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Exposure assessment of treated greywater reused for irrigation

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ABSTRACT

The aim of the study was to assess the risk of exposure of treated greywater reused for irrigation. It consisted of untreated and treated greywater quality analysis and heavy metal exposure assessment. After treatment with a local conventional stabilization pond, parameters such as BOD, COD, Zn, and Pb dropped to 35.63, 9.95, 0.011, and 0.001 mg/L, respectively. Total coliform and fecal coliform decreased to 23,417 and 5,666 MPN/100 mL, respectively. These results showed almost all detected contaminants in treated greywater were minimized and did not exceed Thailand's standard and World Health Organization guidelines. After that, treated greywater was reused for irrigation. There were three vegetable plots: treated greywater, tap water with fertilizer, and tap water. There were seven edible plants, four leafy and three fruiting vegetables, cultivated in each plot. Zn, Pb, Cd, Cu, and Hg were detected depending on the kind of plant; for example, the maximum concentration was found in leafy vegetables. However, the concentrations of contaminants did not exceed the Codex standard for contaminants and toxins in foods and Thailand's standard. Risk characterization showed no risk with a hazard quotient far less than 1. Therefore, treated greywater reuse could be considered to promote sustainable water management in the community.

Key words: conventional stabilization pond, exposure assessment, greywater, greywater reuse, plant irrigation

HIGHLIGHTS

- This study is of one of the greywater management systems applied in a real field in a community where has water scarcity problem, especially in agricultural areas.
- A local conventional stabilization pond for greywater treatment could be suitable for such community with approximately 80–100 houses.
- Heavy metal exposure assessment of greywater reused for plant irrigation and risk of exposure, including chronic daily intake and hazard quotient, are estimated for public health information.
- Plants may be chosen to cultivate, for example, bulb vegetables, flowering and ornamental plants or plants that are not eaten fresh.
- Cultivation should be done with caution and the community should monitor the consumer's health in long term cultivation.

1. INTRODUCTION

Wastewater produced in a household is divided into two types: greywater and blackwater. Greywater is wastewater produced from kitchens, showers, sinks, and laundry services (WHO 2006; Ajit 2016; Noutsopoulos *et al.* 2018). The amount of greywater is approximately 55%–75% of the total household wastewater, which produces up to 20–60 L/person/day or may reach several hundred litres a day depending on the level of water consumption (Carr *et al.* 2004; Ridderstolpe 2004; Shaikh & Younus 2015). Greywater could be one of the water resources used to solve water scarcity problems, such as groundwater recharge, landscaping, and plant irrigation, for integrated water management (Vuppaladadiyam *et al.* 2019; Boano *et al.* 2020). However, greywater should be properly treated before reuse or recycling (Masi *et al.* 2016). The general treatment system – for example, septic tanks, trickling filters, bioreactors, constructed wetland systems, or stabilization ponds – is usually set to minimize or eliminate some contaminants in greywater (Winblad & Simpson-Hébert 2004; Ramprasad & Philip 2016). The contaminants in greywater vary and depend on household activities. They can be divided into three types: physical, chemical, and biological contaminants. These factors can also contaminate the environment and become not only an environmental quality problem but also a public health problem. Thus, greywater should be managed to minimize and eliminate risks (WHO 2006). Exposure assessment is one of the methods that the WHO, FAO, and other international organizations usually use to estimate risk and reduce the risk of hazard exposure. Hazards are chemical, biological, or physical agents or a set of conditions that present a source of risk and have the potential to cause harm (Noutsopoulos *et al.* 2018; Vuppaladadiyam *et al.* 2019). Furthermore, the risk of exposure is evaluated to estimate the magnitude of the public health problem and to evaluate variability and uncertainty. As mentioned above, greywater is used for irrigation, discharge to surface water or percolation to groundwater (Al-Jayyousi 2003; Travis *et al.* 2010). Although the quality and use of greywater has been reported (Finley *et al.* 2009; Mah *et al.* 2009; Mohamed *et al.* 2013), exposure assessment should be a considerable process, especially in the reuse of greywater for crop irrigation (Ganoulis 2012). In addition, Carr *et al.* (2004) reported that the assessment of risks associated with wastewater use in agriculture should be first established to develop realistic guidelines for using wastewater in agriculture. Therefore, exposure assessment is the main objective of this study. This is used to assess the risk of the treated greywater reused for irrigation to prevent public health and environmental risks and promote water management for sustainability.

2. AREA DESCRIPTIONS

The Ban Lalom Mo community was selected due to the occurrence of water scarcity problems, especially in agricultural areas, and the existence of a greywater treatment system. Ban Lalom Mo is in Moo 4, located in Kokkruat Subdistrict, Muang District, Nakhon Ratchasima Province, in northeastern Thailand. Ban Lalom Mo has 413 houses: 332 houses within the area of Kokkruat Subdistrict Municipality and 81 houses within the area of the Subdistrict Administrative Organization. The total population is 1,665 and is divided into 887 women and 778 men. The population in the area of the Subdistrict Administrative Organization is approximately 250, divided into 130 women and 120 men. The main career path is agricultural services, such as rice or cassava farming.

This community lies on Lamtakong canal flowing from Lamtakong Dam. This canal is used for all activities of the community, such as agricultural and household activities and water supply production. The water supply under the Kokkruat Subdistrict Municipality produces approximately 4,800 m^3/day, while the water usage in Ban Lalom Mo village is 5,600 m^3/month or 187 m^3/day. The estimated consumption of water in the community is approximately 112 L/person/day. The water supply is sufficient for this community. However, water scarcity for cultivation is still a major problem, especially during dry and hot summers.

