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Effects of field stockpiling on leaching calcareous constituents from recycled concrete aggregate to be used as unbound base course/subbase material

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Highlights

- Carbonation of RCA occurs because of aging in contact with atmospheric CO_2.
- Aging in the field is slow but it affects the leaching behaviour of Ca.
- Ca release becomes half of initial concentration within few months of aging.
- RCA should be aged in the field for at least six to twelve months.

Abstract

Understanding the potential of recycled concrete aggregate (RCA) to precipitate calcareous constituents requires investigating the changes in solid and liquid chemistry of material throughout its service life including the stockpiling period. This study focuses on stockpiling freshly produced RCA in the field and monitoring the changes in concentrations of calcium and sulfur (as dominant ions), as well as pH, total dissolved solids, and carbonate content for about two years. The results showed that the carbonate content of solid particles increases with time, which causes a noticeable reduction in total dissolved solids in the leachate. Aging also slightly decreases the pH; however, this decrease is very slow. The leached concentration of calcium showed a noticeable reduction with aging, indicating the effect of the formation of secondary carbonated minerals with lower solubility than calcium hydroxide and calcium-silicate-hydrate. On the contrary, sulfur (S) release (which is leached as sulfate ion) showed little effect of aging and carbonation process on the long-term leaching of S. The results obtained from the aging of the field RCA pile created in this study were also compared with the previously published laboratory aging of RCA and the comparison provided evidence supporting the field observations.

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1. Introduction

In this day and age where sustainable practices are acknowledged and appreciated by the general public, recycled concrete aggregate (RCA) is becoming a commonly considered aggregate source for constructing transportation infrastructure including roads, highways, and airport runways in the U.S. [1-5]. However, concerns regarding the potential of RCA to precipitate calcareous constituents (also known as tufa) have caused implementation restrictions in several States when the construction includes an underground drainage system with geotextile filter fabric [6-7]. The precipitation and accumulation of calcareous constituent may lead to chemical clogging of the geotextile used in subdrains as opposed to physical clogging which is propagation and deposition of fine particles within geotextile fibers due to seepage force [8-9]. Precipitation of calcium carbonate from the RCA leachate has been previously reported from field and laboratory observations [10-14]. However, previous field studies present conflicting results when it comes to severity. In some cases, abundances of calcareous constituents were observed on the geotextile surface, and in other cases very little was observed as pointed out by Abbaspour and Tanyu [15]. Therefore, understanding the leaching behavior of different elements from RCA under different conditions carries the utmost importance.

Different aspects of leaching behavior (e.g. pH-dependent release of elements) were studied by various researchers in the past [16-19]. Several field test sections were also constructed with RCA and were monitored under service for more than several years (e.g. in Ohio [11], Oslow [20], and Wisconsin [3]). However, leaching of the calcareous constituents (i.e. calcium carbonate and calcium sulfate)

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from field sites have not been properly studied as these constituents are not hazardous and do not pose any environmental threat. The concern still remains that excessive release of such components can lead to precipitation of tufa and undermining the serviceability of the subdrains and consequently the whole infrastructure.

Additionally, the potential of RCA to produce calcareous constituents over time should be evaluated starting from the time RCA is produced (and stockpiled) not after it is placed in the ground during construction. This is because most often, RCA is not placed in the ground right away. For example, in the U.S., it is not uncommon for the RCA to be stored in a stockpile after production for about a year (and in some cases even more than a year) before it is used in the construction. It is known that one of the hydrated cement paste (HCP) degrading processes in the hardened concrete in contact with atmospheric carbon dioxide is carbonation [21]. The carbonation process leads to the formation of an outer layer of carbonated material, which has a lower pH than the uncarbonated HCP [21].

