Report for Blue Mountains World Heritage Institute in relation to the contamination of Upland Swamps and Waterways: *Water from concrete leachate*

Wright Ian A[1, 2] and Nettle Holly[2]

1. School of Science. Western Sydney University, Locked Bag 1797, Penrith NSW, 2751.

2. Research Associate, Blue Mountains World Heritage Institute

Abstract

This study investigated water quality changes resulting from recirculating water through portland concrete, polymer concrete and blue metal gravel aggregate materials. Water was sourced from an unmodified high conservation-value wetland (Temperate Highland Peat Swamps on Sandstone (THPSS)) within the Greater Blue Mountains World Heritage Area. The conservation value of many wetlands in the region in catchments containing urban development have impaired water quality. It has been hypothesised that a major contributing source of contamination of urban THPSS are urban concrete materials. Use of recycled concrete materials in local urban areas has recently been growing. This study used water from a pristine wetland unaffected by urban development. The water was dilute (17.8 µS/cm), acidic (pH 5.97), was poorly buffered with only detectable major ions of sodium, bicarbonate and chloride. Water was recirculated in the laboratory for 60 minutes through four treatments of recycled concrete aggregates (RCA) of different fragment sizes (10mm, two sources of 20mm and 60mm). One additional treatment used a traditional blue metal basalt gravel. Another additional treatment used polymer concrete aggregates. Post-recirculation the concrete treatments increased salinity, pH and also increased calcium, potassium, bicarbonate and sulphate concentrations. Water exposed to concrete materials also increased the concentration of several metals. Aluminium, arsenic, copper, lead and zinc were detected at levels hazardous for aquatic species. The basalt (blue metal) and polymer concrete aggregates also modified water quality to a less hazardous extent than did the RCA materials. Results from this study add further support to the hypothesis that concrete materials can be an environmentally hazardous materials when exposed to water. We recommend that RCA materials be used with caution in settings, such as in urban settings, upstream of Blue Mountains THPSS. Extreme caution is required when RCA materials are exposed to water, such as in drainage infrastructure, where runoff flows into Blue Mountains poorly buffered waterways within the Greater Blue Mountains World Heritage Area.

Urban degradation of Blue Mountains Temperate Highland Peat Swamps

Temperate Highland Peat Swamps on Sandstone (THPSS) are high conservation-value freshwater swamps that have developed over thousands of years, within sandstone geology, in the Blue Mountains and Southern Highlands regions of NSW. The swamps are sometimes termed 'Blue Mountains Upland Swamps' contain a distinct vegetation community that is dominated by sedges and shrubs (Mooney & Martin, 2016). They are listed by the Commonwealth Government as an 'endangered ecological community' under the EPBC Act 1999 (BMCC, 2016; Commonwealth of Australia, 2014). They have a limited geographic distribution and are exposed to risks of ongoing threats associated with human activities. They have important biodiversity values within the Greater Blue Mountains World Heritage Area. Many THPSS are found in the Blue Mountains region of NSW, at altitudes from 500m ASL to 1000m ASL. They are scattered in many small swamps with a combined area of less than 2000 ha. Urban land uses cause several different pressures that threaten their health and survival (Christiansen et al. 2019). This includes encroachment within swamps, the generation of urban runoff, water pollution, proliferation of weeds and other invasive species (Hensen & Mahony, 2010). It has been estimated that almost one third of THPSS are adversely impacted by urbanization (Hensen & Mahony, 2010), and many swamp catchments occur along the fringe or fall outside the World Heritage boundary in urban areas across the higher elevation sections of the Blue Mountains, making them vulnerable to disturbance from urbanisation.

Water quality and the ecological condition of Blue Mountains THPSS has been substantially degraded in catchments at least partly occupied by urban land uses (Fryirs et al. 2014; Belmer et al. 2015). The environmental damage inflicted by urban development on THPSS includes erosion and channelisation from modifications to the hydrology of urban catchments (Fryirs et al. 2014; Cowley et al. 2016, 2020). Ecological impairment of urban wetlands includes proliferation of invasive weed species and changes in microbial decomposer communities (Hensen & Mahony, 2010; Christiansen et al. 2019; Carroll et al., 2020). Swamp invertebrate communities have also been disturbed in urban catchments, with loss of abundance of families sensitive to poor water quality (Belmer et al. 2018).

Water quality of undisturbed and urban Blue Mountains Temperate Highland Peat Swamps

Water and sediment quality of Blue Mountains swamps has also been strongly modified (Fryirs et al. 2014; Purdy 2019). THPSS Swamps in catchments at least partially covered by urban development reveals a strongly modified sediment and water quality (Belmer, et al. 2015; Purdy, 2019; Carroll et al., 2020; Carroll et al., 2022). In contrast, water quality of THPSS in naturally vegetated and undisturbed catchments is very different and of a much higher quality with low concentrations of pollutants. The ecological condition and water quality of THPSS is largely protected by management of the Greater Blue Mountains World Heritage Area (Belmer et al. 2015, 2018).

Water quality of undisturbed Blue Mountains peat swamps

Undisturbed THPSS that are unaffected by urban development have water that is dilute with very low levels of salinity (generally <40 µS/cm) (Belmer, et al. 2015; Purdy, 2019; Carroll et al., 2020). They are also acidic, with pH generally <6.5 and some swamps have surface water pH <5.0. The pH of the water in THPSS is important as they have developed a naturally acidic environment by accumulating peat from decomposition of wetland plant material. The pH of the waterlogged swamps is very important for the microbial community that supports their ecological health (Christiansen et al. 2019).

Water quality of undisturbed THPSS also has a distinct ionic composition, with a domination by sodium and chloride ions (Purdy, 2019; Carroll et al. 2020). The metallic elements in undisturbed swamps are dominated by iron, aluminium and manganese. All of these metals are naturally abundant in the Blue Mountains sandstone geology (Carroll et al. 2020).

Water quality of Blue Mountains peat swamps in urban catchments

Blue Mountains THPSS in catchments that include urban development have water that is more saline (generally >75 µS/cm) with salinity mostly two to three times higher than in undisturbed swamps (Belmer, 2015; Purdy, 2019; Carroll et al. 2020). The highest mean salinity reported from an urban THPSS swamp was reported in an urban catchment swamp at Bullaburra in 2014-15 and followed major disturbance from road works and construction of a new stormwater system (associated with the Great Western Highway upgrade) (Belmer et al. 2015). This swamp recorded the highest salinity ever reported (471 µS/cm) in any Blue Mountains THPSS.

Urban THPSS swamps are also generally acidic, with pH generally 6.0 to 6.5. But surveys of pH including both natural and urban THPSS swamps always reports that urban swamps have higher pH, usually about 1 to 1.5 pH units higher than the undisturbed swamps located within protected areas (Belmer, 2015; Purdy, 2019; Carroll et al. 2020).

Water quality of urban THPSS have a distinct urban ionic composition (Belmer, 2015; Purdy, 2019; Carroll et al. 2020).. Their cations are often dominated by calcium, with sub-dominant sodium, potassium and magnesium ions. The anions in urban THPSS are strongly dominated by bicarbonate anions, with subdominant chloride and sulfate ions. A study of water quality in 2014-15 by Belmer et al. (2015) reported very high calcium and bicarbonate concentrations at an urban swamp affected by recent urban development in the catchment of the swamp.

Belmer et al. (2015) addressed the much higher concentrations of calcium and bicarbonate (Figure 1) in the water collected from the Bullaburra urban swamp: '*We suggest that the higher calcium and bicarbonate levels in this BMUS are linked to recent highway construction in this catchment with associated installations of new concrete stormwater infrastructure which capture and direct highway runoff to the head of the BMUS'.*

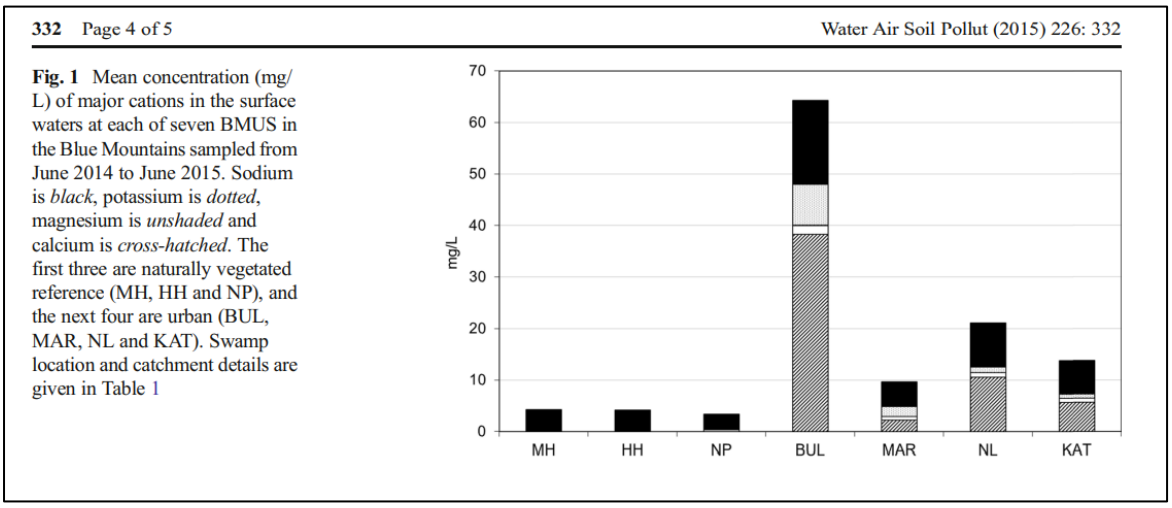

Figure 1. Extract of major cations from a survey of seven THPSS swamps (three undisturbed and four urban) by Belmer et al. (2015).

One of the clearest graphical illustrations of the widely contrasting water quality in urban versus undisturbed non-urban Blue Mountains THPSS swamps is provided by the principal coordinates analysis (PCA) biplot provided by Purdy 2019 (Figure 2). This biplot illustrates the combination of urban water quality attributes

(higher pH, salinity (EC), and higher concentrations of ion (calcium, bicarbonate, potassium, magnesium, chloride, sodium) and metals (barium, strontium). In contrast, the non-urban swamps have elevated dissolved oxygen and elevated aluminium. This biplot shows the major separation in the THPSS water quality between the two groups (undisturbed versus urban; Figure 2).

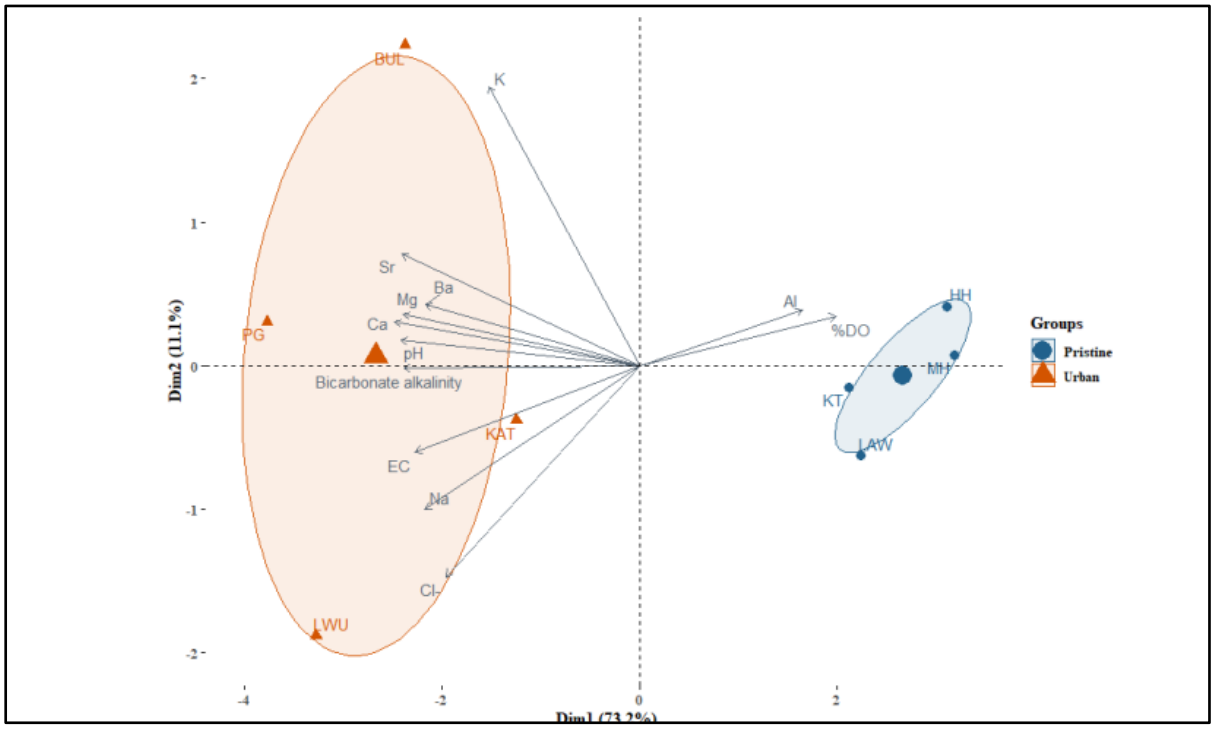

Figure 15. Biplot of the first two principal components accounting for 84.3% of the variance between naturally vegetated/pristine (blue) and urban (orange) swamps with 95% confidence ellipses, based on averages calculated from the water quality and metal and ion concentration data collected.

Figure 2. Biplot using water quality data from a 2019 survey of eight THPSS swamps (four undisturbed and four urban) by Purdy 2019.

Another illustration of the geochemical contrasts in the ionic composition between urban and natural non-urban THPSS is the ternary diagram from Carroll et al. 2020 (Figure 3). This helps to highlight one of the fragile elements of the natural geochemistry of the Blue Mountains swamps – with often undetected or very low calcium and magnesium concentrations providing little to no buffering capacity. This is when water has zero or very low hardness. This is very important as only minor geochemical changes that change natural pH can cause substantial shifts in pH. Figure 4 and 5 also show the contrasting ionic composition between water quality of natural and urban THPSS.