2.1. Greywater treatment system in the community

The greywater from the 81 houses within the area of the Subdistrict Administrative Organization is usually treated by the local conventional stabilization pond. This system has been in operation since 1996. It is composed of five ponds connected continuously, as shown in Figure 1. The volumes of ponds 1, 2, 3, 4, and 5 are 6 m^3, 15 m^3, 45 m^3, 450 m^3, and 1,350 m^3, respectively. The influent flow rate is approximately 6.48 m^3/day. The flow rates from pond numbers 1 to 2, 2 to 3, 3 to 4 and 4 to 5 are approximately 2.88, 3.89, 2.88, and 2.59 m^3/day, respectively.

3. MATERIALS AND METHODS

3.1. Greywater characterization

The characteristics of untreated and treated greywater were determined during every week of cultivation; thus, 13 samples of each type of greywater were analyzed. The parameters were pH, temperature, DO, conductivity, turbidity, COD, BOD, N, P, K, TS, TDS, TSS, grease and oil, organochlorine pesticides, Zn, Cu, Cr, Cd, Pb, Hg, As, total coliforms, and fecal coliforms (The National Environmental Board of Thailand 1994; WHO 2006). The treated greywater quality was compared with a standard of surface water quality used for agriculture advised by the National Environmental Board of Thailand (1994) and WHO (2006) guidelines for the safe use of wastewater, excreta, and greywater.

In addition, the tap water quality in the water supply system was monitored two times a year by the Water Supply Division in Kokkruat Subdistrict Municipality and the Laboratory of the Department of Health, Ministry of Public Health. All monitored parameters were compared with the water supply quality standard of the Department of Health, Ministry of Public Health, Thailand (Table 1). The results showed that not all parameters, especially heavy metals such as Zn, Cu, Cr, Cd, Pb, Hg, and As or enteric bacteria, were found, and the others did not exceed the standards of the Department of Health and were safe for the consumer.

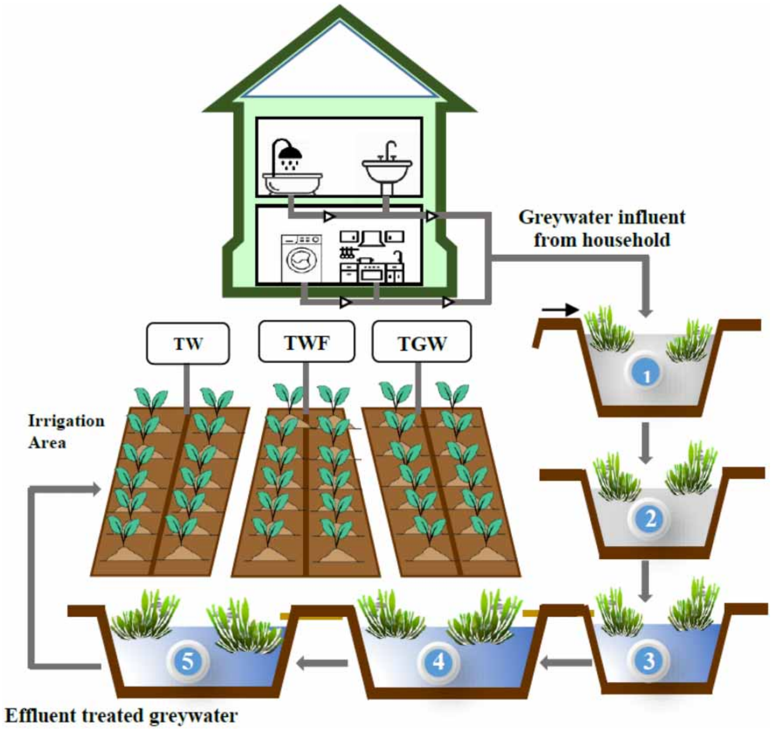

Figure 1 | Schematic diagram of the conventional stabilization pond system and plant irrigation area (no. 1 to no. 5: number of ponds 1– 5; TGW: treated greywater irrigation plot, TWF: tap water with fertilizer irrigation plot, TW: tap water irrigation plot).

Table 1 | Tap water quality in the community

Parameters	Values	Tap water quality criteria[a]
pH (pH at 25 °C)	8.0	6.5–8.5
Color (platinum cobalt)	<5	<15
Turbidity (NTU)	0.58	<5
Hardness (mg/L)	141	<500
TDS (mg/L)	216	<1,000
Fe (mg/L)	ND	<0.5
Mn (mg/L)	ND	<0.3
Cu (mg/L)	ND	<1.0
Zn (mg/L)	ND	<3.0
Pb (mg/L)	ND	<0.01
Cr (mg/L)	ND	<0.05
Cd (mg/L)	ND	<0.003
As (mg/L)	ND	<0.01
Hg (mg/L)	ND	<0.001
Sulfate (mg/L)	37	<250
Chloride (mg/L)	20	<250
Nitrate (mg/L)	0.48	<50
Fluoride (mg/L)	0.13	<0.7
Coliform bacteria (MPN/100 mL)	ND	ND
Fecal coliform bacteria (MPN/100 mL)	ND	ND

ND, not detected.
[a]Tap water quality criteria reported by the Department of Health, Ministry of Public Health, Thailand (Department of Health 2010).

3.2. Plant irrigation plots

The plant irrigation area was located beside the local conventional stabilization pond (Figure 1). The seven edible plants were four leafy vegetables, namely, coriander, dill, green shallot, pak-choi, and three fruiting vegetables, namely, winged bean, eggplant, and lady's finger. There were three vegetable plots, i.e., the effluent from the stabilization pond, called the treated greywater reuse plot (TGW), the tap water with fertilizer plot (TWF), as a comparative sample, and the tap water plot (TW), as a control. All vegetable plots had a length of 12 m and a width of 1 m. The total amount of water sample for each plot was approximately 200 L, two times a day. The nutrients in soil, such as N, P, K, and plant growth, were determined before and after cultivation.

