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Recirculating Water through Concrete Aggregates Rapidly Produced Ecologically Hazardous Water Quality

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Abstract: The use of recycled concrete aggregates as a construction material is growing and this study was conducted to investigate the potential water quality issues that arise when concrete aggregates are exposed to water. The water used in the study was from a high-conservation-value wetland. It was dilute ($17.8 \mu\text{S cm}^{-1}$), acidic (pH 5.97) and poorly buffered. The ionic composition comprised sodium, bicarbonate and chloride ions. Water was recirculated for 60 min through a control treatment and three treatments containing recycled concrete aggregates (RCAs) of different fragment sizes (10 mm, 20 mm and 60 mm). The fragment size influenced the final water quality, but the response patterns varied between the attributes tested. Post-recirculation, the RCA treatments increased the electrical conductivity by 6 to 12 times; pH by 2.3 to 3.8 pH units; and concentrations of calcium, potassium, bicarbonate and sulphate. The water exposed to RCA materials also increased the concentrations of several metals (aluminium, arsenic, copper, lead and zinc), resulting in hazardous concentrations for aquatic species according to ecological water quality guidelines. Strontium concentrations in water exposed to RCAs increased by 30 to 120 times background levels. The results from this study added further support to a growing body of evidence that the exposure of concrete materials to water can produce environmentally hazardous water quality.

Keywords: concrete contamination; water chemistry; urban water salinisation; metals



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1. Introduction

Urbanisation is increasing across the globe and causing complex physical, chemical and biological modifications to the waterways draining urban areas [1]. Urban lands typically have greater coverage of artificial impervious surfaces, including roads, roofs, paving and carparks [2,3]. Urban areas also contain engineered stormwater drainage systems, which alter the natural hydrology of landscapes with increased volume and velocity of surface runoff that transports nutrients, sediment and a wide array of pollutants from urban areas into waterways. Such modification of waterways by urban development is associated with a complex series of symptoms involving physical, chemical and biological factors, which is collectively termed the urban stream syndrome [1]. The association between catchment urbanisation and increased coverage by impervious surfaces causes the degradation of both urban water quality and urban stream ecosystems in a complex web of environmental impairment symptoms [4–6]. The ecological modifications typically reported from urban streams include the loss of sensitive aquatic species and the proliferation of tolerant species, which often include invasive species [1,3,4].

The Blue Mountains Temperate Highland Peat Swamps on Sandstone (THPSS) are unique freshwater wetlands that occupy a total area of less than 2000 ha within and surrounding the high-conservation value Greater Blue Mountains World Heritage Area [7,8]. They contain endemic and endangered species of flora and fauna and provide essential ecosystem services, including water filtration, element cycling and carbon sequestration [9–11]. The THPSS are listed as an endangered ecological community under the Environment Protection and Biodiversity Conservation Act 1999 (Commonwealth), as they

have a restricted geographic distribution and are vulnerable to ongoing threats [12,13]. Despite this legislative protection, the ecological condition of many THPSS wetlands was substantially degraded in catchments containing urban development [9,10,13,14].

The conservation outlook for the THPSS wetlands and the surrounding Greater Blue Mountains World Heritage Area was downgraded due to concerns about the impacts of climate change, invasive pests, urban stormwater pollution, and frequent and intense wildfires [8]. The physical, chemical and biological degradation of the THPSS due to the influence of urban development was termed the ‘urban swamp syndrome’ [13,15]. The ecological degradation of the urban THPSS has included the spread of invasive weeds [9] and a reduction in the abundance and taxonomic richness of aquatic invertebrates [13]. Previous water quality studies established that the unmodified Blue Mountains THPSS have naturally acidic (<5.8 pH) water with dilute electrical conductivity (EC; <40 $\mu\text{S cm}^{-1}$) and major ions dominated by sodium and chloride [10,15,16]. In contrast, the THPSS wetlands that drain urban areas have a characteristic signature of contamination that includes higher pH; increased EC; increased concentration of major ions (particularly calcium and bicarbonate); and highly elevated concentrations of several metals, notably barium and strontium [10,13,16]. Previous studies suggested that this chemical signature in the urban THPSS wetland sediment and water is indicative of concrete contamination [10,16]. It was estimated that almost one-third of the THPSS are adversely impacted by urbanisation [9].

The weathering of urban concrete materials is one of several anthropogenic processes that contribute to the geochemical modification of urban waterways [6–19], urban wetlands [10,15,16] and urban riparian soils [20,21]. Concrete materials are one of the most widely used structural materials used in urban areas and modern urban drainage systems. Urban runoff drains through concrete drainage structures that include pipes, culverts, gutters and canals [22–24]. The widespread use of calcareous concrete urban materials has been termed ‘urban karst’, which can rapidly weather when exposed to low-pH precipitation [6,19]. Experimental recirculation of water through concrete pipes has revealed that urban concrete materials can rapidly (in less than 60 min) cause substantial modification of water quality [22]. Chemical changes include increased pH and EC, in addition to increased concentrations of major ions, including calcium, potassium and bicarbonate [21,22]. Studies over broad geographical scales reported that weathering of concrete urban materials has a widespread impact on the geochemistry of urban stream water quality [25]. Research reported an association between higher pH, EC and concentrations of major ions (including calcium, potassium and bicarbonate) in urban streams linked to increased coverage of catchments by impervious surfaces [2,6,23,25]. A similar association with coverage of catchments by artificial impervious surfaces was also reported in the urban Blue Mountains THPSS water quality [10,13].

RCA materials are rapidly growing in popularity across the world as construction materials. As the world faces growing shortages of non-renewable natural resources, RCA materials that were once disposed of as concrete wastes in landfills can be reused, helping the construction industry to become more sustainable [26–29]. They are widely used as a substitute for crushed rock aggregates and virgin concrete materials [26]. There is an increasing trend towards using RCAs as coarse aggregates within different formulations of concrete [27]. The beneficial use of RCAs is broadly regarded as a more sustainable option than crushed rock aggregates and virgin concrete materials [28]. RCA materials are popular and versatile products that are used in a variety of settings, such as landscape fill material, road bases, bedding for paving and structuring embankments [29]. RCAs are also widely used for backfilling trenches for laying utilities, including water, stormwater and sewer pipes. In Australia, the NSW Environment Protection Authority (EPA) recommends that RCAs are suitable as road and paving bases, as landscaping fill and as a ‘drainage medium for backfilling drainage structures’ [30]. There has also been a growing usage of RCAs for road bases and backfilling service trenches in urban development in the Blue Mountains area (personal observation).

Whilst the influence of urban concrete drainage materials, such as pipes and gutters, on water quality in urban streams and rivers is becoming well known [19,22,31], there is growing concern about water quality impacts from the growing use of RCA materials. This was demonstrated in a field study where leachate draining from RCA stockpiles was found to be highly alkaline and enriched with metals, namely, arsenic, chromium, lead and selenium [32]. There were several laboratory leaching studies that examined the leaching behaviour of metals from RCA under different settings [33–35]. Based on the potential for short-term exposure (one hour) of runoff to RCA materials in field settings, a short-term recirculation laboratory study evaluated water quality changes [36]. This demonstrated rapid changes in EC and pH and reported increased major ion concentration and increased metal concentrations after 60 min of recirculation. A longer laboratory study that involved a 96 h immersion of RCA materials of different fragment sizes in water revealed that particle size influenced the concentration of cations, with the smaller RCA particle size generating the largest cation concentrations [37].

