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Cover photo: Stormwater collecting at the base of recycled concrete stockpiles. Photo taken at an unannounced inspection of Girard Resources & Recycling LLC in Kent, Washington by Department of Ecology.
Photo taken by Tamara Welty in 2017.

Recycled Concrete Aggregate Leachate

A Literature Review

by
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Table of Contents

Page

List of Figures

List of Tables

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Executive Summary

Concrete is a hydrated, heterogeneous composite of cementitious paste[1] and aggregates (sand, gravel, and/or crushed stone). The aggregates in concrete are generally considered fillers or inert material. Concrete is an important material to the construction industry, because it is versatile, durable, and inexpensive for global usage in diverse formats.

Modern day concrete structures generally last no longer than 50 to 100 years. A survey of demolition waste from 19 non-residential projects in the Pacific Northwest revealed an average of 66% by weight of concrete material (EPA, 1998)[2]. According to the Washington State Department of Ecology (Ecology) Solid Waste Management Program as well as the U.S. Geological Survey, Washington State collected 1.2 million tons of concrete rubble and sold or used 4.2 million metric tons of concrete aggregates[3] in 2017 (USGS, 2020a).

Recycled concrete aggregate (RCA) is defined by United States Department of Transportation Federal Highway Administration as a granular old portland cement concrete that has been removed from service and purposefully crushed to produce concrete aggregates for re-use (RCW 70A.205.700[4]; FHWA, 2004). In 2009, the Washington State Department of Transportation (WSDOT) recommended the consideration of RCA as a substitute for natural aggregates in western Washington infrastructure projects (WSDOT, 2009).

Sites with stockpiles of pre-approved and evaluated sources of RCA are provided by the WSDOT at their Qualified Products List website[5]. The intent of the Qualified Products List is to aid, promote, and encourage the use of RCA by Washington State's construction industry. RCA material is recommended for use to reduce carbon dioxide emissions, preserve limited resources, provide economic benefits to the industry, and produce less waste.

The uses of RCA include landfill material, bound and unbound aggregates in road projects, incorporation into recycled aggregate concrete, as well as other uses. Historically, the use of RCA has been limited because of uncertainties and unfamiliarity in technical requirements, performance, durability, and environmental concerns of recycled aggregate concrete. Several recent studies (e.g., Bestgen et al. 2016b; Chen et al. 2019; Engelsen et al. 2017) have attempted to document mechanical characteristics and alleviate environmental concerns of recycled concrete materials.

This report is not a review of the concrete or concrete recycling industry, but rather a literature review of the physical and chemical characteristics of RCA and RCA leachate, as well as potential environmental concerns when the material is not properly managed. The sensible management of stockpiles, implementation of best management practices (BMPs), and use of

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[1] In this report, the phrases "adhered cement-paste", "cement-paste", and "cementitious-paste" will refer to a hardened, hydrated cementitious binder material around aggregates. The word "cement" refers to a hardened, hydrated cementitious material without aggregates. The phrase "cement clinker" refers to the finished product discharged from a kiln and combined with other materials; the "cement clinker" is dry and non-hydrated. All of these cementitious materials do not necessarily need to be composed of portland cement.

[2] The survey of 19 non-residential buildings demolished by R.W. Rhine, Inc., Tacoma, WA and reported to EPA, (1998) contained 167,200 tons of demolition waste. Residential buildings concrete percentage varied, but were lower than non-residential buildings.

[3] Concrete aggregates here is all concrete material including concrete sand.

[4] Washington State RCW 70A.205.700: Solid Waste Management – Reduction and Recycling: Develop and establish objectives and strategies for the re-use and recycling of construction aggregate and recycled concrete materials.

[5] The Washington State Department of Transportation Qualified Product List website of pre-approved and evaluated sources of recycled concrete aggregate is: https://wsdot.wa.gov/Business/MaterialsLab/QPL.htm.

treatment technologies may reduce the negative environmental impacts while retaining the net benefits of recycling concrete.

The Sand and Gravel General Permit

The Washington State 2021 *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* (Sand and Gravel General Permit) has 19 different sand and gravel activities, including the activity ECY002: Concrete Recycling. Ecology added activity ECY002 to the Sand and Gravel General Permit in 2016, prior to the ECY002 designation the concrete recycling activities were lumped in with another activity code.

The definition of activity code ECY002 in the 2021 Sand and Gravel General Permit is the processing of hardened concrete to produce a reusable concrete product. Facilities commonly stockpile the RCA outdoors, exposing the RCA to the elements. Water, following exposure to RCA, may runoff to and/or infiltrate into waters of the State[6].

For compliance to the Sand and Gravel General Permit, the discharge of stormwater and process/mine dewatering water to waters of the State must be between or below effluent limits for the designated sand and gravel activity[7]. Ecology manages the permits of 153 active concrete recycling facilities[8]. The dozen counties surrounding Puget Sound and Salish Sea contain over half of the facilities.

Concrete recycling facilities with a Sand and Gravel General Permit must comply with all monitoring, reporting, and requirements outlined in the permit. BMPs, construction and design control, soil neutralization capacity, and the dilution factor could enable a facility with concrete recycling identified as an activity[9] to discharge an effluent below the State and Federal water quality criteria[10] for the majority of pollutants (Engelsen et al. 2012; Snyder et al. 2018).

The Sand and Gravel General Permit requires a Site Management Plan to be developed by all permitted facilities. The Site Management Plan must contain a site map, erosion and sediment control plan, monitoring plan, stormwater pollution prevention plan, and spill control plan.

One of the intended purposes of this report is to identify the pollutants of environmental concern potentially leaching from RCA to better inform Ecology's Water Quality Program for the development of appropriate requirements for concrete recycling.

Carbonation and Leaching Processes

Carbonation of concrete is a complex exothermic, mainly diffusion-based, and environmental process, progressing from the exposed concrete surface inwards. The carbonation of a stockpile

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[6] The waters of the State include those waters as defined by "waters of the United States" in 40 CFR Subpart 122.2 within the geographic boundaries of Washington State and "waters of the State" as defined in Chapter 90.48 RCW. This includes groundwater, lakes, rivers, ponds, streams, wetlands, inland waters, salt waters and all other surface waters and watercourses within the jurisdiction of the State of Washington.

[7] See the effluent limits and monitoring requirements in the 2021 release of the Sand and Gravel General Permit. The requirements for pH, turbidity, total suspended solids, oil sheen, and total dissolved solids are provided in detail (effluent limits, sampling frequency, etc.).

[8] Represents the number of active facilities with concrete recycling included in their permit as of 2/19/2021.

[9] The word "facilities" and "facility" will now carry the meaning "actively permitted facilities with concrete recycling identified as a Sand and Gravel General Permit activity," located in Washington State.

[10] "State water quality criteria" refers to Washington State Chapter 90.48 RCW: Water Pollution Control, Chapter 173-201A WAC: Water Quality Standards for Surface Waters, Chapter 173-200 WAC Water Quality Standards for Groundwaters, and Chapter 173-204 WAC Sediment Management Standards of the State of Washington. "Federal water quality criteria" refers to 40 Code of Federal Regulation (CFR) Part 131 – Water quality standards; Available here.

of cement-based materials is affected by the geometry, size, and shape of the stockpile, partial pressure of carbon dioxide in the atmosphere, relative humidity, temperature, particle size, porosity, and quality of the cement-based material, among other factors (AzariJafari et al. 2021; Lagerblad, 2005). The carbonation process affects the physical properties (e.g., porosity, density) and chemical composition of the RCA (i.e. carbonated complexes) (Engelsen et al. 2005; Sanger et al. 2019).