3.3. Exposure assessment analysis

The contaminants in plants, such as Zn, Cu, Cr, Cd, Pb, Hg, As, and organochlorine pesticides, were analyzed. To assess the exposure rates, the exposure of metal contaminants was also determined to analyze the consumption intake in consumers using Equation (1) (USEPA 1989):

$$CDI = \frac{C \times IR \times EF \times ED}{BW \times AT} \tag{1}$$

where
CDI = chronic daily intake, mg/kg·d;
C = concentration of heavy metals in each plant, mg/kg;
IR = intake rate, mg/day (0.4 kg/day, (WHO 2003));
EF = exposure frequency, days/year (350 days/year);[1]
ED = exposure duration, years (30 years);[1]
BW = body weight, kg (70 kg);[2]
AT = average time, days (10,950 days).[1]

Finally, the heavy metal risk characterization was analyzed to estimate the risk of exposure and the impact on consumer health, and the hazard quotient was determined (Equation (2)). The hazard quotient calculation was defined as the estimated exposure divided by the reference doses or RfD (Table 2); therefore, HQ $\leq$1 indicates an acceptable risk, whereas HQ $>$1 indicates a possible risk of concern (USEPA 1989):

$$HQ = \frac{CDI}{RfD} \tag{2}$$

where
HQ = hazard quotient;
CDI = average daily intake from the exposure calculation, mg/kg·d;
RfD = reference doses, mg/kg·d.

Table 2 | Reference dose values of heavy metals

Heavy metals	RfD (mg/kg/day)
Zn	0.3[a]
Pb	0.0035[a]
Cd	0.001[a]
Cu	0.04[a]
Hg	0.002[b]

[a]USEPA (2005); Ghosh *et al.* (2012); Khan *et al.* (2013).
[b]IPCS (2003).

[1] The EPA recommended standard values.
[2] The average bodyweights used for an adult, which are the EPA recommended standard values for daily intake calculations.

3.4. Sampling and analysis

The quality of untreated and treated greywater was collected by grab sampling once a week until the end of the experiment. All samples were analyzed in triplicate. Physical characteristics, including the pH, temperature, electrical conductivity (EC), turbidity, total solid (TS) content, total dissolved solid (TDS) content, and total suspended solid (TSS) content, were analyzed. The pH, temperature and EC were measured by a multi-probe meter (Consort C532 T). Turbidity was measured by a turbidity meter (Hach 2100p). TS, TDS, and TSS were determined according to the *Standard Methods for the Examination of Water and Wastewater* (APHA/AWWA/WEF 1995) and thus were analyzed by drying at 103–105 °C. The chemical characteristics, including dissolved oxygen (DO), biochemical oxygen demand (BOD), chemical oxygen demand (COD), grease and oil, total nitrogen (total Kjeldahl nitrogen, TKN), total phosphorus (P), total potassium (K), and organochlorine pesticide content, were determined according to the *Standard Methods for the Examination of Water and Wastewater* (APHA/AWWA/WEF 1995). DO was measured by a DO meter (Hach Sension 6), whereas BOD and COD were analyzed by a BOD five-day test and the closed reflux method, respectively. The grease and oil components were analyzed by the Soxhlet extraction method. TN and TP were analyzed by the Kjeldahl method and ascorbic acid method, respectively. The organochlorine pesticides were analyzed by gas chromatography with an electron capture detector (GC-ECD). In addition, potassium and heavy metals such as cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), zinc (Zn), mercury (Hg), and arsenic (As) were also analyzed according to the *Standard Methods for the Examination of Water and Wastewater* (APHA/AWWA/WEF 1995) by an atomic absorption spectrometer (AAS). Biological characteristics such as enteric bacteria indicators, for example, total coliform bacteria and fecal coliform bacteria, were also determined according to Standard Method 9221 (APHA/AWWA/WEF 2005) by a multiple tube fermentation technique (MPN).

In addition, three samples of the edible part of each plant were collected randomly for chemical determination. Therefore, 21 samples of edible plants were prepared for analysis. Each sample was washed with deionized water, dried for 24 h in a hot air oven at 80 °C and then milled with a mortar and pestle. After that, three prepared samples of each edible plant were thoroughly mixed to form composite samples for further analysis. Acid digestion was performed for heavy metal analysis, whereas organic solvents or liquid–liquid extraction was performed for organochlorine pesticide analysis. These contaminants in the prepared samples were analyzed in triplicate. Heavy metals (Cd, Cr, Cu, Pb, Zn, Hg and As) were analyzed by atomic absorption spectrometry (AAS), and organochlorine pesticides were analyzed by GC-ECD (Jones & Case 1990; Kalra 1998; Lekkhong 2004).

Soil samples were also collected in the area of plants growing in each plot and mixed for composite sampling for nutrient analysis. The nutrients in the soil were determined before and after plantation as follows: N, P, and K were analyzed by the Kjeldahl method, Bray II method, and AAS, respectively (Department of Agriculture 2002).

Plant growth was observed and determined when plants grew by height measurement for leafy vegetables and fresh weight measurement for fruiting vegetables until harvesting.

4. RESULTS AND DISCUSSION

4.1. Characterization of untreated and treated greywater

The characteristics of the untreated greywater and treated greywater were identified and compared with the standards of Thailand and the WHO guidelines (Table 3). The untreated greywater exhibited a very low concentration of dissolved oxygen but exhibited very high concentrations of BOD and COD. This means that the greywater was contaminated with a very high concentration of organic matter. In addition, coliform bacteria can use oxygen to break down organic materials for their growth (Department of Industrial Works 2002; Pollution Control Department 2002; WHO 2006). The EC of greywater indicated the presence of very high values of inorganic matter, including heavy metals, solids, and nutrients such as nitrogen, phosphorus and potassium. High concentrations of nitrogen are usually considered indicators of urine contamination, but the source of phosphorus is detergent soaps from dishwashers and laundry (Leonard *et al.* 2016; Noutsopoulos *et al.* 2018; Vuppaladadiyam *et al.* 2019). These nutrients in greywater can cause eutrophication and algae blooms, which decrease dissolved oxygen and cause a severe reduction in water quality (Metcalf & Eddy 2013). Therefore, the greywater must be treated properly before discharge to the environment and compared with the country standard recommendation, especially for reused greywater (WHO 2006).