In this current study, we aimed to investigate the water chemistry changes that resulted from exposing water sourced from a natural and unmodified THPSS to a range of RCA materials. We chose to use a wider range of commercial RCA materials of different aggregate sizes to investigate whether the aggregate size influences the nature and scale of water quality modification. In particular, we wanted to test whether RCA materials exposed to water for a realistic amount of time (60 min) resulted in the production of leachate containing environmentally hazardous concentrations of metals and other water quality attributes [26,36].

2. Materials and Methods

2.1. Study Area

Water for this experiment was sourced from a Blue Mountains THPSS near Mt Hay located in a protected conservation reserve (Blue Mountains National Park), Leura, NSW (−33.669806, 150.347170). The swamp and its immediate catchment are naturally vegetated and free of urban development [15]. Water from this swamp has been monitored for several years in multiple investigations and is regarded as being indicative of a near-pristine swamp with sound ecological health [13,15,16,36]. Water emerging from this swamp has the characteristic low pH (<5.8), low EC (<35 $\mu\text{S cm}^{-1}$) and ionic composition domination by sodium and chloride ions that are typical of a pristine THPSS [10,15].

2.2. Collecting Water from the THPSS

Surface water was collected from the exit stream of the Mt Hay swamp on 22 September 2022 early on the same day that all the laboratory recirculation experiments were conducted. Water physiochemical properties were recorded in the field, including pH (pH units), electrical conductivity (EC; $\mu\text{S cm}^{-1}$) and temperature ($^{\circ}\text{C}$), using a calibrated TPS Aqua-CP/A waterproof conductivity–TDS–temperature meter (supplied by TPS PTY LTD, Brendale, Queensland, Australia). Water for the experiments was collected (unfiltered) in several cleaned, acid-washed and rinsed 5 L plastic containers. The containers were filled, with care taken to ensure minimal air headspace in the container to limit gas exchange between the water and air.

2.3. Preparation of Aggregates

All RCA materials were commercially sourced from the Western Sydney region and their fragment size and location of purchase were as follows:

- RCA 10 mm (Turtle Nursery and Landscape Supplies, Rouse Hill, NSW);
- RCA 20 mm (Turtle Nursery and Landscape Supplies, Rouse Hill, NSW);
- RCA 60 mm (Raygal, Castlereagh, NSW).

All aggregates were soaked for several minutes before being rinsed several times in deionised water to remove any fine contaminants. The washing revealed non-concrete contaminants, such as timber, stones, wire, plastics, ceramic tiles and terracotta pipe

fragments. Non-concrete aggregates were manually removed. The aggregates were then dried in the laboratory in an incubator (Labec incubator S4218) for 48 h at 38.5 °C prior to the water recirculation experiments. The aggregates were carefully weighed into batches for testing (1000 ± 5 g) using a laboratory electronic balance (AND FX-3200).

2.4. Recirculation Experiment

All treatments used clean, previously unused PVC pipes that were 100 mm in diameter and 500 mm long (supplied by Bunnings, Pymble, NSW, Australia). The lower end had a PVC cap fitted, with a small 4 mm hole drilled, and was supported in the laboratory at an angle of approximately 15° to allow approximately 200 mL of water to drain per minute (Figure 1). All PVC pipes, beakers and measuring cylinders were acid-washed and then rinsed in deionised water prior to use.

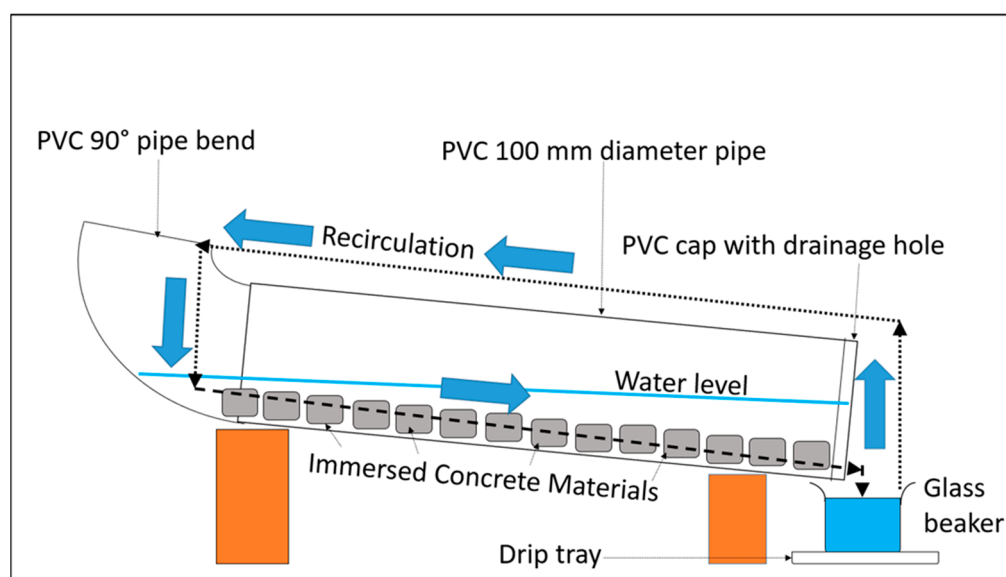


Figure 1. Testing apparatus used to recirculate water through a clean PVC pipe containing concrete and other aggregate materials.

All four treatments used an identical experimental setup, but the control treatment did not contain any aggregates and the other three treatments contained different sizes of RCA aggregate materials (Figure 1). The three treatments containing the RCA all had 1 kg of cleaned and dried aggregates that were evenly spread to cover the bottom of the PVC pipe to an average depth of 40 mm. For each of the four treatments, 1000 mL of source water was continuously recirculated for 60 min. The 1000 mL of water was poured from a measuring cylinder into the higher end of the PVC pipe through an upturned 90° PVC collar (Figure 1). This allowed the water to flow through the length of the pipe through the submerged aggregate material. The flow was restricted by the small exit hole drilled in the PVC end cap before draining into one of the two 300 mL beakers, which were used to return the water to the higher end of the PVC pipe. The rate of flow was maintained at about 200 mL per minute over the 60 min recirculation period. The experiment was repeated a second time for each treatment using identical methods.

The water quality was monitored throughout the experiment with a TPS AQUA-Cond-pH meter, with probes immersed in one of the beakers used to collect flow emerging from the lower end of the PVC pipe. The pH (pH units), salinity as electrical conductivity 'EC' ($\mu\text{S cm}^{-1}$) and water temperature (°C) were measured. The probes were rinsed, tested and calibrated before each experiment. The EC and pH measurements were taken at 1 min intervals for the first 5 min of the recirculation period and every 5 min thereafter. These intervals were chosen based on previous studies [21,22,36]. The intervals were also confirmed through an earlier pilot study conducted by the authors that demonstrated that

the pH and EC changes for the RCA treatments were very rapid in the first five minutes of the recirculation experiment. Prior to the recirculation experiments and in between replicates, the hand-held meter and the PVC pipes and beakers were all thoroughly rinsed using deionised water to ensure that the probes were not contaminated by previous trials.

Replicate 200 mL anion, cation and metal samples of the Mt Hay swamp THPSS water were collected before the experiment commenced. These were classified as ‘before’ samples. Similarly, replicate water samples for testing (anion, cation and metals) were collected after each 60 min recirculation experiment. These were considered the ‘after-recirculation’ samples. These ‘before’ and ‘after-recirculation’ samples were sent to Envirolab, Chatswood NSW, which is a commercial, NATA-accredited laboratory, for the determination of major ion and metal concentrations. Concentrations of a suite of metals were measured using inductively coupled plasma–mass spectrometry, and ion concentrations were evaluated using inductively coupled plasma–atomic emission spectroscopy [38].

