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Quality of roof-harvested rainwater – Comparison of different roofing materials

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ABSTRACT

The objective of the study reported in this paper was to assess the quality of harvested rainwater on the basis of the roofing materials used and the presence of lichens/mosses on the roofing surface. Four pilot structures with different roofing materials (i.e., wooden shingle tiles, concrete tiles, clay tiles [*Gi-Wa*] and galvanized steel) were installed in a field. The galvanized steel was found to be the most suitable for rainwater harvesting applications, with their resulting physical and chemical water quality parameters meeting the Korean guidelines for drinking water quality (e.g., pH (5.8–8.5), TSS <500 mg/L, $NO_3^- < 10$ mg/L, $SO_4^{2-} < 200$ mg/L, Al < 0.2 mg/L, Cu < 1 mg/L, Fe < 0.3 mg/L, Pb < 0.05 mg/L, Zn < 1 mg/L, and *E. coli (No detection)*). In the galvanized steel case, the relatively high water quality was probably due to ultraviolet light and the high temperature effectively disinfecting the harvested rainwater. It was also found that the presence of lichens and mosses may adversely affect the physical, chemical and microbiological quality of rainwater.

1. Introduction

In recent years, urbanization has contributed to increased surface runoff flooding and runoff contamination. Rainwater harvesting is one of the best available methods for establishing sustainable water cycles in urban developments (Lye, 2009; Kim et al., 2005; Pazwash and Boswell, 1997). Effective rainwater harvesting strategies are essential to meet the escalating demand for good quality water in sufficient quantities in urban areas that experience urban stream depletion and water shortages (Farreny, 2011; Fletcher et al., 2008; Van Roon, 2007; Forster, 1998). However, several studies have reported that rainwater harvesting may pose a public health risk because of its potential to carry microbial pathogens (Ahmed et al., 2008, 2011; Simmons et al., 2011). Most guidelines for rainwater utilization suggest that bacterial pathogens such as total coliforms and *Escherichia coli* (*E. coli)* are not detectable at counts <1 CFU/100 mL (Ahmed et al., 2011; WHO, 2004). The World Health Organization (WHO, 2004) suggests that the total coliform count for a water resource should be <10 CFU/100 mL in 95% of samples taken. For levels >20 CFU/100 mL, it recommends requiring further treatment for drinking water (Ahmed et al., 2011; WHO, 2004). Other studies have suggested that harvested rainwater used for drinking water should be assessed by monitoring the presence of fecal indicators and bacterial pathogens (Ahmed et al., 2010, 2011). The quality of harvested rainwater has also been found to be dependent both on the roof type and environmental conditions, i.e., the local climate and atmospheric pollution (Lee et al., 2010). Representative potential sources of nonpoint pollution on a rooftop are classified according to whether they are external or internal. External sources include airborne pollutants and organic substances from human activity, leaves and bird waste. Pathogens are found primarily in the feces of birds and mammals that have access to the rooftop. Internal sources of nonpoint pollution originate in the roofing materials themselves. Rainwater reacts physico-chemically with roof materials, and the presence of lichens and mosses on the roof also influences water quality over the long term. Numerous studies have analyzed the quality of harvested rainwater based on microbiological, physical and chemical parameters (see Table 1 for details). Lee and Jones (1982), for example, showed that roofing materials have no significant impact on the quality of such water.

The main objective of the study reported herein was to examine the effect of roofing materials on the chemical and microbiological quality of rainwater harvested for domestic use. This paper also provides guidelines for the selection of roofing materials that will aid in the harvesting of clean rainwater.

2. Materials and methods

2.1. Experimental design and sample collection

Conventional pilot-scale roofs were constructed with wooden shingles (Seoul, South Korea), concrete tiles (Seoul), clay tiles (or *Gi-Wa*; E-cheon, Gyeunggi-do,

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Table 1
Review of significant factors for harvested rainwater quality estimates.

Parameter, Reference	Physical[a]	Chemical[b]	Micro-biological[c]
Lee et al. (2010), Simmons et al. (2011)	O	O	O
Nicholson et al. (2009), Despins et al. (2009), Chang et al. (2004), Kingett Mitchell Ltd (2003), Spinks et al. (2003), Van Metre and Mahler (2003), Ariyananda and Mawatha (1999), Forster (1999), Steuer et al. (1997), Chang and Crowley (1993), Good (1993), Quek and Forster (1993), Thomas and Greene (1993), King and Bedient (1982)	O	O	–
Ahmed et al. (2008, 2011)	–	–	O
Lee and Jones (1982)	–	–	–

[a] Turbidity, TSS and temperature.
[b] pH, TOC, NO_3^-, PO_4^{2-}, Fe, Al, Cu, Cr, As, Pb, Zn and Cd.
[c] Fecal indicators and bacterial pathogen.

South Korea), and galvanized steel (zinc alloy coated steel, Seoul). The roofing materials were left in their natural condition for one year before the pilot-scale roofs were constructed. The roofs were angled 20.5° from the horizontal with a catchment area of 2.55 m^2 (length = 1.5 m, width = 1.7 m). The channel, gutter and downpipe systems were manufactured from PVC (SAMIK Co. Ltd). The gutter shape was half round formation. Fig. 1 shows a schematic diagram of the pilot-scale design. To divert the first flush, the inside of the downpipe was a floating ball and its position is located at the top of the first flush downpipe when it is full in the first flush tank. The runoff rainwater from rooftop flows into the rainwater tank. Harvested rainwater samples were collected from the four pilot-scale roofs made of different materials on 10 occasions in 2009 and 30 in 2010. Each sample was collected in a 500 mL sterilized bottle and subjected to chemical and microbiological analyses. The time of sample taking were July (2 events), August (3 events) and September (5 events) in 2009 and May (3 events), June (5 events), July (5 events), August (6 events), September (8 events) and October (3 events) in 2010.