After treatment, the quality of the treated greywater showed that the important parameters in terms of BOD, COD, total nitrogen, solid content, Pb, Zn, and coliform bacteria decreased, and none of them exceeded the standard recommendation except BOD and coliform bacteria, whereas that of dissolved oxygen was slightly increased but did not meet the standard.

Table 3 | The characteristics of the untreated and treated greywater and their related standards

Characteristics	Average concentration[a] Untreated GW	Treated GW	Standard recommendation[b,c]
pH	7.23	7.28	5.0–9.0[b]
Temperature (°C)	26.26	24.65	<3 °C from natural temperature[b]
Dissolved oxygen, DO (mg/L)	0.85	1.49	4.0[b]
Electric conductivity, EC (μS/cm)	596.00	586.39	–
Turbidity (NTU)	38.57	18.2	–
Chemical oxygen demand, COD (mg/L)	29.74	9.95	–
Biochemical oxygen demand, BOD (mg/L)	106.99	35.63	2.0[b]
Total nitrogen, N (mg/L)	18.98	14.75	5–30[c]
Total phosphorus, P (mg/L)	0.330	0.339	–
Total potassium, K (mg/L)	1.06	1.07	–
Total solids, TS (mg/L)	408.98	353.70	<500[c]
Total dissolved solids, TDS (mg/L)	345.61	333.80	<450[c]
Total suspended solids, TSS (mg/L)	47.10	7.60	<50[c]
Grease and oil (mg/L)	20.24	3.30	–
Organochlorine pesticides (mg/L)	ND	ND	0.05[b]
Cadmium, Cd (mg/L)	0.0005	0.0005	0.05[b]
Chromium, Cr (mg/L)	ND	ND	0.05[b]
Copper, Cu (mg/L)	ND	ND	0.1[b]
Lead, Pb (mg/L)	0.002	0.001	0.05[b]
Zinc, Zn (mg/L)	0.021	0.011	1.0[b]
Mercury, Hg (mg/L)	ND	ND	0.002[b]
Arsenic, As (mg/L)	ND	ND	0.01[b]
Total coliforms (MPN/100 mL)	24,000	23,417	20,000[b]
Fecal coliforms (MPN/100 mL)	6,132	5,666	4,000[b] Relaxed to <10,000 for high-growing leaf crops or drip irrigation[c]

ND, not detected.
[a]The average concentration during 13 weeks of cultivation.
[b]The standard of the surface water quality used for agriculture notified by the National Environmental Board of Thailand (1994).
[c]*Guidelines for the Safe Use of Wastewater, Excreta, and Greywater* (WHO 2006).

This means that the local conventional stabilization pond could operate as one of the greywater treatment systems for a community.

To reuse the treated greywater for cultivation, heavy metal concentrations have been carefully considered. The order of the detected heavy metal concentrations was Zn > Pb > Cd; however, it was found that all of them were less than the standards (Table 3). Similar results in other research reported that Cd, Cr, Cu, Pb and Zn were detected at low levels and did not exceed their standards (Kananke *et al.* 2014; Noutsopoulos *et al.* 2018). Cd, Pb and Zn are classified as priority pollutants; however, they are necessary for the growth of biological life (Metcalf & Eddy 2013). The FAO (2003) also reported that Pb and Hg are not readily absorbed and translocated to edible vegetables. The amounts of heavy metal contamination depend upon community activities (WHO 2006). The most common sources of Cd are municipal sewage sludge applications, lead and zinc mines, phosphate fertilizers, fungicide, cadmium-containing water tap pipes and zinc and lead contents in nature (McBride 1995; Adesuyi *et al.* 2015; WHO 2019b). It can accumulate in the kidneys and liver (Divrikli *et al.* 2006). Pb is a natural element that is persistent in water and soil. Pb in the environment, however, largely comes from anthropogenic sources such as mining, refining, lead-containing water pipes or industrial sewage sludge or wastewater application (WHO 2019a). It is very concerning due to the possible impacts on multiple body systems, including the neurological, hematological, cardiovascular, and renal systems, especially in children, that influence their learning abilities and behavior (IPCS 1995; WHO 2019a). On the other hand, Zn is an essential element in the human diet and is required to maintain the functioning of the immune system. However, the excessive ingestion of zinc may lead to vomiting, renal damage, and cramps in humans (Alexander *et al.* 2006; Ladipo & Doherty 2011).

In addition to heavy metals, WHO guidelines state that organic matter should be at appropriate levels. Organic matter in the form of BOD at 110–400 mg/L improves soil fertility and increases crop productivity; in addition, coliform bacteria, especially fecal coliform, should be considered before reuse of the treated greywater because this is the most commonly used correlation with enteric pathogens (Keely *et al.* 2015; Benami *et al.* 2016; Boano *et al.* 2020), in which 1,000/100 mL is the level for the unrestricted concentration for irrigation of crops eaten raw, although this is relaxed to 10,000/100 mL for high-growing leaf crops or drip irrigation (WHO 2006). However, the risk from fecal coliform bacteria can be significantly reduced if foods are eaten after thorough cooking (WHO 2006). As a result, TGW was still contaminated with some organic compounds and coliforms, but these contaminants were adequately minimized, and some of them did not exceed the standard.