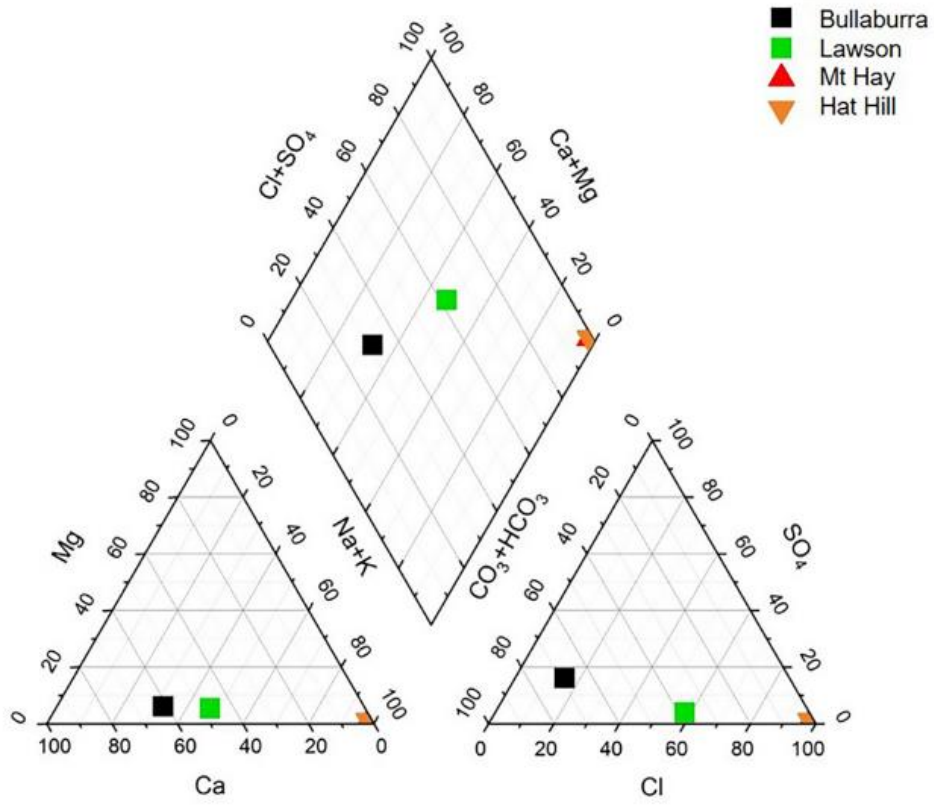

Fig. 2. Ternary diagram represents the ionic composition of water samples collected from each of the four wetlands including the two urban catchment wetlands (Bullaburra and Lawson) and the two naturally vegetated catchment wetlands (Mt Hay and Hat Hill). The Mt. Hay and Hat Hill symbols overlap due to their similar ionic composition. This diagram indicates that the urban wetland do not share ionic composition properties with the natural catchment wetlands.

Figure 3. Ternary diagram using water quality data from a 2018 survey of four THPSS swamps (two undisturbed and two urban) by Carroll 2020.

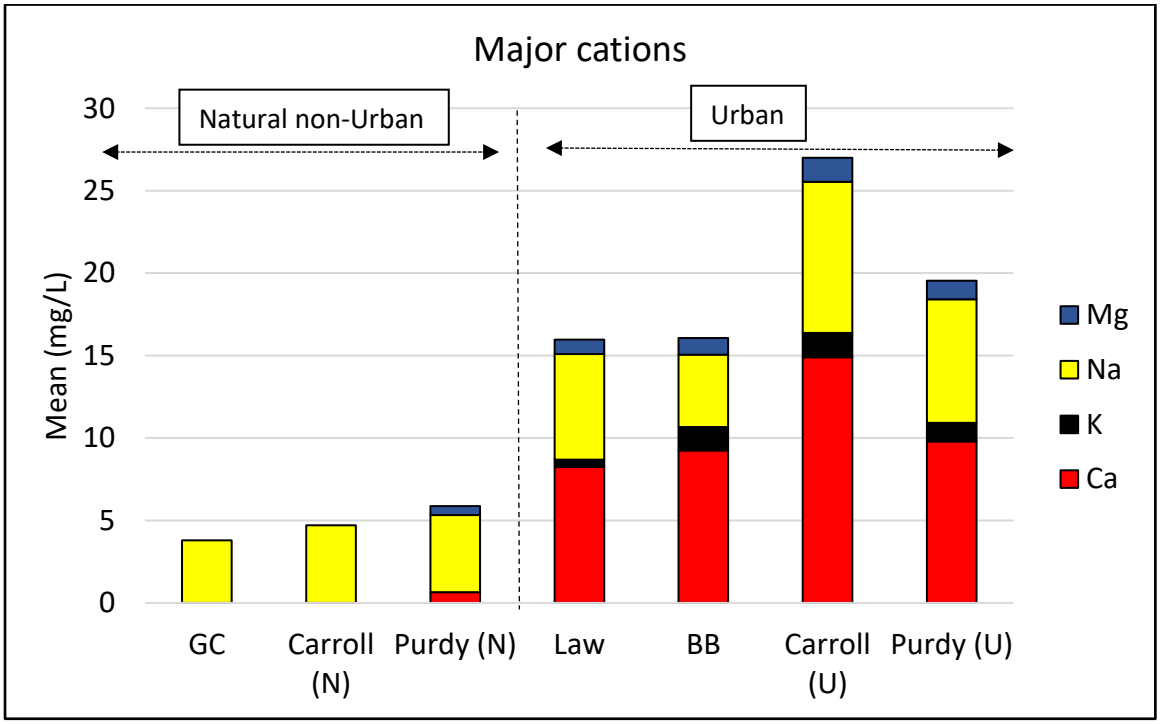

Figure 4. Mean concentration of major cations recorded from three swamps 2021-2022 by BMWHI (GC=natural urban; Law = urban; BB=urban) compared to earlier surveys (2017, 2019) of multiple urban (U) and natural (N) swamps by Carroll (et al. 2019) and Purdy (2019).

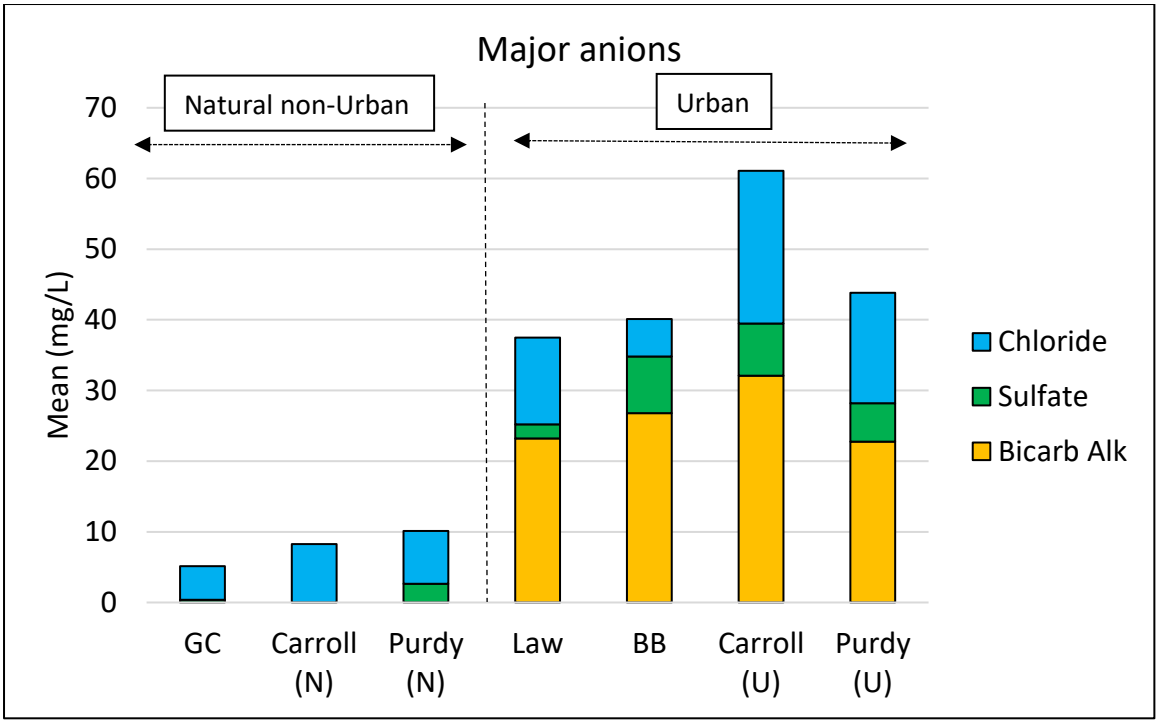

Figure 5. Mean concentration of major anions recorded from three swamps 2021-2022 by BMWHI (GC=natural urban; Law = urban; BB=urban) compared to earlier surveys (2017, 2019) of urban (U) and natural (N) swamps by Carroll (et al. 2019) and Purdy (2019).

The metallic elements in urban THPSS swamps are also dominated by iron and aluminium and to a lesser extent, also manganese (Figure 6, 7). Some urban swamps, at times, have extraordinarily large concentrations of iron. For example, the urban swamp at Lawson recorded iron at a mean concentration of more than 4000 µg/L in 2021/22 (Unpublished BMWHI data). This was about 20 times higher than the typical range of iron concentration recorded in non-urban swamps (Purdy 2019; Carroll et al. 2020).

Barium and strontium (Figure 6) are two metals that are often highly abundant in urban swamps (Purdy 2019; Carroll et al. 2020). This contrasts with natural swamps where the concentration of barium and strontium are recorded at much lower concentrations. Carroll et al (2020) recorded a mean concentration of barium in urban swamps of 17.25 µg/L, more than four times that in undisturbed swamps (mean 3.75 µg/L). Purdy (2019) also reported barium at higher concentrations in urban swamps, compared to natural swamps (mean 15.5 µg/L compared to 5.56 µg/L). More recently, in 2021/22 an unpublished survey of swamps by BMWHI has recorded a mean barium contents in two urban swamps of 12.1 µg/L and 13.5 µg/L. The same study recorded a barium content in water from the undisturbed swamp that was more than 90% lower (mean 1 µg/L).

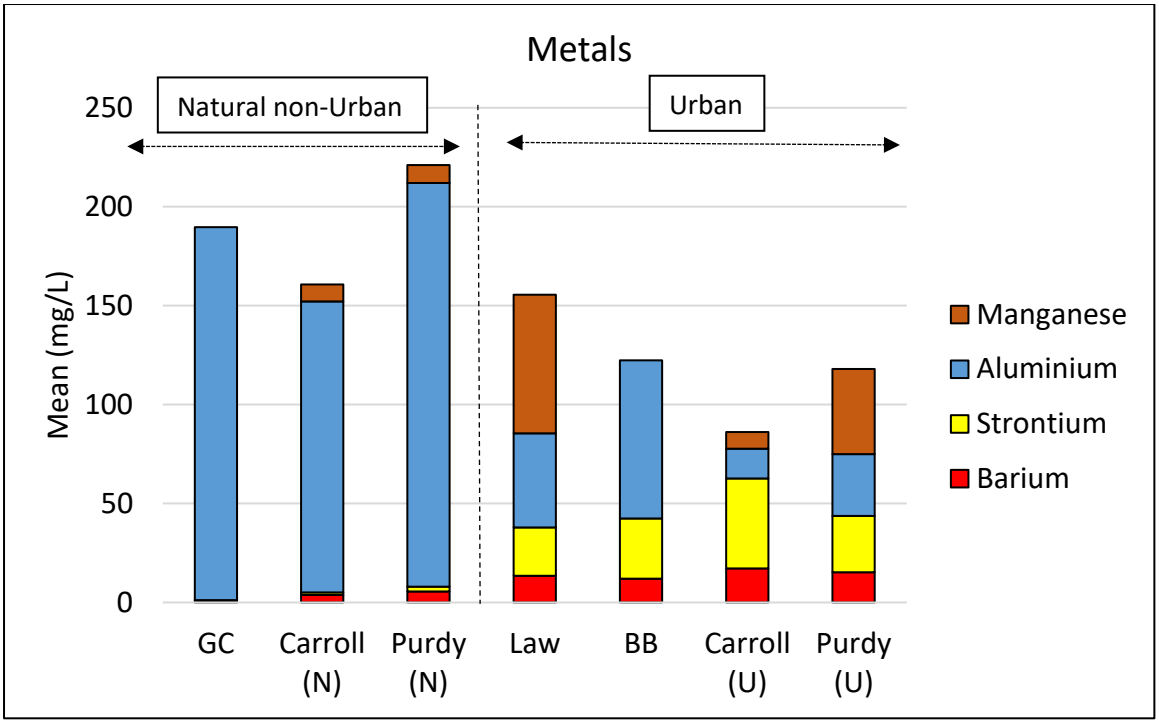

Figure 6. Mean concentration of four metals recorded from three swamps 2021-2022 by BMWHI (GC=natural urban; Law = urban; BB=urban) compared to earlier surveys (2017, 2019) of urban (U) and natural (N) swamps by Carroll (et al. 2019) and Purdy (2019).