2.2. *Sample analysis*

Table 2 summarizes the analytical parameters, methods and equipment used (APHA, 1995).

3. Results and discussion

3.1. *Quality assessment*

3.1.1. *Physical parameters*

3.1.1.1. pH. The average pH of the harvested rainwater from all of the pilot-scale roofs was in the near-neutral range (pH 6.0–7.9). The pH of the samples taken from the wooden shingle, concrete tile and clay tile roofs was higher than that from the galvanized steel roof. The rainwater harvested from the concrete tile roof had the highest level, with an average pH of 7.2 found in both the first flush and rainwater tanks, due to the reaction of pure rainwater (pH 4.8–5.9) to the alkaline components of the tiles. The pH values are shown in Fig. 2.

3.1.1.2. Total Suspended Solids. The average TSS concentrations of the concrete tile (309 mg/L) and galvanized steel (285.8 mg/L) samples were significantly higher than those of the two other roofing material samples in the first flush tanks: 213.9 mg/L for the wooden shingle and 219.3 mg/L for the clay tile. The average TSS concentrations of 35.65 mg/L for the wooden shingle roof (ρ-value < 0.05), 45 mg/L for the concrete tile roof (ρ-value < 0.05) and 41.6 mg/L for the clay tile roof (ρ-value < 0.05) were higher than the average TSS concentration of 15.1 mg/L for the galvanized steel roof after the first flush. Interestingly, the lower TSS concentration measured for the galvanized steel after the first flush displayed an almost total wash-off in the first flush stage. The other roofing materials, in contrast, exhibited no significant differences. The TSS found in the water from the concrete and clay tile roofs were mainly inorganic materials (i.e., air dust and roofing debris), whereas those found in that from the wooden shingle tile roof also included organic materials (i.e., lichens, mosses and plants). It was further observed that lichens, mosses and plants frequently colonized the wooden shingle tile roof. The U.S. Environmental Protection Agency's (USEPA, 2004) suggested guideline for non-potable urban water reuse is that sedimentation should not exceed 5 mg/L. The levels found in the harvested rainwater from the rainwater tanks of all four pilot-scale roofs in this study failed to exceed 5 mg/L within 2–3 h of rain cessation. The TSS values are shown in Fig. 3.

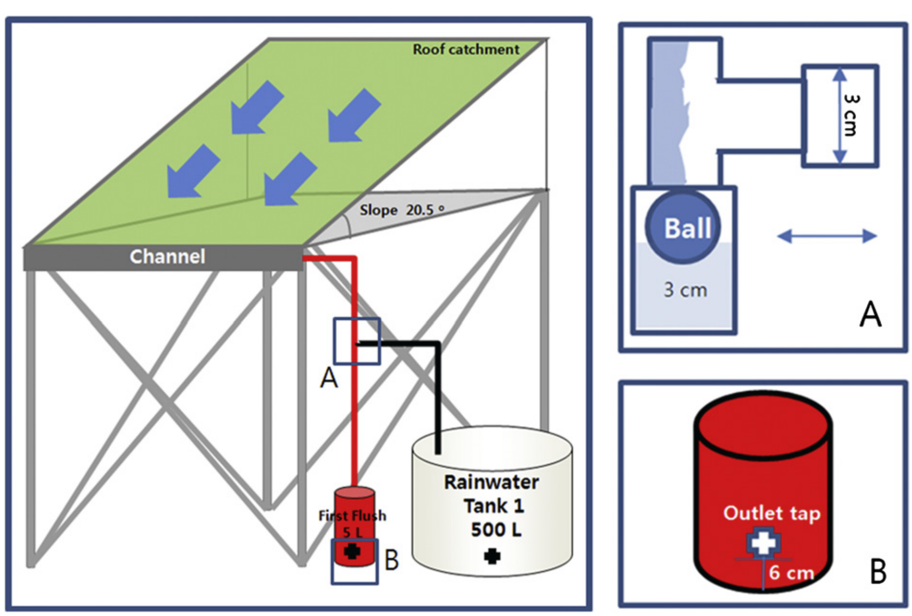

Fig. 1. Schematic diagram of the pilot-scale design.

Table 2
Review of analytical parameters and methods.

	Parameter	Method and equipment
Physical	pH	Metrohm, Model 826 series
	TSS	APHA, 1995
Chemical	NO_3, SO_4	DIONEX ICS 3000
	Metal	SHIMADZU AA-7000
		APHA, 2005
	TOC	GE-Sievers 5310 C, USA
Micro-biological	TC, *E. coli*	ISO method 9308-1
	Enterococci	ISO 7899-2
	Pseudomonas spp.	APHA, 1995
	Salmonella spp.	
	Cryptosporidium spp.	

3.1.2. Chemical parameters

3.1.2.1. Total Organic Carbon. The TOC concentrations of the water from the different roofing materials are given in Fig. 4. The highest concentration (average = 49.7 mg/L) was found in the first flush and rainwater tanks of the wooden shingle roof, no doubt due to the weathering of the roofing material, which is itself composed of organic material. Additionally, the presence and growth of lichen was also observed mainly in the wooden shingle roof. The other roofing materials displayed no significant differences in TOC concentrations: 32.9 mg/L for the concrete tiles, 35.6 mg/L for the clay tiles and 31.8 mg/L for the galvanized steel, as measured in the first flush tanks (ρ-value $<$ 0.05). It should be noted that the drinking water guidelines promulgated by South Korea, the USEPA and the European Commission have no stated limits for TOC concentrations.