4.2. Soil nutrient and plant growth

The average nutrient concentrations in soil before and after cultivation are reported (Table 4). Nitrogen, phosphorus and potassium are macronutrients used by plants that can be found in greywater. These nutrients act as necessary fertilizer sources for plant productivity. The most beneficial and the most frequently excessive nutrient in reclaimed water is nitrogen (Metcalf & Eddy 2013). The nitrogen concentration was increased by approximately 300, 200 and 100 mg/kg when irrigated with TWF, TW and TGW, respectively. In soil, an appropriate concentration for cultivation should be in the range of 100–200 mg/kg (Maneepong 1994). The WHO (2006) study reported that a concentration of nitrogen between 5 and 30 mg/L in greywater does not affect soil but rather promotes rapid plant growth, as shown by the results of this study (Table 3). However, restrictions on use have been made if the concentration is 30 mg/L in greywater. On the other hand, total phosphorus concentrations exhibited very low levels in all plots. Phosphorus is stable in soils and is considered beneficial with no impacts on plants. However, if accumulation occurs over time, it can reach surface water through soil erosion and runoff and can cause eutrophication (WHO 2006). Mohamed *et al.* (2013) revealed that phosphorus in soil irrigated with greywater was higher than that in non-irrigated greywater soil, similar to the results of this study. In contrast, total potassium slightly decreased in all plots. In addition, the concentration of phosphorus can be in the range of 6–20 mg/L in greywater and increase plant productivity, while a normal content of potassium in greywater also increases plant productivity (WHO 2006). Therefore, the optimum concentration of nutrients, especially nitrogen, in soil was found in the TGW irrigation plot. In addition to the sources of fertility, the reuse of greywater for irrigation is still one of the water management strategies not only for water scarcity areas but also for minimizing the environmental impact of domestic greywater discharges.

As a result, all vegetables grew very well and rapidly when irrigated with TGW, similar to the TWF, which grew better than the TW. For the leafy vegetables, the greatest height of coriander and dill was found in the plot irrigated with treated greywater (Figure 2(a) and 2(b)). Similarly in other studies, it was found that leafy vegetables such as spinach and edible tubers such as potato and capsicum irrigated by greywater grew very rapidly and better than those irrigated by tap water (Zavadil 2009; Mzini & Winter 2015; Disha *et al.* 2020). The greatest heights of pak-choi and green shallots were found in the TWF plot; however, cultivation with TGW revealed greater growth than the others during two or three weeks depending on the plant species (Figure 2(c) and 2(d)). For the fruiting vegetables, winged beans and eggplants cultivated in tap water with fertilizer showed a greater weight than those cultivated in TGW and TW, whereas lady's fingers cultivated in treated

Table 4 | Nutrient concentrations in soil before and after irrigation

	Total nitrogen (mg/kg)			Total phosphorus (mg/kg)			Total potassium (mg/kg)		
Soil	TWF plot	TW plot	TGW plot	TWF plot	TW plot	TGW plot	TWF plot	TW plot	TGW plot
Before irrigation	100	100	100	0.54	0.12	0.31	36.67	46.33	53.67
After irrigation	400	300	200	0.26	0.02	0.9	35.67	45.33	50.67

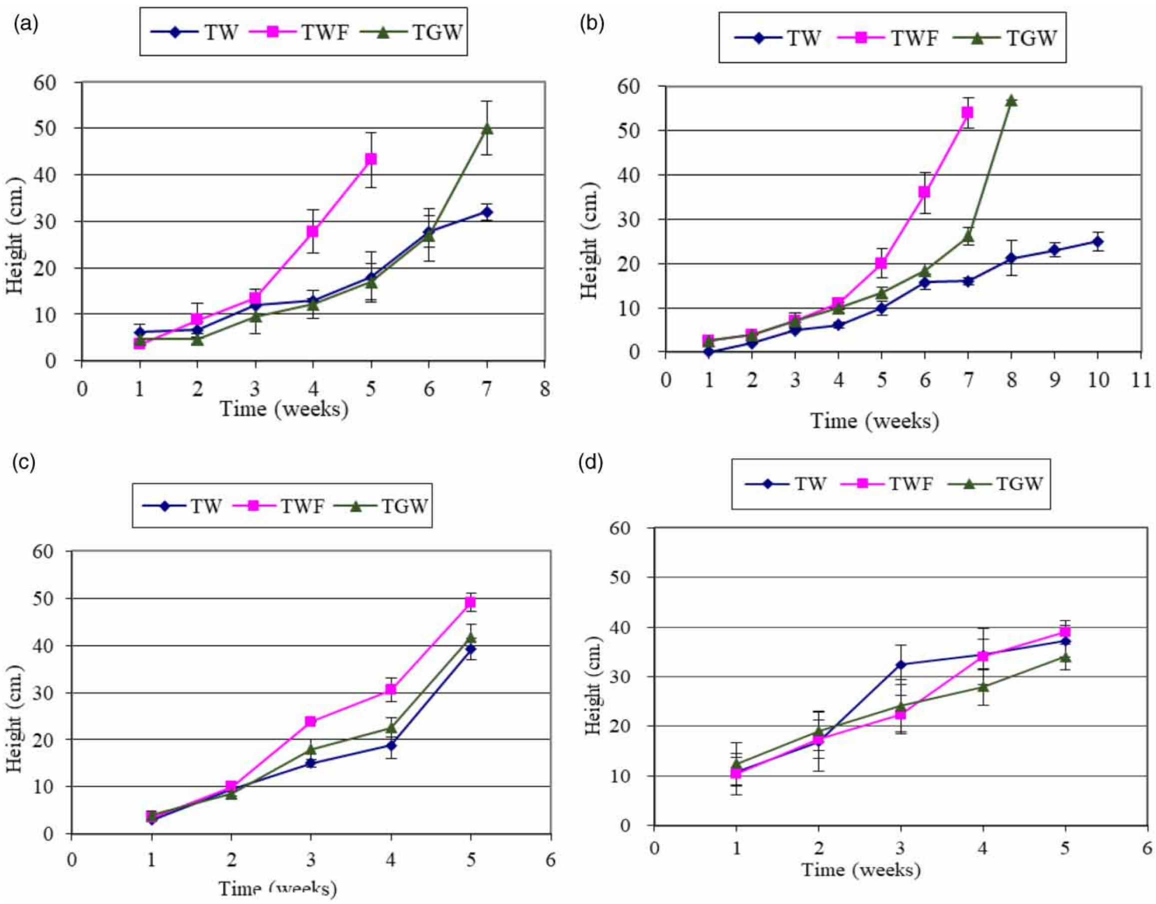

Figure 2 | Growth of (a) coriander, (b) dill, (c) pak-choi, and (d) and green shallot.

greywater exhibited a heavier weight than the others (Figure 3). This might be due to the nitrogen in treated greywater providing the appropriate amount for these plants. However, soil with treated greywater irrigation should be monitored throughout cultivation to observe the nutrient quantities that accumulate in the soil.