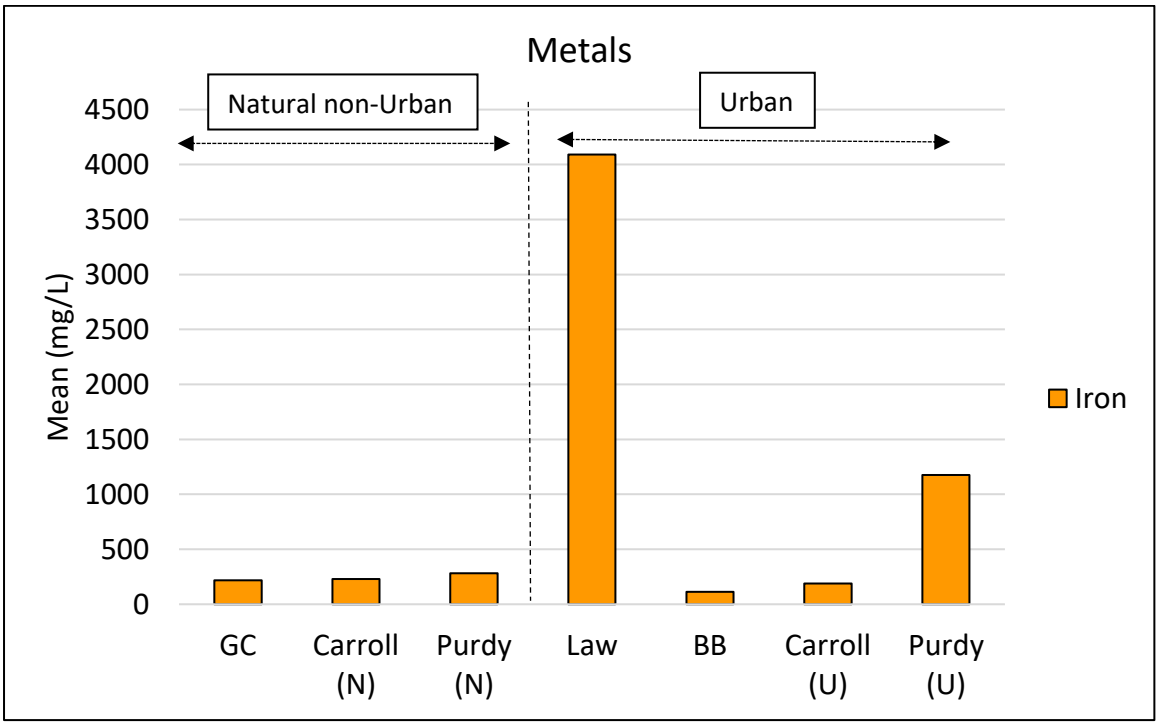

Figure 7. Mean concentration of iron recorded from three swamps 2021-2022 by BMWHI (GC=natural urban; Law = urban; BB=urban) compared to earlier surveys (2017, 2019) of urban (U) and natural (N) swamps by Carroll (et al. 2019) and Purdy (2019).

Materials and Methods

Study location

Water for this experiment was sourced from a Blue Mountains Upland Swamp (BMUS) near Mt Hay located in a protected conservation reserve (Blue Mountains National Park), Leura, NSW (-33.669806, 150.347170). Water from this swamp has been monitored for several years and is regarded as being indicative of a near-pristine swamp in excellent ecological health (Belmer et al. 2015; Belmer et al. 2018; Purdy 2019; Carroll et al. 2020).

The 'Mt Hay' BMUS was selected to be the water source for these tests as this swamp is considered pristine, since 100% of its catchment is naturally vegetated and it exhibits the characteristic low pH (<5), low EC (<30 µs/cm) and domination by sodium and chloride ions as typical of non-urbanised BMUS (Belmer *et al.* 2015; Carroll *et al.* 2020).

Surface water was collected from the exit stream of the Mt Hay swamp on two occasions, 22 September 2022 and 6 October 2022 (Figure 8,9). Water physiochemical properties were recorded in the field, including pH (pH units) and electrical conductivity (EC; µS/cm) and temperature (◦C) using a calibrated TPS Aqua-CP/A waterproof conductivity–TDS-temperature meter (supplied by TPS PTY LTD, Brendale, Queensland). Water was collected (unfiltered) in several sterilized 5-L plastic containers. Containers were filled, with care taken to ensure minimal air headspace in the container to limit gas exchange between the water and air.

Water pH was measured in the field on 22 September and was 5.73. This was almost exactly one pH unit higher to that recorded from three non-urban BMUS which had a mean pH of 4.7 in an earlier study (Belmer et al. 2015). The mean pH of non-urban swamps reported by Carroll et al. 2020 was just below 5 pH units, and Purdy (2019) reported a mean of just above 5 pH units.

Figure 8. Collecting water from an exit stream emerging from the undisturbed Mt Hay THPSS.

Figure 9. Collecting water that was used in recirculation experiments from the exit stream flowing from the THPSS wetland 'Mt Hay' near Leura. The five litre containers were filled with minimal air 'headspace' in the containers.

Preparation of aggregates

All aggregates were commercially sourced from the western Sydney region. The only exception was the polymer concrete that was only available as a solid testing cylinder. It was broken into 10-40 mm fragments using a 1.4 kg lump hammer.

- RCA RBG (Sourced from Fairfield Council as 'RMS spec' road base) 20mm
- RCA 10mm (Turtle Nursery)
- RCA 20mm (Turtle Nursery)
- RCA 60mm (Raygal, Castlereagh)
- Blue Metal gravel (10mm) (supplied by Parklea Soil and Sand, South Windsor, NSW)

All aggregates were soaked for several minutes and then rinsed several times in deionised water to remove any muddy contaminants. The washing also helped to reveal non-concrete contaminants, such as timber, stones, wire, plastics, ceramic tiles and terracotta pipe fragments. The aggregates were then dried in the laboratory in an incubator (Labec incubator S4218) for 48 hours at 38.5 degrees prior to the water recirculation. Before being carefully weighed (1000 +/- 5 grams) using laboratory scales each batch was carefully examined to remove non-concrete material manually removed from the RCA aggregates. There was no foreign contaminants in the polymer aggregates, and similarly there were no non-blue metal contaminants in the blue metal gravel aggregates.

Recirculation experiment

The first treatment, which is considered the procedural control, was a clean, empty PVC pipe, 100 mm wide and 2500 mm long (supplied by Bunnings, Pymble, NSW). It contained a PVC cap with a small 5 mm hole drilled and was secured in the laboratory at an angle of about 15 °, prior to use it was calibrated to allow about

200 mL of water to drain per minute (Figure 10-12). All PVC pipes, beakers, measuring cylinders were acid rinsed and then deionised water rinsed prior to use.

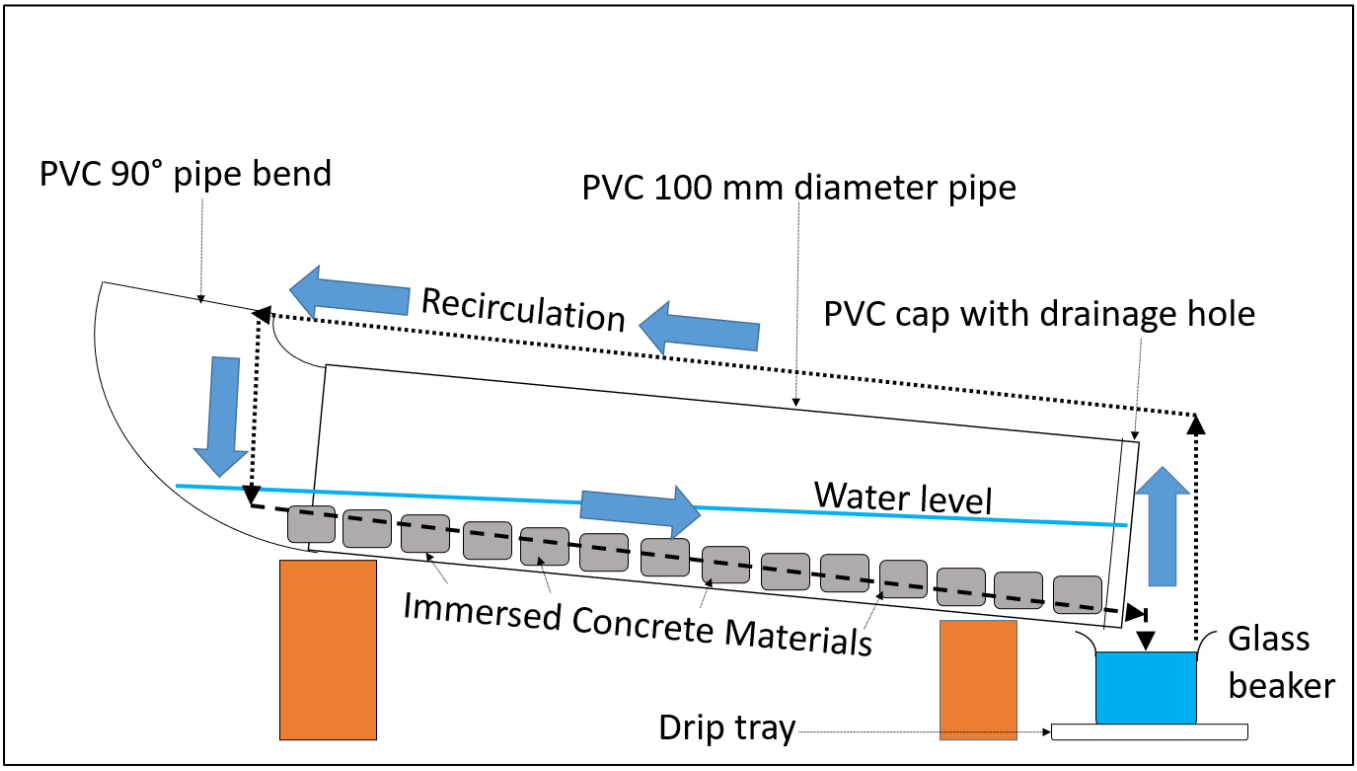

Figure 10. Testing apparatus used to recirculate water through a clean PVC pipe containing concrete and other aggregate materials.

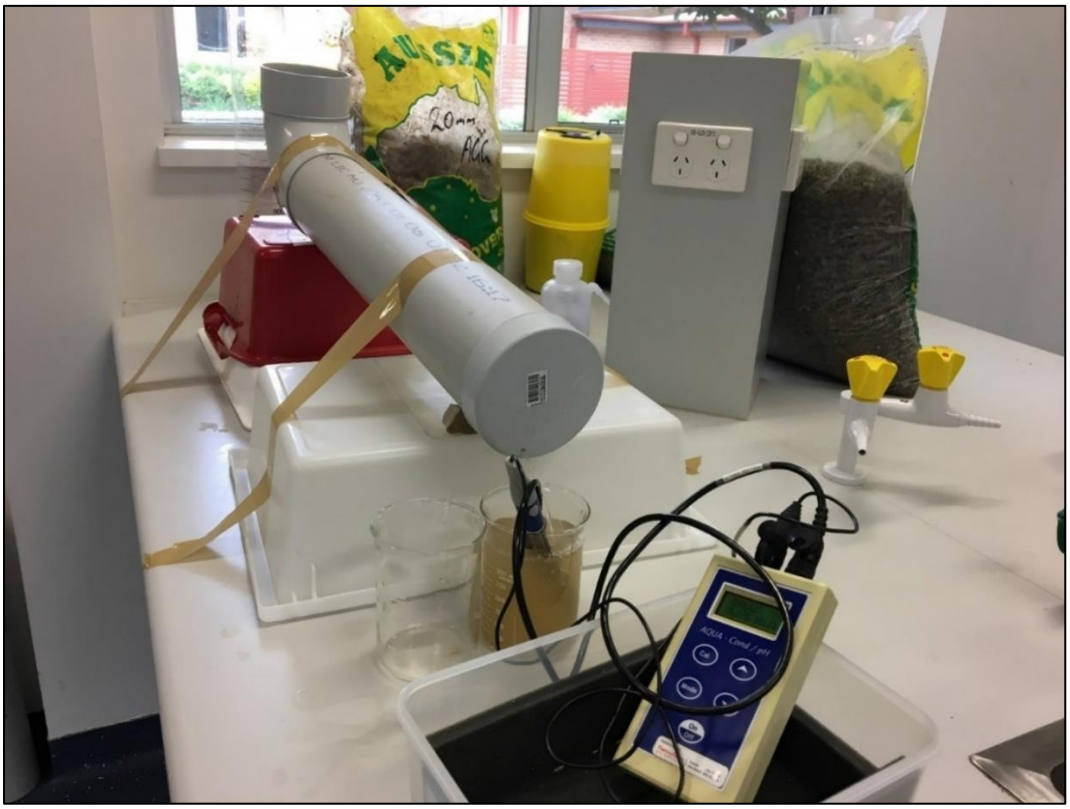

Figure 11. A pilot study that was used to recirculate water through a PVC pipe containing concrete and other aggregate materials. After this pilot study all aggregates were soaked and rinsed in deionised water to remove 'muddy' contaminants.

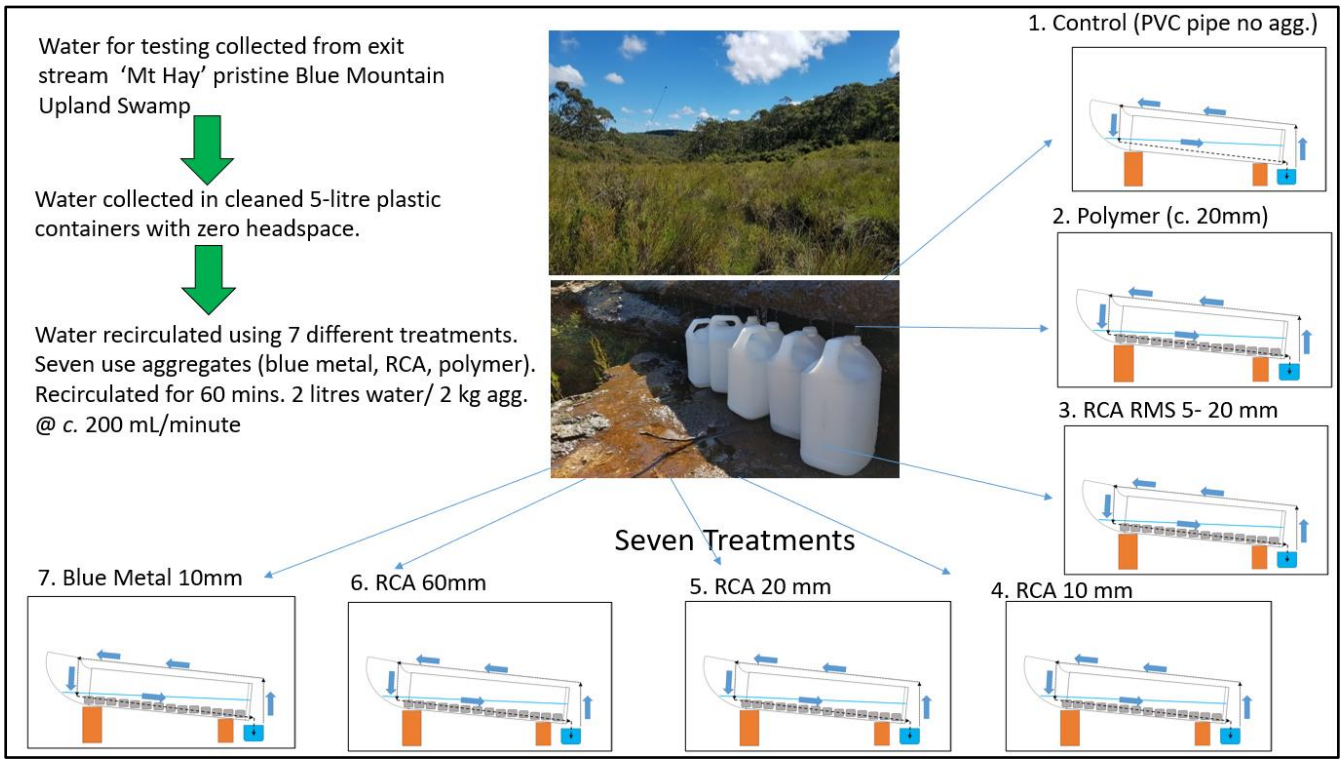

Figure 12. The seven treatments used to test water recirculation through concrete aggregate materials.