3.1.2.2. Nitrate and sulfate. The nitrate (NO_3^-) and sulfate (SO_4^{2-}) concentrations of the rainwater samples from the different roofing materials are displayed in Fig. 5. These concentrations were expected due to the presence/growth of lichens, animal waste and wet/dry deposition on the rooftops. The average concentrations found in the wooden shingle sample (3.3 NO_3^- mg/L and 5.57 SO_4^{2-} mg/L) were higher than those for the three other roofing materials in the first flush tank (ρ-value $<$ 0.05): 2.55 NO_3^- mg/L and 3.64 SO_4^{2-} mg/L, 1.89 NO_3^- mg/L and 3.1 SO_4^{2-} mg/L, and 2.8 NO_3^- mg/L and 2.87 SO_4^{2-} mg/L for the concrete tile, clay tile and galvanized steel samples, respectively. This result is unsurprising, given the different characteristics of the four roofing materials. Wooden shingle roofs, in particular, have a relatively high degree of porosity compared to the other three, and colonies of lichens and mosses are frequently found on this type of roof. The associated microbiological processes produced higher levels of nitrate and sulfate. The results from the rainwater tank measurements showed average concentrations of 0.02 NO_3^- mg/L and 0.11 SO_4^{2-} mg/L in water from the galvanized steel roof, significantly lower concentrations than those found for the three other roofing materials: 0.30 NO_3^- mg/L and 0.65 SO_4^{2-} mg/L for the wooden shingle tiles, 0.28 NO_3^- mg/L and 0.38 SO_4^{2-} mg/L for the concrete tiles, and 2.81 NO_3^- mg/L and 2.87 SO_4^{2-} mg/L for the clay tiles. The water in neither the first flush

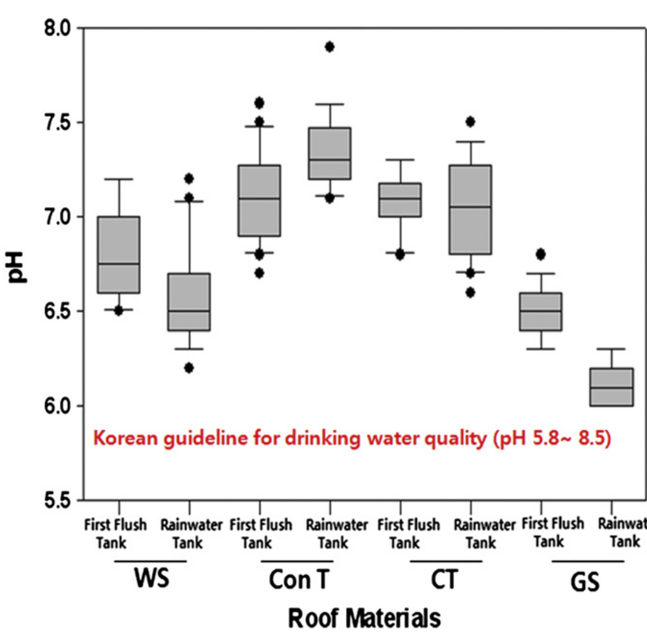

Fig. 2. Box plot diagram of the pH values of rainwater harvested from the four roofing materials (total events [$n = 40$]). Note: WS = Wooden Shingle; Con T = Concrete Tile; CT = Clay Tile (*Gi-Wa*); GS = Galvanized Steel.

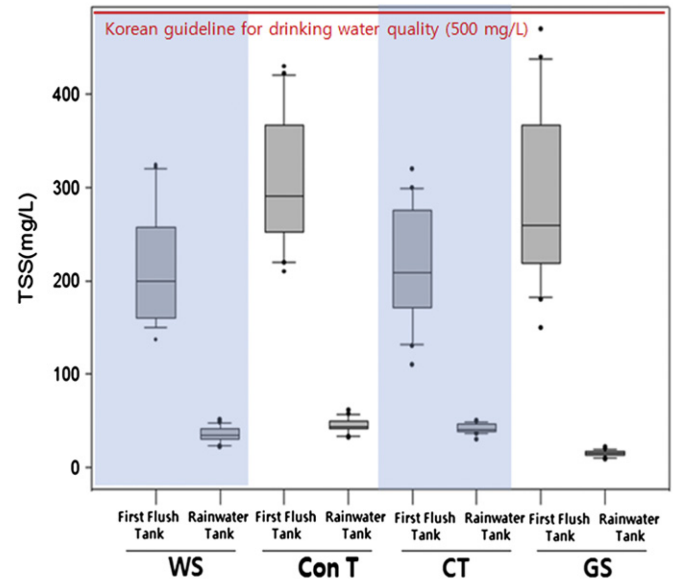

Fig. 3. Box plot diagram of the TSS concentrations in the rainwater harvested from the four roofing materials (total events [$n = 40$]).

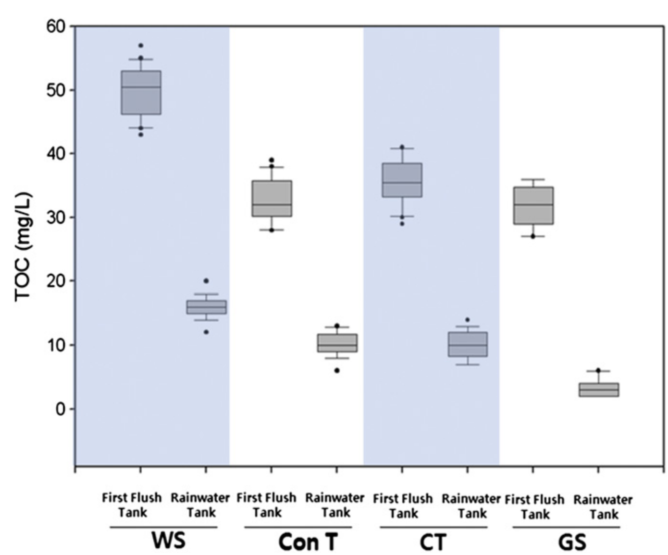

Fig. 4. Box plot diagram of TOC concentrations in rainwater harvested from the different roofing materials (total events [$n = 40$]).