In the past studies, some of the observed variations in RCA leaching behavior such as a decrease in pH of the leachate, was attributed to carbonation [20, 22]. However, these studies neither investigated the effect of carbonation on leaching behavior of RCA with time nor did they provide measurements on changes in carbonate content of RCA. This is most likely because either the objectives of those studies were not necessarily to investigate the carbonation effect or RCA was not being tested for aging reflecting actual field conditions. In this regard, Abbaspour et al. [1, 23] conducted a one-year study in which a batch of RCA material was aged by the mean of spreading RCA over a flat surface in a room with constant temperature and humidity (18°C and 80%, respectively) and conducting cycles of wetting and drying by using synthetic rainwater produced based on the average ionic concentrations that are measured in the rainwater collected in Washington D.C. metropolitan area. It was shown that although aging has little effect on the physical properties of RCA, the carbonation reduces the leached concentrations of various elements such as aluminum (Al), calcium (Ca), chloride (Cl), potassium (K), sulfur (S leached as sulfate) and copper (Cu) and the released concentrations approach to a steady state in terms of changes in concentrations. The aging also reduces the pH of the leachate (which represents the pH of the pore water) from values in the order of 11 to 10 reducing the potential to leach out calcareous constituents with time.

The observations in Abbaspour et al. [1, 23] studies pertain to a relatively small quantity of RCA material aged in laboratory conditions that were constant and adjusted to favor the progress of the carbonation process and aging. However, the conditions in an actual RCA stockpiled in the field can be affected by many variables that are not simulated in the laboratory such as but not limited to variation in temperature, humidity, rain events, the occurrence of freeze/thaw cycles, etc. The quantity of the material stockpiled in the field can also have an impact as only the material on the surface of a large stockpile is exposed to sunlight, atmospheric carbon dioxide (CO_2), and excessive variation of temperature. Considering all these variables, it is necessary to evaluate the effect of field stockpiling on RCA leaching characteristics before the material is placed as an unbound base/subbase layer.

To understand such an effect, in this study an RCA pile was created in the field and monitored for about two years. Changes in total leached concentrations of the dominant elements known to contribute to RCA tufa precipitation (Ca and S) [15, 24-25], pH, total dissolved solids (TDS), and carbonate content of solid samples were monitored over time. The obtained data from this study is then compared with the findings of laboratory aging of RCA under a controlled environment [23].

2. Material

A small stockpile with an approximate volume of 15 m^3 was created from a freshly produced RCA in a facility located in northern Virginia, U.S.A. This facility produces RCA regularly (at least once a year) from demolished concrete, which is obtained from various sources such as buildings, curbs, and gutters, pipes, and washouts. In addition to the variation in the source, there is an inherent inhomogeneity with regard to mortar content (MC) of RCA particles as the MC varies for different particle sizes and even for particles with a similar diameter. Considering the inhomogeneity of the concrete source and MC, a detailed random sampling procedure was followed when collecting RCA samples and creating an RCA stockpile for this study. This approach was also followed to avoid having biased data as a result of the concrete source on its chemical and physical properties. From the freshly produced RCA (about one week after production), randomly determined 7 different locations (per ASTM standard D3665 [26]) were identified to gather RCA. At each location, segregated material from the surface were removed and the remaining material were mixed with each other. The combined pile was set aside as RCA stockpile and left open to atmospheric conditions for aging. RCA samples for laboratory evaluation were also collected following ASTM standard C702 [27].

The physical and index properties of collected RCA can be seen in Table 1 The material classified as silty and well-graded sand (SW-SM) based on a unified soil classification system. The parent material of RCA was obtained after acid treatment and examined for chemical composition. Geologic examination identified the parent rock as predominantly diabase with traces of granite, quartzite, and hornfels, which are expected based on the geology of the rocks in northern Virginia. Table 2 summarizes the measured total elements in RCA and parent material.

When MC of the RCA is evaluated, it is known that calcium silica hydrate (C-S-H) and calcium hydroxide (CH) (which take up to 70-80% of the HCP in the MC of RCA) are by-products of the hydration of alite (C_3S) and belite (C_2S) in clinker [28]. Considering that RCA is produced from crushed concrete, it is not surprising that cement minerals are present during hydration. The remaining of the HCP consists of calcium aluminates as well as AFt/AFm (tri- and mono-sulfoaluminate) phases [29-30].