All treatments used an identical set-up to the procedural control, except included 1 kg of different aggregate materials (Figure 10, 11, 12). The other six treatments used the same PVC pipe specifications (all were new and used sections of pipe of identical dimensions (Figure 12). The second treatments contained 1kg of commercially sourced blue metal/basalt aggregates with fragment sizes of 10mm. The other four treatments used different grades of commercially sourced RCA aggregates (Figure 12). One had fragment size of 10mm (third treatment), 20mm (fourth treatment; supplied by Turtle Nursery and Landscape Supplies, Rouse Hill, NSW) and 40-70mm (fifth treatment) (supplied by Raygal Pty Ltd, Cranebook, NSW). The sixth treatment was a 20mm RCA road based that is supplied by Fairfield Council, which they advertised as recycled concrete road base 'RMS spec'. In this report this is labelled 'RMS'. Although the size grade has not been measured, the RMS aggregates appeared to include very fine grade crushed concrete, probably representative of the 2mm to 20mm particle sizes.

The treatments that included aggregates evenly spread the aggregates so that it covered the PVC pipe to an average depth of 40 mm (Figure 10). 200 mL of water was emptied into one of the two 250 mL glass beakers that were used for each 60-minute experiment (Figure 11). When the recirculation experiment commenced, 800mL of water was then manually poured from a measuring cylinder into the higher end of the PVC pipe, using a 90 ° PVC collar (Figure 13). This allowed the water to flow through the length of the pipe through the submerged aggregate material and flow was restricted by the small exit hole drilled in the PVC end cap, draining into one of the two beakers. The flow rate was estimated to be 5.2 mL/second.

The water was continuously recirculated manually, using one of the two beakers, for 60 min, for each of the five treatments. Water quality was monitored throughout the experiment with a TPS AQUA-Cond-pH meter, with probes immersed in one of the beakers. pH (pH units) and salinity was measured as electrical conductivity EC (μS/cm) and temperature (◦C) readings were taken at 5-min intervals throughout the experiment for each treatment.

The probes were rinsed, tested and calibrated before each experiment. These 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 by (Davies et al. 2010; Purdy 2019; Purdy et al. 2020) and were also confirmed through an earlier pilot study conducted by the authors which demonstrated that the pH and EC changes for the treatments were very rapid in the first 5 minutes of the recirculation. Thus, a shorter sampling interval was required for just the first five minutes. For each treatment, the experiment was repeated a second time using the above methods. Prior to 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 200mL anion, cation and metal samples of the Mt Hay swamp THPSS water were collected before the experiment commenced. These were classified as 'before-recirculation' samples. Similarly, replicate anion, cation and samples were also taken for each experimental treatment at the end of each recirculation experiment (Figure 14). These were considered the 'after-recirculation' samples. These 'before-recirculation' and 'after-recirculation' samples were sent to a commercial, NATA-accredited laboratory for 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.

ANZECC guidelines for ecosystem protection

The water-quality results obtained at the completion of the 60-min recirculation experiments were compared with ANZECC (2000) water-quality guidelines. This was undertaken to provide guidance on potential adverse impacts to aquatic species if water exposed to materials in the experimental treatments flowed into a THPPS. The guidelines assessed metals / metaloids aluminium, arsenic, copper, lead, zinc and nickel by using the highest level of protection

(protection of 99% of species), given the conservation significance of THPSS wetlands. The guidelines used a salinity (as EC) of 55 µS/cm, based on south-eastern upland streams in eastern highlands (Table 3.3.3 in Chapter 3 Aquatic Ecosystems; ANZECC 2000). There are no water quality guidelines specification recommended for THPSS wetlands, and pH is such an important attribute (Christiansen et al. 2019; Carroll et al., 2020). Given the importance of pH, and the acidic nature of undisturbed THPSS wetlands, an interim recommendation of <6 pH is recommended.

Table 1. Trigger values adopted for THPSS for protection of 99% of aquatic species, based on ANZECC (2000).

Indicator	ANZECC (2000)
Salinity	<55 µS/cm
pH	<6 (based on previous research by Belmer et al. 2015; Carroll et al. 2020)
Aluminium	<27 µg/L (if pH >6.5)
Arsenic	<1.0 µg/L
Copper	<1.0 µg/L
Lead	<1.0 µg/L
Nickel	<8.0 µg/L
Zinc	<2.4 µg/L

Statistical analysis

All statistical analyses were performed using Microsoft Excel. Changes in pH, EC, metal, metalloid and major anion and cation 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. Some water chemical attributes were not detected because they were lower than laboratory detection limits. In these cases, for data-analysis purposes, the result was assumed to be half of the detection limit (Moore et al. 2017). P-values of less than 0.05 were considered significant.

Figure 13. Measuring 1 litre water prior to starting the recirculation experiment. All PVC pipes and laboratory glassware was cleaned and acid-rinsed then deionised water rinsed prior to each recirculation.

Figure 14. Preparing water samples, collected before and after the 60-minute recircluation experiments, for major ion and metal testing by a NATA accredited laboratory.

Strontium

There are no ANZECC (2000) guidelines available for strontium. Strontium is regarded as one of the key indicators of the 'urban geochemical signature' in urban THPSS and a major contributor is thought to be urban concrete materials (Carroll et al. 2020). Strontium was found by Carroll et al. (2020) at low background concentration in natural non-urban THPSS swamps (mean 1.4 µg/L) and more than 40 times greater in urban swamps (mean 45.5 µg/L; Carroll et al. 2020). Similar results were reported by Purdy (2019) with urban swamps recording more than 10 times more highly elevated strontium (mean 28.5 µg/L) in urban swamp water compared to natural swamps (mean=2.4 µg/L). Purdy (2019) also reported strontium accumulating in urban swamp sediment at 175% higher concentrations, compared to natural swamp sediment.

The source water for the recirculation experiments (from non-urban swamp at Mt Hay) contained strontium at 1.3 µg/L. After water was recirculated through the seven treatments (Figure 15), the polymer fragments results in the lowest strontium concentration (mean = 5 µg/L). The strontium content steeply increased for all other aggregates, ranging from RCA 10mm (mean 44.7 µg/L) to RMS RCA (mean = 360 µg/L; Figure 15).

Caution is recommended for interpretation of any environmental risks of elevated strontium in THPSS wetlands affected by urban development. A previous study has documented the bioaccumulation of barium and strontium in plant tissue, for plants grown in water with elevated barium concentrations (Morrison et al. 2019). Strontium and barium are two of the most abundant metals found in urban THPSS and further investigation is needed to fully understand the hazards of elevated concentrations of both on these high conservation-value wetlands.

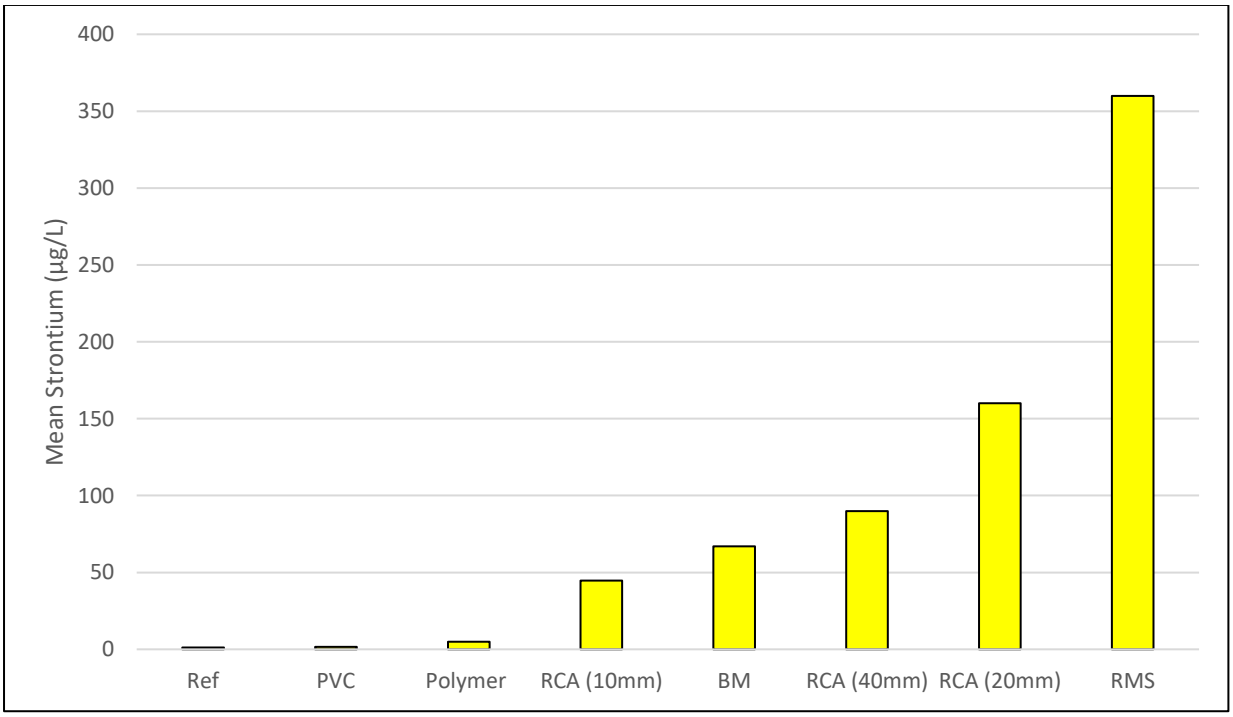

Figure 15. Mean concentration of strontium before (Ref) and after 60-minutes of recirculation through seven treatments. As there is no ANZECC (2000) guideline for strontium the mean result for each treatment is coloured yellow.

Barium

There are no ANZECC (2000) guidelines available for barium. Barium has previously been detected at low background concentration in natural non-urban swamps (mean 3.75 µg/L) and more than four times greater in urban swamps (mean 17.25 µg/L; Carroll et al. 2020). Very similar results, with greater barium detected in urban swamps, was reported by Purdy (2019). Purdy (2019) also reported barium accumulating in urban swamp sediment.

The source water for the recirculation experiments (from non-urban swamp at Mt Hay) contained barium at 2.5 µg/L. After water was recirculated through the four RCA treatments for 60 minutes, significant increases in barium were observed (Figure 16). They increased above background levels (mean concentration before = 2.5 µg/L) by at least 4 times (mean 10.7 µg/L for 20mm RCA) to more than 17 times (mean 43 µg/L RMS RCA; Figure 16).

Barium recorded the mildest increase in concentration, after the 60-minute recirculation tests, when recirculated through the blue metal aggregate treatment. It recorded a mean barium concentration of 4.8 µg/L. The barium concentration, after recirculation through polymer fragments, was mildly greater than the blue metal treatment (mean 5.0 µg/L).

Caution is recommended for interpretation of any environmental risks of elevated barium in THPSS wetlands affected by urban development. A previous study has documented the bioaccumulation of barium in plant tissue, for plants grown in water with elevated barium concentrations (Morrison et al. 2019). Barium if one of the most abundant metals found in urban THPSS and further investigation is needed to fully understand the hazards of elevated barium.