The most notable effect of carbonation on RCA is the decrease of available, leachable calcium hydroxide from the RCA surface. This is a result of the release of calcium hydroxide and its transformation to calcium carbonate, which consequently reduces the leachate pH. Leachate is a solution obtained by exposing a solid to a liquid, resulting in the solid-liquid partitioning between the solid material and the aqueous phase. The leachate contains soluble, diffusible, and available pollutants of concern.

The release of chemical[11], inorganic, and organic pollutants of concern from the exposed material surface depends heavily on the leachate pH, electrical conductivity (or ion concentration), and concentration gradients between the solid and liquid phases (Clark et al. 2013; Kamali et al. 2008). In addition, the partitioning into the leachate from the material depends on the physical and chemical properties of the material being leached (e.g., porosity, particle size, degree of carbonation). The leachability of pollutants of concern from the solid does not only depend on the composition, but primarily on the solubility and availability of the minerals present on the exposed surface (Barbudo et al. 2012; Del Rey et al. 2015; Galvín et al. 2013).

Monitoring Challenges

The inherent physical and chemical heterogeneities of the RCA encountered at permitted facilities in Washington State restrict the assignment of a singular value to describe all concrete materials. In addition to other factors, the RCA physical properties depend on the concrete's original use, original water-to-cementitious material ratio, type of aggregate used, and recycling framework employed (e.g., crusher type, maximum diameter selected). Concrete may also incorporate several reactive/non-reactive filler materials (e.g., gravel, sand, natural Pozzolan ash[12] or other supplementary cementitious material) to occupy space and save cement.

The major inorganic constituents of cement-based materials are relatively consistent (e.g., calcium, silicon, aluminum, iron). Heavy and trace inorganic constituents (e.g., barium, arsenic, lead, selenium) in cement-based materials vary depending on the raw materials, cement manufacturing processes, and exposure to other materials during original- and post-use. Organic constituents potentially present in/on concrete include polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls, (surface-modified) carbon nanotubes, phenols, and concrete admixtures (e.g., set-accelerating or set-retarding admixtures, air-entraining admixtures, superplasticizers).

Laboratory leaching[13] methods potentially fail to incorporate critical physical and chemical parameters or processes encountered in the field that may render the laboratory leaching results useless or irrelevant. The expense and challenge of measuring, incorporating, and reproducing

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[11] Chemical parameters include, but are not limited to, pH, turbidity, total solids (suspended and dissolved), hardness, oxidation-reduction potential, and electrical conductivity.

[12] A natural siliceous (or siliceous-aluminous) material commonly added to cement clinker to consume excess lime (CaO) generating additional cement-phase minerals and increases strength, density, and durability.

[13] Laboratory leaching tests include: batch, column, and lysimeter leaching methods (e.g., Method 1311, 1312, 1313, 1314, 1315, and 1316).

the environmental field parameters in the laboratory are recognized as a reason for discrepancy between the laboratory and field leaching results (Maia et al. 2019; Sanger et al. 2020; van der Sloot and Dijkstra, 2004).

The results from a field leaching study provide a more representative release of pollutants of concern from RCA than laboratory leaching methods due to the incorporation of environmental field parameters. Regardless, batch laboratory leaching methods[14] have proven to be consistent, inexpensive, and simple for regulators, researchers, and facility site managers to implement for evaluation of a material's potential environmental impact.

Environmental Concerns

In this report, the chemical, inorganic, and organic constituents in RCA leachate reported in the reviewed literature are compared to the Washington State water quality criteria. The results of this literature review should be viewed judiciously with the knowledge the RCA material assessed has a wide degree of heterogeneities. In addition, this assessment evaluates studies using different leaching methods from an international and domestic perspective.

This literature review concludes that the pH, or the negative logarithm of the hydrogen ion concentration, is essential to determining the environmental risk posed from the RCA. Freshly crushed, non-carbonated concrete material is extremely alkaline, with a pH between 13 and 14 standard units (S.U.) (Cabrera et al. 2019; Engelsen et al. 2017). The degree of carbonation the concrete material has experienced is critical to determining the leachate pH, as a highly carbonated concrete produces a near-neutral leachate pH.

The pH results of fully carbonated to non-carbonated RCA for all leaching methods reviewed ranged between 7.23 to 13.2 S.U. (Butera et al. 2015b; Roque et al. 2016). The runoff leachate pH immediately following contact with concrete material may be above 8.5 S.U., or the upper effluent limit in the Sand and Gravel General Permit[15]. This pH-impacted water has been shown to discharge from RCA for at least the first-year in field scenarios (Engelsen et al. 2012, 2017; Sanger et al. 2020).

The total dissolved solid[15] and total suspended solid[15] effluent limits in the Sand and Gravel General Permit were exceeded at least once in every study reviewed reporting the parameters (Rodriguez et al. 2017; Sadecki et al. 1996). The leachate turbidity[15] effluent limit was exceeded once in the beginning for a fine-grained RCA (less than 4.75 mm in diameter) stockpile leachate study. This was the only study reporting turbidity (Sadecki et al. 1996).

Several pollutants of potential environmental concern were identified by direct comparison of State inorganic water quality criteria to the RCA laboratory or field leachate concentrations in the reviewed literature. Several of the identified inorganic pollutants of concern (e.g., chromium, sulfur (as sulfate), arsenic) may be sourced from raw materials used to manufacture the concrete. Another possible source of these pollutants in/on RCA is exposure to a foreign material (i.e. physical contaminants) during primary use (e.g., asphalt covering, steel reinforcement, deicing salt, pavement surface pollutants). Physical contaminants should be removed prior to concrete recycling.

RCA regularly releases antimony, arsenic, chromium (VI), copper, nickel, and selenium in excess of the State surface water quality criteria (Chapter 173-201A WAC). The releases of

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[14] Batch laboratory leaching methods are typically an approved laboratory extraction procedure used to determine hazard characteristics of a material using a single standard leaching scenario (e.g., Method 1311, 1312, 1313, and 1316).

[15] Requirement in S2 of the *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* 2021.

lead, mercury, and zinc from RCA do not commonly exceed the State surface water quality criteria, but the criteria were exceeded at least once in the studies reviewed.

Arsenic, chromium, and selenium are regularly released from RCA in excess of the State groundwater quality criteria (Chapter 173-200 WAC). The releases of barium, iron, lead, and manganese from RCA do not commonly exceed the State groundwater quality criteria, but the criteria were exceeded at least once in the reviewed studies. For a short period, the chloride concentrations may be released above, before decreasing to below, the State surface water and groundwater quality criteria. Sulfur (as sulfate), although seldom released from RCA above the State groundwater quality criterion, has been observed in the RCA leachate at concentrations exceeding the criterion by a factor between 2.7 and 23.

The polycyclic aromatic hydrocarbon (PAH) concentration in the RCA leachate was above the State groundwater and surface water quality criteria in the only study discovered evaluating PAHs in the RCA leachate.

Polychlorinated biphenyls have been observed in the RCA composition at low but non-negligible concentrations (Dhir et al. 2019). The polychlorinated biphenyls present in the RCA leachate did not exceed 0.01 ug/L, or the study's apparent analytical detection limit (Strufe et al. 2006; Engelsen et al. 2002).

Alkaline pH Neutralization and Dilution

The carbonation process produces physical, chemical, and mineralogical changes to the RCA that influences the release of POC and the material pH. In a field leaching scenario, the highest leachate pH is observed immediately after the leachate leaves the RCA material. The RCA alkaline pH may be neutralized and potential risks minimized depending on the specific conditions existing on-site (i.e. infiltration rate, organic matter content of soils).

Surface water pH neutralization of the alkaline leachate occurs primarily from the presence of excess hydrogen ions and the carbonate system. Whereas subsurface pH neutralization mechanisms occur via interaction of the leachate with soil and groundwater acidity, soil vapor carbon dioxide, and carbonation. The propagation of the alkaline pH front though the soil is directly proportional to the mineralogy of the soil (Chen et al. 2020). Silt (ML14) or clay (CL25 and CH38) classified soils[16] are shown to retard the propagation of the alkaline pH front compared to clayey sand (SC10).