2.5. Water Quality Guidelines for Ecosystem Protection

The mean water quality results obtained at the completion of the 60 min recirculation experiments were compared with the ANZECC [39] Australian water quality guidelines (Table 1). This was done to provide guidance on the potential adverse impacts to aquatic species of water exposed to harmful contaminants, assuming the experimental treatments flowed into a THPPS. The guidelines included EC and metals/metalloids comprising aluminium, arsenic, lead, nickel and zinc. The THPSS wetlands are threatened communities with very high conservation significance, and thus, the highest level of ecological protection (protection of 99% of species) was selected from the guidelines as a basis for comparison with our results. The guidelines used a salinity (as EC) target of $55 \mu\text{S cm}^{-1}$, which was based on south-eastern upland streams in the eastern highlands (Table 3.3.3 in Chapter 3 Aquatic Ecosystems) [39]. There are no water quality guideline specifications recommended for the THPSS wetlands, and pH is an important attribute [16,40]. Given the importance of pH and the acidic nature of the undisturbed THPSS wetlands, an interim recommendation of <5.8 pH was recommended based on previous studies that reported the pH of the undisturbed THPSS [13,15,16].

Table 1. Guideline values for protecting 99% of aquatic species in the THPSS based on the ANZECC [39] Australian water quality guideline trigger values and previous THPSS investigations [15,16]. EC—electrical conductivity.

Indicator	THPSS Guidelines
EC	$<55 \mu\text{S cm}^{-1}$
pH	<5.8 pH units [15,16]
Aluminium	$<27 \mu\text{g L}^{-1}$ (if pH > 6.5)
Arsenic	$<1.0 \mu\text{g L}^{-1}$
Lead	$<1.0 \mu\text{g L}^{-1}$
Nickel	$<8.0 \mu\text{g L}^{-1}$
Zinc	$<2.4 \mu\text{g L}^{-1}$

2.6. Data Analysis

Statistical Analysis

All statistical analyses were performed using Microsoft Excel. Changes in pH and EC, as well as metal, metalloid and major ion concentrations, were compared ‘before’ and ‘after’ 60 min of recirculation through the different treatments. Changes in water chemical attributes ‘before’ versus ‘after’ recirculation were evaluated using Student’s *t*-test assuming unequal variance and using log-transformed data. Some water chemical attributes were not detected because they were lower than the laboratory detection limits.

In these cases, for data analysis purposes, the result was assumed to be half of the detection limit [41]. Any p -values less than 0.05 were considered significant. The ionic composition of the THPSS source water was compared with the ionic composition of water after 60 min of recirculation through the three RCA treatments using a Piper diagram.

3. Results

3.1. Water Chemistry

3.1.1. Water pH and EC

The THPSS water was acidic and within the expected ranges before the commencement of the experiment (mean 5.76 pH units). Following the 60 min recirculation period, the pH of the water in the RCA treatments increased significantly ($p < 0.001$; Table 2, Figure 2). This contrasted with the control treatment, whose pH increased by 0.86 pH units to a mean of 6.62. All RCA treatments had an increased mean pH, with increases of 2.87 pH units to 8.63 (20 mm), 3.07 pH units to 8.83 (60 mm) and the largest was 4.2 pH units to 9.96 (10 mm). The steepest pH increases occurred within the first 30 min of water recirculation with the RCA treatments (Figure 2). The EC also increased significantly ($p < 0.001$) after recirculation through each RCA treatment (Table 2, Figure 3). The mean EC of the reference water before recirculation was $17.64 \mu\text{S cm}^{-1}$, increasing to $20.65 \mu\text{S cm}^{-1}$ after the control treatment. The mean RCA post-recirculation EC values increased by 9.5 times ($184.1 \mu\text{S cm}^{-1}$; 60 mm), 14 times ($263.6 \mu\text{S cm}^{-1}$; 10 mm) and 16.5 times ($308 \mu\text{S cm}^{-1}$; 20 mm RCA).

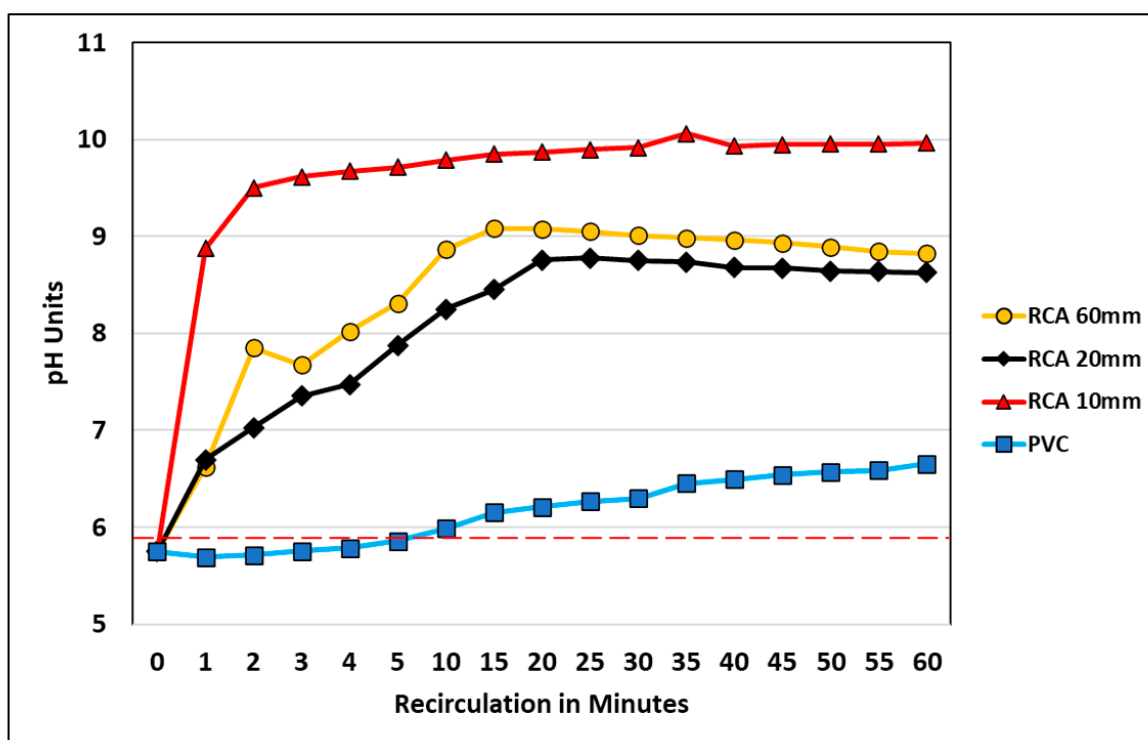


Figure 2. Mean pH of water recirculated for 60 min through control (PVC) and treatments containing concrete aggregate materials. pH values are provided for before the exposure (time = 0), then at 1 min intervals for the first 5 min, and then at five-minute intervals up to 60 min. A dotted red line indicates a guideline of pH of 5.8 (see Table 1).

Table 2. Summary statistics (minimum–maximum (mean)) of the pH, EC, temperature, major ion, hardness, metalloid, metal and major cation results collected from the ‘before’ (reference water from the THPSS) compared with the water ‘after’ 60 min recirculation through one of four treatments (PVC control, 10 mm recycled concrete aggregate (RCA) material, 20 mm RCA material, 40 mm RCA material). The results are in bold if they were statistically different according to Student’s *t*-test of ‘before’ versus ‘after’. * $p < 0.05$, ** $p < 0.001$, *** $p < 0.0001$, NS—not significant. ND—not detected. Alk.—alkalinity. EC—electrical conductivity. SI—saturation index.