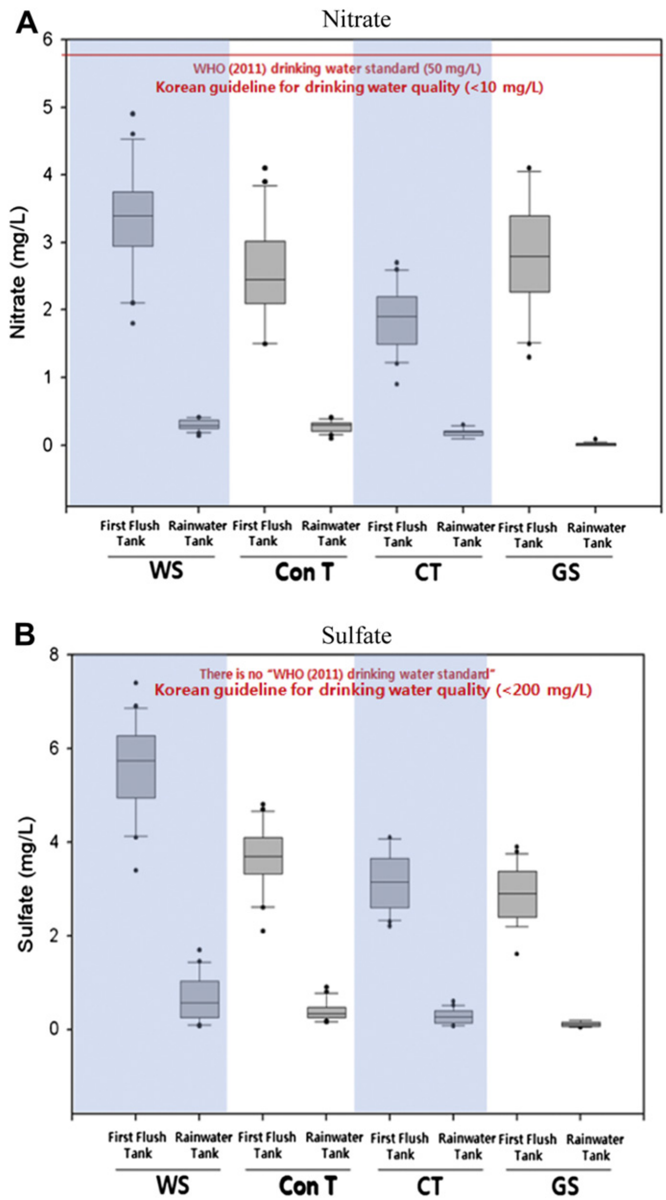

Fig. 5. Box plot diagram for the nitrate (NO_3^-) and sulfate (SO_4^{2-}) concentrations in the water collected from the different roofing materials (total events [$n = 40$]).

nor rainwater tanks exceeded the maximum contaminant level (MCL) recommended in the USEPA (10 NO_3^- mg/L) and European Commission guidelines (50 NO_3^- mg/L and 250 SO_4^{2-} mg/L) (Mendez et al., 2011; Nicholson et al., 2009; European Commission, 1998).

(A) Nitrate
(B) Sulfate

3.1.2.3. Metals. The total metal concentrations (i.e., Al, Cu, Fe, Pb and Zn) are shown in Fig. 6. The average concentration of total aluminum was found to be 227 μg/L for the wooden shingles, 535 μg/L for the concrete tiles, 243 μg/L for the clay tiles and 622 μg/L for the galvanized steel in the first flush tank (Fig. 6-A). Following the first flush, the average concentration of total aluminum in the rainwater tank was measured at 43 μg/L for the wooden shingles, 99 μg/L for the concrete tiles, 36 μg/L for the clay tiles and 33 μg/L for the galvanized steel (Fig. 6-A). The samples from the galvanized steel roof had strikingly higher total aluminum levels than those from the other roofs in the first flush tank (ρ-value < 0.05). Given that a galvanized steel roof is composed of zinc and iron, it may be that the aluminum levels found in this study originated in atmospheric dust and dry deposition. It should also be noted that the water in the rainwater tank after the first flush did not exceed the USEPA's recommended MCL for aluminum in drinking water (200 μg/L).

The average concentration of total copper in the first flush tank was measured at 34 μg/L for the wooden shingles, 58 μg/L for the concrete tiles, 37 μg/L for the clay tiles and 59 μg/L for the galvanized steel (Fig. 6-B). The average concentration of total copper in the rainwater tank after the first flush fell to 9 μg/L for the wooden shingles, 15 μg/L for the concrete tiles, 12 μg/L for the clay tiles and 16 μg/L for the galvanized steel (Fig. 6-B). In sum, the average total concentration of copper in the water samples from the concrete tile and galvanized steel roofs was notably higher than that in those from the wooden shingle and clay tile roofs. Atmospheric deposition may be the source of the copper we found. Wooden shingles and clay tiles are densely porous materials, and the lower copper concentrations measured for them here is likely the result of pollutants becoming trapped within their porous sites. None of the rainwater samples exceeded the USEPA's MCL for copper in drinking water (1300 μg/L).

