacceptable such as rapid decrease in dissolved oxygen (DO) level and simultaneous increase of total coliform (TC). Storing grey water for a long time can result in high strength grey wa-

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ter which possesses high biochemical oxygen demand (BOD) levels, and this can potentially change soil properties resulting in poor aeration and can lead to breakdown of soil structure (Schneider, 2009). Another significant issue is that if raw grey water is stored for a long time and released into the environment, contamination of surface and groundwater resources could occur with low DO levels, leading to death of many aquatic organisms due to anaerobic conditions (Schneider, 2009). Moreover, grey water will decompose at a much faster rate than the blackwater. A study from Sweden has shown that after 5 days of storage, balckwater only reached 40 % of its total decomposition, whilst grey water achieved 90 % of decomposition (Karlgren *et al*., 1967).

Storage is a vital element in any grey water recycling process, and grey water needs to be stored at some point if it is to be reused. Storing grey water for more than 24 hours is a very common error that many people make due to the lack of information provided on grey water (Oasis Design, 2009), and the water becomes too dirty to reuse. If grey water could be stored for more than 24 hours it would be of great benefit, because households could utilise it more efficiently. Having a longer storage time would also mean that no grey water would go to waste, and therefore the amount of grey water used on a daily basis could be controlled. According to current law, grey water needs to be used within 24 hours; this could result in excess irrigation which is not good for plants and soils, and if it is not used at all, it goes to waste. In Western Australia, grey water should be maintained below 20 mg/L BOD, 30 mg/L suspended solids and disinfection should achieve TC <10 cfu coliforms/100mL for unrestricted use such as surface spray irrigation (Department of Health, 2002).

Grey water also contains pathogens and there is potential for spreading illness. They can cause a range of diseases, lung infections, ingestion, gastroenteritis, eye and skin infection, pneumonia, pulmonary disease and toxic shock syndrome (Winward, 2007). Faecal coliforms are a type of bacteria that originate in warm blooded animals, and they can be used as an indicator for the presence of pathogenic microbes or can also be used to indicate the level of disinfection needed (Schneider, 2009). However there is substantial evidence that bacterial growth occurs with increasing storage time. One study showed that an initial TC count (grey water stored for 0 hours) was 100 cfu/100mL but at 72 hours storage the measured count was $8.4*10^6$ cfu/100mL (March and Gual, 2009).

Various treatment technologies have been employed to store grey water for irrigation purposes. The common treatment technologies are physical treatment (sand filtration, membrane filtration) and biological process followed by chemical treatment (disinfectants). Different disinfectants have been employed to store the grey water for a long time and they are chloramine, chlorine, hydrogen peroxide, UV light and oils. The chemical and physical quality of the grey water will heavily influence what type of disinfection method is most suitable (Winward, 2007). For example, presence of organic matter and suspended solids in grey water can affect efficiency of disinfection and disinfectant demand. Organic material generally reacts with disinfectant and therefore a greater initial dose is needed to achieve total inactivation of bacteria (Ronen *et al.*, 2010). Asano *et al.* (2007) also found that a greater initial dose of chlorine is needed to overcome its interaction with organic matter. It was also found that larger particles can help shield bacteria from disinfection (Winward *et al.*, 2007).

The chemical and physical characteristics of grey water are greatly affected by the local environment: type of detergent used, from where it was generated etc. Therefore, prior to employing any technology to store grey water, it is necessary to understand the grey water quality and suitability of treatment processes. This study investigated the effectiveness of chemical and combination of physical and chemical treatments to increase grey water storage time. For the chemical treatment, common disinfectants; chlorine, chloramine and hydrogen peroxide were assessed at the same concentration.

2.0 MATERIALS AND METHODS

2.1 Grey Water Sampling

The sample was collected from a washing machine in a typical urban home. A 50 mL volume of 'Radiant Micro Max' front loader liquid detergent was utilised for each wash. The contents of the washing machine were a combination of different clothing from each wash to simulate a typical urban laundry (i.e. underwear, t-shirts, pants, towels etc). The grey water was a combination of all three discharges from the first washing cycle. The sample had been collected in a 10 L High Density Poly-ethylene container. Then, the sample was transported as soon as possible under dark conditions to Curtin University, Western Australia for analysis to reduce storage time before testing.