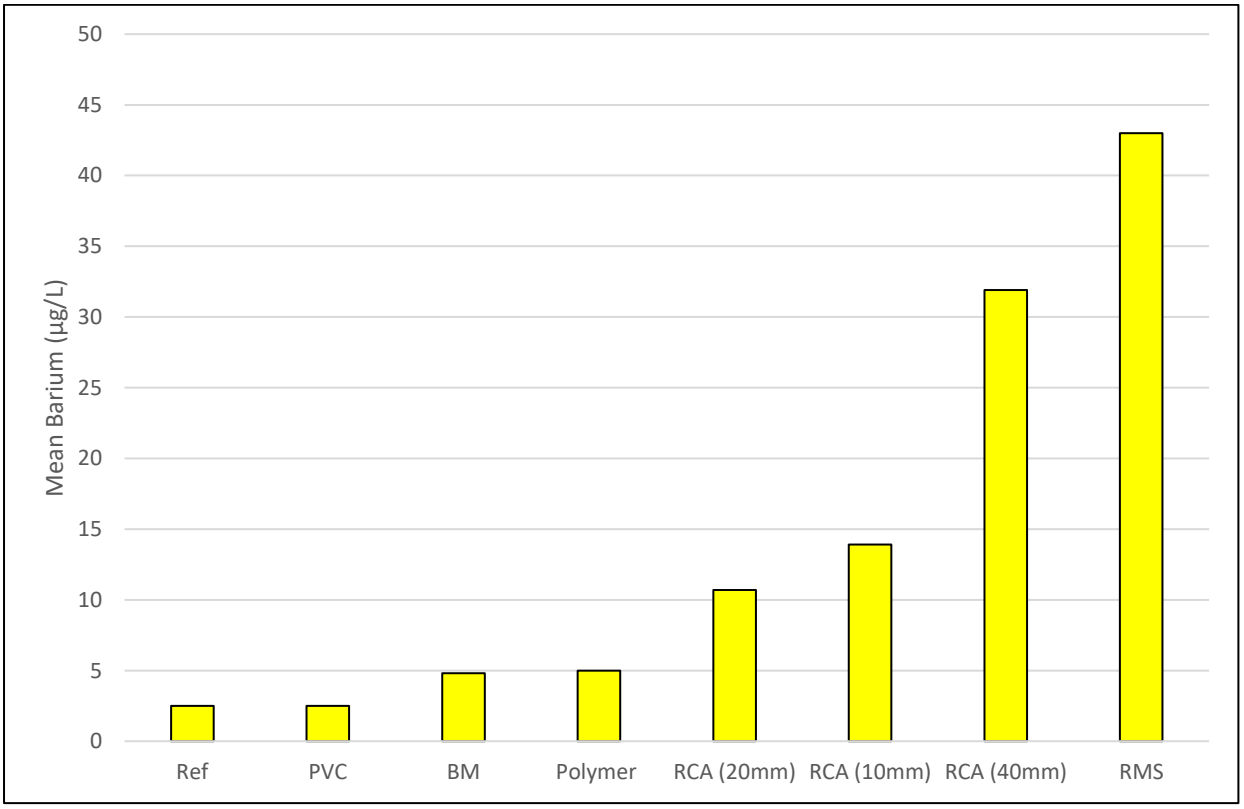

Figure 16. Mean concentration of barium before (Ref) and after 60-minutes of recirculation through seven treatments. As there is no ANZECC (2000) guideline for barium the mean barium result for each treatment is coloured yellow.

Aluminium

The ANZECC (2000) water quality guideline for aluminium for protecting 99% of species is <27 µg/L if pH is >6.5 pH units. Although aluminium was commonly detected at high background concentrations in natural swamps (mean 147.5 µg/L; Carroll et al. 2020) it only occasionally exceeded the ANZECC guidelines as pH of natural swamps is generally less than 6.5 pH units. Aluminium was overall the sub-dominant metal in natural swamps, with an overall lower mean concentration in comparison to the more abundant iron (mean 190 µg/L) at natural THPSS (Carroll et al. 2020).

The source water for the recirculation experiments (from the natural THPSS swamp at Mt Hay) contained aluminium at 115 µg/L, with a pH of 5.73 and as a consequence did not exceed the aluminium ANZECC guidelines (<6.5 pH units; Figure 17). After the Mt Hay water was recirculated through the aggregate treatments for 60 minutes, significant increases in aluminium were observed. As all water had pH of at least 7.5 pH units, all aluminium concentrations, after recirculation through aggregates, exceeded the ANZECC guideline (Figure 17).

Aluminium concentrations, after 60-minutes recirculation through aggregates exceeded the initial concentration by at least 3 times (mean 380 µg/L for 20mm RCA) to more than 40 times (mean 5000 µg/L RMS RCA). The concentration of aluminium increased to 1322 µg/L after recirculation through the blue metal aggregate treatment and 720 µg/L after recirculation through polymer fragments (Figure 17).

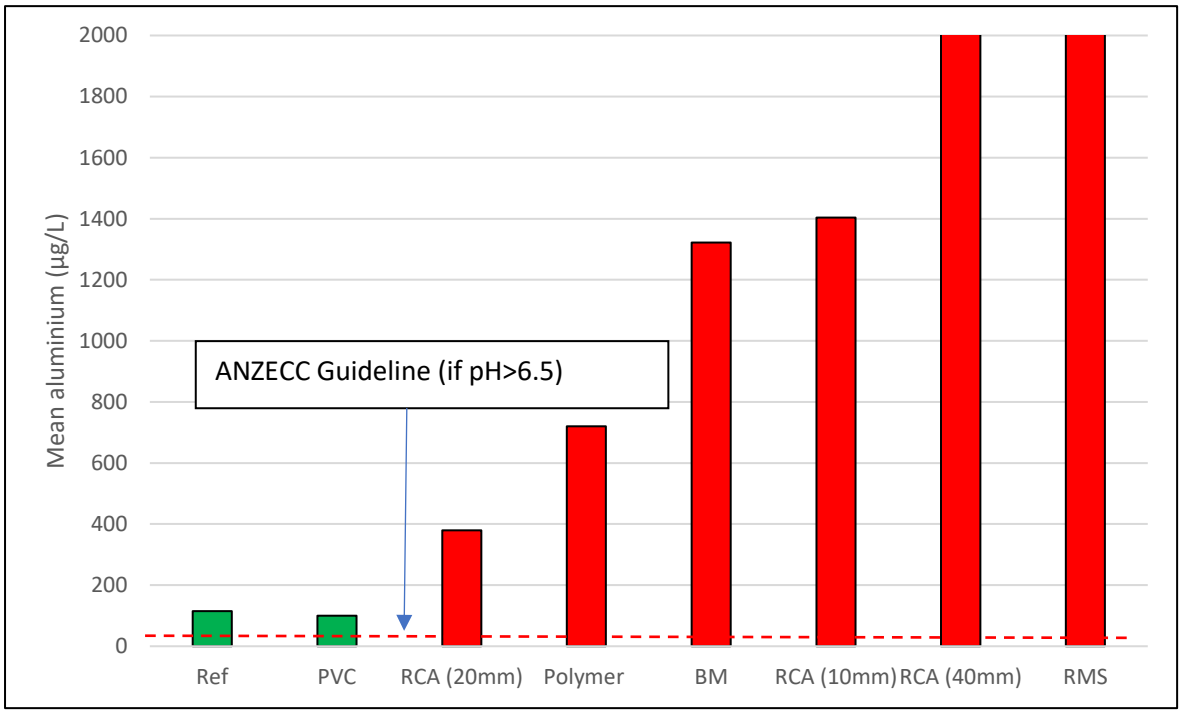

Figure 17. Mean concentration of aluminium before (Ref) and after 60-minutes of recirculation through seven treatments. The ANZECC (2000) guideline for 99% protection of species is shown (<27 µg/L, if pH is >6.5). The result for that treatment is coloured green if compliant with the guideline, orange if it is borderline and red if it exceeds the guideline. Please note –aluminium in water recirculated through RCA (40mm) was 2366 µg/L and RMS was 5000 µg/L and were not fully displayed in this graph.

Arsenic

The ANZECC (2000) water quality guideline for arsenic for protecting 99% of species is <1 µg/L. There is very limited data available on arsenic in any THPSS. Purdy (2019) reported that arsenic was undetected in three of four natural swamps in her 2019 survey. In one natural swamp it was detected at 4 µg/L. In addition, it was detected in three of four urban swamps with a mean concentration of 4.42 µg/L (Purdy, 2019). Unpublished data from a BMWHI survey of THPSS has never detected arsenic in natural swamps, and only ever in two samples (at 1 µg/L) in an urban swamp. The source water for the recirculation experiments (from pristine BMUS at Mt Hay) did not contain a measurable concentration of arsenic.

Before the recirculation, no detectable arsenic was found in any replicate sample. After the Mt Hay water was recirculated through the seven treatments, measurable arsenic was only detected in five treatments. Three (BM, 10mm RCA, 20mm RCA) were measured at concentrations under the guideline (1 µg/L). The RMS RCA treatment had a mean concentration at 1 µg/L, equal to the guideline. The RCA 40mm treatment exceeded the arsenic guideline, by more than two times, with a mean concentration of 2.2 µg/L.

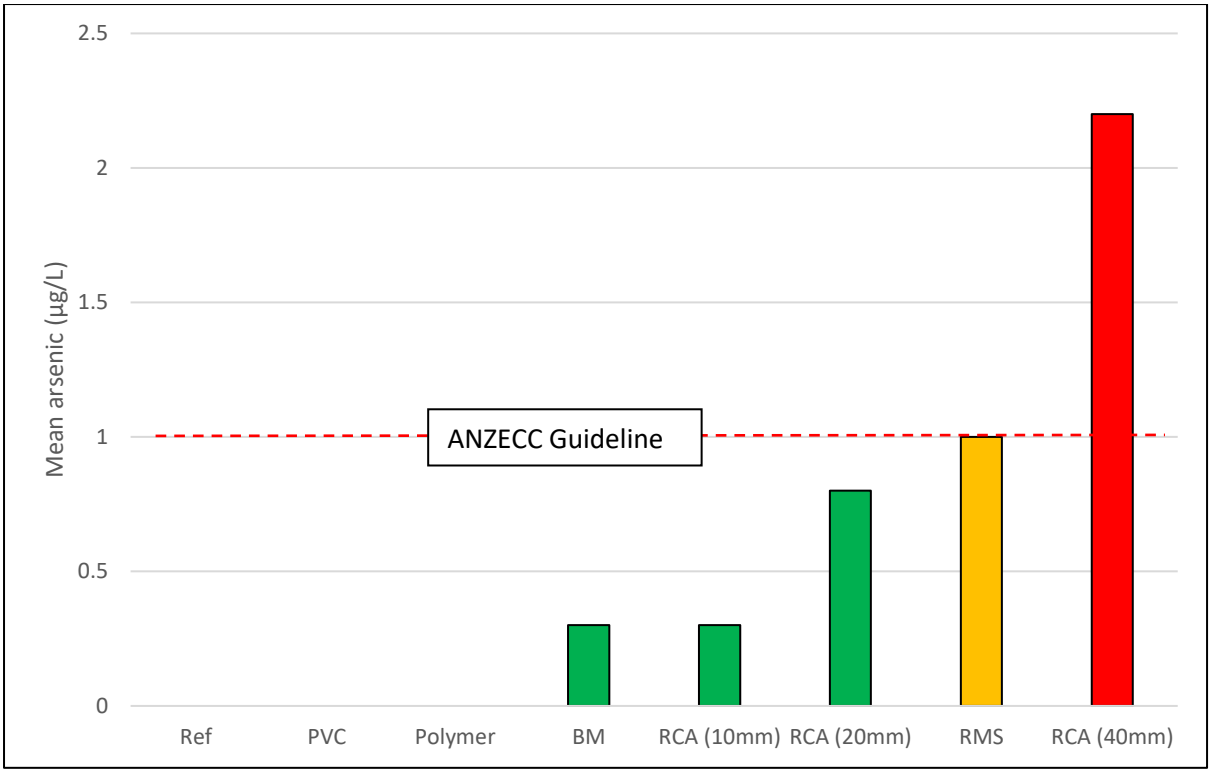

Figure 18. Mean concentration of arsenic before (Ref) and after 60-minutes of recirculation through seven treatments. The ANZECC (2000) guideline for 99% protection of species is shown (<1 µg/L). The result for that treatment is coloured green if compliant with the guideline, orange if it is borderline and red if it exceeds the guideline.

Copper

The ANZECC (2000) water quality guideline for copper for protecting 99% of species is <1 µg/L. There is limited data available on copper in any THPSS. Purdy (2019) reported that the mean copper concentration was slightly higher at natural swamps (3.76 µg/L) compared to urban swamps (1.13 µg/L) in her 2019 survey, however the differences were not statistically significant. She also reported that copper was found at higher (164%) concentration in urban swamp sediments, than natural swamp sediments, and the difference was statistically significant (Purdy, 2019). Carroll et al. (2020) did not detect copper in any THPSS water samples. A survey of two urban swamps by BMWHI in 2021/2 has detected copper in some samples, ranging from undetectable levels to 3 µg/L. In contrast, the survey has never detected copper in the natural swamp.

The copper concentrations in the Mt Hay water used in the current recirculation study were unexpectedly highly elevated (mean 22.25 µg/L). The cause of the elevated copper concentration is not known. It is possible that the elevated copper was associated with the 2019/20 wildfire, and heavy rains in 2021/22. Several researchers have reported that metals present in soil, including copper, can be mobilised by wildfires (Fernandez-Marcos, 2022).

The recirculation experiment recorded a very low copper concentration after the polymer recirculation treatment (mean 3 µg/L), and a very high concentration (mean=64.2 µg/L) in the blue metal treatment, after 60-minute recirculation (Figure 19).

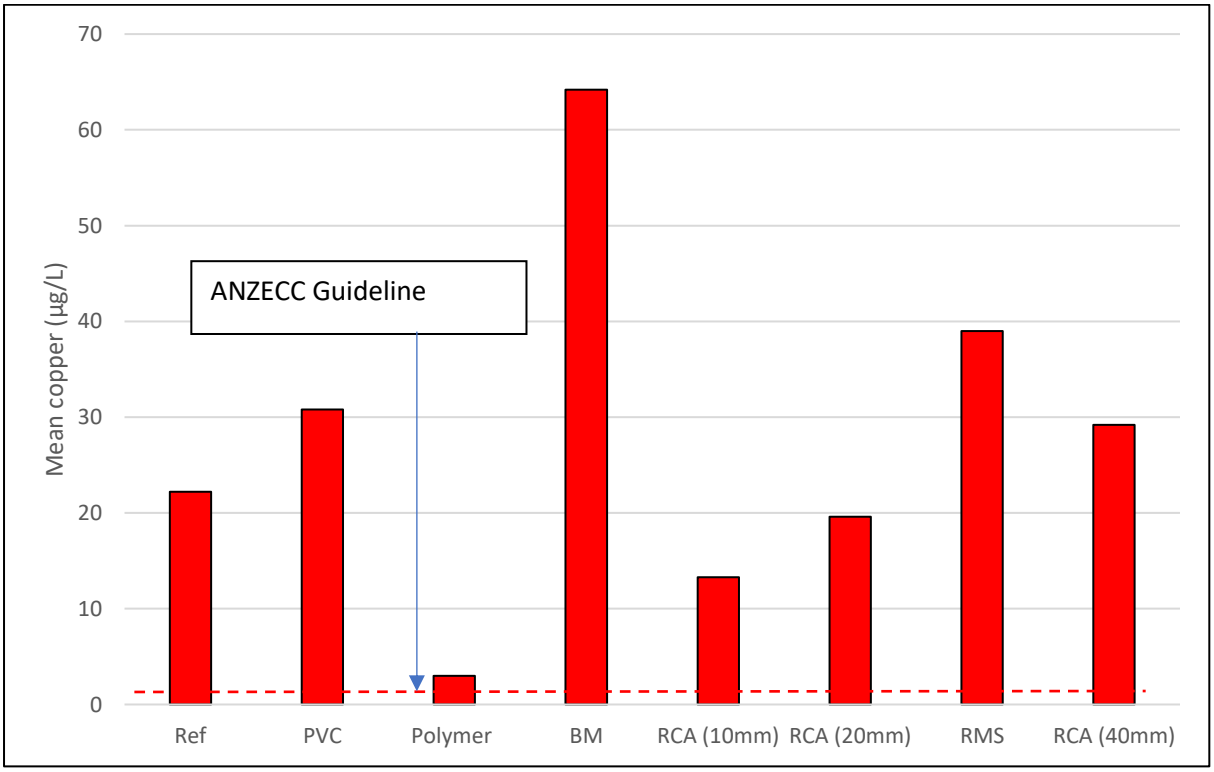

Figure 19. Mean concentration of copper before (Ref) and after 60-minutes of recirculation through seven treatments. The ANZECC (2000) guideline for 99% protection of species is shown (<1 µg/L). The result for that treatment is coloured green if compliant with the guideline, orange if it is borderline and red if it exceeds the guideline.