The average concentration of total iron was measured at 154 μg/L for the wooden shingle, 160 μg/L for the concrete tiles, 155 μg/L for the clay tiles and 302 μg/L for the galvanized steel in the first flush tank (Fig. 6-C). The average concentration of total iron in the rainwater tank after the first flush was measured at 23 μg/L for the wooden shingles, 48 μg/L for the concrete tiles, 24 μg/L for the clay tiles and 27 μg/L for the galvanized steel (Fig. 6-C). The galvanized steel samples had higher average total iron concentrations than the other three in the first flush tank (ρ-value < 0.05). The source of this iron may be the galvanized steel itself and atmospheric deposition. With the exception of the water from the first flush tank of the galvanized steel roof, none of the samples exceeded the USEPA's MCL guideline for iron in drinking water (300 μg/L).

The average concentration of total lead was measured at 10 μg/L for the wooden shingles, 14 μg/L for the concrete tiles, 11 μg/L for the clay tiles and 12 μg/L for the galvanized steel in the first flush tank (Fig. 6-D). After the first flush, the average concentration of total lead in the rainwater tank was measured at 3 μg/L for the wooden shingles, 5 μg/L for the concrete tiles, 3 μg/L for the clay tiles and 3 μg/L for the galvanized steel (Fig. 6-D). The average total lead concentrations found in the concrete tile and galvanized steel samples in the first flush tank were higher than those in the samples from the other two roofing materials. The source of the lead may be atmospheric deposition and the galvanized steel itself. With the exception of the water from the first flush tank of the concrete tile roof, the harvested rainwater in all of the tanks met the USEPA's MCL guideline for lead in drinking water (15 μg/L).

Finally, the average concentration of total zinc was measured at 135 μg/L for the wooden shingle, 196 μg/L for the concrete tile, 131 μg/L for the clay tile and 428 μg/L for the galvanized steel samples in the first flush tank (Fig. 6-E). The average concentration of total zinc in the rainwater tank after the first flush was measured at 18 μg/L for the wooden shingle, 38 μg/L for the concrete tile, 19 μg/L for the clay tile and 74 μg/L for the galvanized steel roofs (Fig. 6-E). The average total zinc concentration of the water samples from the galvanized steel roof was higher than those from the other three roofing materials in the first flush tank (ρ-value < 0.05). The source of the zinc could be the galvanized steel itself and atmospheric deposition. The rainwater harvested from all of the tanks met the USEPA's MCL for zinc in drinking water (5000 μg/L).

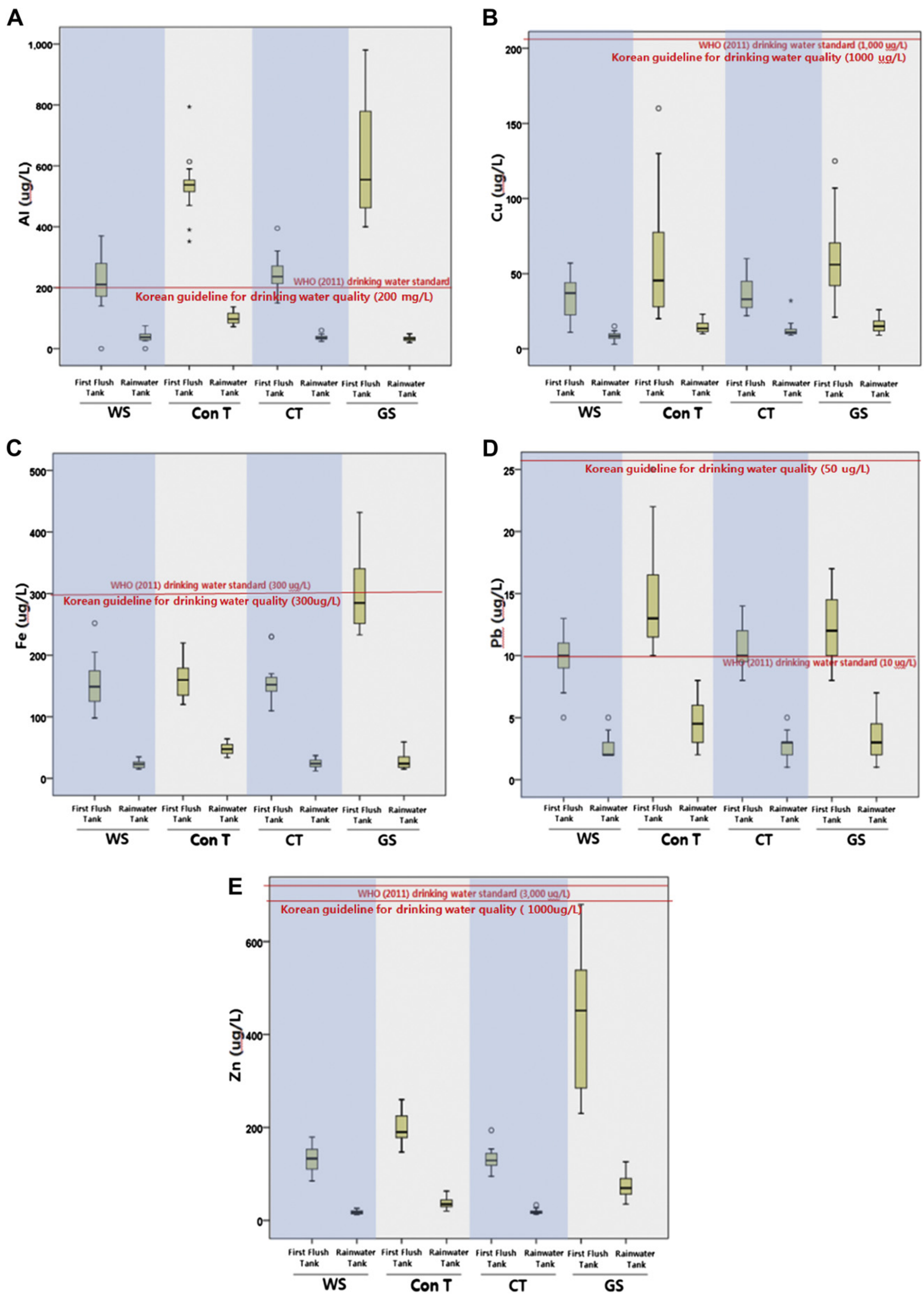

Fig. 6. Box plot diagram for the total metal (Al, Cu, Fe, Pb and Zn) concentrations in the water from different roofing materials (total events [$n = 40$]).