2.2 Experimental Design

The experiments were divided into two sets. In the first set of the experiment, grey water collected in 10 L High Density Poly-ethylene container. Then the water was divided into two portions. The first portion (named raw water) was stored at temperature (20.0±2.0^{o}C) in a dark place. The second portion, named filtered sample, was stored at 20.0±2.0^{o}C after filtering through the 11 μm filter paper to understand the effect of big suspended solids. Consequently, chemical parameters (dissolved organic carbon (DOC), ammonia, nitrite, nitrate, pH and DO) and TC were monitored to understand how the grey water quality changes over a week of storage. TC was monitored only three times, 0, 24 and 168 hours of storage.

In the second set of the experiment, three different disinfectants (total chlorine, chloramine and hydrogen peroxide) were assessed for its ability to extend the storage time of both filtered and raw water samples prepared for first set. The filtered and unfiltered (raw) samples were filled up in 500 ml polyethylene terephthalate bottles. A 13.2 mg/L of hydrogen peroxide, chlorine and chloramine were dosed in both filtered and raw samples. All the samples were prepared in duplicate. Chloramine residual was adjusted using stock solutions of ammonium chloride (500 mg-N/L) and sodium hypochlorite (500 mg-Cl/L). Chloramine dose contained total chlorine to ammonia-nitrogen ratio at 4.5:1, and it is expected ammonia in grey water would reduce this ratio further. Chlorine was adjusted using 500 mg-Cl/L stock solution of sodium hypochlorite whereas 30% hydrogen peroxide stock solution was employed to maintain a 13.2 mg/L of hydrogen peroxide. Samples were incubated in a dark water bath maintaining a constant temperature (20^{o}C). Total chlorine, DOC and pH were continuously monitored in both samples and average of them was reported. TC was monitored only three times as reported in the first set of the experiment.

2.3 Analytical Procedures

The Aquakem 200, a high precision wet chemistry automated analyser, was employed to measure ammonia, nitrite and NOx (summation of nitrite and nitrate) concentrations. Ammonia and NOx were measured spectrophotometrically according to the methods described in EPA (1981a). Nitrite was measured by the sulphanilamide method (4500-NO_2^- B) (APHA *et al*, 1998). The analyser has a high detection limit for ammonia, nitrite and NOx levels of 0.002 mg-N/L. The experimental errors were 1.5% (95% confidence level) for ammonia, nitrite and NOx measurement. Total chlorine residuals were measured by DPD colorimetric method using a HACH pocket colorimeter. Total chlorine measurement had an experimental error of ±0.03 mg/L. The TOC and DOC were measured using Sievers 5310C Laboratory TOC analyzer; an experimental error for DOC was ±5%. To measure DOC, sample was filtered through 0.45 μm filter paper. The sample was diluted 25 times with the MilliQ water (18 MΩ) before measuring DOC. A portable multimeter (HACH 40d) was used to measure pH and DO. The measurement error was ±0.1 for pH and DO.

TC was measured in an external laboratory (Silliker Laboratory in Western Australia). The TC sample was collected in a 100mL flask containing a Sodium Thiosulphate tablet to remove all disinfectant in an instant to ensure that chlorine/chloramine did not continue to inactivate and kill bacteria. The sample was transported in a 4^{o}C esky as soon as the samples were collected so that it can be analysed within a 24 hour period according to Australian standards. They employed membrane filtration method (APHA *et al*, 1998) to measure TC.

3.0 RESULTS AND DISCUSSION

3.1 Changes in Water Quality during Storage of Raw and Filtered Samples, the First Set of Experiment

Common chemical parameters were measured at various time period of grey water storage at 20^{o}C and are presented in Figure 1. The DO level dropped to 1.0±0.2 mg/L from 6.5 mg/L within 24 hours of storage proving the quick deterioration of grey water quality as reported elsewhere (March and Gual, 2009; Oasis Design, 2009; Winward *et al*., 2007). Afterwards, DO slowly decreased till 7 days of storage (Figure 1A) resulting in an anaerobic condition. The low DO level (0.3±0.2 mg/L) could be due to exposure to air during measurement. During storage, the DOC decreased substantially. The decreased DOC would have converted to cells under anaerobic condition and some would have changed to carbon dioxide due to the presence of low DO levels achieved during opening/closing for sampling. In addition, sulphate which would have been present in the original sample could have acted as an electron acceptor for the degradation of organic carbon. Either produced carbon dioxide or anaerobic conversion of organics or reduced sulphate (resulting in hydrogen sulphide production) would resulted in decrease of pH from 7.8 to 7.0 (Figure 1A).