Table 1. Index properties of collected RCA material

Property	Average Value
Specific Gravity for Coarse Aggregate (per ASTM C127)	
Apparent Specific Gravity	2.79
Water absorption (%)	5.09
Specific Gravity for Fine Aggregate (per ASTM C128))	
Apparent Specific Gravity	2.75
Water absorption (%)	13.06
Grain size distribution (per ASTM D422)	
Percent Passing 3/4" (< 19mm)	93.5
Percent Passing 3/8" (< 9.5mm)	68.9
Percent Passing #4 sieve (< 4.75mm)	55.8
Percent Passing #10 sieve (< 2mm)	37.7
Percent Passing #40 sieve (< 0.43mm)	14.9
Percent Passing #200 sieve (< 0.074mm)	6.2
Atterberg Limits (per ASTM D4318)	
Liquid Limit (%)	28
Plastic Limit (%)	NP
Plastic Index	NP
Mortar Content (%)	36.07

Notes:

- Mortar content of RCA was measured using acid treatment procedure per Akbarnezhad et al. [31]
- NP = Non plastic.

Table 2. Measured total elements of RCA and parent material

Element (> 0.1% by weight)	RCA	Parent Material
Iron	4.32	7.75
Calcium	6.92	6.95
Magnesium	2.31	5.7
Aluminium	5.54	7
Sodium	1.31	1.29
Potassium	1.03	0.43

3. Methods

3.1. Field aging of RCA

The aging process in the field occurred naturally as the RCA stockpile was left uncovered and exposed to natural conditions fluctuating based on seasonal changes such as (but not limited to) precipitation, freeze/thaw, and sunlight. The meteorological data for the area where RCA was piled was obtained from a nearby weather station (Station name: CENTREweather, Station Code: KVACENTR1, Coordination: 38.845 -77.455) as reported by [32].

Figure 1 shows the monthly average for recorded precipitation, temperature, and relative humidity during the duration of the stockpiling for this study. Also, the analyzed average annual chemical composition of rainwater in northern Virginia and the District of Columbia (D.C.) area can be seen in Table 3 (data obtained from NADP [33]). Samples from the RCA stockpile were collected every month (about every 30-40 days). In order to avoid the segregated material on the surface of the RCA pile, all samples were collected from at least 30 cm below the surface and then mixed and reduced to the desired sample size. These samples were then used to obtain moisture content, pH, TDS, leached concentrations of elements (via USGSLT method), and carbonate content (via per ASTM standard D4373 [34]).

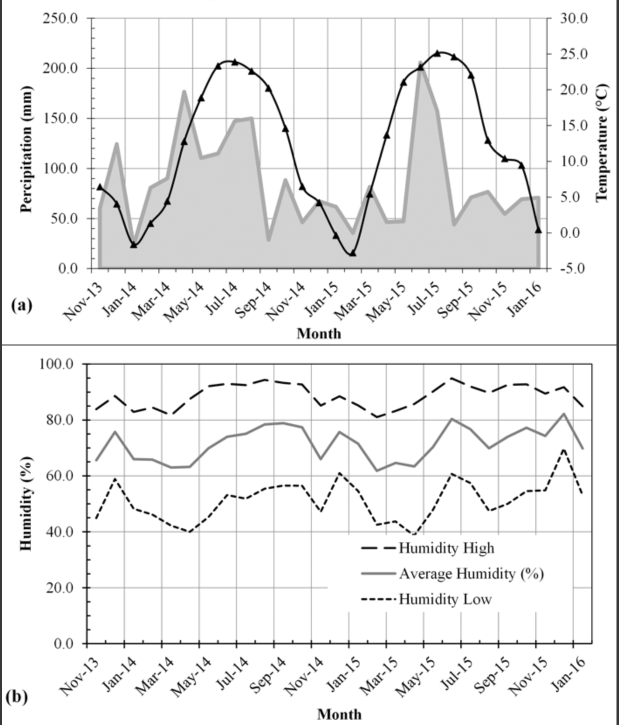

Figure 1. Meteorological data of the area nearby the RCA pile. a) total monthly precipitation and monthly average temperature vs. time, and b) measured relative humidity vs. time (Data after Tanyu and Abbaspour [35]).