3.1.3. Microbiological parameters

3.1.3.1. Fecal indicators. To determine the suitability of harvested rainwater as a source of drinking water, the samples were tested for total coliform count, *E. coli* and enterococci. These fecal indicators may be fecal in origin, but can also originate from dust and plants. In the samples from the first flush tanks, we detected average total coliform counts of 131 CFU/100 mL for the wooden shingle, 197 CFU/100 mL for the concrete tile, 76 CFU/100 mL for the clay tile and 70 CFU/100 mL for the galvanized steel roofs (Fig. 7-A). In those from the rainwater tanks after the first flush, we found average total coliform counts of 12 CFU/100 mL for the wooden shingle, 12 CFU/100 mL for the concrete tile, 2 CFU/100 mL for the clay tile and <1 CFU/100 mL for the galvanized steel roofs (Fig. 7-A). The result for the galvanized steel roof samples from the rainwater tank is particularly notable: 82.5% of these samples had no measurable total coliform count because the total coliforms were completely washed away by the first flush. The WHO (2004) recommends a total coliform count <10 CFU/100 mL in 95% of samples collected from a particular drinking water source (see also Ahmed et al., 2011).

The average *E. coli* counts in the first flush tank were 14 CFU/100 mL for the wooden shingle roof, 18 CFU/100 mL for the concrete tile roof, 8 CFU/100 mL for the clay tile roof and 4 CFU/100 mL for the galvanized steel roof (Fig. 7-B). In the samples from the rainwater tanks after the first flush, in contrast, we found average *E. coli* counts of 1 CFU/100 mL for the wooden shingle roof, 2 CFU/100 mL for the concrete tile roof, <1 CFU/100 mL for the clay tile roof and zero CFU/100 mL for the galvanized steel roof (Fig. 7-B). All of the samples (not shown) taken from the rainwater tank of the galvanized steel roof were negative for *E. coli* because any that had been present was completely washed away by the first flush. The Australian Drinking Water Guidelines (ADWG, 2004) state that the *E. coli* count should be zero CFU/100 mL (see also Ahmed et al., 2011).

The average enterococci counts in the first flush tank were measured at 1 CFU/100 mL for the wooden shingle roof, 2 CFU/100 mL for the concrete tile roof, <1 CFU/100 mL for the clay tile roof and <1 CFU/100 mL for the galvanized steel roof (Fig. 7-C). Of these first flush tank samples, 88% of those from the wooden shingle roof, 77% of those from the concrete tile roof, 92% of those from the clay tile roof and 92% of those from the galvanized steel roof (none shown) had no measurable levels of enterococci. In the rainwater tank, 100% of the samples from all four roofing materials