Ammonia-nitrogen increased substantially within 48 hours of raw water storage (Figure 1B) reinforcing the fact that hydrolysis of organic nitrogen, which was possibly present in the grey water. Then, ammonia-nitrogen gradually decreased, probably due to assimilation into microbial cells as DOC gradually decreased. However, there were no significant changes in nitrate and nitrate profiles (Figure 1B), although minor nitrite production indicated the availability of low level oxygen. Similar observation was made in other three sets of experiments.

Filtered samples experienced faster DOC reduction (Figure 1A) indicating that removal of suspended solids actually helps in accelerating the degradation of organic matter. The suspended solids are expected to consist of particulate organic matter rather than the clumps of bacteria since the grey water was collected fresh from the washing machine. If any bacteria are to be present, they are likely to be present in suspension. Moreover, the filter adopted for filtration was 11μm which achieved only larger particles removal rather than bacterial removal. What is removed by filtration is therefore mainly organics or other impurities originating from the clothes which were further confirmed by measuring TC (Figure 1C, at 0 hour). It is therefore possible that slower degradation in the raw water sample was due to the contribution of suspended solids to the DOC as microbial degradation of organic matter takes place. At 24 hours, significant difference in TC count was noted. Filtered sample had 10 cfu/100 mL while raw water achieved >8000. However, after 7 days both were more than 8000 cfu/mL (Figure 1C) in both samples (absolute value was not determined).

3.2 Chlorine and Chloramine Decay Profiles in Filtered and Raw Grey Water Samples, the Second Set of Experiment

As shown in Figures 2 and 3, rapid chloramine and chlorine losses were noted within 24 hours of spiking in grey water samples. The chloramine levels had dropped from 13.2 to 5.2 and 4.0 mg/L in the filtered and raw samples, respectively after 24 hours whereas total chlorine residual dropped to 3.0 mg/L in both filtered and raw samples. Subsequently, both chlorine and chloramine slowly decreased. Chlorine completely decayed (dropped to 0.0 mg/L) in the raw grey water after 4 days of storage (Figure 3). The accelerated chlorine loss in the raw samples compared to chloramine could be due to its highly reactive characteristics. Moreover, both chlorine and chloramine decayed at a faster rate in raw grey water samples than in the filtered ones, although chloramine decay profile was only slightly different. This is a further evidence that filtration slows decay since there were less impurities that react with disinfectants. The slower decay rate of chloramine than chlorine does actually have benefits in preventing bacterial growth for long storage periods.