Variable (Unit)	THPSS (Before)	After 60 min Recirculation			
		PVC (Control)	10 mm RCA	20 mm RCA	60 mm RCA
pH (pH units)	5.71–5.78 (5.76)	6.5–6.69 (6.62) **	9.82–10.1 (9.96) ***	8.58–8.68 (8.63) ***	8.82–8.85 (8.83) ***
EC ($\mu\text{S cm}^{-1}$)	17.52–17.77 (17.64)	20.31–20.92 (20.65) **	250.5–274 (263.6) ***	302–316 (308) ***	186–191.6 (184.1) ***
Temperature ($^{\circ}\text{C}$)	17.1–17.5 (17.3)	19–19.1 (19.08) ***	19.8–20.0 (19.9) ***	19.5–19.8 (19.7)	19.2–19.6 (19.4) ***
Calcium (mg L^{-1})	ND	0.7–0.8 (0.77) ***	16–21 (19) ***	35–40 (37.8) ***	24–25 (24.4) ***
Potassium (mg L^{-1})	ND	ND	12–12 (12) ***	4–4 (4) ***	5.1–5.4 (5.2) ***
Sodium (mg L^{-1})	3	3 NS	18–21 (20) ***	12–14 (13) ***	8–8.6 (8.2) ***
Magnesium (mg L^{-1})	ND	ND	ND	1–1 (1) ***	0.8–0.9 (0.84) ***
Hardness (mg L^{-1})	ND	ND	41–53 (47.9) ***	93–110 (99.6) ***	62–66 (64.3) ***
Bicarbonate (mg L^{-1})	ND–6 (4.3)	5–7 (6.16) *	26–49 (31) ***	26–30 (28) ***	37–39 (38.2) ***
Carbonate (mg L^{-1})	ND	ND	13–21 (13.0) **	ND	ND
Total Alk. (mg L^{-1})	ND–6 (4.3)	5–7 (6.16) *	39–68 (47) ***	26–30 (28) ***	37–39 (38.2) ***
Sulphate (mg L^{-1})	ND	ND	44–58 (51.7) ***	77–81 (79) ***	36–38 (37.1) ***
Chloride (mg L^{-1})	3	3 NS	8–10 (9.1) ***	13–14 (13.4) ***	5–7 (5.9) ***
Arsenic ($\mu\text{g L}^{-1}$)	ND	ND	2–3 (2.2) ***	ND–1 (0.8) **	ND–1 (0.64) NS
Aluminium ($\mu\text{g L}^{-1}$)	100–130 (115)	90–110 (100) *	1500–3700 (2367) ***	290–450 (380) ***	970–2200 (1404) ***
Barium ($\mu\text{g L}^{-1}$)	2–3 (2.5)	2–3 (2.5) NS	18–49 (31.9) **	9–12 (10.7) **	12–17 (13.9) ***
Chromium ($\mu\text{g L}^{-1}$)	ND	ND	23–30 (27) ***	5–6 (5.8) ***	6–16 (11.4) ***
Lead ($\mu\text{g L}^{-1}$)	ND	ND–1.0 (0.75) **	6–18 (11.1) ***	3–4 (3.8) ***	3–4 (3.3) ***
Lithium ($\mu\text{g L}^{-1}$)	ND	ND	24–27 (25.6) ***	3–4 (3.3) ***	11–13 (12) ***
Manganese ($\mu\text{g L}^{-1}$)	ND	ND–6.0 (3.5) *	30–85 (55.8) ***	10–14 (11.9) ***	28–44 (34.8) ***
Molybdenum ($\mu\text{g L}^{-1}$)	ND	ND	6–9 (7.1) ***	3–4 (3.6) ***	3–9 (6.1) ***
Selenium ($\mu\text{g L}^{-1}$)	ND	ND	ND	ND	ND
Strontium ($\mu\text{g L}^{-1}$)	ND–1.9 (1.5)	1.1–2.0 (0.95) NS	78–100 (89.9) ***	150–170 (160) ***	40–48 (44.7) ***
Titanium ($\mu\text{g L}^{-1}$)	ND–1.5 (0.9)	ND–1.0 (0.95) NS	43–230 (106.7) ***	7.7–11 (9.4) ***	25–76 (47.9) ***
Vanadium ($\mu\text{g L}^{-1}$)	ND	ND	20–32 (24.6) ***	3–4 (3.6) ***	4–7 (5.3) ***
Zinc ($\mu\text{g L}^{-1}$)	1–11 (4.25)	4–10 (7.33) *	13–29 (20.8) **	10–17 (13.1) **	10–16 (12.9) **
Uranium ($\mu\text{g L}^{-1}$)	ND	ND	ND	ND	ND
Ionic Strength ($\mu\text{mol L}^{-1}$)	116	266	3211	4024	2788
Ca(OH) ₂ SI	–17.9	–14.1	–6.2	–8.5	–8.3
Calcite SI	–7.7	–4.0	1.3	0.3	0.7

3.1.2. Ionic Composition

The concentration of major ions was measured after the completion of the 60 min recirculation through the four treatments (Table 2, Figures 4 and 5). The initial THPSS water had an overall ionic strength of $116 \mu\text{mol L}^{-1}$ and this increased to between 2788 and $4024 \mu\text{mol L}^{-1}$ with the addition of the RCAs. After 60 min, there were substantial changes in the major ion composition and concentration (Table 2, Figure 4). Significant ($p < 0.0001$) increases in bicarbonate were observed, rising from 4.3 mg L^{-1} (before) to 28 mg L^{-1} (20 mm), 31 mg L^{-1} (10 mm) and 38.2 mg L^{-1} (60 mm). A smaller increase in mean bicarbonate was measured after the control treatment. Chloride concentrations also increased (at the $p < 0.0001$ level) for all RCA treatments, with mean levels rising from 3 mg L^{-1} (before) to 5.9 mg/L (60 mm), 9.1 mg L^{-1} (10 mm) and 13.4 mg L^{-1} (20 mm). Sulphate was not detected in the ‘before’ samples or after the control treatment, but the

mean concentrations after recirculation through the RCA treatments were 37.1 mg L^{-1} (60 mm), 51.7 mg L^{-1} (10 mm) and 79 mg L^{-1} (20 mm).