3.2. U.S. geological survey leach test (USGSLT)

A quick leachate extraction method described by Hageman [36] is used in order to obtain duplicate leachate samples in the field as this method provides an easy way to obtain liquid samples without the need for specific laboratory equipment or space. In order to have a better representative sample, first collected RCA samples were sieved through a 19 mm sieve and then reduced to 50 g as required by the test method. Oversize particles (larger than 19 mm) had to be discarded because each larger particle had a mass range of 15 to 35 g and when included took up most of the 50 g sample set aside for testing. This condition would have caused biased and unrepresentative samples. The discarded larger particles made up only 6.5% of the whole RCA gradation. After samples were created, each 50 g solid sample was placed into a 1-liter plastic bottle, which then was filled with 1000 ± 0.5 ml DI water (Solid to liquid ratio of 1:20). After vigorously agitating the sample for 5 minutes and letting it sit for 10 minutes, 50 ml of leachate was filtered through 0.45µm nylon filters using plastic syringes and acidified to pH<2 using trace metal concentration of HNO_3 (32%) and stored at 4ºC for chemical analysis. The rest of the unfiltered sample was used for determining the pH and TDS. The average value of duplicate tests is reported.

3.3. Chemical analyses

pH and TDS of the leachate solutions obtained from USGSLT were measured using pH and conductivity meter probes. Concentrations of dissolved metals (Ca and S) were measured using inductively coupled plasma mass spectrometry (ICP-MS).

Solid samples obtained from RCA were also used to determine the elemental content of RCA and to monitor the change in carbonate content during the field aging process. The concentrations of elements in duplicate solid samples were measured using the ICP-MS method, where the samples were prepared by the acid digestion method. Carbonate content of RCA was measured by the treatment of triplicate solid particles smaller than 0.425 mm using hydrochloric acid in a sealed reaction cylinder and measuring the changes in the produced carbon dioxide gas pressure (per ASTM standard D4373 [34]). Reported values in this study are the average of duplicate and triplicate tests

Table 3. Annual average rainwater chemical composition of Northern Virginia and D.C. area

Ion	mg/L	Ion	mg/L
Na^+	0.1	Mg^{2+}	0.025
Ca^{2+}	0.1	Cl^-	0.2
K^+	0.03	NO_3^-	0.6-0.75
NH_4^+	0.2-0.25	SO_4^{2-}	1

Note: Average measured pH of rainwater = 4.8

4. Results and Discussions

4.1. Effect of field aging on carbonation degree

Figure 2 illustrates the changes in carbonate content of RCA during aging. The existing fluctuation in the recorded data can be attributed to the variation in the collected samples on different dates, which cannot be avoided due to the inherent inhomogeneity of the material. The range of fluctuations during the first year seems to be much higher than the range of fluctuations during the second year (about 67 mg/g calcite equivalent for the first year compared to about 38 mg/g calcite equivalent for the second year). One of the reasons for observing such behavior, in addition to the existing variation in the RCA stockpile, may be attributed to the existence of unevenly carbonated RCA within the pile. This variation appears to decrease with time as the carbonation process continues to convert decalcified C-S-H in the MC into calcite and other carbonated species. Overall, the carbonate content results show about a 16% increase in the carbonated solid mass with time (from about 110 mg/g calcite equivalent at the beginning of the aging to about 128 mg/g calcite equivalent at the end of aging). A linear regression of the data also confirms this overall behavior.