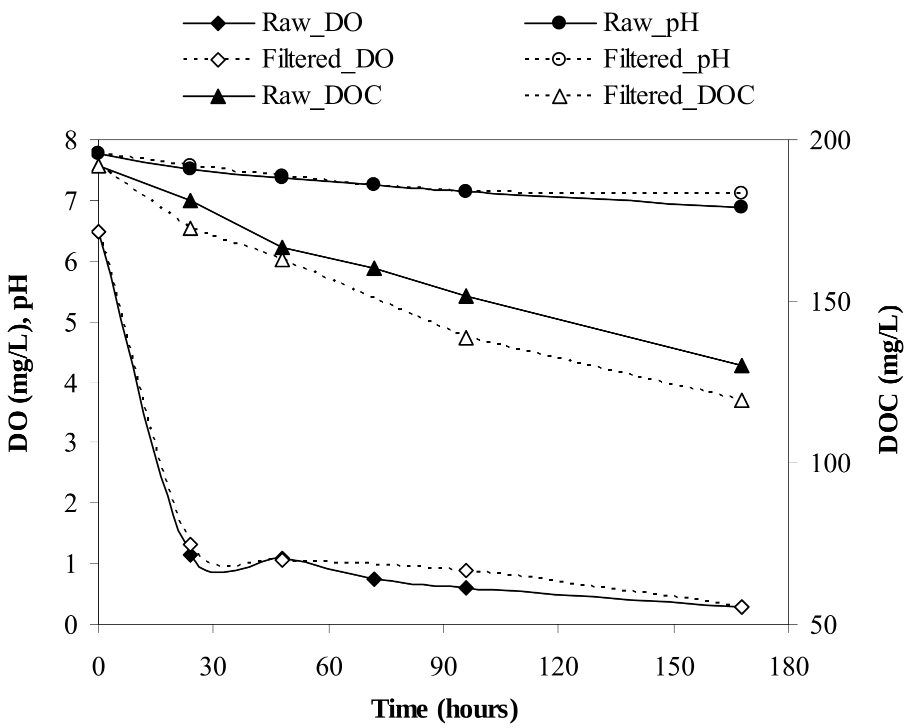

Figure 1A

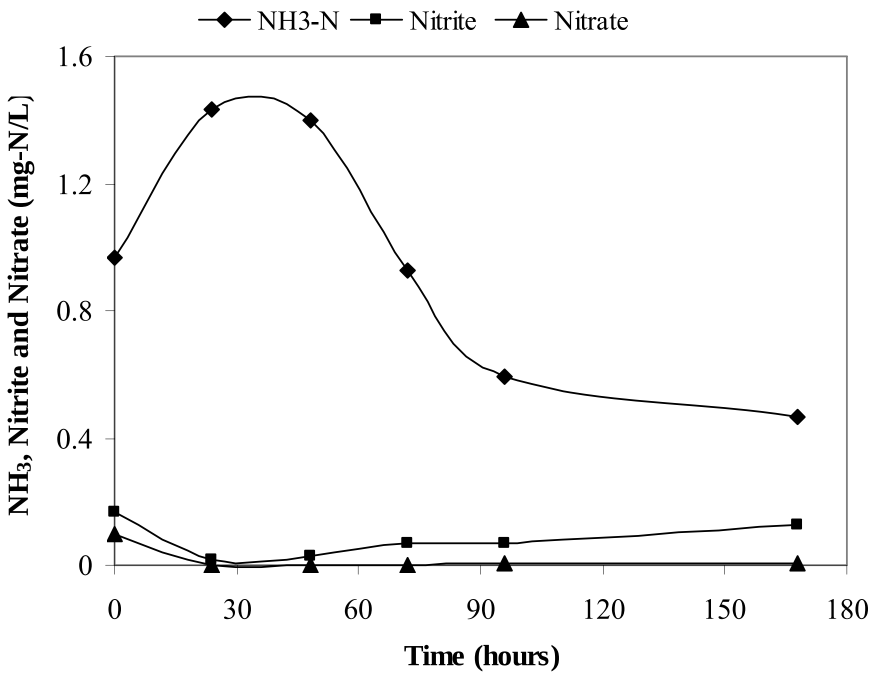

Figure 1B

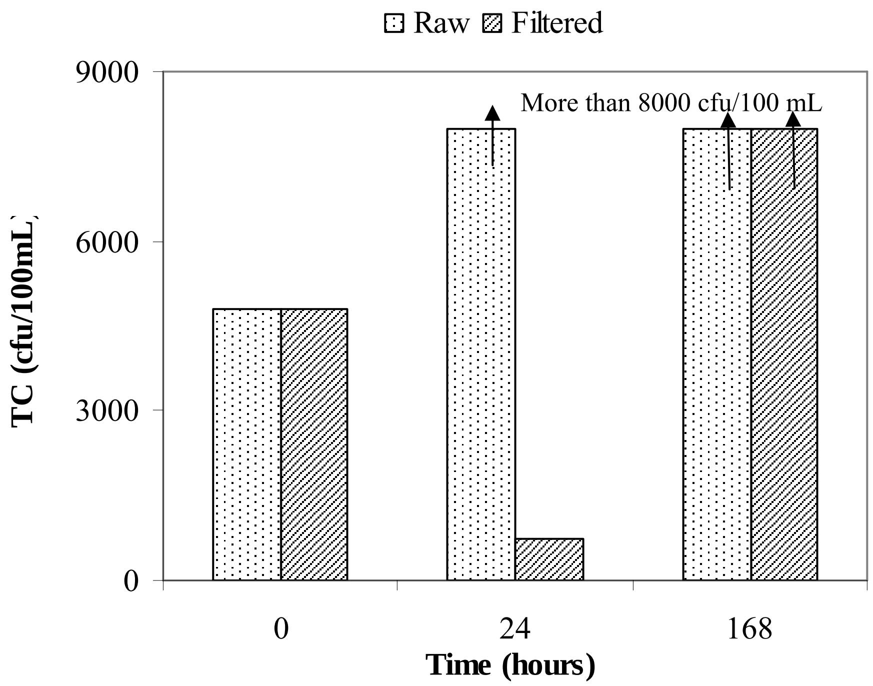

Figure 1C

Figure 1 Chemical parameters and total coliform (TC) of raw and filtered (11 μm filter) grey water at different time period of storage at temperature (20.0±2.0°C). **1A**: pH, DO and DOC profiles. **1B**: Nitrogenous compounds profiles in raw grey water. **1C**: TC profiles