Lead

The ANZECC (2000) water quality guideline for protecting 99% of species is <1 µg/L. There has been limited data available on lead in THPSS. Purdy (2019) reported that lead was detected at slightly greater concentrations in natural swamps (mean = 1.47 µg/L) compared to urban swamps (mean = 1.01 µg/L). The difference was not statistically significant. In addition, Purdy (2019) also found that the concentration of lead in urban swamp sediment was 79% greater than that in natural swamp sediments, but again, the difference was not significant. Carroll et al. (2020) did not detect lead in her survey of water from natural and urban swamps. An unpublished survey of three THPSS swamps by BMWHI (2021/22) did not detect any lead in the natural or urban swamps.

No lead was detected in the Mt Hay water that was used as the raw water for the recirculation experiments. A trace was detected in the PVC control treatment. After the Mt Hay water was recirculated through the treatments, all four RCA treatments recorded lead at concentrations above the ANZECC guideline (Figure 20). No lead was detected in the BM treatment. The polymer treatment recorded 5 µg/L of lead, after 60-minutes. The highest concentration of 12 µg/L was detected in the RCA 10mm treatment, after 60-minutes of recirculation (Figure 20).

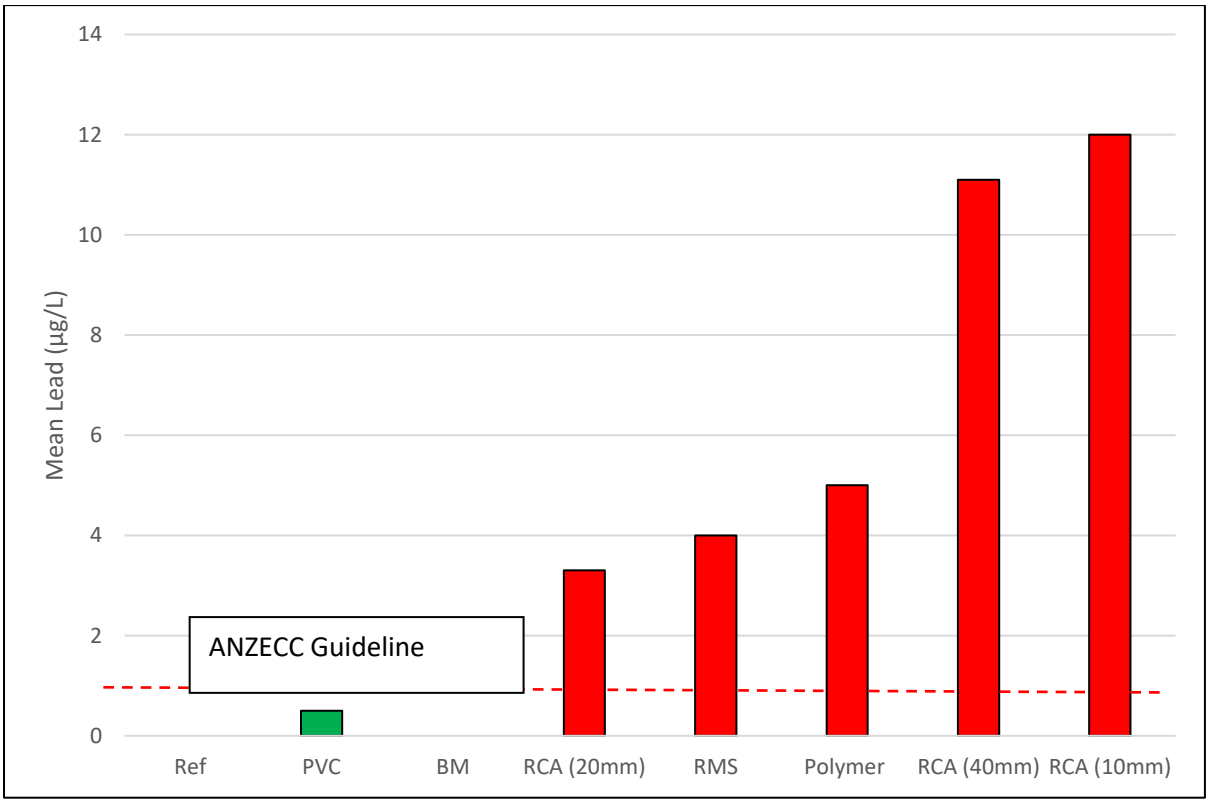

Figure 20. Mean concentration of lead before (Ref) and after 60-minutes of recirculation through seven treatments. The ANZECC (2000) guideline for 99% protection of species is shown (<1 µg/L). The result for that treatment is coloured green if compliant with the guideline, orange if it is borderline and red if it exceeds the guideline.

Zinc

The ANZECC (2000) water quality guideline for zinc for protecting 99% of species is <2.4 µg/L. There is limited data available on zinc in THPSS. Purdy (2019) reported that zinc was measured at greater concentrations in natural swamps (mean = 11.69 µg/L) compared to urban swamps (mean = 6.38 µg/L). The difference was not statistically significant. In addition, Purdy (2019) also found that the concentration of zinc in urban swamp sediment was 633% greater than that in natural swamp sediments, the difference was statistically significant.

Carroll et al. (2020) detected low concentrations of zinc in her survey of water from natural and urban swamps (natural=2.5 µg/L; urban =2.25 µg/L). Similarly, a survey of three THPSS swamps by BMWHI (2021/22) recorded slightly lower zinc concentrations in the natural (mean=2.3 µg/L) and urban swamps (mean 3.5 and 3.1 µg/L respectively).

The mean concentration of zinc (4.25 µg/L) in the Mt Hay water that was used as the raw water for the recirculation experiments was nearly twice the ANZECC guideline. After the Mt Hay water was recirculated through the treatments, all treatments recorded zinc at concentrations well above the ANZECC guideline and above the concentration recorded in the water from Mt Hay (Figure 21). The polymer treatment recorded 7 µg/L of zinc, after 60-minutes of recirculation. The RCA treatments all recorded the highest zinc concentrations, starting with 12.9 µg/L in the 10mm RCA treatment. The highest concentration of zinc of 20.8 µg/L was found in the RCA 40mm treatment, after 60-minutes of recirculation (Figure 21).

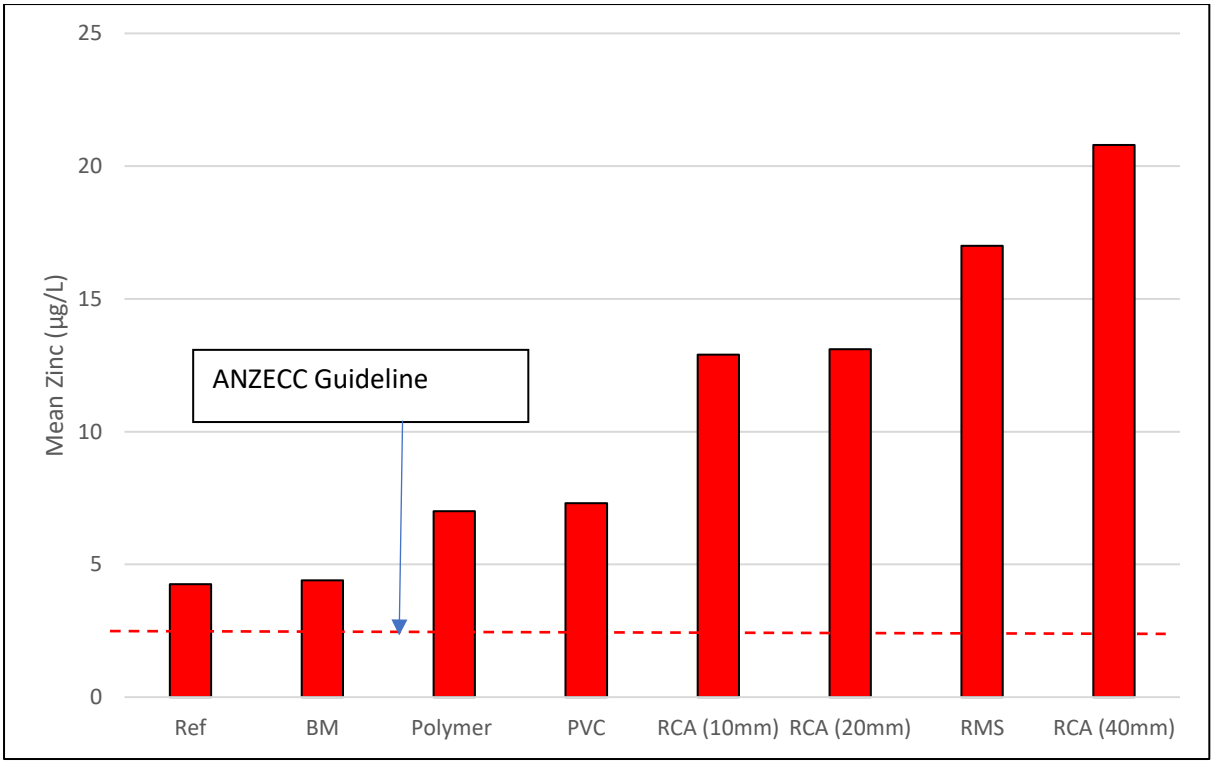

Figure 21. Mean concentration of zinc before (Ref) and after 60-minutes of recirculation through seven treatments. The ANZECC (2000) zinc guideline for 99% protection of species is shown (<2.4 µg/L). The result for that treatment is coloured green if compliant with the guideline, orange if it is borderline and red if it exceeds the guideline.

Combination of minor metals mobilised by recirculation

Figure 21 shows the mean combined concentration of minor metals in water samples that had been recirculated through different aggregate treatments. This graphic excludes iron and aluminium, which were often recorded in such large concentrations that tend to obscure the presence of other metals.

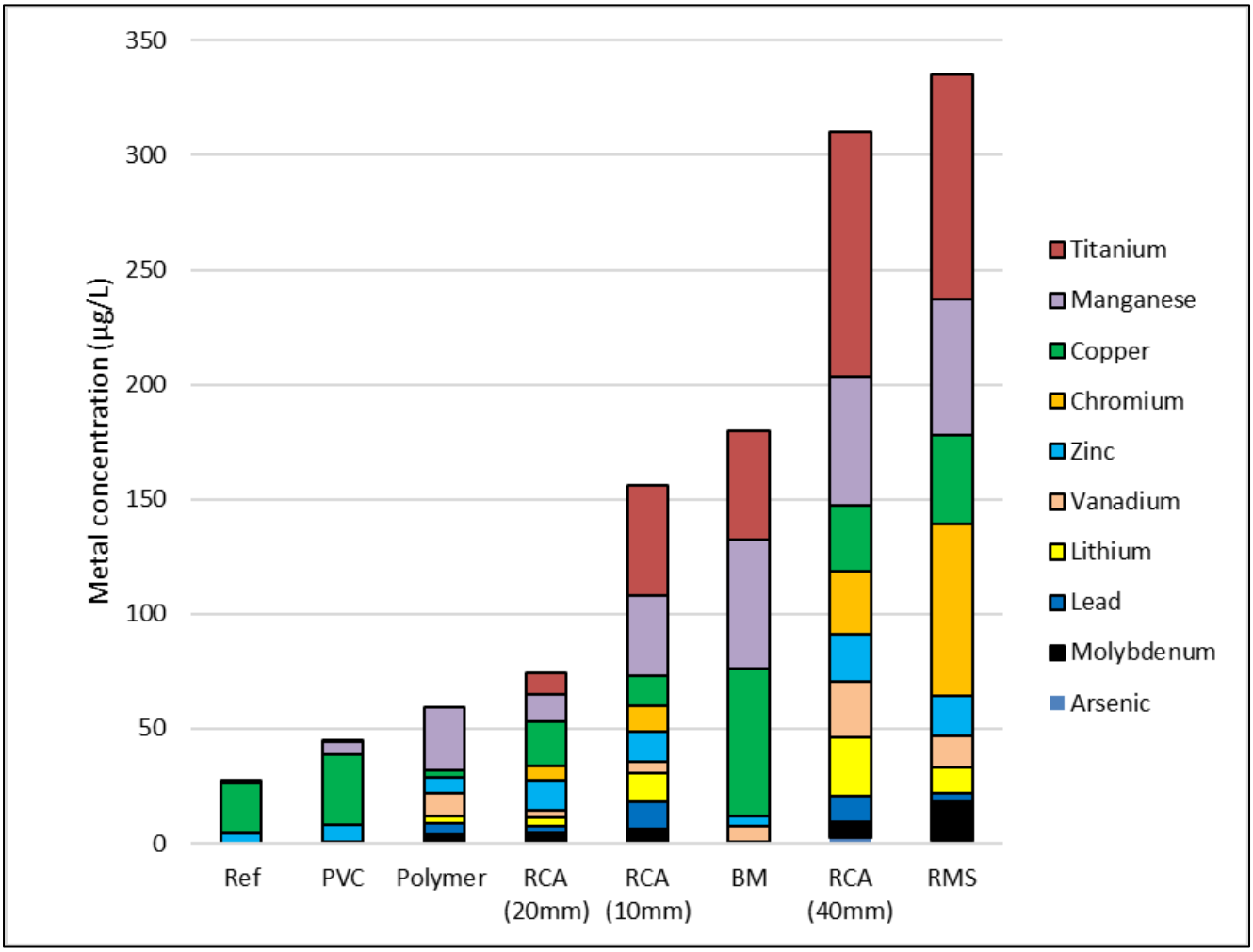

Figure 22. Stacked bar graph showing the combined mean concentration of metals (excluding iron) in water before (Ref) and after 60-minutes of recirculation through the seven treatments.

pH

The raw water from Mt Hay was used in the recirculation experiment was acidic, with a pH of 5.8 pH units. This was higher than an earlier survey of four natural BMUS that had a mean pH of just under 5 pH units (mean pH = 4.96; Carroll et al. 2020). There are no ANZECC (2000) guidelines that apply to THPSS wetlands, but the ANZECC (2000) approach recommends derivation of local guidelines, and the 80th percentile of pH at natural THPSS of 6 pH units is based on previous research (Carroll et al. 2020; Purdy, 2019; Belmer et al. 2015).