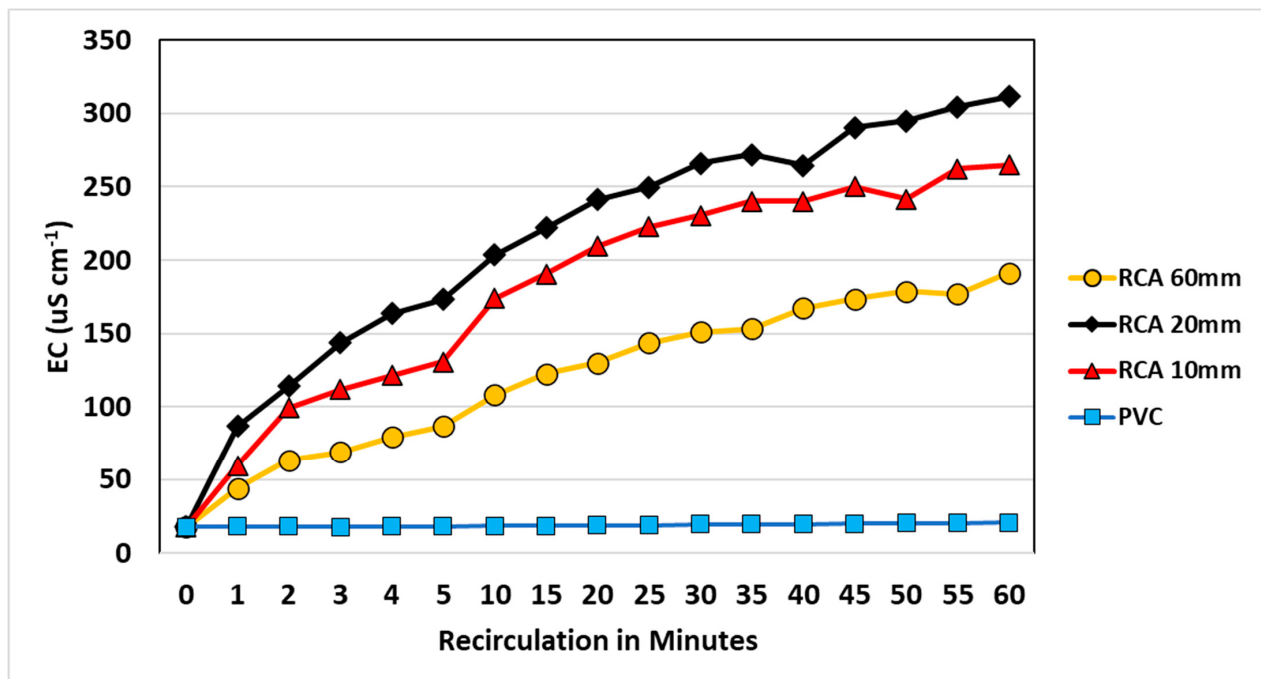


Figure 3. Mean EC of water recirculated for 60 min through control (PVC) and treatments containing concrete aggregate materials. EC values are provided for before exposure (time = 0), then at 1 min intervals for the first 5 min, and then at five-minute intervals up to 60 min. A dotted red line indicates a guideline for EC of $55 \text{ } \mu\text{S cm}^{-1}$ (see Table 1).

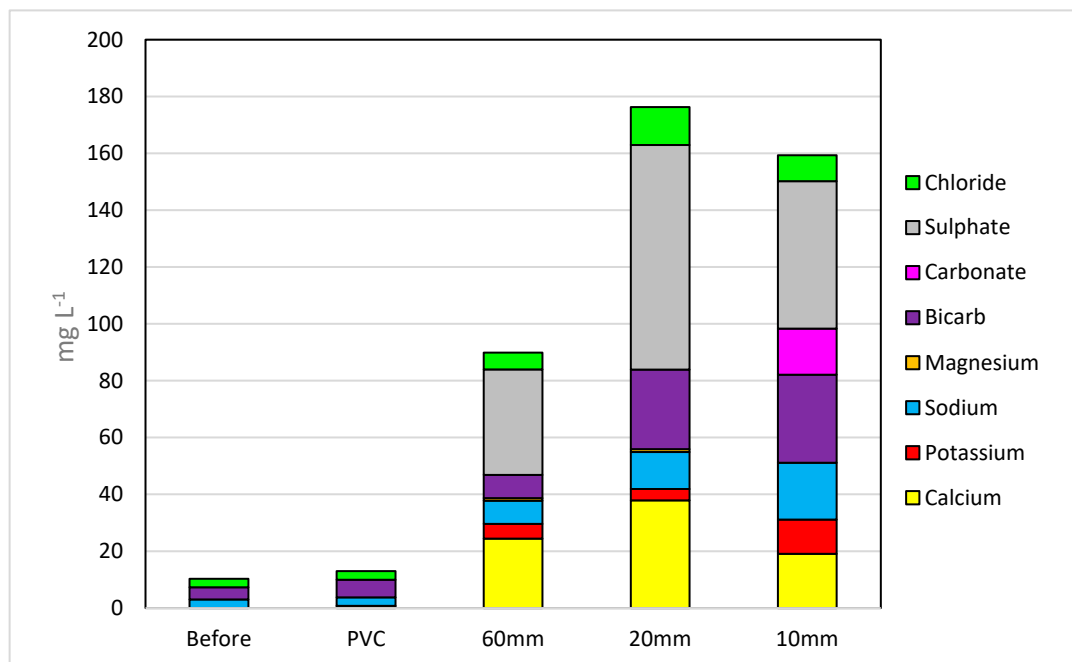


Figure 4. Mean major ion composition (stacked) of water before and after it was recirculated for 60 min through the four treatments. They include a control (PVC) and three RCA treatments, each containing different sizes of concrete aggregate materials (60 mm, 20 mm, 10 mm).

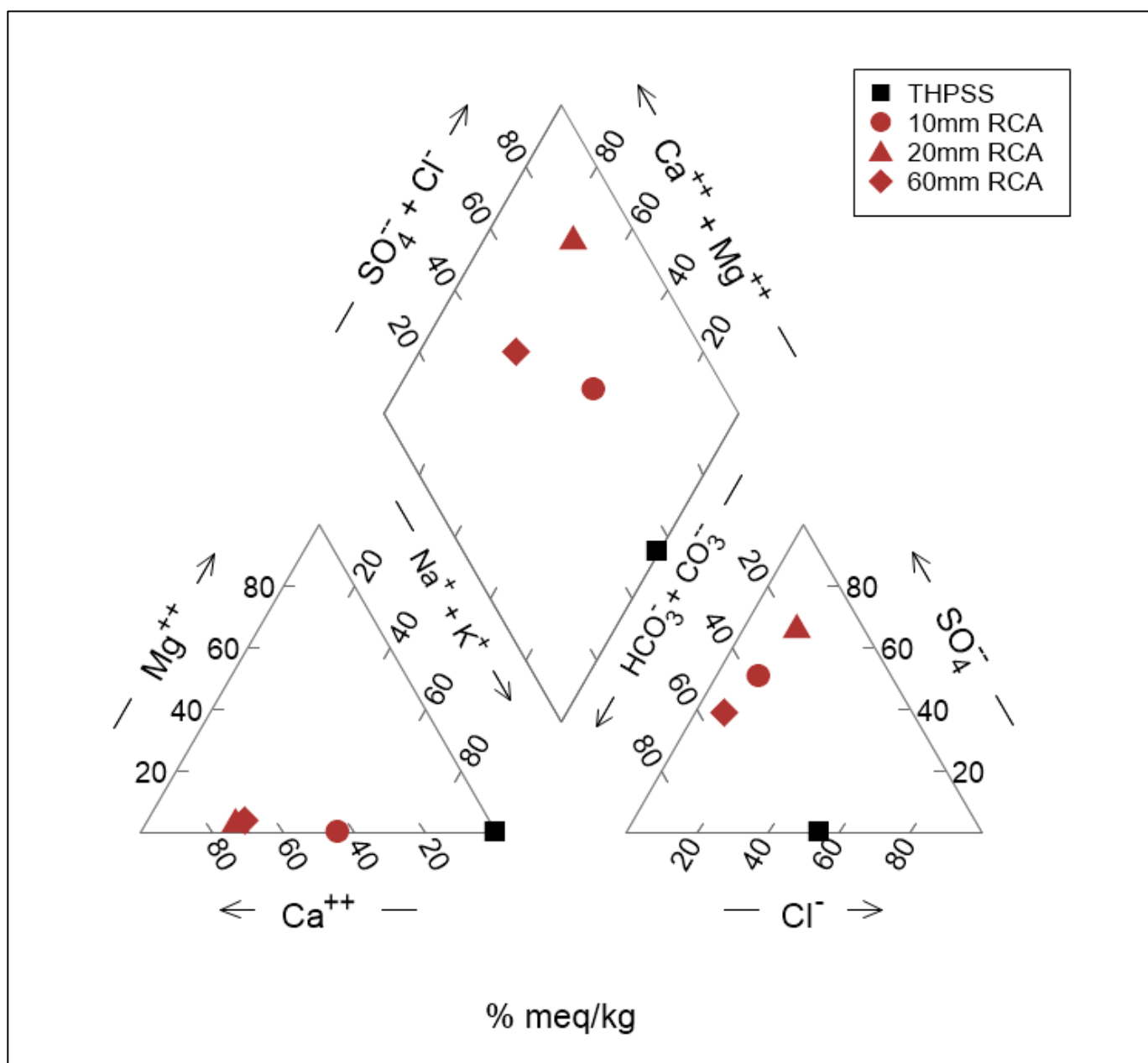


Figure 5. Piper diagram of water before (THPSS) and after the water was continuously recirculated for 60 min through three RCA treatments, each of which contained different sizes of concrete aggregate materials (60 mm, 20 mm, 10 mm).