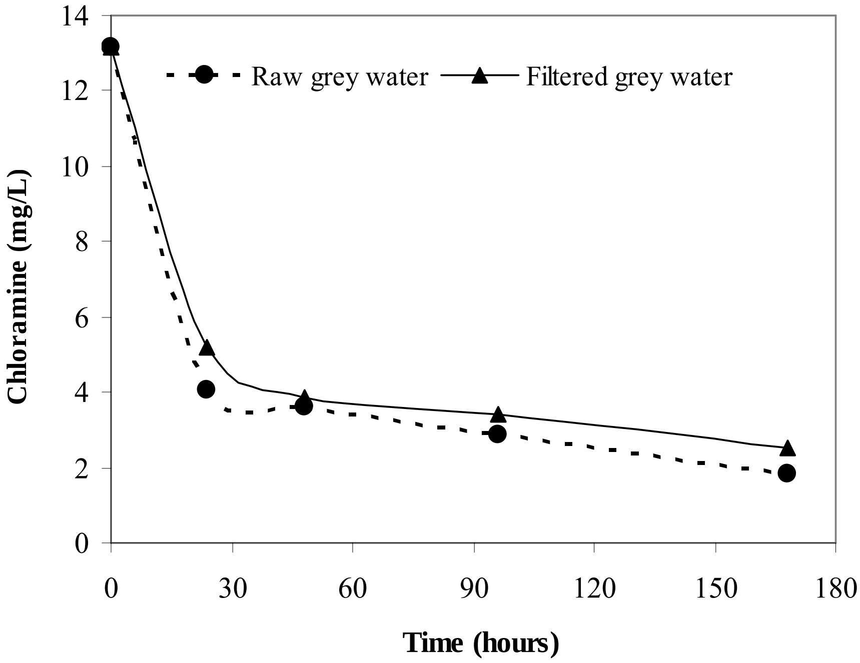

Figure 2 Chloramine decay profiles in raw and filtered grey water samples

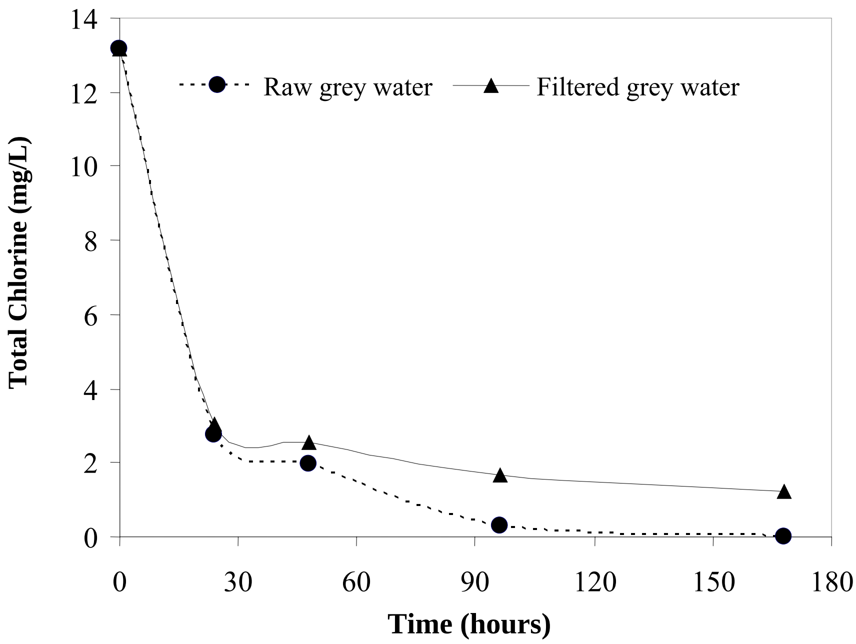

Figure 3 Total Chlorine decay profiles in raw and filtered grey water samples

3.3 Dissolved Organic Carbon (DOC) Levels in Disinfectant used Grey Water Samples, the Second Set of Experiment

DOC level was measured over a week of storage period, as shown in Figure 4, giving an indication of bacterial activity subject to different disinfectants. The DOC levels were consistent in the chloraminated (raw and filtered) and chlorinated filtered samples over the storage period. This result demonstrated that 13.15 mg/L chloramine is adequate for inhibiting microbial degradation of grey water whereas the same amount of total chlorine is effective only for filtered sample, as DOC degradation was seen in raw water (Figure 4) after chlorine had dropped to zero levels (Figure 3). It is commonly known that solid particles shield the microbial cells and protect from the disinfectants, such microbes would have found their way to grow after the chlorine residuals have reached zero mg/L.