There is potential for RCA stockpile water to runoff into nearby waters of the State, if minimum stockpile setbacks from State water bodies are not maintained (Snyder et al. 2018). Dilution of the RCA effluent discharge results in a pollutant of concern concentration that are substantially lower than the non-diluted effluent concentration. The impacted groundwater could intercept a surface water body and be further diluted by the ambient surface water (Engelsen et al. 2020).

The use of RCA as road-base results in a short-term alkaline pH discharge as opposed to a longer-term discharge associated with RCA stockpiles at processing facilities. For subsurface discharge, the soil acidity will become exhausted over a long-term period at a high liquid-to-solid ratio. After the soil acidity is expended and depending on site-specific conditions (i.e. soil organic matter), the soil may use carbonation to sufficiently buffer against the alkaline pH (Oliveira et al. 2020).

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[16] Classified by the Unified Soil Classification System.

Conclusions

Future work should include an investigation of "Washington State-specific RCA leachate" both in composition and in acute and chronic whole effluent toxicity on "Washington State-specific target organisms." This is due to the fact that untreated RCA leachate has "suspected toxicity because of apparent damage to aquatic biota" and vegetation at low concentrations of the leachate (Brás et al. 2018; Rodrigues et al. 2017; WAC 173-205-040(1)(f)[17]).

Additionally, correlation between RCA leachate presented in the domestic and international reviewed literature to Washington State-specific RCA is needed to establish if the pollutants of concern previously identified may or may not leach from Washington State-specific RCA at concentrations of concern.

Conventional and innovative construction controls, site layout, and operational management strategies can provide environmental protection from RCA leachate when these mitigation strategies are properly employed and maintained. Redundant, innovative, and conventional best management practices implemented by the concrete recycling industry have been shown to mitigate environmental concerns (Snyder et al. 2018).

Concrete recycling is recognized as beneficial to the construction industry, with benefits ranging from a decrease in greenhouse gas emissions to saving raw materials. Concrete recycling process controls must be monitored to ensure environmental risks are minimized, while retaining the net benefits of concrete recycling.

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[17] WAC 173-205-040– Whole Effluent Toxicity Testing and Limits: Determining the need for effluent characterization.

Introduction

The Washington State Department of Ecology's (Ecology) issuance of the *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* (general permit; provided in Appendix A) initially went into effect April 1, 1994. The permit has undergone several revisions and reissuances to include emerging operations discharging wastewaters to waters of Washington State (State). The general permit provides Ecology with the regulatory mechanism to limit the discharge of pollutants to waters of the State (33 U.S Code 1251[18]; Chapter 90.48 RCW[19]).

Since 2010, Ecology has used the general permit to regulate the discharge from any facility with concrete recycling identified as an activity[20]. The general permit defines concrete recycling as the processing of hardened concrete to produce a reusable concrete product. The recycled concrete aggregate (RCA) is commonly stockpiled outdoors where it is exposed to environmental stressors. The general permit prohibits the effluent from a facility to cause or contribute to a violation of the State or Federal water quality criteria[21].

The United States Department of Transportation Federal Highway Administration refers to RCA as portland cement concrete that is purposefully crushed to a desired gradation to produce concrete aggregates capable of re-use (RCW 70A.205.700; FHWA, 2004). Generally, concrete product is processed at a permitted facility and stockpiled on-site in various particle sizes until the materials are reused. Concrete recycling processing includes, but is not limited to, the crushing, fracturing, sorting, stockpiling, grading, and washing of concrete material to produce a reusable product.

The potential environmental concerns posed by RCA stockpile leachate has prompted Ecology's Water Quality Program to support a RCA literature review. Articles presented in this review focus on the physical properties, carbonation, and release of pollutants of concern (POC), as well as the environmental hazards associated with RCA. The environmental hazards posed by the RCA leachate include effluent above State water quality criteria as well as potential toxicity on aquatic organisms. Best management practices (BMPs) and application of known and available methods of prevention, control, and treatment at facilities may be capable of mitigating environmental concerns of RCA leachate.

Several domestic and international authors contribute to the foundational understanding of the construction and demolition waste industry, with particular focus on cementitious materials. Despite environmental issues potentially associated with the recycling of concrete material, Ecology encourages the re-use of the material to preserve virgin aggregate sources, reduce greenhouse gas emissions, and lessen the disposal in landfills, as well as for economic reasons.

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[18] 33 U.S. Code § 1251 – Congressional declaration of goals and policy

[19] Chapter 90.48 RCW – Water Pollution Control

[20] The word "facilities" and "facility" will now carry the meaning "actively permitted facility(ies) with concrete recycling identified as a Sand and Gravel General Permit activity," located in Washington State and identifies with the activity code "ECY002."

[21] "State water quality criteria" refers to Washington State Chapter 90.48 RCW: Water Pollution Control, Chapter 173-201A WAC: Water Quality Standards for Surface Waters, Chapter 173-200 WAC Water Quality Standards for Groundwaters, and Chapter 173-204 WAC Sediment Management Standards of the State of Washington. "Federal water quality criteria" refers to 40 Code of Federal Regulation (CFR) Part 131 – Water quality standards.

This report is not a review of the concrete or concrete recycling industry, but rather a literature review of the characteristics of RCA and RCA leachate, as well as potential environmental concerns when the material is not properly managed. The sensible management of stockpiles and implementation of BMPs may reduce the negative environmental impacts while retaining the net benefits of recycling concrete.

Report Goals

The purpose of this report is to present Ecology's Water Quality Program with the current science of the release of POC from RCA. Specific POC released from RCA may exceed (not meet) the State water quality criteria[22] in laboratory tests and/or in field leaching tests.

Direct exposure of RCA stockpiles to sources of water identified in the general permit potentially allows RCA exposed leachate to runoff to a surface water body and/or infiltrate into a subsurface aquifer. The sensible management of stockpiles and implementation of BMPs will reduce the negative environmental impacts while retaining the net benefits of recycling concrete.

The main goals of this report are to:

- Describe the life-span of concrete materials,
- Identify the physical properties and general chemical components of RCA,
- Explain the carbonation process of cement-based/concrete materials,
- Present a brief description of leaching methodologies discussed in the literature,
  - Define the effect of the leachate aggressiveness on the solid-liquid partitioning,
- Review factors controlling the POC released by RCA; including the dependence on:
  - pH,
  - degree of carbonation,
  - liquid-to-solid ratio,
  - particle size,
  - other potential release factors,
- Report environmental concerns presented by water contact with RCA, and
- Briefly describe BMPs available to mitigate the environmental concerns of concrete recycling.

Methods

Literature presented in this literature review are sourced from peer-reviewed journal articles, textbooks, and reports authored by academic, scientific and governmental groups. Personal opinion, biased work, and data not from a reputable source are excluded from this literature review. Raw data, unpublished literature, and preliminary data is avoided for failing to meet project data quality standards.

There is a tremendous amount of literature published on cementitious materials. More than 1,000 documents were assessed to a degree for use in this literature review. These documents were found using keywords (i.e. recycled concrete aggregate, concrete material, construction and demolition waste) and references from other RCA literature.

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[22] Washington State Chapter 90.48 RCW: Water Pollution Control, Washington State Chapter 173-201A WAC: Water Quality Standards for Surface Waters of the State of Washington, and Chapter 173-200 WAC Water Quality Standards for Groundwaters of the State of Washington. "Federal water quality criteria" refers to 40 Code of Federal Regulation (CFR) Part 131 – Water quality standards. These regulations will be considered the "water quality criteria."