Figure 2 also presents the changes in carbonate content of RCA samples aged in the laboratory. Laboratory aging was conducted by performing cycles of wetting and drying during a period of one year under constant environmental condition (temperature and relative humidity) that was in favor of carbonation. Also, laboratory samples did not experience freezing and thawing as field samples do. Details of the laboratory aging are provided in Abbaspour et al. [1, 23]. A comparison of laboratory and field aging was possible in this study because both of the RCA samples were collected at the same time from the same location by the same researchers and the aging tests were started around the same time. The comparison shows that the carbonate content of RCA (depicted as calcite equivalent) aged both in the laboratory and field provide similar results; however, the carbonation rate in the laboratory was two times faster than the rate in the field (reaching very similar calcite equivalent values after one vs. two years in the laboratory vs. field respectively). The observed slower rate of carbonation in the field may be mainly attributed to the fluctuation in temperature, amount of precipitation, and relative humidity. In the laboratory, these conditions were kept constant at a level, which was favorable for the carbonation process [1, 23].

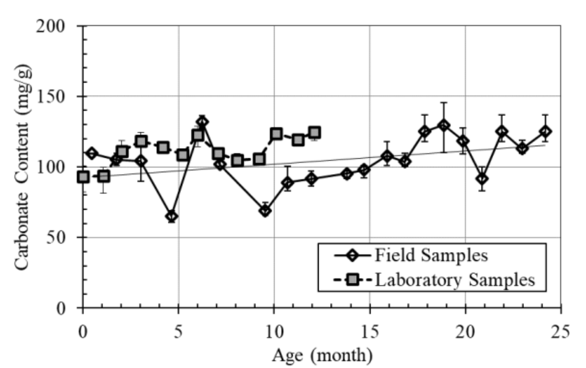

Figure 2. Changes in Carbonate Content of RCA Samples with Time (Data modified after Tanyu and Abbaspour [35])

4.2. Effects of stockpiling on chemical properties of leachate

Changes in measured pH and TDS of the leachate with the age of the material are depicted in Figure 3. Figure 3a shows the changes in pH values in the field with time. Since the process of extraction is done in closed tubes and the solution was not exposed to atmospheric carbon dioxide, the measured pH values are representative of the changes of the pH of the pore water of material. The data from the field shows fluctuations, which similar to carbonate content is attributed to the variation in the collected samples on different dates and inherent inhomogeneity of the material. As can be seen, despite the existing fluctuation in the recorded pH values throughout the aging, the overall pH values do not show noticeable changes for field aged material. A linear regression line shows a slight decrease in the pH values during two years of aging. However, in the laboratory, the recorded pH values with time decrease much more noticeable compared to the pH values from the field even though the aging duration in the field was twice the laboratory. The existing difference between the field and laboratory results supports that the RCA in the laboratory aged more than the RCA in the field despite the shorter aging duration.

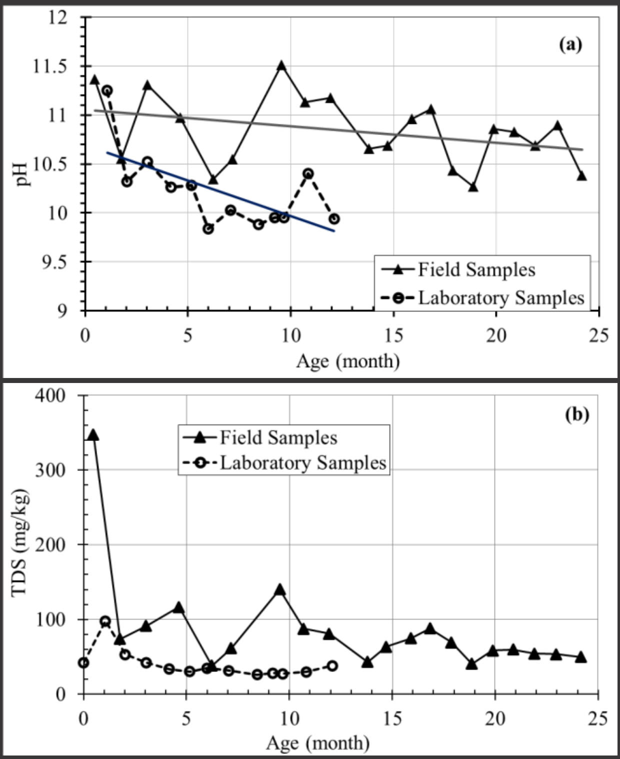

Figure 3. Changes in Leachate Chemistry with Aging Time (a) pH and (b) Electric Conductivity (Data modified after Tanyu and Abbaspour [36]).