Out of all disinfected samples, the DOC levels were the first to decrease in the hydrogen peroxide samples (both raw and filtered) and continued to drop thereafter which indicated that bacterial consumption continued. However, the degradation in the non-disinfectant dosed samples was similar to that in the hydrogen peroxide sample (Figures 1A and 4), indicating that hydrogen peroxide has a limited ability to control heterotrophic bacteria that consumes DOC in the grey water. Similar to what was observed in the first set, faster loss of DOC in filtered sample can be observed as discussed in section 3.1. Moreover, hydrogen peroxide seems not effective like other two disinfectants in both raw and filtered samples (Figure 4) which was further confirmed by TC count presented in the following section.

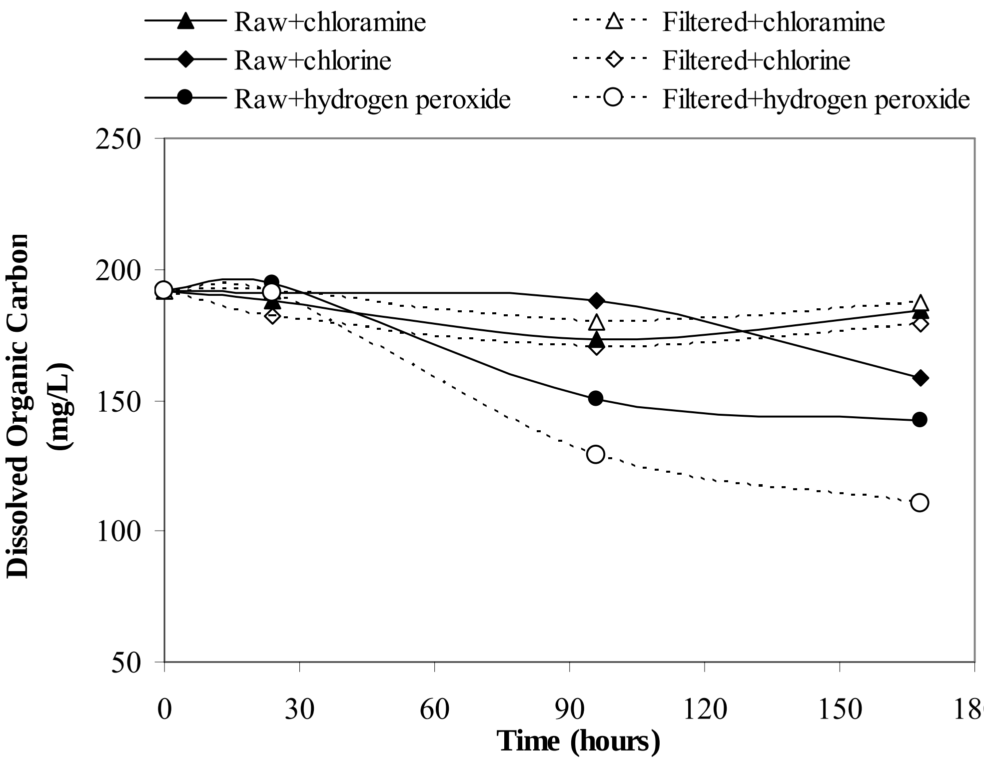

Figure 4 DOC levels varying with storage time for grey water samples containing various disinfectants

3.4 pH Profiles in Disinfectant Used Grey water Samples, the Second Set of Experiment