The 60-minute recirculation experiments increased the pH of all treatments, including within the procedural control treatment, a PVC pipe that contained no aggregate materials. The water in the PVC control increased pH by 0.8 pH units over the 60-minute recirculation (Figure 23). The second smallest increase in pH was after the 10 mm blue metal gravel recirculation treatment which increased pH by 2.3 pH units, to a final pH of 8.1 units (Figure 23). The largest change in pH was recorded for the RMS RCA treatment, with pH increasing by 5.2 pH units to pH of 11.05 at the conclusion of the 60-minute recirculation. A similar, but slightly smaller, increase in pH increase of 5 pH units (final pH of 10.85) was recorded after 60 minutes recirculation through polymer concrete fragments.

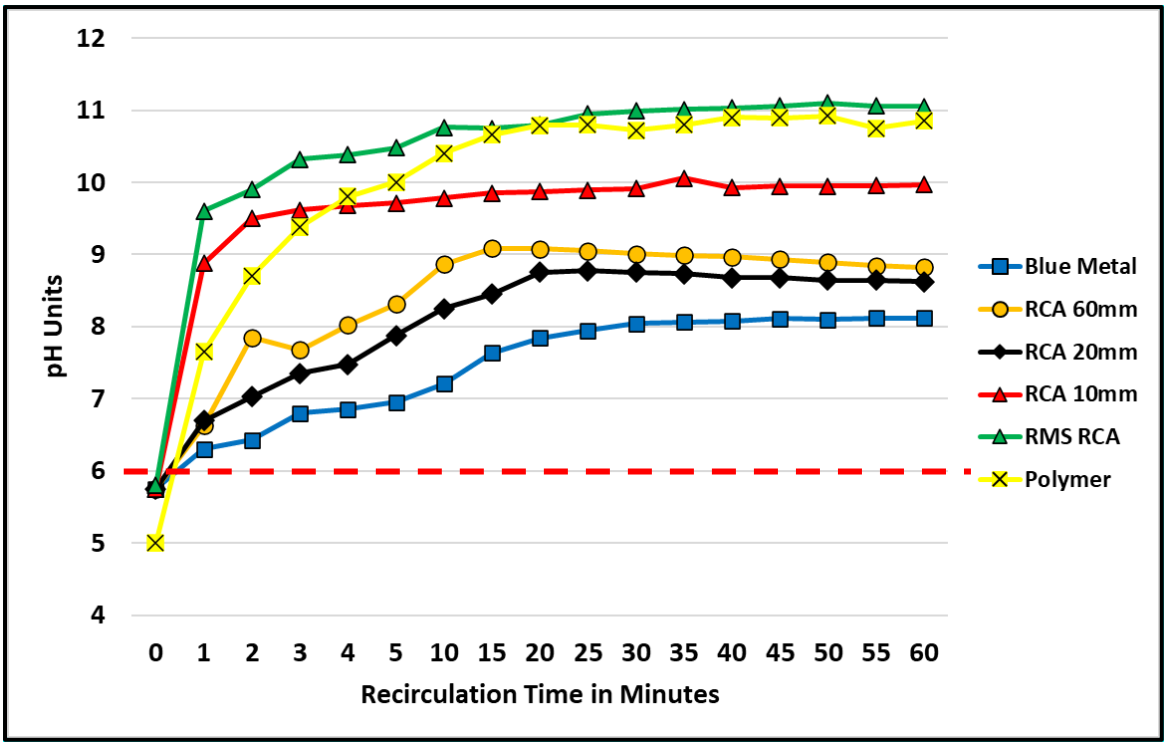

Figure 23. Time series graph showing the pH of as water is continuously recirculated through treatments containing concrete and other aggregate materials. Before (time=0) and pH is provided at 1 minute intervals for the first 5 minutes, and then at five minute intervals until 60 minutes has elapsed. A dotted red line indicate a pH of 6, which indicates an approximate 80th percentile of pH values from natural THPSS (Carroll et al. 2020; Purdy, 2019; Belmer et al. 2015).

The pH of soil and wetland water can have far-reaching effects due its influence on bioavailability of major and minor nutrients. In many ways pH can act as a gateway. It has been hypothesised that one of the major human modifications, via urban development, is the increased pH of urban THPSS. See Figure 23 which shows how pH can modify the bioavailability of several major and minor nutrients. Carroll et al. (2020) has hypothesised that increased pH of THPSS has influenced bioavailability of major nutrients (Figure 23, 24), to the favour of invasive weed species in urban THPSS.

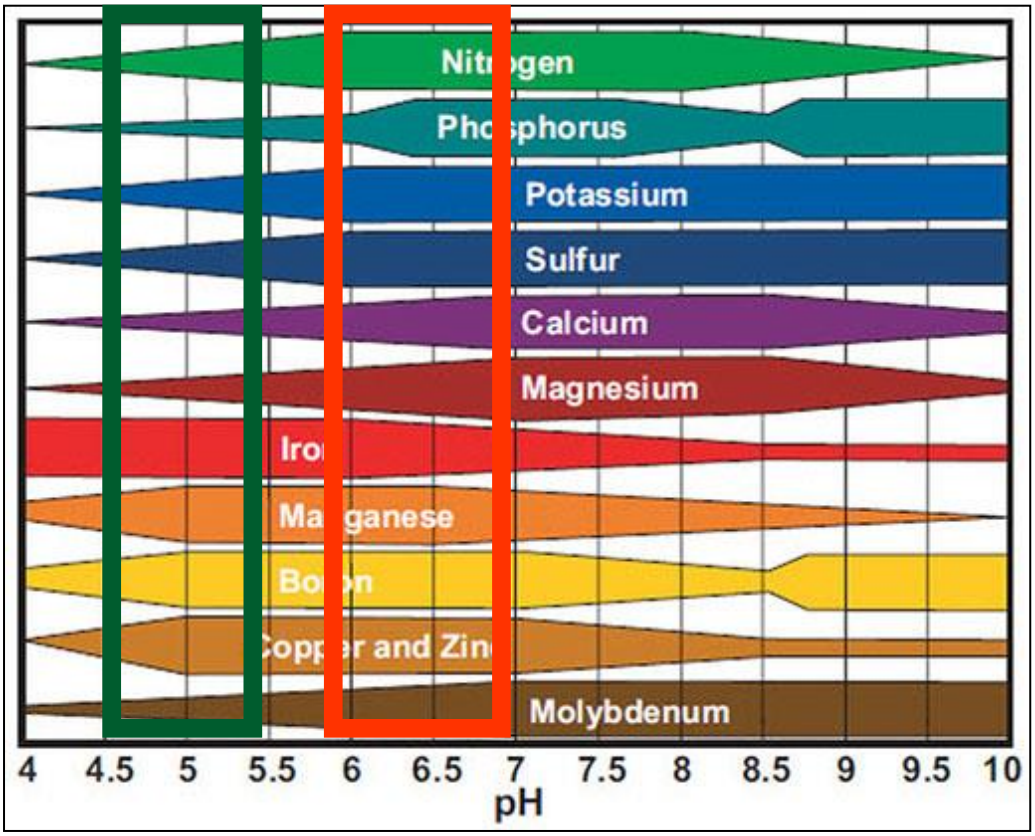

Figure 23, Bioavailability of elements by plants at different pH values. The green lines indicates the range of pH recorded at natural Blue Mountains THPSS and the red lines indicates the range of pH recorded at urban THPSS (Carroll et al. 2020). The base graphic is from 'The growing season 2013'.

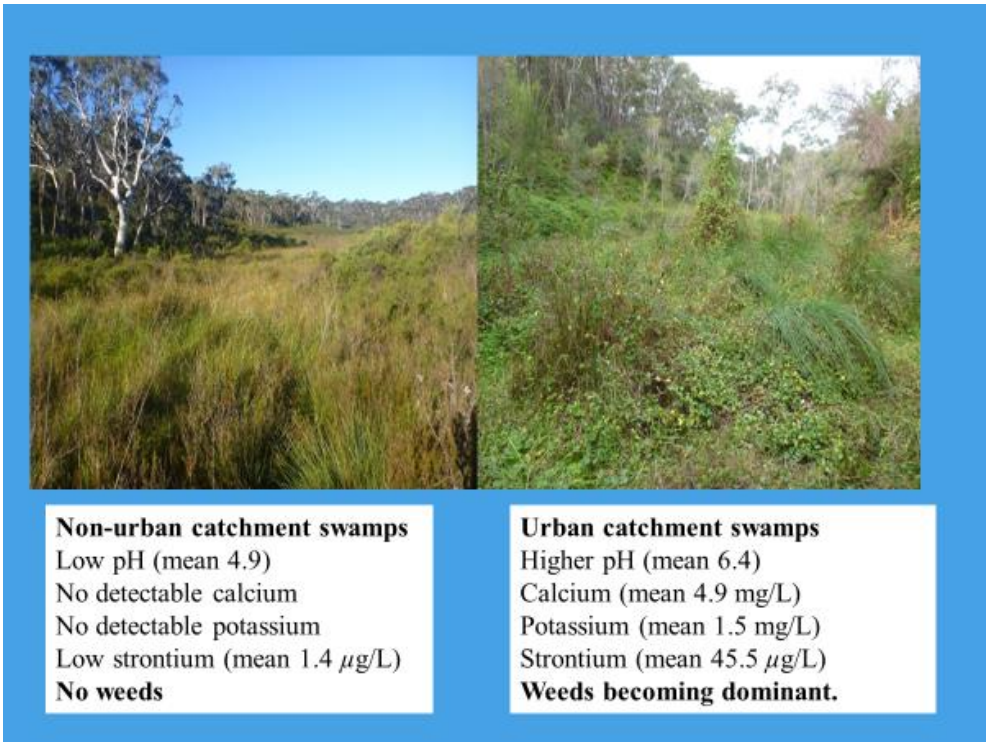

Figure 24, Summary of key geochemical differences in urban and natural swamps and the relative abundance of invasive weeds in urban and non-urban Blue Mountains THPSS (Carroll et al. 2020).

Salinity

The salinity in water from Mt Hay that was used in the recirculation experiment was dilute, with a electrical conductivity of just under 18 µS/cm. This was less saline than the mean salinity revealed in earlier survey of four natural BMUS (mean salinity = 28.5 µS/cm; Carroll et al. 2020). The Mt Hay water was collected in clean and acid-washed 5-litre bottles, filled with zero head space on the same day of the recirculation experiments.

The 60-minute recirculation experiments increased the salinity in all treatments, including within the procedural control treatment, a PVC pipe that contained no aggregate materials. The water in the PVC control increased salinity by 2.7 µS/cm, at the end of the 60-minute recirculation (Figure 25). The second smallest increase in salinity resulted from the recirculation through 10 mm blue metal aggregates which doubled the initial salinity to 43.6 µS/cm, after 60 minutes (Figure 25).

The largest change in salinity was recorded for water recirculated through the RMS RCA treatment. It resulted in a 50-fold increase in salinity to 916 µS/cm, after 60 minutes (Figure 25). The second largest increase in salinity of 415 µS/cm was recorded after 60 minutes recirculation through polymer fragments.