An attempt was made to focus exclusively on RCA. Occasionally reports on cement-based materials are referenced. Cement-based materials are relevant to the discussion of RCA because cement is a major component of RCA. Concrete laboratory reference materials, fresh concrete, and RCA leachate results are assessed in this literature review.

Internationally and domestically, RCA is considered any material containing no less than 90% concrete-based material. Construction and demolition debris, or "demolition waste," is an all-inclusive name for construction and demolition waste generated from civil infrastructure projects. Demolition waste was not used as a search criterion in this literature review as it consists of, but is not limited to: bricks, bituminous materials, wood, and other non-concrete products (WAC 173-304-100[23]).

Concrete material with known exposure to contamination such as: chemical, physical, biological, or radiological substances are not included in this literature review. Contaminated concrete is excluded because the conclusions may be a result of the contaminated substances and atypical of concrete expected at a permitted concrete recycling facility. Albeit mentioned, RCA with documented replacements (e.g., coal bottom and fly ash, Pozzolan ash[24], and blast furnace slag), were excluded from the assessment.

The use of RCA extends beyond the authority of Ecology, and as such, any discussion on topics beyond the authority of Ecology will be limited. This includes asphalt-bound sub-base and the production of recycled aggregate concrete (RAC) using RCA as the aggregate source[25, 26]. A brief discussion of the use of RCA by Washington State Department of Transportation (WSDOT) is included.

Around 200 pieces of literature, related to the life cycle, carbonation, physical and chemical properties, leaching methods, leachates, environmental effects, and BMPs of concrete, RCA, and cementitious material met the goals and objectives for inclusion in this literature review.

The literature reviewed by Ecology must be identified and categorized if the agency takes significant action (i.e. support for future permitting decisions) as a result of this report (RCW 34.05.272[27]). The References section lists the 11 categories of 'sources' for the literature reviewed.

Background

Concrete refers to a man-made conglomerate of aggregates (gravel, sand, and/or crushed stone) bound by a cementitious paste. Around 4 billion tons of cement and over 11 billion tons of concrete are produced on a global annual basis (Meyer, 2009; USGS, 2020b). The global concrete material generation increases by almost 5% per year and there is a rising need of good-quality concrete aggregates for use in construction projects (Dhir et al. 2019).

In 2017, according to the Ecology's Solid Waste Program, approximately 1.2 million tons of concrete material was collected in the State of Washington. Washington State also sold or used 4.2 million metric tons of concrete aggregates[28] in 2017 (USGS, 2020a). The WSDOT began

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[23] WAC 173-304-100 Minimum Functional Standards for Solid Waste Handling: Definitions.

[24] A natural siliceous (or siliceous-aluminous) material commonly added to cement clinker to consume excess lime (CaO) generating additional cement-phase minerals and increases strength, density, durability.

[25] Recycled aggregate concrete (RAC) is fresh concrete produced using recycled concrete aggregates as the primary aggregate source.

[26] Bestgen et al. (2016b) will be included in this literature review. They assess recycled aggregate concrete curing time effect on the concrete leachate concentration.

[27] RCW 34.05.272 Administrative Procedure Act: Department of ecology—Significant agency action—Identification and categorization of sources of information used.

[28] Concrete aggregates here is all concrete material including concrete sand.

their RCA recycle and re-use initiative in 2016. Since 2017, less than 7% of the total tonnage of material used on State construction projects was RCA (WSDOT, 2018a, 2018b, 2019).

The collected concrete material is typically stored in stockpiles at a facility until its potential re-use (e.g., disposal as a fill material in a landfill, reincorporation into concrete) (Butera et al. 2015a; Leigh and Patterson, 2004). In February 2021, Washington State has 153 actively permitted facilities with concrete recycling identified as an activity, as shown in Figure 1. Over half of the facilities are within the 12 counties surrounding Puget Sound and Salish Sea.

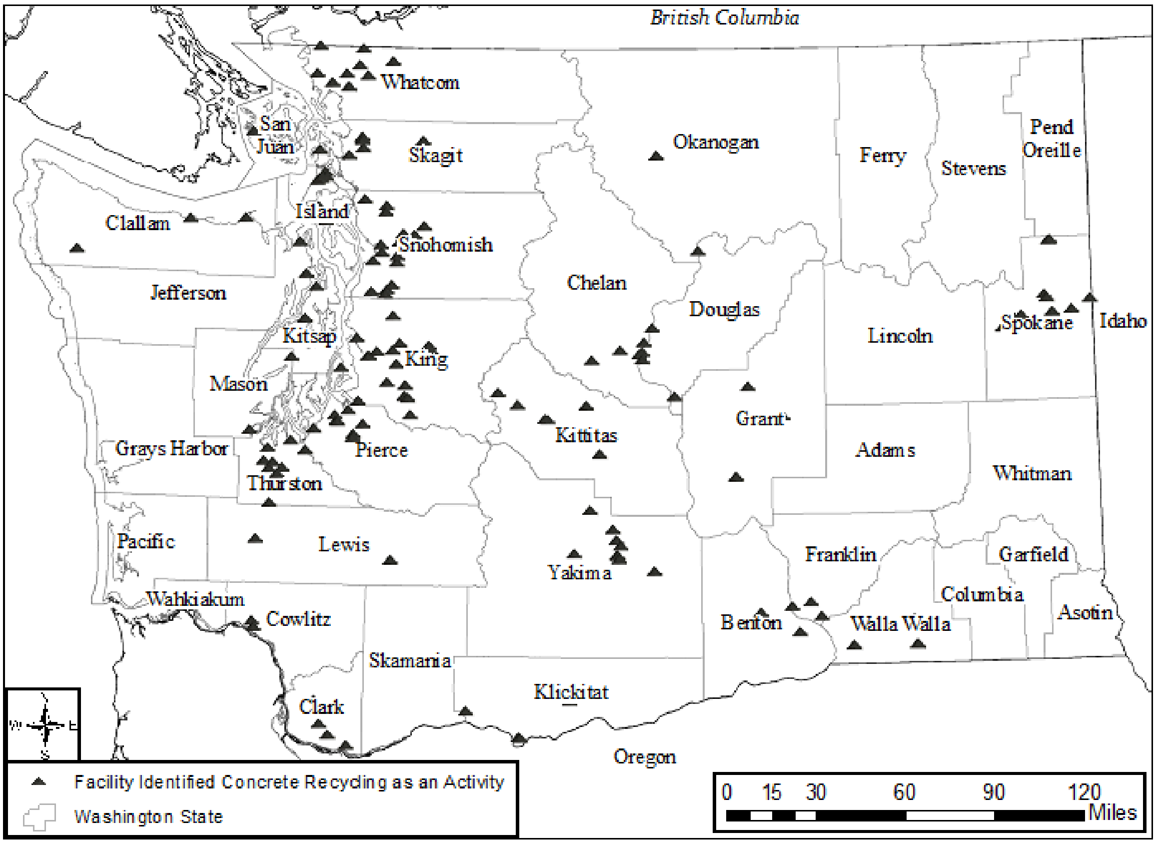

Figure 1: The locations of facilities across Washington State identifying concrete recycling as an activity as of 2/19/2021.
The source of the locations is Ecology's Permitting and Reporting Information System (PARIS) database.

Puget Sound is a biologically productive fjord-estuary in Washington State. Changes to water chemistry in waters of the State, including Puget Sound, may result in adverse effects to aquatic life. The majority of the State permitted concrete recycling facilities discharge to groundwater and seldom to surface water bodies.

Figure 2 shows surface water runoff and groundwater discharge as a result of precipitation onto a recycled concrete aggregate (RCA) stockpile; note this figure does not represent current concrete recycling facility practice (i.e. best management practices, collection and treatment of RCA leachate). A facility's water quality discharge from a RCA stockpile to waters of the State are measured for compliance to the *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* 2021 (general permit; provided in Appendix A).