Calcium was not detected in the initial THPSS water but was detected (mean 0.77 mg L^{-1}) after the PVC control treatments (Table 2, Figures 4 and 5). After recirculation through the RCA treatments for 60 min, significant ($p < 0.0001$) increases in calcium concentration were measured, increasing from $<0.5 \text{ mg L}^{-1}$ (before) to 19 mg L^{-1} (10 mm), 24.4 mg L^{-1} (60 mm) and 37.8 mg L^{-1} (20 mm). The hardness of the water increased after recirculation through the three RCA treatments, where the mean hardness was 47.9 mg L^{-1} (10 mm), which is classified as ‘soft’ (Table 2). The water hardness was classified as moderately hard after 60 min of recirculation through the other two RCA treatments (mean 64.3 mg L^{-1} for 60 mm and mean 99.6 mg L^{-1} for 20 mm).

The saturation indices (Table 2) indicate that the initial THPSS water was undersaturated with respect to both calcium hydroxide and calcite. Following the mixing of the THPSS water with RCAs, the saturation index for the calcite surpassed the equilibrium

and approached saturation with the PVC control remaining undersaturated. The calcium hydroxide index remained undersaturated with decreasing levels.

3.1.3. Metals

With only one exception (arsenic, 10 mm), there were statistically significant increases (mostly $p < 0.0001$) in the mean concentrations of metals measured after versus before the water recirculation through the three RCA treatments (Table 2, Figures 6 and 7). Barium increased from $2.5 \mu\text{g L}^{-1}$ (before) to means of $10.7 \mu\text{g L}^{-1}$ (20 mm), $13.9 \mu\text{g L}^{-1}$ (60 mm) and $31.9 \mu\text{g L}^{-1}$ (10 mm) after the recirculation (Figure 6). Large increases in mean strontium concentrations were recorded, rising from $1.3 \mu\text{g L}^{-1}$ before to $44.7 \mu\text{g L}^{-1}$ (60 mm), $89.9 \mu\text{g L}^{-1}$ (10 mm) and $160 \mu\text{g L}^{-1}$ (20 mm) afterwards. Similar to barium and strontium, large increases in concentrations of lithium, titanium and vanadium were measured in the water after the recirculation through the three RCA treatments (Figure 6).

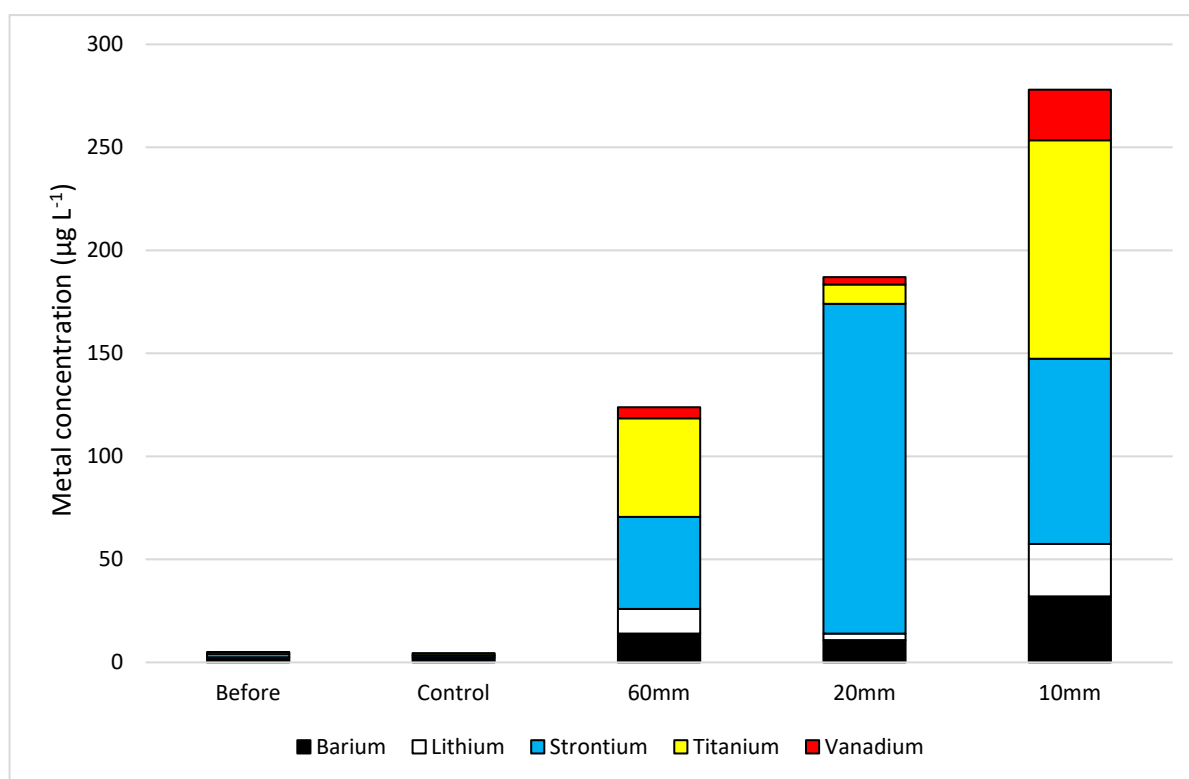


Figure 6. Mean stacked concentration for five metals (barium, lithium, strontium, titanium, vanadium) in the water before and after it was recirculated for 60 min through the four treatments. These treatments included a control (PVC) and three RCA treatments, each of which contained different sizes of concrete aggregate materials (60 mm, 20 mm, 10 mm).

The largest mean increases for any metal (in the RCA treatments) were detected for aluminium, increasing from $115 \mu\text{g L}^{-1}$ (before) to $380 \mu\text{g L}^{-1}$ (20 mm), $1404 \mu\text{g L}^{-1}$ (60 mm) and $2367 \mu\text{g L}^{-1}$ (10 mm; Table 2, Figure 7) after 60 min of recirculation.

Seven metals had undetectable concentrations in the before samples: $<5 \mu\text{g L}^{-1}$ for manganese and $<1 \mu\text{g L}^{-1}$ for arsenic, chromium, lead, lithium, molybdenum and vanadium (from the THPSS; Table 2). The largest increase in the mean concentrations for this group of metals was manganese, which was undetected ($<5 \mu\text{g L}^{-1}$) before but increased to $11.9 \mu\text{g L}^{-1}$ (20 mm), $55.8 \mu\text{g L}^{-1}$ (10 mm) and $34.8 \mu\text{g L}^{-1}$ (60 mm) after 60 min of recirculation.