All salinity readings are well above the guideline value of 55 µS/cm. Contact of water with concrete materials causes a rapid increase in salinity (Figure 25) which helps explain why Belmer et al. (2015) record such highly elevated salinity levels in a THPSS wetland that was receiving stormwater runoff from a new concrete stormwater system in 2014/15,

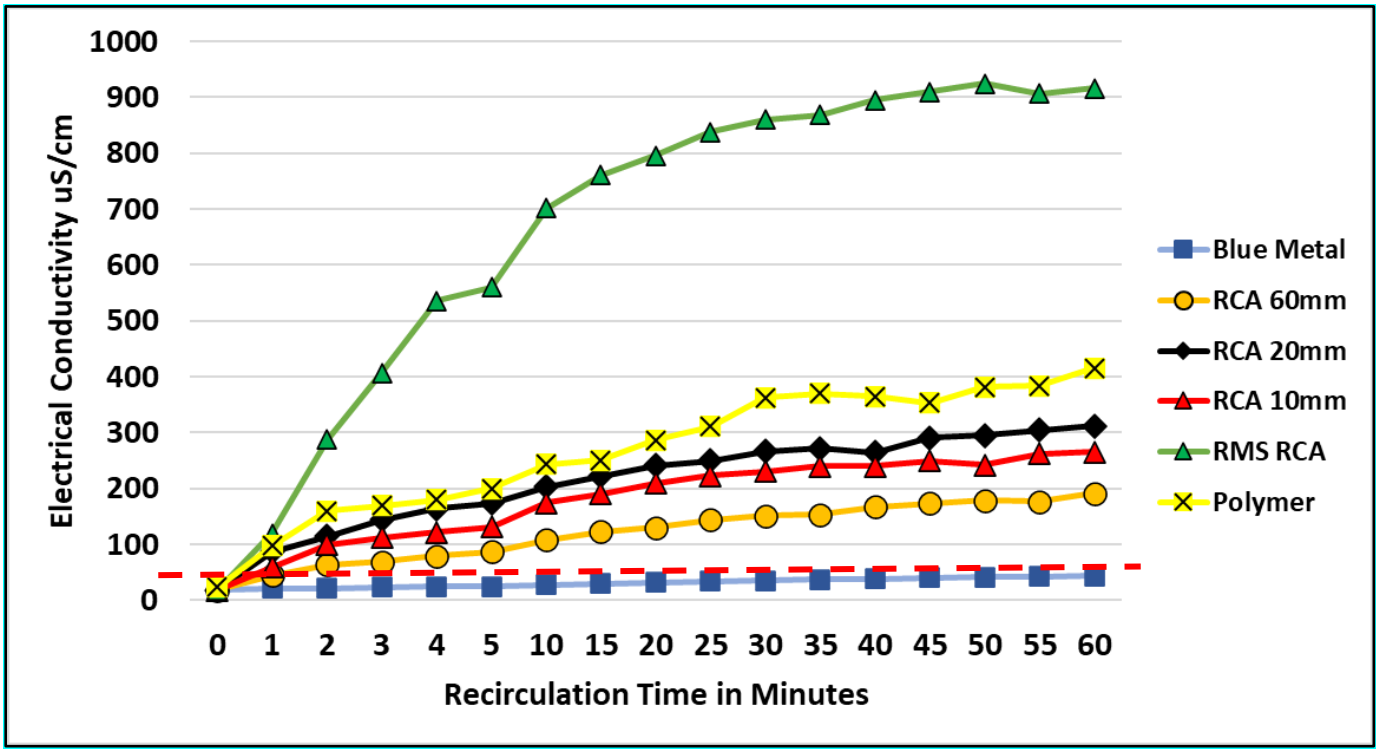

Figure 25. Time series graph showing the salinity (as electrical conductivity 'EC') as water is continuously recirculated through treatments containing concrete and other aggregate materials. Before (time=0) and EC is provided at 1 minute intervals for the first 5 minutes, and then at five minute intervals until 60 minutes has elapsed. A dotted red line indicate an EC guidelines of 55 µS/cm..

Major ions (minerals)

The ionic composition of water changed after being recirculated through the different treatments. The scale of the modification was largest for the four RCA aggregates.

Cations (sodium, potassium, sodium and magnesium).

The Mt Hay water, that was used as the reference water for the experiment, only had sodium cations detectable (mean 3 mg/L). The concentration of sodium was greater in water after it had been recirculated through blue metal and all RCA aggregates (Figure 26).

Calcium was not found at detectable concentrations in Mt Hay reference water, but it was the dominant cation in water recirculated through blue metal and all RCA treatments (Figure 26). Potassium was only found in water recirculated through aggregates (blue metal and RCAs).

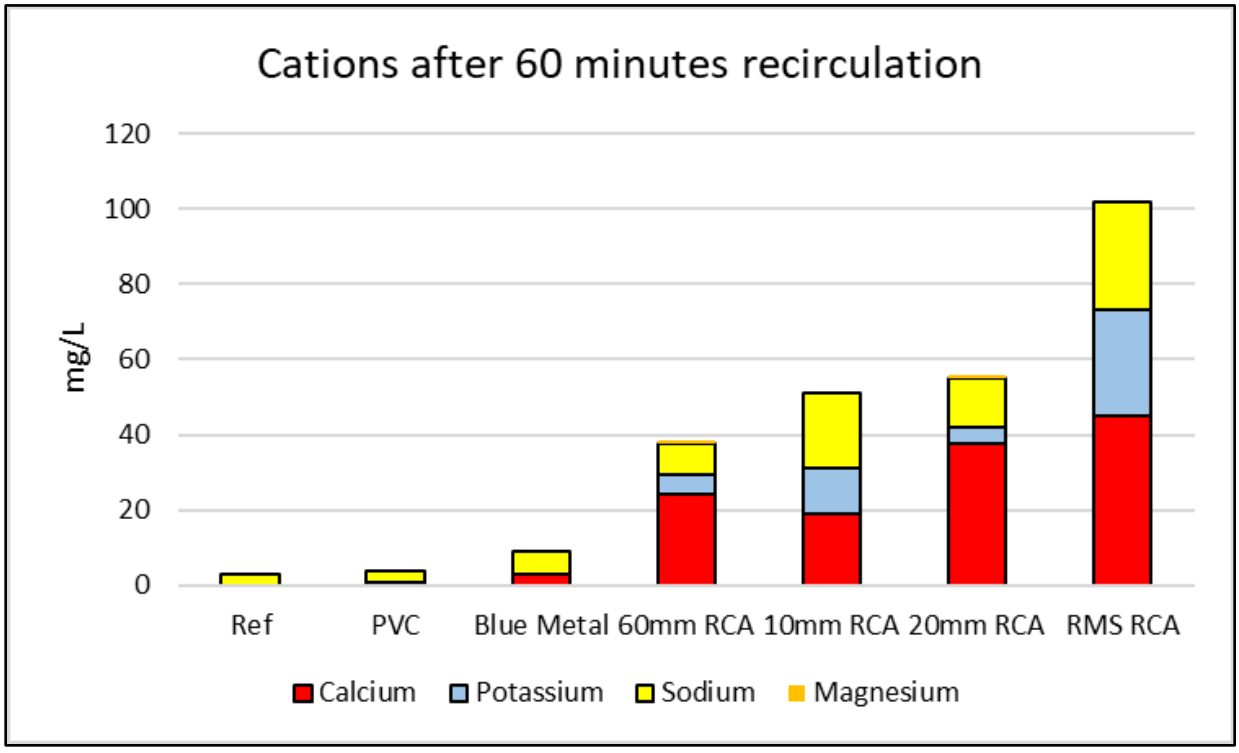

Figure 26. Stacked graph showing mean cation concentration before (Ref) versus after 60 minutes recirculation through different treatments.

Anions (sodium, potassium, sodium and magnesium).

The Mt Hay water, that was used as the reference water for the experiment, only had bicarbonate and chloride anions detectable.

The concentration of chloride was found to increase after the recirculation through all treatments (Figure 27). It was greater in water recirculated through blue metal and all RCA aggregates.

The bicarbonate concentration was higher than Mt Hay in all treatments, with the exception of RMS RCA, where it was not detected (Figure 27). The RMS RCA treatment had hydroxide and carbonate alkalinity, probable due to the very high pH affecting the solubility of bicarbonate.

Sulfate was only found in water recirculated through RCA aggregates, with the largest concentration in the 20mm RCA treatment (Figure 27).

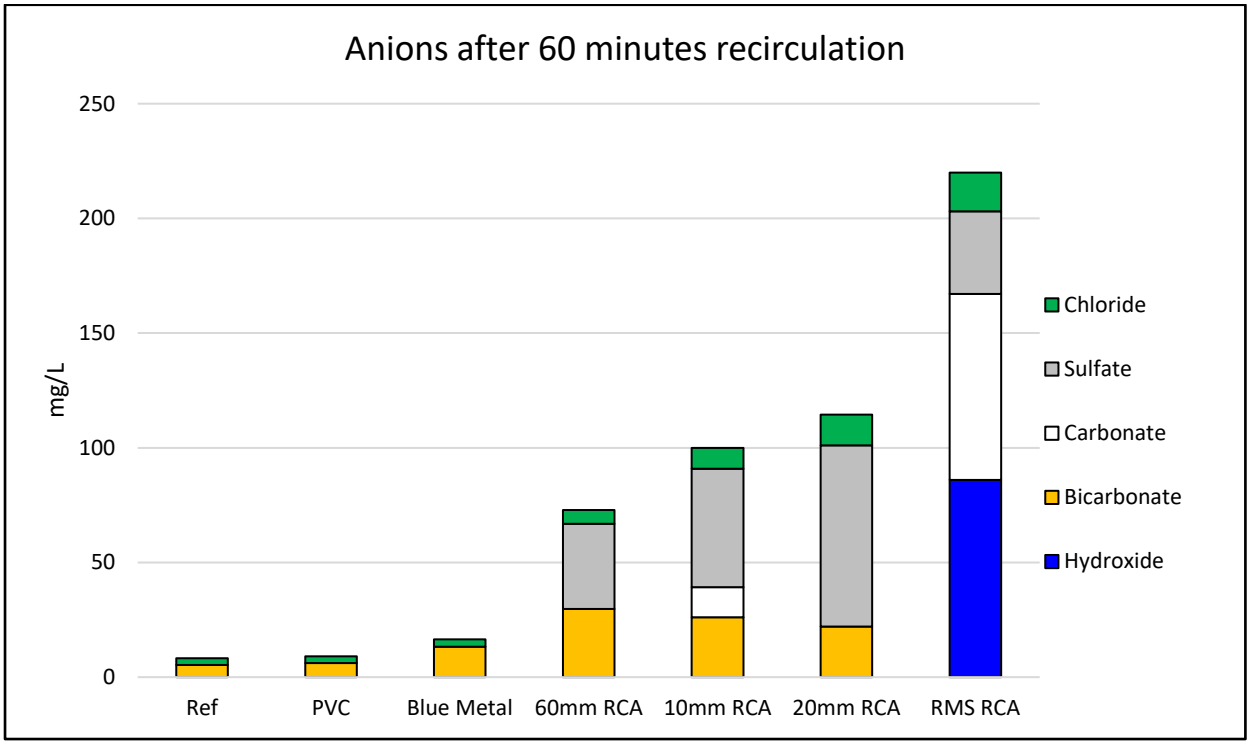

Figure 27. Stacked graph showing mean anion concentration before (Ref) versus after 60 minutes recirculation through different treatments.

Conclusion

This study demonstrates how concrete materials, particularly in the form of RCA aggregates, that become exposed to water can rapidly modify water chemistry. This includes the dissolution of contaminants into water at concentrations that could be ecologically hazardous to aquatic and terrestrial species. This investigation builds upon previous studies that revealed how water in contact with concrete materials can trigger rapid and substantial increases in water pH, EC, calcium, potassium and aluminium (Davies *et al.* 2011; Grella *et al.* 2014; Wright *et al.* 2018; Purdy et al. 2020). The current study showed that water recirculated through concrete aggregate materials develop a range of modified ionic composition with elevated concentrations of calcium, potassium, bicarbonate, carbonate and sulfate. The results of this investigation add further supporting information to the hypothesis that exposure of water to concrete materials can contribute to the water chemistry differences between rivers and streams flowing in urban versus undisturbed non-urban catchments (Wright *et al.* 2011; Connor *et al.* 2014; Tippler *et al.* 2014).

Ecologically hazardous concentrations of aluminium were detected in water recirculated through both concrete and blue metal aggregates. After recirculation through aggregates the aluminium concentrations ranged from 380 µg/L (20mm RCA) to 2366 µg/L (60mm RCA). Similar results were also achieved in an earlier water pollution study of concrete aggregates (Purdy et al. 2020). Post-recirculation solution in each of the four aggregate treatments had pH ranging from 7.54 (blue metal) to 9.77 (10mm RCA). The combination of elevated aluminium with alkaline pH contributed to failure of ecological guidelines. Guidelines for aluminium for protection of 99% of species: <27 µg/L if pH is >6.5; ANZECC, 2000). In the current study all of the water exposed to aggregate treatments failed to comply with aluminium guideline (<27 µg/L) by 14 times to 87 times.

Lead was also measured at ecologically hazardous concentrations, after 60-minutes of recirculation, through concrete aggregate treatments. Lead was undetected (<1 µg/L) at the outset of the experiment, which is consistent with a previous study of BMUS (Carroll et al. 2020). After recirculation through concrete aggregates the mean lead concentrations were 3.3 µg/L (20 mm RCA), 11.1 µg/L (60 mm RCA) and 12 µg/L (10 mm RCA). These lead concentrations exceeded Australian guidelines for protecting aquatic ecosystems (ANZECC 2000). These guidelines have a lead trigger value of 1.0µg/L for a protection of 99% level of protection. Similar lead results were also reported from an earlier pollution study of concrete aggregates (Purdy et al. 2020).

The dissolution of metals into solution through recirculation through aggregates in this study is influenced by water pH. This finding supports Engelsen *et al.* (2010) who reported that several metals are leached from crushed concrete (< 1mm size) in water due to strong pH dependence for several elements (Cr, Cu, Mn, Mo, Pb, V and Zn).

The current study confirms the observations by Wu *et al.* (2016) that RCA composition differs markedly from batch to batch. Our assessment of commercial RCA mixtures revealed that many contained non-concrete materials. These foreign materials included ceramic, brick, plastics, wood and wire. It seems likely that the observed differences water chemical attributes (such as pH, EC, major ions and metals) between the three size grades of RCA aggregates could be influenced by variations in RCA batches.

It is likely that the leaching of metals in this investigation was influenced by fly-ash in the concrete materials used in this study. Concrete products in Australian use fly-ash as an ingredient (ADDA 2009). In Australia, it is common that concrete contains up to 5% fly-ash (ADDA 2009). It is likely that RCA with lesser or

negligible fly-ash in the original concrete formulation would probably result in lower metal concentrations.

Additionally, the water chemical changes are also influenced by the surface area water/concrete particles that the smaller concrete fragments (such as 10mm) compared to the larger fragments (20mm and 60 mm). We support recommendation of Wu *et al.* (2016), Rahman *et al.* (2014) and Purdy et al. 2020 that further testing of multiple RCA products and batches to investigate potential environmental contamination risks associated with the exposure of RCA to water.

Given the potentially unforeseen water contaminations risks associated with RCA in this study, stricter guidelines may need to be developed by local, regional and national councils and government bodies to regulate the use of RCA. Of particular concern is its use as a drainage material and as a road base and fill material in waterlogged locations in or near waterways riparian zones and in proximity to sensitive ecosystems such as BMUS. The current study shows that concrete materials can strongly and rapidly modify water quality, including the triggering of the release contaminants into solution.

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