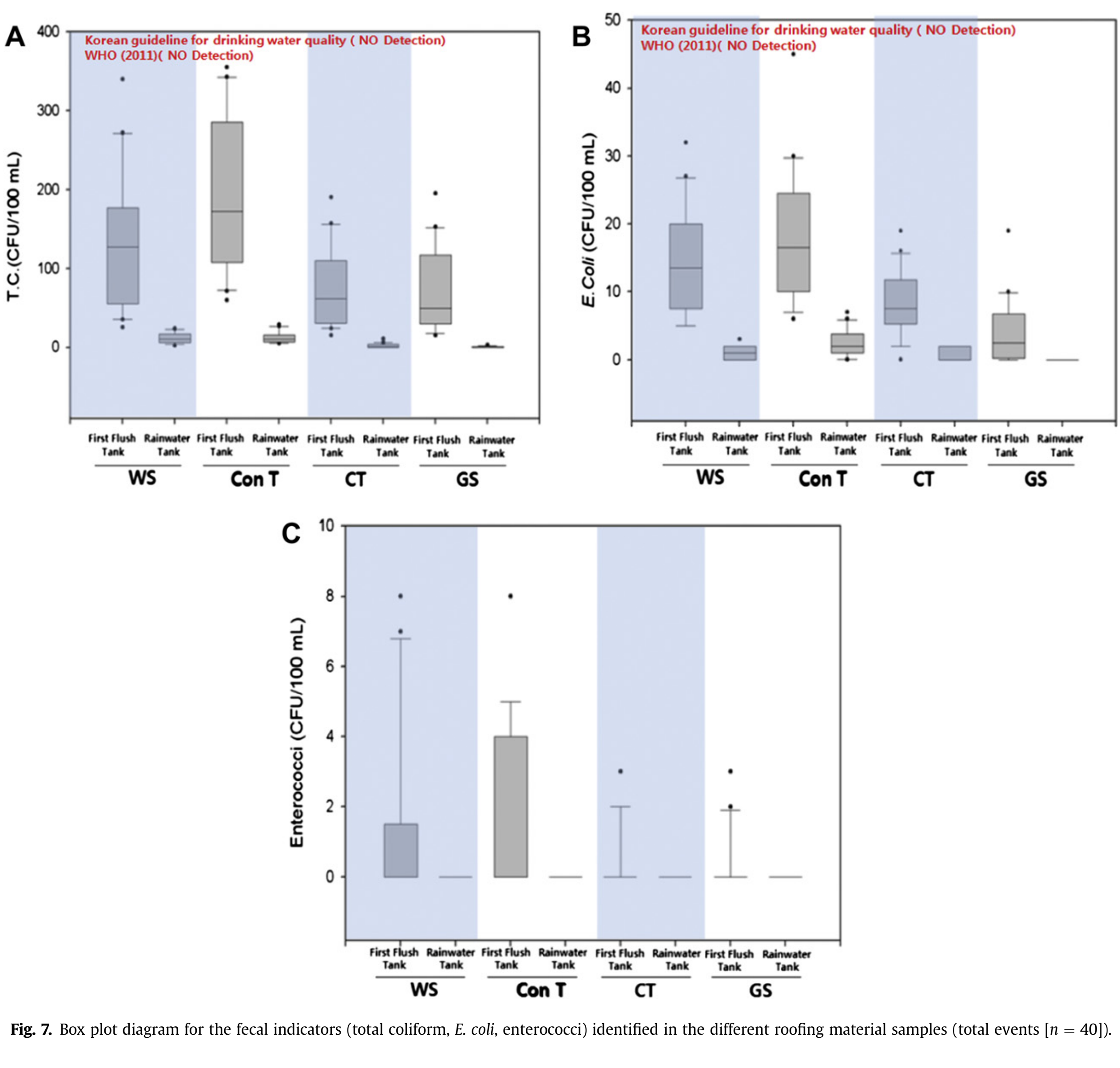

Fig. 7. Box plot diagram for the fecal indicators (total coliform, *E. coli*, enterococci) identified in the different roofing material samples (total events [$n = 40$]).

Table 3
Percentage of samples testing positive for bacterial pathogens (*Pseudomonas* spp., *Salmonella* spp., *Cryptosporidium* spp.) by roof material (total events [$n = 40$]).

	% of samples testing positive for bacterial pathogens					
	Pseudomonas spp.		*Salmonella* spp.		*Cryptosporidium* spp.	
	F.F.T.[a]	R.T.[a]	F.F.T.	R.T.	F.F.T.	R.T.
WS	12.5	ND	5	ND	ND	ND
Con T	7.5	ND	5	ND	ND	ND
CT	2	ND	ND	ND	ND	ND
GS	ND	ND	ND	ND	ND	ND

[a] F.F.T. = First Flush Tank; R.T. = Rainwater Tank; ND = not detected.

tested negative for enterococci (Fig. 7-C). Thus, all of the harvested rainwater in this study met the recommended enterococci level (zero CFU/100 mL) in the ADWG (2004; see also Ahmed et al., 2011).

3.1.3.2. Bacterial pathogens. The harvested rainwater was also analyzed for bacterial pathogens, i.e., *Pseudomonas* spp., *Salmonella* spp. and *Cryptosporidium* spp. (see Table 3). *Cryptosporidium* spp. was not detected in any of the water samples. The percentages of samples that tested positive for *Pseudomonas* spp. in the first flush tank were 12.5% for the wooden shingle roof, 7.5% for the concrete tile roof, and 2% for the clay tile roof and 0% for the galvanized steel roof. The percentages for *Salmonella* spp. were 5% for the wooden shingle roof and 5% for the concrete tile roof. No *Salmonella* spp. was found in the clay tile or galvanized steel samples. The absence of bacterial pathogens in the water samples from the galvanized steel roof may be due to the high temperatures of these roofs, which reach 75 °C–85 °C in the summertime. In addition, galvanized steel concentrates ultraviolet sunlight, which acts as a disinfectant against bacterial pathogens. The highest number of bacterial pathogens was found in the wooden shingle roof samples, mostly likely because of the greater presence and growth of lichens, mosses and plants on this roofing material.

3.2. Analysis of Spearman's correlation coefficient (r)

Table 4 shows that the correlation coefficient (r) between the antecedent dry day (ADD) and TSS has a significantly positive value of 0.964 ($p < 0.01$). The ADD also has strong relationships with TOC ($r = 0.636$, $p < 0.01$), NO_3^- ($r = 0.634$, $p < 0.01$), SO_4^{2-} ($r = 0.725$, $p < 0.01$), Al ($r = 0.841$, $p < 0.01$), Cu ($r = 0.823$, $p < 0.01$), Fe ($r = 0.847$, $p < 0.01$), Pb ($r = 0.681$, $p < 0.01$), Zn ($r = 0.937$, $p < 0.01$), total coliforms ($r = 0.758$, $p < 0.01$), *E. coli* ($r = 0.743$, $p < 0.01$) and enterococci ($r = 0.623$, $p < 0.01$). In addition, TSS has positive relationships with TOC ($r = 0.580$, $p < 0.01$), NO_3^- ($r = 0.342$, $p < 0.01$), SO_4^{2-} ($r = 0.436$, $p < 0.01$), Al ($r = 0.653$, $p < 0.01$), Cu ($r = 0.627$, $p < 0.01$), Fe ($r = 0.553$, $p < 0.01$), Pb ($r = 0.680$, $p < 0.01$), Zn ($r = 0.631$, $p < 0.01$), total coliforms ($r = 0.647$, $p < 0.01$), *E. coli* ($r = 0.651$, $p < 0.01$) and enterococci ($r = 0.424$, $p < 0.01$). Interestingly, TSS has a stronger relationship with inorganic components (i.e., Al, Cu, Fe, Pb and Zn) than organic components (i.e., TOC, NO_3^- and SO_4^{2-}), demonstrating that the inorganic components originate in dust and sand as dry deposition, whereas the organic components originate in animal waste, lichens and mosses on the roof. Relative to the correlation coefficient for the metals, TOC enjoys a strongly positive relationship with NO_3^- ($r = 0.671$, $p < 0.01$) and SO_4^{2-} ($r = 0.733$, $p < 0.01$). The total coliform count also has a strongly positive relationship with NO_3^- ($r = 0.695$, $p < 0.01$) and SO_4^{2-} ($r = 0.618$, $p < 0.01$), as does *E. coli* ($r = 0.675$ for NO_3^-, $p < 0.01$; $r = 0.659$ for SO_4^{2-}, $p < 0.01$) and enterococci ($r = 0.553$ for NO_3^-, $p < 0.01$; $r = 0.514$ for SO_4^{2-}, $p < 0.01$). On the basis of these results, we can conclude that TOC, NO_3^- and SO_4^{2-} concentrations in samples of water for domestic use can be indicators of the potential presence of microbes.