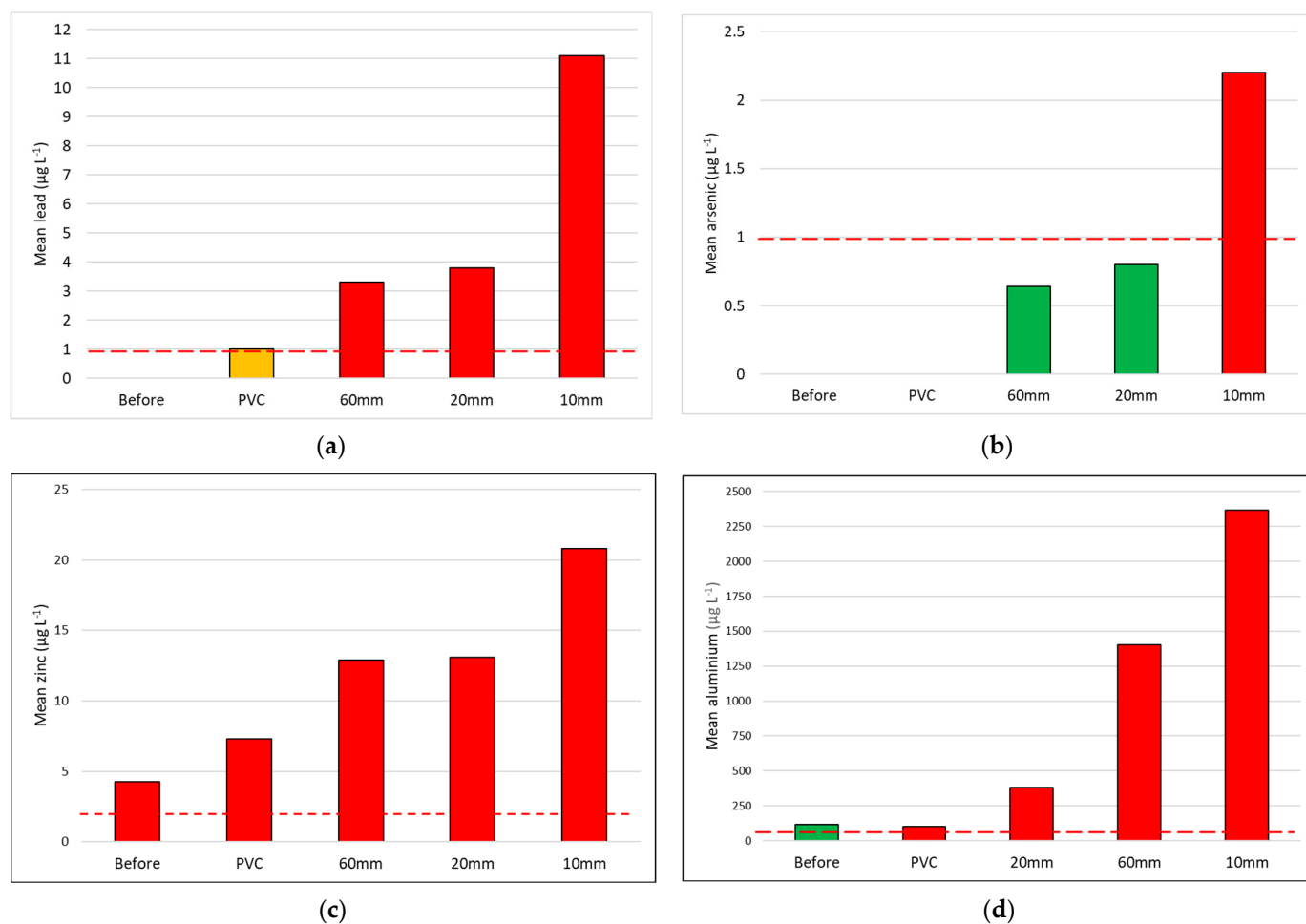


Figure 7. Mean metal concentration for lead (a), arsenic (b), zinc (c) and aluminium (d) in the THPSS water before and after the recirculation for 60 min through the control (PVC) and three RCA treatments (60 mm, 20 mm, 10 mm). The bar is green if the mean concentration complied with the ANZECC (2000) trigger value for 99% species protection. The bar is orange if it was equal to the trigger value. It is red if the mean concentration exceeded the trigger value. The dotted line indicates the trigger value.

4. Discussion

The results from this research add to the growing body of knowledge that weathering of concrete materials can cause substantial modification of urban water chemistry [6,17–19,22,23,25]. This investigation showed that accelerated weathering of ‘urban karst’ concrete materials [19], in the form of fragmented concrete materials, could trigger rapid and potentially harmful changes in water quality when exposed to water. The dissolution of RCA materials immersed in water could rapidly create a cocktail of elevated pH and EC and a modified ionic composition enriched with calcium, bicarbonate, sulphate and several metals [18,19]. The current study adds to our understanding of the geochemical signature that can be triggered by the accelerated weathering of urban karst materials. Urban streams and wetlands often have higher salinity [6] and alkalinity and a modified ionic composition [2,3,5]. The current study also demonstrated that the dissolution of RCA materials in water can release a broad suite of metals, including ecologically hazardous concentrations of arsenic, lead, aluminium and zinc.

Our research highlighted the potential for the production of impaired water quality when concrete materials are in a fragmented form (such as RCAs) and are exposed to water. The changes in water chemistry occur rapidly and are primarily driven by the dissolution of alkaline materials within the RCA. The results of this study added further support to the

hypothesis that weathering of urban concrete materials can make substantial contributions to geochemical differences between urban and non-urban waterways [23,25,42]. The higher pH in an urban THPSS was identified as one of the signatures of urbanisation [16]. This research showed that RCA materials can rapidly impart higher pH to water after a relatively short period of contact. All RCA treatments in the current study increased the pH by more than 2 pH units for water recirculated through RCA materials. The most alkaline pH (mean 9.96) occurred after recirculation through the smallest (10 mm) RCA treatment, compared with the coarser fragment sizes that reached 8.83 (60 mm) and 8.63 (20 mm) after recirculation. Increases in the pH of this magnitude could contribute to the contamination and ecological degradation of the Blue Mountains THPSS wetlands. The pH of pristine THPSS wetlands is typically less than 5.75, with an increased pH of urban wetlands reported to be approximately 1.2–1.5 pH units higher than natural non-urban wetlands [10,15].

An increased pH of water exposed to concrete materials can potentially have far-reaching effects on the conservation of the critically endangered Blue Mountains THPSS ecosystems. Research found that the elevated pH of an urban THPSS was associated with an impaired ecological condition of the wetlands through the modification of wetland microbial communities [40]. An increased pH of an urban compared with a non-urban THPSS [10] was similar to the increased pH of urban riparian soils in the Georges River catchment [21]. Both the Blue Mountains and Georges River share similar sandstone geologies and are located about 70 to 80 km apart. Increased urban riparian soil pH was identified as one of the key factors associated with increased growth in invasive riparian weeds across a gradient of urbanisation in the Georges River catchment, south of Sydney [21]. Weed invasion is also a major contributor to the impaired ecological condition of an urban THPSS [7,9,10].

The results from this current research added to the previous research [36,37], which reported that smaller fragment sizes of concrete aggregates that are exposed to water generally increased the nature and magnitude of water quality modification. For example, previous research [37] recorded steeper and greater increases in pH, EC and major ion concentrations when water was exposed to smaller concrete aggregates (less than 30 mm³) compared with larger concrete aggregates (30–80 mm³) in a static 96 h laboratory experiment. However, the current study found that this was not always the case for all water quality attributes. For example, in this study, the mean aluminium (1404 µg L⁻¹) concentration for water recirculated through the largest particle size (60 mm RCAs) treatment was more than three times larger than the mean concentration of aluminium (380 µg L⁻¹) in water recirculated through the smaller 20 mm RCAs. This was likely influenced by the individual differences in the make-up of the raw concrete materials that were crushed to produce RCAs. This followed the observation by [43] that RCA composition differs markedly from batch to batch, with some mixtures containing much higher proportions of ceramic, brick or concrete.