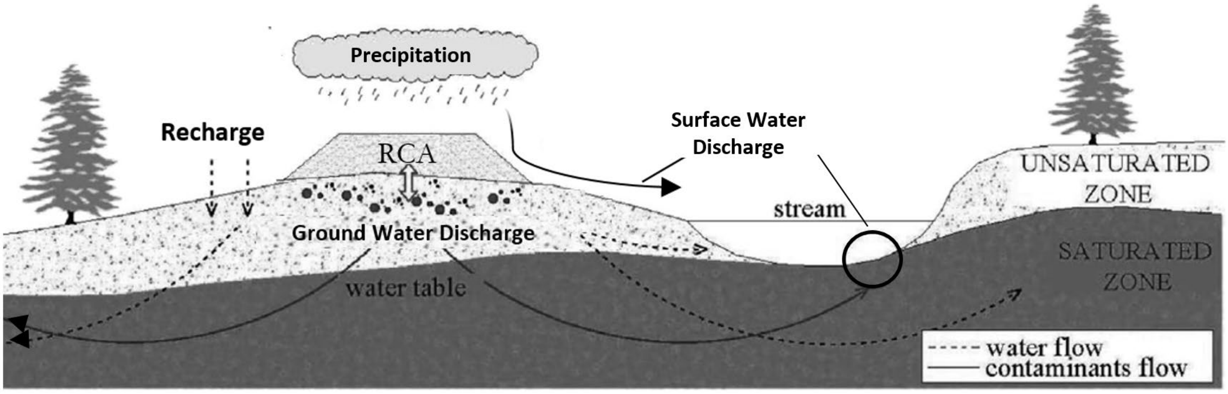

Figure 2: An illustrative field scenario of a recycled concrete aggregate (RCA) stockpile. *The transport flow paths for the pollutants of concern are indicated as solid lines (modified from Galvín et al. 2014a). The figure does not represent current concrete recycling facility practice (i.e. best management practices, collection and treatment of RCA leachate).*

The general permit requires the pH of the effluent discharge from permitted facilities to be between 6.5 and 8.5 S.U. In Washington State, a solid or liquid with a pH equal to or above 12.5 S.U. or below 2 S.U. characterizes the material as a dangerous corrosive waste (WAC 173-303-090; 40 CFR 261.22[29]).

The pore water pH of concrete, and RCA, decreases from around 13 to 8 S.U. over time (Engelsen et al. 2012; Kosson et al. 2002). RCA is considered an alkaline material (Dhir et al. 2019). Carbonation of the cementitious matrix of RCA is an exothermic, mainly diffusion-based process, progressing from the exposed RCA surface inwards (Ai et al. 2019; Engelsen et al. 2005; Sanger et al. 2020; Walton et al. 1997). A result of RCA carbonation is a neutralization of the leachate pH to near neutral (van der Sloot et al. 2011).

In addition to carbonation from the atmosphere, other environmental leachate pH neutralization mechanisms exist and must be taken into account (Chen et al. 2020; Gupta et al. 2018). The degree of carbonation the RCA has experienced prior to leaching dictates the leachate pH and thus the solubility of the POC released from the material (Ai et al. 2019; Engelsen et al. 2009; Mulugeta et al. 2010). The pH dependent releases of POC from RCA material are critical to evaluating the environmental risk (Dhir et al. 2019; Engelsen et al. 2006, 2009, 2010, 2012, 2017; Maia et al. 2018).

The RCA leachate produced from water contact with RCA creates a mixture of POC with an alkaline pH, which may act synergistically on the environment. Laboratory ecotoxicity assessments indicate adverse effects to aquatic biota following exposure to laboratory created concrete leachate (Brás et al. 2018; Choi et al. 2013; Rodrigues et al. 2017).

The environmental risks posed by the RCA leachate can be mitigated by the implementation of best management practices (BMPs) at the facility. BMPs include redundant conventional and innovative apparatuses for management of total dissolved solids and pH of the RCA leachate, facility layout, design, and planning, as well as employees of the facility maintaining their training.

The statewide quantity of RCA produced annually and potential of the RCA leachate violating State water quality criteria has generated environmental concerns. This report attempts to inform the reader of concrete production and use, physical and general chemical properties of RCA, carbonation of cementitious materials, and leaching of POC from RCA. This report concludes by identifying leachate POC from RCA with the greatest likelihood of exceeding the

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[29] U.S. Code of Federal Regulations Title 40 Chapter 261.22: Characteristic of corrosivity.

water quality criteria according to the cited literature, employable best management practices at a facility, and Washington State-specific RCA future studies.

Except for pH, the metal and organic parameters reported in the RCA leachate did **not** exceed the dangerous waste toxicity characteristics listed in WAC 173-303-090[30] (Clark et al. 2013; Poon et al. 2006). This determination was performed using the U.S. Environmental Protection Agency (EPA) Method 1311[31, 32] (TCLP) leaching method, which determines if a solid waste is considered a dangerous waste (Chapter 173-303 WAC).

Previous RCA Literature Reviews

In the following sections, observations and conclusions from three existing RCA literature reviews are presented. These recently published RCA literature reviews vary in scope and objective, but together provide a solid foundation of knowledge regarding RCA research. The three literature reviews cover the current state of the RCA chemistry and physical properties, RCA leachate chemistry, and leaching methods (Dhir et al., 2019; Maia et al. 2018; Sanger et al. 2020).

Sanger et al. (2020) – Recycled concrete aggregate in base course applications: Review of field and laboratory investigations of leachate pH

Sanger et al. (2020) reviewed numerous reports and methods employed by authors of RCA leachate investigations for evaluation of the leachate pH. The authors discuss the general chemical properties of RCA that influence the pH leaching behavior.

Sanger et al. (2020) compares the leachate pH observed in the reviewed studies to the leaching method used (i.e. batch, column, or field leaching method). They develop general conclusions and limitations of the leaching methods applied. The importance of specific RCA properties (i.e. carbonation, particle size) as well as leaching parameters are also presented.

General Chemical Characteristics

Sanger et al. (2020) state the following general characteristics of RCA:

- RCA is created from a concrete monolith after the crushing process produces fine- and coarse-grained aggregates. Where a fine-grained aggregate is any particle with a diameter less than 4.75 mm and coarse-grained aggregate has a diameter greater than 4.75 mm (WSDOT, 2009).
- The primary cement hydrate phases existing in RCA are calcium carbonate, ettringite (hydrous calcium aluminum trisulfate), calcium hydroxide ($Ca(OH)_2$), calcium-silicate-hydrate (CSH), as well as trace amounts of unreacted cement (e.g., alite (C_3S [33]), belite (C_2S), celite (C_3A), and felite (C_4AF)).

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[30] WAC 173-303-090 – Dangerous Waste Regulations: Dangerous waste characteristics.

[31] Hazardous Waste Test Methods – SW-846: Test Method 1311: Toxicity Characteristic Leaching Procedure (TCLP). The TCLP is the only leaching method used to determine if a solid waste exhibits the characteristic of toxicity for a list of contaminants available in WAC 173-303-090.

[32] The EPA leaching methods Toxicity Characteristics Leaching Procedure (TCLP) and Synthetic Precipitation Leaching Procedure (SPLP) methods will be referred to as Method 1311 and Method 1312, respectively. Also, the EPA Leaching Environmental Assessment Framework (EPA LEAF Method 1313, 1314, 1315, and 1316 will be referred to as such (i.e. Method 1315) for the rest of the report.