4. Conclusion

Four types of roofing materials (wooden shingles, concrete tiles, clay tiles [*Gi-Wa*] and galvanized steel), which are widely used in South Korea, were analyzed to determine their suitability for use in the harvesting of rainwater for domestic use. The monitoring parameters were physical (pH and TSS), chemical (TOC, NO_3^-, SO_4^{2-} and metal ions) and microbiological (total coliforms, *E. coli*, enterococci, *Pseudomonas* spp., *Salmonella* spp. and *Cryptosporidium* spp.).

In summary, the galvanized steel was found to be the most suitable for rainwater harvesting applications after the first flush, with their resulting physical, chemical and biological water quality parameters meeting the Korean and WHO guidelines for drinking water quality (e.g., pH 5.8–8.5, TSS < 500 mg/L, NO_3^- < 10 mg/L, SO_4^{2-} < 200 mg/L, Al < 0.2 mg/L, Cu < 1 mg/L, Fe < 0.3 mg/L, Pb < 0.05 mg/L, Zn < 1 mg/L, and *E. coli (No detection)*).

With regard to the physical and chemical parameters, the findings of this study show that the type of roofing material used has some influence on the quality of harvested rainwater. The TSS and metal concentrations in the samples from the galvanized steel roof were at higher levels in the first flush tank. The three other

Table 4
Spearman correlation coefficients between the water quality and microbiological parameters for the full set of roofs (total events [$n = 40$]).

	pH	ADD	TSS	TOC	NO_3	SO_4	Al	Cu	Fe	Pb	Zn	TC	EC	ENT
pH	1.000	0.417**	0.547**	0.384**	0.363**	0.574*	0.238**	−0.251	−0.233	0.125	−0.256*	0.381*	0.313**	0.351
ADD[a]		1.000	0.964**	0.636**	0.634**	0.725**	0.841**	0.823**	0.847**	0.681*	0.937**	0.758**	0.743**	0.623**
TSS			1.000	0.580**	0.342**	0.436**	0.653**	0.627**	0.553**	0.680*	0.631**	0.647**	0.651**	0.424**
TOC				1.000	0.671**	0.733**	0.342**	0.279**	0.218**	0.202*	0.265**	0.889**	0.860**	0.713**
NO_3					1.000	0.697**	0.544**	0.533**	0.533**	0.408	0.441**	0.695**	0.675**	0.553**
SO_4						1.000	0.495**	0.487**	0.484**	0.345	0.478**	0.618**	0.659**	0.514**
Al							1.000	0.769**	0.633**	0.677**	0.781**	0.431**	0.346**	0.254*
Cu								1.000	0.547**	0.650**	0.619**	0.337**	0.380**	0.233*
Fe									1.000	0.622*	0.689**	0.368**	0.403**	0.275
Pb										1.000	0.787	0.686**	0.781*	0.255
Zn											1.000	−0.191	-0.189*	−0.112
TC												1.000	0.960**	0.649*
EC													1.000	0.656*
ENT[b]														1.000

*$p < 0.05$; **$p < 0.01$.
[a] ADD = Antecedent Dry Day.
[b] ENT = Enterococci.

roofing materials under study have a relatively higher degree of porosity than galvanized steel, which serves to trap pollutants. Lichens and mosses were frequently found on the wooden shingle tile, concrete tile and clay tile roofs examined here, with the wooden shingle tiles boasting the greatest degree of colonization. The concentrations of TOC, NO_3^-, and SO_4^{2-} in the water samples taken from the first flush tank of the wooden shingle tile roof were relatively high. However, the rainwater harvested from all four types of roofs became acceptable for domestic use after the first flush.

Microbiological issues, in contrast, merit careful consideration in the selection of roofing materials for rainwater harvesting. The findings of this study suggest that galvanized steel and clay tiles are appropriate for rainwater harvesting applications. The concentrations of fecal indicators (total coliforms, *E. coli* and enterococci) in the water samples taken from the pilot roofs made of these two types of materials were measured at lower levels in the first flush tank relative to those from the other two. Additionally, no bacterial pathogens (*Pseudomonas* spp., *Salmonella* spp. and *Cryptosporidium* spp.) were detected in the water samples taken from the first flush tank of the galvanized steel roof, possibly because ultraviolet light and the high temperatures acted as disinfection agents.

Correlation analysis showed the TOC concentration in the harvested rainwater samples to have a strong, positive relationship with total coliforms, *E. coli* and enterococci. In conclusion, roofing material selection requires careful analysis to determine the effects of these materials on the quality of harvested rainwater. Although the results of this study clearly demonstrate that quality to improve after the first flush, the water quality in the first flush tank could also be improved through better tank design (i.e., a calm inlet) and effective maintenance.

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