Roy [21] showed that there is an inverse relationship between the carbonation degree of HCP and the pH of the pore solution. Despite the fact that the measured carbonate content of particles finer than 0.425 mm for the field samples reaches the same level as the laboratory samples after two years of aging, the measured pH values from field RCA clearly shows that the carbonation degree of RCA in the field is not same as the laboratory samples. Based on the linear regression of the recorded pH values of leachate from field samples, it can be predicted that the field RCA will reach the same carbonation level as the laboratory aged RCA within 3.5 years.

When the results from Figure 3b were evaluated, it is seen that the TDS of the solution decreases drastically during the first month of aging from about 350 mg/kg at the beginning of aging and then stays constant at a level below 150 mg/kg in the upcoming months. This observed trend is also consistent in the laboratory. The main reason for observing such a trend in TDS is believed to be that, at the beginning of the aging, the uncarbonated C-S-H and CH become exposed due to crushing procedure, which releases a higher concentration of constituents as they have higher solubility compare to the carbonated calcium species (calcite, aragonite, magnesite, and dolomite) [37].

4.3. Leaching of elements contributing to tufa

Changes in total leached concentrations (TLC) of Ca and Mg are presented in Figure 4. The TLCs of Ca and Mg were determined every 4 months during the first year of aging and once in a month after that. The reason for a low number of tests during the first year is because, at the time, it was considered sufficient resolution of data in comparison to the long-term period of the aging program. However, as drastic changes were observed from test results, it was decided to monitor the TLC of constituents on monthly basis.

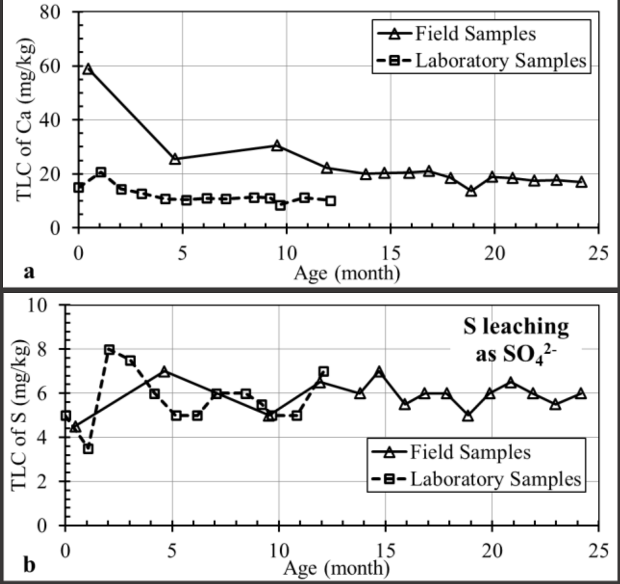

Figure 4. Changes in the leached concentration of calcium (Ca) and sulfur (S leached as sulfate ion SO_4^{2-}) with aging.

As illustrated in Figure 4a, the release of Ca in the field is at its peak at the beginning of the aging and then reduces noticeably with aging but the rate of this reduction in released Ca decreases with time and approaches a value of about 20 mg/kg by the end of the second year. This behavior is in agreement with the observed behavior for the measured TDS. One explanation for this kind of behavior may be based on the carbonation model developed by Suzuki et al. [38]. In this model, the carbonation of synthetic C-S-H is explained in four stages. The first stage of this model is the formation of C-S-H, calcite, and modified silica gel. The second stage includes the formation of excess calcite from residual calcium ions with a very high reaction rate. In the third stage, C-S-H starts to decalcify and releases calcium, which forms more calcite and silicate ions, and finally, in the fourth stage, the modified silica gel completely changes into pure silica gel and C-S-H disappears. In the liquid phase (pore water), calcium ion decreases continuously during all stages but more rapidly during the second stage as the residual Ca^{2+} ions react as part of carbonation. The behavior observed in this study is in fair agreement with the model provided by Suzuki et al. [38]. However, one should always keep in mind that this model was developed for synthetic C-S-H with no impurities whereas the existence of other chemical compounds in RCA potentially can change any of the aforementioned stages. Another explanation for this kind of behavior may be that exposed CH and C-S-H transform with time into calcite and other carbonates as part of carbonation. The solubility of calcite is lower than CH and C-S-H [37-38]; therefore, the leached concertation of Ca reduces with time and approaches to a steady value, which probably indicates that the solubility of secondary mineral (in this case calcite) is controlling the release of Ca as suggested by Kitamura et al. [39].