[33] Cement Chemist Notation (CCN) are presented in italics. As an example, alite, or tricalcium silicate, is referred to as C_3S, where C = CaO; S = SiO_2; A = Al_2O_3; F = Fe_2O_3; H = H_2O; $\bar{S}$ =SO_3; $\bar{C}$ =CO_2; m = mono; t = tri.

- The amount of unreacted cement present in the RCA depends heavily on the water-to-cementitious material (*w/cm*) ratio[34] used in the original formation of the concrete.
- The degree of carbonation the RCA has experienced is directly correlated to the pH of the RCA leachate.
- Carbonation is (*mainly*) a diffusion-based mechanism occurring on the surface and progressing inwards. Carbonation of RCA requires the contact of water containing carbon dioxide. In this process, the *CSH* and calcium hydroxide present in/on the RCA reacts with the aqueous carbonic acid to form calcium carbonate in calcite and vaterite forms[35].
- The dissolution of calcium hydroxide provides the release of hydroxide ions, creating the high pH observed in RCA leachate.
- The formation of calcium carbonate creates a protective layer on the surface of the RCA, restricting mass transport and dissolution of minerals in the liquid-phase[36].
- The degree of carbonation and thus the leachate pH is dependent on the amount of calcium hydroxide, calcium carbonate, and unreacted, unhydrated cement-paste present in/on the RCA.

Leaching Methodologies

Sanger et al. (2020) present an assessment of valuable leaching method information, which should be considered when designing a laboratory- or field-scale RCA pH leachate project:

- A common, inexpensive, and reproducible method employed by researchers to investigate the RCA leachate is the use of **batch** leaching tests.
  - **Batch** leaching methods typically vary at least one parameter (e.g., liquid-to-solid (L/S) ratio, exposure times, pH) followed by extraction of the leachate (or solution) for analysis.
- The RCA batch leachate extract pH ranges from 9.9 to 13 S.U., in the reviewed literature.
- The pH of fresh, non-carbonated RCA leachate is between 11.5 and 12.7 S.U., while the pH of the carbonated RCA leachate ranges from 9.9 to 11.8 S.U.
- Limitations of using the batch leaching method on RCA include the following:
  - Particle abrasion, disrupting the protective carbonate layer,
  - The exposure times for the leaching tests,
  - The continuous saturation is not reflected in the field leaching of RCA. Precipitation allows for periods of intermittent wetting and drying enhancing the rate of carbonation[37].
- A more representative leaching method of environmental conditions is the **column** leaching method.
  - The **column** leaching method is a standard leaching procedure using a column apparatus packed with a material exposed to an aqueous solution flowing in either an upward or downward direction (ASTM, 2014). The volume of aqueous solution the

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[34] The water-to-cementitious material (*w/cm*) ratio refers to the mass of the water to the mass of the dry cementitious material. The volumetric water-to-cementitious material ratio will not be referred to in this literature review.

[35] As carbonation progresses, cement hydrates (i.e. calcium-silicate-hydrate) other than calcium hydroxide may start to react with aqueous carbonic acid.

[36] The carbonation of concrete can lead to tensile stress buildup potentially resulting in subsequent cracking, facilitating the mass transport and dissolution of POC-containing minerals into the liquid phase.

[37] Intermittent wetting and drying cycles enhance the rate of carbonation of concrete via a wick action that accelerates the ingress of water and carbonic acid.

material is exposed to is commonly referred to as either the liquid-to-solid (L/S) ratio or pore volumes of flow (PVF).

- The results of upward or downward flow in RCA column leaching tests have been shown to be similar.
- The column leaching method preserves the protective carbonated surface, allowing for more similar leaching to field studies.
- When intermittent wetting and drying cycles are applied in RCA column leaching methods, the results are more representative of environmental conditions.
- The leachate pH of RCA from column tests is between 10 and 12.5 S.U.
- Studies using a continuously saturated column test, do not account for the intermittent wetting and drying cycles the RCA experience in the field.
- The RCA column leachate pH is therefore considered the maximum pH observed in a field study.
- **Long-term field** leaching investigations are the most representative of environmental conditions RCA experiences while stockpiled, with the potential exception of data collection[38].
  - Typical **long-term field** studies use a high-density polyethylene liner to collect the discharge from a RCA base course or stockpile into either a flow-through cell or a collection chamber where the pH is measured instantaneously or periodically, respectfully.
- Following an initial high pH period, the RCA leachate approaches a neutral pH, around 8 S.U., between the first and second year of exposure to the environment (assuming no addition of freshly crushed RCA material).
- After the leachate is no longer in contact with the RCA material, diffusion of atmospheric carbon dioxide into the leachate decreases the leachate pH over time and causes mineralization of POC in the RCA leachate.

Methods Assessment Conclusions

Sanger et al. (2020) conclude with a discussion about lessons learned from their review of laboratory and field leaching methods with respect to RCA leachate pH.

- The observed discrepancy between the field and laboratory leaching methods is attributable to the difficulty in measuring environmental parameters, e.g., the frequency and duration of precipitation, pavement drainage design, degree of saturation, temperature, heterogeneity in the sub-base soil geology, and traffic loads. Complicating the issue, these environmental parameters vary in space and time.
- They suggest that when carbonated RCA is encountered, particle abrasion should be avoided to preserve the protective carbonate layers on the exterior of the RCA particle, as the carbonate layer influences the leaching behavior of the RCA.
- When designing a RCA leachate laboratory investigation or in providing guidance for the use of RCA as a base course material certain aspects must be considered. Aspects to consider include the abrasion of the particle, an accurate model of the degree of carbonation

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[38] In long-term field leaching investigations, the RCA leachate should be collected, assessed, and analyzed immediately. If the RCA leachate is stored for a period of time before collecting for analysis, diffusion of carbon dioxide from the atmosphere into the RCA leachate will occur. Additionally, this may cause the recrystallization of pollutants of concern into colloids, minerals, and carbonated complexes. They therefore recommend an instantaneous or near instantaneous chemical parameter collection interval (i.e. a pH probe in the RCA effluent continuously collecting data, a sample fractionator and collector).

the RCA has experienced, and investigating of how the pH changes when the leachate is no longer in contact with the RCA.

- They suggest future research is required to investigate the evolution of the leachate pH after the leachate leaves the RCA base-course layer, or stockpile. The evolution of the leachate pH was investigated as a function of the soil classification the leachate travels through after leaving the RCA, in a publication with Morgan Sanger as a co-author (Chen et al. 2020).

Dhir et al. (2019) – Sustainable Construction Materials: Recycled Aggregates

The textbook *Sustainable Construction Materials: Recycled Aggregates* by Dhir et al. (2019) is one of the more comprehensive reviews of recycled aggregates to date.

Dhir et al. (2019) expand their book beyond RCA and into construction and demolition (C&D) waste aggregates. Among others, they present several chapters detailing the use, mechanical, physical, and chemical properties of recycled aggregates. Chapter 13 details the environmental impact produced by recycled aggregates, including RCA.

General Composition of RCA

The general concepts and principals included in Chapter 13 of Dhir et al. (2019) include:

- The primary elemental constituents of RCA are silicon, calcium, and aluminum, with significant concentrations of iron, potassium, sodium, and magnesium present in RCA.
- The abundance of trace elements in RCA vary significantly depending on the quality of sorting and exposure to other substances.
- Trace elements found in RCA include manganese, barium, phosphorus, strontium, lead, vanadium, zinc, chromium, lithium, nickel, copper, arsenic, cobalt, molybdenum, antimony, cadmium, and selenium.
- Polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs) are present in RCA in low but non-negligible concentrations.

Table 1 (next page) provides the minimum, maximum, and average elemental composition of RCA, the organic and inorganic carbon content is reported.