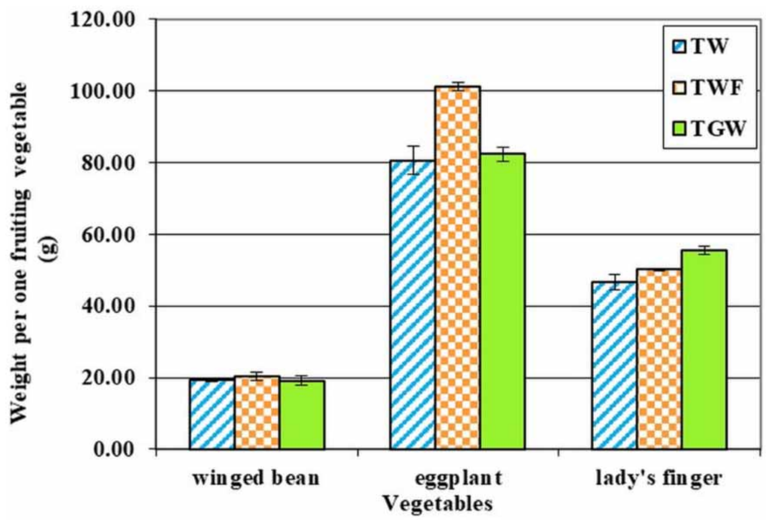

Figure 3 | Weight of vegetables after harvest (TW: vegetables irrigated with tap water; TWF: vegetables irrigated with tap water and fertilizer; TGW: vegetables irrigated with treated greywater).

4.3. Exposure assessment

The exposure assessment should be estimated based on the toxic chemicals found in the greywater, especially that reused for irrigation (Vigneswaran & Sundaravadivel 2004; Australian Health Ministers' Conference 2006; WHO 2006; USEPA 2012). The accumulation of contaminants affecting vegetables was determined (Table 5). The results showed that Cr and As were not found in any vegetables, whereas high levels of Zn and Cu were detected in all vegetables, although Cu was not found in tap water and treated greywater, as mentioned above. This may be because Cu can be found in nature and accumulates in green leafy vegetables, edible tubers and fruiting vegetables. However, Cu can be toxic to a number of plants at concentrations of 0.1–1.0 mg/L, and Zn can be toxic to many plants at widely varying concentrations, especially at 10 mg/L in reclaimed water, which requires short-term use (USEPA 2012). This is similar to other research, which found that Zn and Cu were detected in lettuce tissue at relatively high levels compared with other elements (Eregno *et al.* 2017).

Pb accumulated at high levels in coriander in all plots. Similar to other studies, leafy vegetables such as spinach, mint, and coriander tend to accumulate more heavy metals (Ghosh *et al.* 2012; Avci & Deveci 2013; Khan *et al.* 2013). From the results of plots irrigated with tap water and fertilizer, Pb levels were also found in all vegetables except dill. This might be due to the presence of some components in fertilizer that were related to lead or promoted lead accumulation. However, the detected lead in the treated greywater plot did not accumulate in green shallot, pak-choi, or dill plants.

Cd accumulated only in pak-choi at approximately 0.01 mg/kg in all plots, although it was found in the treated greywater. This might mean that almost all vegetables do not absorb and translocate this metal into their cells. In nature, however, cadmium can be found as complexes with Zn and Pb, and in addition, phosphate or phosphorus fertilizer in soil may induce the accumulation of cadmium in the soil and in plants (Alloway 1995). Furthermore, Yang *et al.* (2004) found that the increasing level of Zn accumulation in plants may induce an increase in the level of Cd accumulation in plants.

Hg accumulations were found in vegetables irrigated with tap water. The maximum concentration was found in winged bean. In contrast, Hg was not found in tap water and greywater, but it was still detected in vegetables. Mercury can be found in nature, especially in natural gas in the form of dimethylmercury and diethylmercury (Nriagu 2019). Furthermore, it can also be found as complexes with Cu and Zn (Alloway 1995). For these reasons, Hg could accumulate in vegetables.

Heavy metal accumulation was higher in leafy vegetables than in fruiting vegetables, except for mercury accumulation. Although the concentrations of these contaminants in all vegetables did not exceed the standard recommendations (Codex 1995; FAO/WHO 2001; TACFS 2005), the chronic daily intakes were evaluated (Table 6) to assess the exposure. These values represent consumer exposure from the intake of these chemicals in vegetables throughout their lifetime. The estimated chronic daily intakes of all heavy metals through the consumption of all vegetables were less than the reference dose. This value depends on the actual concentration of heavy metals existing in vegetables. The CDI exhibited the order of $Zn > Cu > Pb > Cd > Hg$, which followed the same order in all plots. In addition to heavy metals, the organochlorine pesticide did not accumulate in all plants, but it accumulated only in winged bean irrigated with tap water, where chronic daily intake was very low at less than 1.37×10^{-5} mg/kg·d.

4.4. Heavy metals risk characterization

To estimate the effect on consumer health, the hazard quotient was determined (Codex 1995; FAO/WHO 2001; TACFS 2005; USEPA 2012). The hazards of leafy vegetables indicated that coriander exhibited higher heavy metal accumulation than the others, with a total hazard quotient of approximately 0.05. In the fruiting vegetables, lady's finger showed a higher hazard quotient of heavy metal accumulation, with a total hazard quotient of 0.02, followed by winged bean and eggplant (Table 6). Other studies have reported that the hazard quotients for Pb and Ni were very high in fruiting vegetables, whereas that of Cd was very high in leafy vegetables in wastewater-irrigated areas (Singh *et al.* 2010). In the present study, the hazard quotient of all metals was far less than 1 in all vegetables; therefore, the consumption of vegetables irrigated with treated greywater did not pose health risk concerns.