When the field data is evaluated for S, as can be seen from Figure 4b, the leaching of S is not much affected by the aging and carbonation process. The source of sulfate in cement paste is mainly due to the addition of gypsum to the raw clinker meal during cement production. The gypsum transforms into ettringite during the setting phase of cement paste hydration; however, ettringite disintegrates back into gypsum with the progress of the hydration process [21, 30]. The gypsum content of the cement paste remains unchanged during the hardening and service life of the concrete for the most part unless concrete was exposed to sulfate attack during its service life [21, 24].

A comparison between TLC of Ca from the field and laboratory shows that Ca release from field samples is much higher than the laboratory RCA throughout the whole aging process, whereas for S, both the laboratory and field RCA samples have comparable released concentrations. The observed behavior of Ca release is another evidence that the field RCA are much less carbonated throughout the whole aging process.

5. Conclusions

The purpose of this study was to investigate the effects of stockpiling of RCA on leaching calcareous constituents. Understanding these effects was of interest because RCA is becoming a popular recycled aggregate to be used in construction and transportation industry in the U.S. and parts of the world. Understanding the formation of calcareous precipitates provides insight to further evaluate the suitability of using RCA up against underdrain systems consisting of geotextile filtration.

An RCA pile was created in the field and monitored for about two years to simulate the stockpiling in practice. Changes in total leached concentrations of Ca and S (leached as SO_4^{2-}) as well as leachate properties such as pH and TDS with aging were measured. Moreover, changes in carbonate content of solid particles were monitored over time to evaluate and quantify the carbonation process and its degree within the aged RCA. The obtained data from this study are compared with findings of a previous study, in which, RCA material was aged in the laboratory under controlled conditions. Below summarize the findings from this study:

1. Carbonation of RCA occurs as a result of aging when RCA is in contact with atmospheric CO_2 and pore moisture. Carbonation is an ongoing degradation process for HCP and RCA, which yields the production and increase of carbonate species in the RCA with time.
2. The pH of the leachate from RCA decreases due to aging and carbonation. The measured pH values, which are representative of the changes in the pH of the pore water of material, stays within the alkaline range throughout the aging process (always pH>9). Based on the chemical composition of HCP and laboratory results, it is expected that the leachate of RCA remains above 9 for the whole service life of RCA.
3. Although carbonation in the field is a slow process and its rate might be affected by the meteorological changes, aging (and carbonation particularly) affects the leaching behavior of Ca (but not S).
4. Ca release decreases drastically within a few months of aging and becomes half of the initial concentration as secondary mineral formation (carbonates with lower solubility) limits the Ca release.
5. S (as SO_4^{2-}) release does not change during the aging as it is controlled by the dissolution of gypsum in HCP, which remains unaffected by the carbonation process.

Based on these findings, it is suggested that if the release of calcium based compounds from RCA is a concern for a given project, it is best to age RCA in the field (create a stockpile and keep it exposed) for six to twelve months before using this material. Longer stockpiling periods could be considered but based on the results observed in this study, most likely there will not be much of benefit in terms of further reducing the release of calcareous constituents appreciably.

Declaration of Interest Statement

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Author Contribution Statement

A. Abbaspour: Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Visualization, Draft writing, Writing – **B. F. Tanyu**: Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Visualization, Draft writing, Writing

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