The reduction in pH is due to the release of carbon dioxide which is produced during the biochemical processes of the bacteria (Dixon *et al.,* 1999). As demonstrated in Figure 5, the hydrogen peroxide samples as well as the raw chlorinated sample experienced a similar drop in pH levels. The hydrogen peroxide sample experienced an immediate reduction in pH at the beginning of storage (7.8 to 6.9), whilst the pH only dropped in the raw chlorinated sample after 100 hours (after total chlorine had dropped to 0 mg/L). These results demonstrate the difficulty in eliminating all bacteria; if coliform or other bacteria had survived bacterial growth would continue and pH levels would continue to drop. Chloramine was shown to be a highly effective disinfectant in both raw and filtered grey water samples as pH levels were consistent (Figure 5).

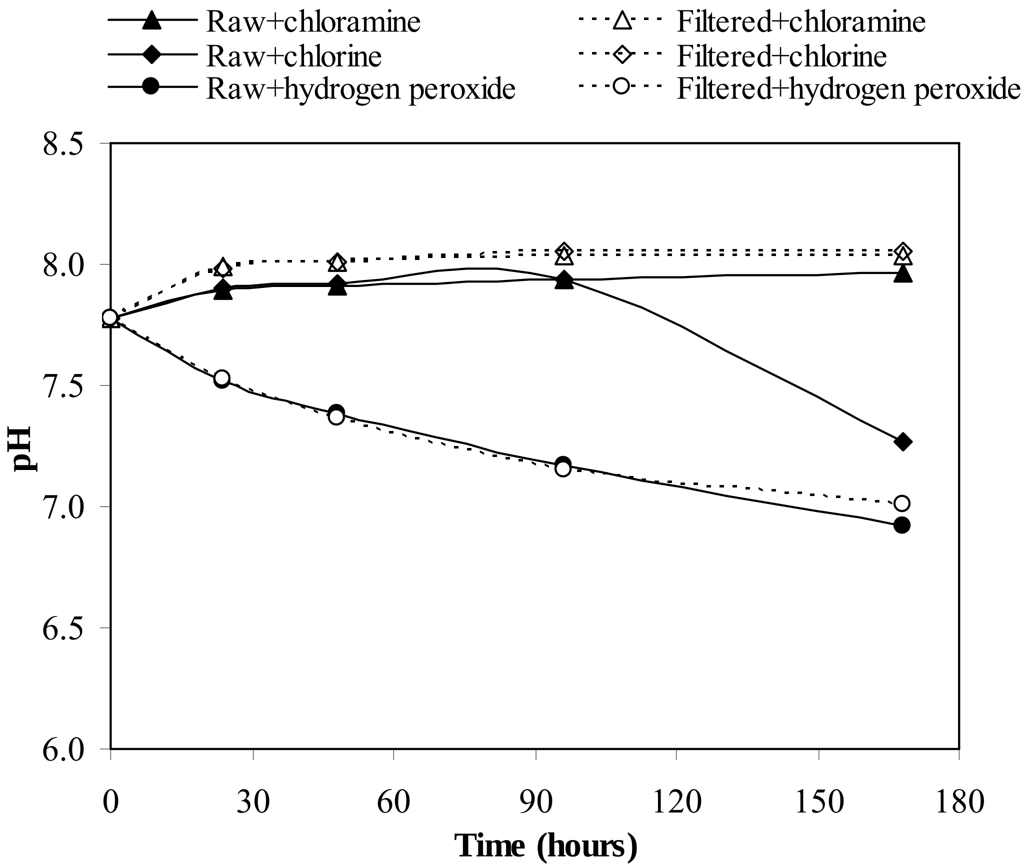

Figure 5 pH levels varying with storage time for grey water samples containing various disinfectants

3.5 Total Coliforms, TC in Disinfectant Used Grey Water Samples, the Second Set of Experiment

As shown in Figure 6, the TC count for all chlorinated and chloraminated samples after 24 hours was 2 cfu/100mL indicating that the disinfectants effectively killed bacteria, since initial TC count was 4800 cfu/100mL (at initial; 0 hours). The results indicate that hydrogen peroxide was the weakest of the three disinfectants since all initial doses were 13.15 mg/L, and hydrogen peroxide did not show any significant inactivation of bacteria, i.e. a 13.15 mg/L dose of hydrogen peroxide is not enough to inactivate bacteria. However, efficiency of disinfectants depend on grey water quality and disinfectant dose, as Pedahzur *et al.,* (2000) found that 50 mg/L of the hydrogen peroxide was enough to achieve less than 1 cfu/100mL for Faecal coliform, whilst Ronen *et al.,* (2010) found that 125 mg/L of hydrogen peroxide was needed to achieve the same level. Hence, hydrogen peroxide is only effective with high initial doses and long contact times.