5. CONCLUSIONS

Greywater is an important resource for areas of water scarcity. It can be used mainly for plant irrigation and groundwater recharge, but some contaminants in greywater can cause risks to public health and the environment. The risk of exposure is necessary knowledge and should be estimated. This study provided important results about the risk of exposure related to treated greywater reused for plant irrigation and concluded the following: (1) the conventional stabilization pond has a good capability to treat greywater; (2) almost all contaminants in treated greywater were not detected, whereas the others,

Table 5 | Average concentration of heavy metals and organochlorine group detection in each plant irrigated with tap water (TW), tap water and fertilizer (TWF), and treated greywater (TGW)

	Zn (mean ± SD) mg/kg			Pb (mean ± SD) mg/kg			Cd (mean ± SD) mg/kg			Cr (Mean ± SD) mg/kg			Cu (mean ± SD) mg/kg			As (Mean ± SD) μg/kg			Hg (mean ± SD) μg/kg			Organochlorine group mg/kg		
Vegetables	**TW**	**TWF**	**TGW**	**TW**	**TWF**	**TGW**	**TW**	**TWF**	**TGW**	**TW**	**TWF**	**TGW**	**TW**	**TWF**	**TGW**	**TW**	**TWF**	**TGW**	**TW**	**TWF**	**TGW**	**TW**	**TWF**	**TGW**
Green shallot	0.08 ± 0.00	0.22 ± 0.00	0.19 ± 0.01	ND	0.08 ± 0.01	ND	ND	ND	ND	ND	ND	ND	0.02 ± 0.00	0.08 ± 0.00	0.07 ± 0.00	ND	ND	ND	0.0005 ± 0.0001	ND	ND	ND	ND	ND
Pak-choi	0.39 ± 0.01	0.39 ± 0.01	0.32 ± 0.02	ND	0.07 ± 0.00	ND	0.01 ± 0.00	0.01 ± 0.00	0.01 ± 0.00	ND	ND	ND	0.06 ± 0.00	0.06 ± 0.00	0.06 ± 0.00	ND	ND	ND	ND	ND	ND	ND	ND	ND
Coriander	0.55 ± 0.01	0.50 ± 0.01	0.53 ± 0.02	0.11 ± 0.01	0.11 ± 0.01	0.12 ± 0.01	ND	ND	ND	ND	ND	ND	0.16 ± 0.01	0.17 ± 0.01	0.17 ± 0.01	ND	ND	ND	0.0002 ± 0.0001	ND	ND	ND	ND	ND
Dill	0.64 ± 0.02	0.52 ± 0.01	0.67 ± 0.02	0.12 ± 0.01	ND	ND	ND	ND	ND	ND	ND	ND	0.11 ± 0.00	0.21 ± 0.01	0.21 ± 0.01	ND	ND	ND	0.0002 ± 0.0001	ND	ND	ND	ND	ND
Winged bean	0.40 ± 0.01	0.42 ± 0.01	0.41 ± 0.01	ND	0.01 ± 0.00	0.02 ± 0.00	ND	ND	ND	ND	ND	ND	0.16 ± 0.01	0.22 ± 0.02	0.19 ± 0.01	ND	ND	ND	0.0009 ± 0.0002	0.0005 ± 0.0001	ND	<0.01 (dieldrin)	ND	ND
Eggplant	0.21 ± 0.01	0.25 ± 0.01	0.23 ± 0.01	0.01 ± 0.00	0.03 ± 0.00	0.02 ± 0.00	ND	ND	ND	ND	ND	ND	0.10 ± 0.00	0.16 ± 0.01	0.13 ± 0.01	ND	ND	ND	ND	ND	0.0002 ± 0.0000	ND	ND	ND
Lady's finger	0.59 ± 0.02	0.51 ± 0.02	0.56 ± 0.02	0.03 ± 0.00	0.03 ± 0.00	0.03 ± 0.00	ND	ND	ND	ND	ND	ND	0.16 ± 0.01	0.17 ± 0.01	0.15 ± 0.01	ND	ND	ND	0.0004 ± 0.0001	ND	0.0003 ± 0.0000	ND	ND	ND
Leafy vegetables[a]	**100**			**0.3**			**0.2**			**2.3**			**20**			**2.0 mg/kg**			**20**			**0.05**		
Fruiting vegetables[a]	**100**			**0.1**			**0.05**			**2.3**			**20**			**2.0 mg/kg**			**20**			**0.05**		

ND, not detected.
[a]The *Codex General Standard for Contaminants and Toxins in Food and Feed* (1995), FAO/WHO Joint Codex Alimentarius Commission (2001), and TACFS Thailand Standards (2005).

Table 6 | Chronic daily intake (mg/kg·d) and hazard quotient of heavy metals in each plant irrigated with tap water (TW), tap water and fertilizer (TWF), and treated greywater (TGW)

	Zn					
	TW		TWF		TGW	
Vegetables	CDI	HQ	CDI	HQ	CDI	HQ
Green shallot	1.10×10^{-4}	4.00×10^{-4}	3.01×10^{-4}	1.00×10^{-3}	2.60×10^{-4}	9.00×10^{-4}
Pak-choi	5.34×10^{-4}	1.80×10^{-3}	5.34×10^{-4}	1.80×10^{-3}	4.38×10^{-4}	1.50×10^{-3}
Coriander	7.53×10^{-4}	2.50×10^{-3}	6.85×10^{-4}	2.30×10^{-3}	7.26×10^{-4}	2.40×10^{-3}
Dill	8.77×10^{-4}	2.90×10^{-3}	7.12×10^{-4}	2.40×10^{-3}	9.18×10^{-4}	3.10×10^{-3}
Winged bean	5.48×10^{-4}	1.80×10^{-3}	5.75×10^{-4}	1.90×10^{-3}	5.62×10^{-4}	1.90×10^{-3}
Eggplant	2.88×10^{-4}	1.00×10^{-3}	3.42×10^{-4}	1.10×10^{-3}	3.15×10^{-4}	1.10 E-03
Lady's finger	8.08×10^{-4}	2.70×10^{-3}	6.99×10^{-4}	2.30×10^{-3}	7.67×10^{-4}	2.60×10^{-3}