Many of the metals detected in water after recirculation for 60 min through RCA treatments were measured at ecologically dangerous concentrations for the protection of aquatic life. Aluminium was recorded in water samples after recirculation through the RCA treatments at concentrations that were particularly hazardous to aquatic life. The mean aluminium concentrations in water after the three RCA treatments ranged from 380 µg L⁻¹ to 2367 µg L⁻¹. These exceeded the Australian guidelines [39] (<27 µg L⁻¹ if the pH is >6.5) for aluminium for the protection of 99% of species by 13 to 86 times, respectively. The raw THPSS water used in this study from a THPSS had a mean aluminium concentration of 115 µg L⁻¹, but with such an acidic pH (mean 5.76), it was compliant (<6.5 pH) with aluminium guidelines [39]. The aluminium concentrations recorded after recirculation with RCA materials were broadly comparable to the levels reported in Nigerian waterways contaminated by an aluminium smelting operation [44]. Our aluminium results found after the RCA recirculation also exceeded the U.S. Environmental Protection Agency (EPA) chronic criteria of 87 µg L⁻¹ [45] for all treatments. Our research findings showed that concrete materials can be a potent source of aluminium in urban waters. Concrete materials

could be added to the known sources of aluminium introduced to urban waters by human activities. This was reviewed [18], where known major sources of aluminium in urban water, including building materials, cars and drinking water coagulants, were listed.

The study discussed here was one of the first to demonstrate that immersing concrete fragments in water for relatively short periods could mobilise ecologically hazardous concentrations of lead. This supports the findings from an earlier study [36] that reported hazardous concentrations of lead in water after it was exposed to RCA materials. The lead results for the RCA treatments all exceeded the Australian water quality guidelines for protecting aquatic ecosystems [39], which provides a lead guideline of $<1.0 \mu\text{g L}^{-1}$ for the 99% level of protection for aquatic species. The water recirculated through the smallest RCA (10 mm) aggregates had particularly dangerous lead concentrations, ranging from 6 to $18 \mu\text{g L}^{-1}$ (mean $11.1 \mu\text{g L}^{-1}$). These results also (10 mm RCA) exceeded the Australian human health guidelines for lead in drinking water ($<10 \mu\text{g L}^{-1}$) [46]. Lead is commonly reported at elevated concentrations in urban environments, including urban waterways. Lead in urban water was associated with paint, fallout from leaded petrol, lead-acid batteries, and lead pipes and solder as major sources of the metal [18,47].

Arsenic and zinc were also recorded at ecologically hazardous concentrations after water was recirculated through RCA materials for 60 min. An earlier study [36] also reported similar hazardous concentrations of arsenic and chromium after water was recirculated through RCA material. However, this research appears to be one of the first to document hazardous concentrations of zinc triggered by RCA materials. There is a rich literature on the metals arsenic, chromium and zinc in urban waterways due to the influence of a wide range of human industries and activities [18,48,49].

The downgrading of the conservation outlook for the natural environmental values of the Greater Blue Mountains World Heritage Area by the IUCN [8] identified several threats to protecting the area's outstanding universal conservation values. The IUCN [8] conservation outlook report highlighted their concerns about the vulnerability of the THPSS. The report included mention of the high-threat impacts from urban land uses that adjoin the protected areas, which are transported as urban runoff and contribute to the impairment of the THPSS. Similar concerns about urban impacts were earlier expressed when the THPSS were listed as vulnerable ecological communities by NSW Scientific Committee [50]. The current study adds further support that runoff from urban concrete materials can pose a potential risk of contamination to the THPSS that receive drainage from urban areas. A suite of the pollutants (pH, salinity, calcium, potassium, bicarbonate, strontium) detected at elevated levels in water that had been recirculated through RCA treatments in the current study matched the suite of pollutants identified as part of the geochemical signature of urbanisation reported in the urban THPSS [10].

Five metals (barium, lithium, strontium, titanium and vanadium) in the water exposed to RCA materials were recorded at concentrations much greater than in the raw THPSS water. In total, the concentration of these five metals in water from the pristine THPSS wetland was $5 \mu\text{g L}^{-1}$, increasing after the recirculation experiment by 23 times in the 60 mm RCAs ($118.8 \mu\text{g L}^{-1}$), 36 times in the 20 mm RCAs ($182 \mu\text{g L}^{-1}$) and by more than 50 times in the 10 mm RCAs ($273 \mu\text{g L}^{-1}$). Two of these metals, namely, barium and strontium, were previously identified in the urbanisation signature that was recorded at elevated concentrations in urban THPSS wetlands [10,16]. This finding adds support to the hypothesis that contamination by urban concrete materials contributes to the water quality impairment of the endangered THPSS in the Greater Blue Mountains World Heritage Area [10,15,16].

It is likely that the leaching of metals in this investigation was influenced by the make-up and amount of fly ash used in the formulation of concrete materials used in the RCA materials. Portland concrete often contains fly ash, which is a by-product of coal-fired furnaces and can be used as an ingredient with noted benefits such as improved strength and durability of the final hardened concrete product [51–53]. Concrete products in Australia frequently use fly ash as an ingredient in their manufacturing formulations [52].

In Australia, it is common for concrete to contain up to 5% fly ash [52]. It is likely that RCAs with lesser amounts of fly ash in their manufactured concrete formulation would likely result in lower metal concentrations.

Elevated pH and EC and higher concentrations of metals and ions can have a variety of environmental implications and further investigation is needed to evaluate the potential environmental impacts of using RCAs, particularly as backfilling material in trenches and drains. As the RCA composition will vary according to the different formulations of concrete materials used, batches can differ in their composition [43]. Further research should investigate the effects on the water of a variety of types or brands of RCAs from different locations. When compared with virgin aggregate materials, RCAs have significant life cycle benefits, such as reducing greenhouse gas emissions and energy consumption in the production of concrete products [32]. Many governments in Australia, Europe, the USA and Japan offer incentives to encourage the use of RCA materials to reduce the amount of concrete waste going to landfills and the use of virgin aggregate materials [30]. Like many other countries, the Victorian government in Australia has a zero-waste policy, which requires that all potentially recyclable wastes, including concrete, are diverted from landfills into beneficial uses [54]. However, because of the hazardous leachate potential of RCAs, the wise use of RCAs also requires their environmentally safe use [33].

5. Conclusions

Given the potentially unforeseen water contamination risks associated with RCA materials documented in this study, stricter guidelines may need to be developed by planning and regulatory authorities regarding their permitted usage. Of particular concern is its use as a drainage material, road base or fill material in locations that could drain into sensitive ecosystems, such as the Blue Mountains THPSS. The water quality changes (pH, EC, ionic composition and metals) triggered by the combination of water and concrete fragments can cause detrimental impacts on aquatic and semiaquatic communities. Given that the remediation of such impacts would probably be almost impossible, the most effective management strategy would be to avoid using RCA materials where contact with water and drainage to sensitive environmental locations could occur. It is also plausible that RCA materials could potentially contribute to the impairment of water resources used as potable water supplies. We support earlier recommendations [36,43,54] that further testing of multiple RCA products and batches be conducted over a range of periods to investigate the potential environmental contamination risks associated with the exposure of RCAs to water. We recommend that testing includes both field and laboratory studies to better understand the potential and actual water quality impacts of the use of RCA materials.

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