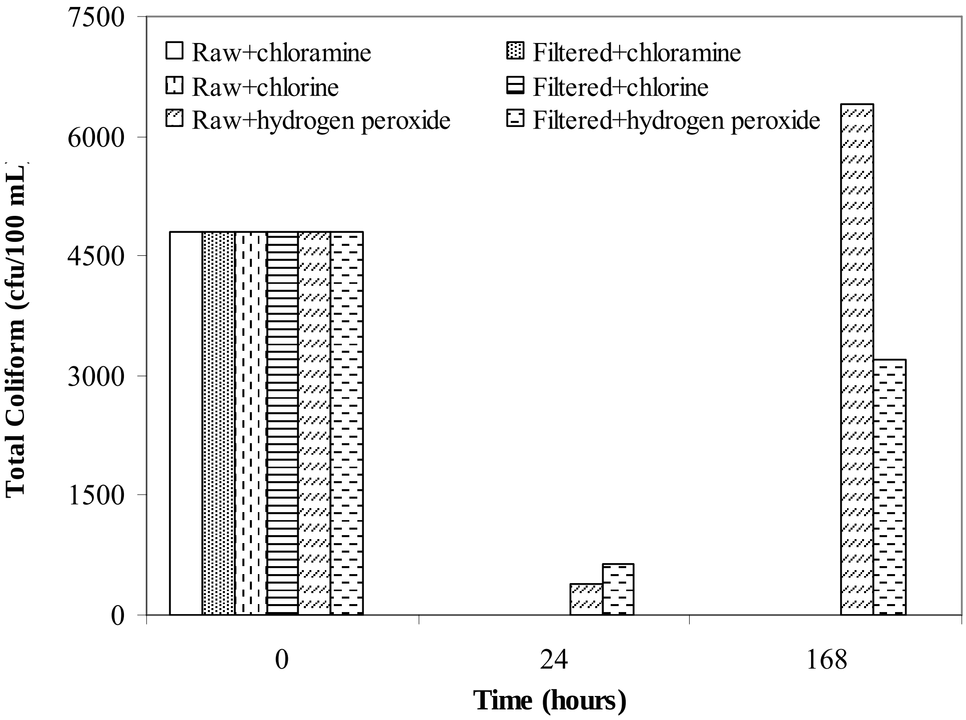

Figure 6 TC count for various samples containing disinfectant at intervals: 0 hours, 1day, and 7 days storage.

The chlorinated and chloraminated samples maintained low TC values after 7 days of storage. The chloraminated raw grey water TC count was 2 cfu/100mL, whilst the chloraminated filtered grey water TC count was 1 cfu/100mL. There was more of a difference in the TC count for the chlorinated samples after 7 days of storage; the TC count was increased in the chlorinated raw grey water to 10 cfu/100mL, whilst the filtered sample maintained 1 cfu/100mL. The results indicate that grey water samples with higher DOC concentration and suspended solids decrease the ability of chlorine to target microbes. This explains why TC levels reached 10 cfu/100mL after 7 days in chlorinated raw grey water samples (greater impurities), whilst filtered samples remained 1cfu/100mL.

4.0 CONCLUSIONS

The importance of physical treatment (filtration) prior to chemical disinfection has been noted in this study. The experimental results concluded that chloramine is the most effective disinfectant for long term storage periods since chloramine achieved the greatest TC inactivation after 7 days, in both filtered and raw grey water. Moreover, it showed that only chemical treatment (addition of chloramine) is sufficient to achieve the TC standard (Australian standard) for irrigation purpose. To obtain the same effluent standard in terms of TC inactivation, combination of filtration and chlorination is needed, signifying that chlorination is less effective than chloramination for storing long time grey water. The TC count in the hydrogen peroxide sample indicated that it is the weakest of all the disinfectants among chlorine and chloramine and it may need very high dose to achieve acceptable standards in both filtered and raw samples.

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