- RCA is considered an alkaline material due to the adhered cement-paste[39] content on the aggregate. An alkaline material has a pH above 7; RCA commonly varies between greater than 10 to below 13 S.U. Although they also state the pH of RCA and recycled masonry aggregates are near the range of natural aggregates or between a pH of 7 and 13 S.U.
- The authors acknowledge water in or recently in contact with RCA has the potential to discharge an alkaline effluent.
- The pH of the concrete depends on the degree of carbonation the RCA has experienced.
    - The authors state coarser-sized particles of RCA have a slightly higher pH value than finer-grain size particles. The particle size diminution allows the rate and uptake of carbonation to increase; due to the decrease in the concretes' volume-to-surface area ratio, other factors are discussed in the Factors Impacting Carbonation section.
- The pH of the material is identified as a significant factor controlling the release of POC.

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[39] "Adhered cement-paste" is the remnant, hardened cement-paste on the recycled concrete aggregate particles following particle size reduction.

Table 1: Maximum, minimum, and average total elemental composition and carbon content of recycled concrete aggregate.

Reproduced from Dhir et al. (2019).

			Total Elemental Composition of RCA			
Element	**Units**	**N[1]**	**Average**	**Standard Deviation**	**Minimum**	**Maximum**
Aluminum (Al)	*g/kg*	76	**40.5**	16.9	3.8	64
Calcium (Ca)	*g/kg*	81	**96.8**	20.1	13.7	>200
Iron (Fe)	*g/kg*	74	**17.9**	6.7	3.2	39
Magnesium (Mg)	*g/kg*	61	**5.5**	3.1	1.8	20
Potassium (K)	*g/kg*	73	**11.2**	7.3	0.8	28
Sodium (Na)	*g/kg*	67	**10.3**	5.4	0.2	19.5
Silicon (Si)	*g/kg*	56	**383**	122.7	240	509
Antimony (Sb)	*mg/kg*	64	**0.73**	0.6	0.001	3.3
Arsenic (As)	*mg/kg*	84	**4.67**	4.1	4.50E-04	33
Barium (Ba)	*mg/kg*	72	**381.7**	244.9	2	689.2
Cadmium (Cd)	*mg/kg*	75	**0.34**	0.3	0.005	2.4
Chromium (Cr)	*mg/kg*	86	**29.5**	26.3	0.239	152
Cobalt (Co)	*mg/kg*	70	**5.12**	2.5	0.201	8.3
Copper (Cu)	*mg/kg*	87	**26.4**	54.8	0.016	546
Lead (Pb)	*mg/kg*	71	**51.1**	100.2	0.321	873
Lithium (Li)	*mg/kg*	50	**15.8**	3.9	8.7	20
Manganese (Mn)	*mg/kg*	57	**453.2**	141.5	180	635
Molybdenum (Mo)	*mg/kg*	71	**11**	1.5	0.006	8.7
Nickel (Ni)	*mg/kg*	71	**20.5**	25.2	0.026	182
Phosphorus (as P)	*mg/kg*	50	**612.5**	211.7	250	1,200
Selenium (Se)	*mg/kg*	61	**5.9**	3.8	0.001	17.4
Strontium (Sr)	*mg/kg*	50	**378.5**	117.1	190	510
Vanadium (V)	*mg/kg*	51	**44.3**	23.1	1.50E-04	130
Zinc (Zn)	*mg/kg*	74	**72.9**	67.2	0.031	553
Carbon Content – Organic and Inorganic Carbon Content of RCA						
Organic Carbon	%	31	**0.5**	0.3	0.1	1.59
Inorganic Carbon	%	23	**2.7**	3.0	0.03	10.7
Polycyclic aromatic hydrocarbons	*mg/kg*	27	**4**	3.1	0.09	7.05
Polychlorinated biphenyls	*ug/kg*	27	**18.6**	62.9	1.45	400

[1] Sample size (N)

Leaching of RCA

The leaching of POC from RCA is categorized into three general patterns with varying pH. These leaching patterns are described below and shown in Figure 3:

- **Cationic pattern**: The POC following the cationic pattern has a maximum leached concentration in the acidic range decreasing into the alkaline range.
- **Amphoteric pattern**: The POC released in an amphoteric pattern has a peak leaching concentration at both the highly alkaline and highly acidic pHs and a valley around the neutral pHs.
- **Oxyanionic pattern**: In the oxyanionic pattern, the leachate concentration varies from a minimum at the mildly acidic pH to a maximum in the slightly alkaline to neutral pH range.

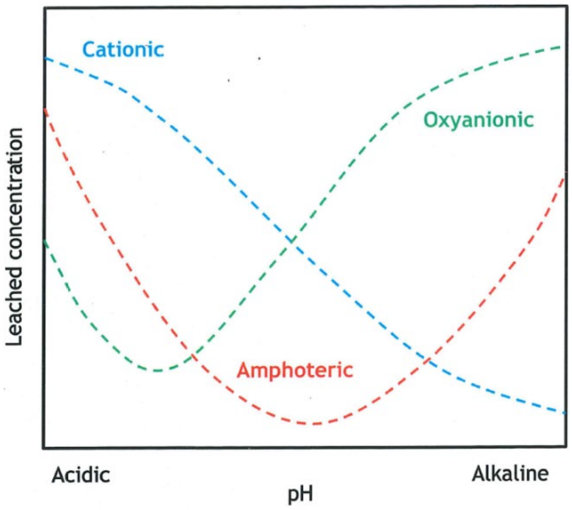

Figure 3: General leaching behavior patterns for pollutants of concern as a function of pH from RCA.
From Dhir et al. (2019).

- Elements are provided with an assigned leaching behavior pattern and pH ranges for when the elements leachate concentration is at a maximum and minimum[40].
- Elements with more than one leaching pattern assigned may leach from RCA in more than one mineral form and/or mechanism, e.g., chromium, zinc, copper.
- In general, the maximum release of POC from RCA typically occur when the leachate is highly acidic, when the pH is alkaline a minimal POC leachate concentration occurs.
- For certain metals, the particle-size affects the leachability of RCA.

Maia et al. (2018) – Toxicity of Recycled Concrete Aggregates: Review on Leaching Tests

In a recent literature review on leaching test methods associated with RCA leachates, Maia et al. (2018) reviewed the reasons leaching tests are performed, results of leaching tests, and ecotoxicological impacts of RCA leachate. The authors identify the following important components of RCA leachates:

- RCA is a heterogeneous material, meaning the cement and aggregates vary between recycling plants. The variance may be sourced from exposure to other materials during original use as well as the facilities management policies.

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[40] The assigned leaching behavior patterns for the elements is shown in Table 12.

- Separation of RCA from other demolition waste is important to reduce the contamination of the material.
- The majority of laboratory leaching methods reduce costs, produce results quickly, and allow for better control over particular leaching parameters.
- The most commonly used leaching methods are batch and column leaching methods including EN 12457-3, EN 12457-4, NEN 7343, CEN/TS 14405, EN 14997[41], and Method 1311 (TCLP).
- The results of leaching methods also depend on the parameters used for the leaching test: e.g., liquid-to-solid (L/S) ratio, exposure time, type of exposure (e.g., column test, end-over-end tumbler [batch leaching method]), particle size, pH, and temperature.
- One of the major advantages of modifying the leachant pH is the ability to observe the leaching behavior of the solid material under varying pH conditions.

Figure 4 is similar to Figure 3. The release behavior of three (3) types of POC from alkaline materials is shown in Figure 4.