	Pb					
	TW		TWF		TGW	
Vegetables	CDI	HQ	CDI	HQ	CDI	HQ
Green shallot	ND	ND	1.10×10^{-4}	3.13×10^{-2}	ND	ND
Pak-choi	ND	ND	9.59×10^{-5}	2.74×10^{-2}	ND	ND
Coriander	1.51×10^{-4}	4.31×10^{-2}	1.51×10^{-4}	4.31×10^{-2}	1.64×10^{-4}	4.70×10^{-2}
Dill	1.64×10^{-4}	4.70×10^{-2}	ND	ND	ND	ND
Winged bean	ND	ND	1.37×10^{-5}	3.90×10^{-3}	2.74×10^{-5}	7.80×10^{-3}
Eggplant	1.37×10^{-5}	3.90×10^{-3}	4.11×10^{-5}	1.17×10^{-2}	2.74×10^{-5}	7.80×10^{-3}
Lady's finger	4.11×10^{-5}	1.17×10^{-2}	4.11×10^{-5}	1.17×10^{-2}	4.11×10^{-5}	1.17×10^{-2}

	Cd					
	TW		TWF		TGW	
Vegetables	CDI	HQ	CDI	HQ	CDI	HQ
Green shallot	ND	ND	ND	ND	ND	ND
Pak-choi	1.37×10^{-5}	1.37×10^{-2}	1.37×10^{-5}	1.37×10^{-2}	1.37×10^{-5}	1.37×10^{-2}
Coriander	ND	ND	ND	ND	ND	ND
Dill	ND	ND	ND	ND	ND	ND
Winged bean	ND	ND	ND	ND	ND	ND
Eggplant	ND	ND	ND	ND	ND	ND
Lady's finger	ND	ND	ND	ND	ND	ND

	Cu					
	TW		TWF		TGW	
Vegetables	CDI	HQ	CDI	HQ	CDI	HQ
Green shallot	2.74×10^{-5}	7.00×10^{-4}	1.10×10^{-4}	2.70×10^{-3}	9.59×10^{-5}	2.40×10^{-3}
Pak-choi	8.22×10^{-5}	2.10×10^{-3}	8.22×10^{-5}	2.10×10^{-3}	8.22×10^{-5}	2.10×10^{-3}
Coriander	2.19×10^{-4}	5.50×10^{-3}	2.33×10^{-4}	5.80×10^{-3}	2.33×10^{-4}	5.80×10^{-3}
Dill	1.51×10^{-4}	3.80×10^{-3}	2.88×10^{-4}	7.20×10^{-3}	2.88×10^{-4}	7.20×10^{-3}
Winged bean	2.19×10^{-4}	5.50×10^{-3}	3.01×10^{-4}	7.50×10^{-3}	2.60×10^{-4}	6.50×10^{-3}
Eggplant	1.37×10^{-4}	3.40×10^{-3}	2.19×10^{-4}	5.50×10^{-3}	1.78×10^{-4}	4.50×10^{-3}
Lady's finger	2.19×10^{-4}	5.50×10^{-3}	2.33×10^{-4}	5.80×10^{-3}	2.05×10^{-4}	5.10×10^{-3}

	Hg					
	TW		TWF		TGW	
Vegetables	CDI	HQ	CDI	HQ	CDI	HQ
Green shallot	6.85×10^{-10}	3.42×10^{-7}	ND	ND	ND	ND
Pak-choi	ND	ND	ND	ND	ND	ND
Coriander	2.74×10^{-10}	1.37×10^{-7}	ND	ND	ND	ND
Dill	2.74×10^{-10}	1.37×10^{-7}	ND	ND	ND	ND
Winged bean	1.23×10^{-9}	6.16×10^{-7}	6.85×10^{-10}	3.42×10^{-7}	ND	ND
Eggplant	ND	ND	ND	ND	2.74×10^{-10}	1.37×10^{-7}
Lady's finger	5.48×10^{-10}	2.74×10^{-7}	ND	ND	4.11×10^{-10}	2.05×10^{-7}

ND, not detected.

such as Cd, Pb, and Zn, were detected but did not exceed the standard; (3) all kinds of plants grew very rapidly when irrigated with TGW, close to the TWF, which grew better than with TW; and (4) exposure assessment revealed that Zn, Pb, Cd, Cu and Hg were detected in all plots, depending on vegetable species. The maximum concentration was found in leafy vegetables, especially in dill. However, the chronic daily intake of heavy metals indicated that Zn was the highest intake throughout the lifetime of a plant, especially when consumed by dill and lady's fingers. The heavy metal risk characterization results also showed no impact on consumer health when compared with the standard that exhibited a hazard quotient of less than 1. Finally, this study suggested that greywater should be treated by the appropriate wastewater treatment system prior to utilization. Plants may be chosen to cultivate, for example, bulb vegetables such as eggplant, husky fruits, flowering and ornamental plants or plants that are not eaten fresh. The cultivation method could be soil irrigation. The contaminants in plant cultivation should be analyzed, especially heavy metals and pathogenic microbes. Cultivation should be done with caution. The community, or anyone who is interested in this matter, should monitor the consumer's health with a physical examination to determine the quantity of residues that could affect consumers in the long term.

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DATA AVAILABILITY STATEMENT

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

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