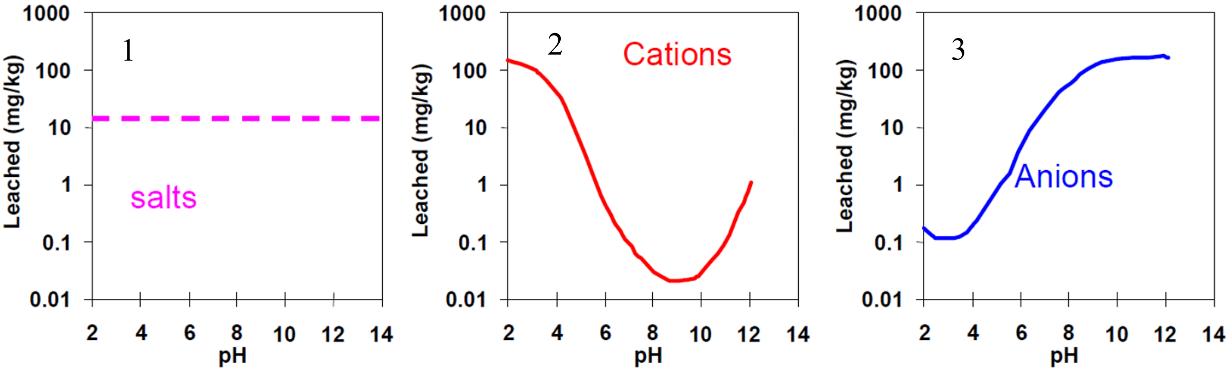

Figure 4: The broad leaching patterns of (1) highly dissolvable salts or dissolvable minerals, (2) cations, and (3) anions and oxyanions as a function of leachate pH.
Modified from van der Sloot and Dijkstra, (2004).

- The leaching test results are RCA-material dependent, in light of the RCA's origins, particle size, strengths, and age or degree of carbonation.
- The pH value of fresh concrete is more than 13 S.U., while fully carbonated concrete commonly has a pH value of less than 10 S.U.
- The observed pH value is less for highly carbonated RCA and also for larger particles compared to finer particles. This is due to an increase in the surface area for carbonation.
- Generally, the RCA chemical composition does not dictate the leachate concentration equilibrium; instead, solubility of the mineral phases present on the RCA surface is the dominant transport mechanism of pollutants into the leachate.
- The critical anions released from RCA materials are chloride (Cl^-) and sulfates (SO_4^{2-}).
- It is reported that above a liquid-to-solid ratio of 5 L/kg, the observed change in pH, conductivity, and other constituents released is negligible.
- It is also reported that the concentration of heavy metals decreases with increasing liquid-to-solid ratio due to dilution. Although, Kosson et al. (2002) indicate it is dependent on the constituent (or POC released from RCA) investigated.

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[41] EN stands for a validated European standard, NEN is a validated Dutch standard, and CEN/TS is a European Committee for Standardization standard and considered a preliminary technical specification that needs to undergo validation.

- The cost of reproducing the environmental conditions experienced by the RCA in the laboratory is very expensive.
- Column leaching methods are superior to batch leaching methods, because they provide results more similar to realistic field conditions.
- The selection of the leaching test method to be used on the RCA should reflect the purpose for evaluating the material.
    - The expected second use of the material should dictate the type of leaching method employed.
- They conclude the leaching method parameters – e.g., the leachant pH, liquid-to-solid ratio (L/S), and particle size – influence the POC release concentrations from the RCA.
- The laboratory leaching methods are not designed to characterize field behavior, rather the methods verify if the leachate concentration is below regulatory levels under the provided test conditions.

Additional Recommended Reading

In addition to Dhir et al. (2019), several textbook chapters contain noteworthy reviews on RCA, recycled aggregate concrete (RAC), recycled aggregates, and demolition waste.

- *Waste and Supplementary Cementitious Materials in Concrete: Characterisation, Properties, and Applications*,
    - Chapter 8: Construction and demolition waste (Ng and Engelsen, 2018).
- *Advances in Construction and Demolition Waste Recycling*,
    - Chapter 21: Leaching Performance of Recycled Aggregates (Engelsen, 2020).
- *New Trends in Eco-efficient and Recycled Concrete,*
    - Chapter 12: Leaching issues in recycled concrete aggregate (Cabrera et al. 2019).
- *Handbook of Recycled Concrete and Demolition Waste*,
    - Chapter 24: Assessing the potential environmental hazards of concrete made using recycled aggregates (RAs) (Tiruta-Barna and Barna, 2013).
- *Lea's Chemistry of Cement and Concrete*; 5th Edition,
    - Chapter 15: Concrete Aggregates (Sims et al. 2019).

Although these five Chapters provide a recent review of the environmental issues and material properties of RAC, RCA and recycled aggregates, several other textbooks discuss recycled aggregates, including concrete (Pacheco-Torgal et al. 2013; Siddique and Cachim, 2018; Youcai and Sheng, 2017).

Previous Leaching Method Literature Reviews

To satisfy the goals and objectives of this literature review, a basic understanding of leaching test methods and the physical and chemical properties affecting the liquid-solid partitioning is required. If additional reading is necessary, the following literature contains a detailed description of the relevant leaching test methods:

- *Toxicity of Recycled Concrete Aggregates: Review on Leaching Tests* (Maia et al. 2018),
- *Laboratory-to-Field Comparison for Leaching Evaluation using the Leaching Environmental Assessment Framework (LEAF)* and *LEAF How-To Guide* (Kosson et al. 2014a; Kosson et al. 2019),
- *An Integrated Framework for Evaluating Leaching in Waste Management and Utilization of Secondary Materials* (Kosson et al. 2002),

- *An Assessment of Laboratory Leaching Tests for Predicting the Impacts of Fill Material on Ground Water and Surface Water Quality* (Ecology, 2003).

The leaching methods used on RCA that met the goals and objectives for inclusion in this literature review were developed by the EPA, European testing and standards community (EN, NEN, CEN), American Society of Testing and Materials (ASTM), United States Geological Survey (USGS), and others.

In the last decade, four new EPA Methods have been developed and added to EPA's Hazardous Waste Test Methods (SW-846)[42]. The brief description of leaching test methods included in this literature review will focus primarily on EPA standard Methods as defined in Kosson et al. (2014a, 2019)[43].

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[42] United States Environmental Protection Agency (EPA): Hazardous Waste Test Methods – SW-846. https://www.epa.gov/hw-sw846

[43] United States Environmental Protection Agency (EPA): The Leaching Environmental Assessment Framework (LEAF) Methods and Guidance. https://www.epa.gov/hw-sw846/leaching-environmental-assessment-framework-leaf-methods-and-guidance

Concrete Lifespan

Concrete is an important material to the construction industry. This is because concrete is versatile, resilient, and economically viable for use in roads, buildings, dams, homes, sidewalks, in addition to other applications. Concrete refers to a mixture of aggregates (sand, gravel, and/or crushed stone)[44] bound together by a cementitious paste.

After initial use, concrete is demolished into rubblized[45] concrete material. Concrete material may be separated, processed, and stored in a stockpile until future use (Leigh and Patterson, 2004). The RCA produced has qualities and properties that are reflective of the original concrete's quality and properties (Dhir et al. 2019; Garber et al. 2019).

According to the U.S. Geological Survey (USGS) Minerals Yearbook, the nationwide sale of RCA increased from 45 to 105 million metric tons from 2016 to 2017 (USGS, 2020a, 2020c). In Washington State, approximately 1.2 million tons of concrete material was collected while 4.2 million metric tons of concrete aggregates were sold or used in 2017 [46](USGS, 2020a).

Despite best efforts to re-use RCA, the structural, chemical, and environmental uncertainty associated with the material have limited its use in infrastructure projects. The storage of RCA in stockpiles have raised environmental concerns (Bestgen et al. 2016a, Chen et al. 2013; Rodrigues et al. 2017; Sanger et al. 2020).

The following section describes in general terms the production, use, destruction, storage, and global carbonation of concrete and should not be mistaken for a life cycle assessment (LCA). A LCA accounts for the environmental exchanges, such as atmospheric emissions and the consumption of energy over the entire life cycle of a product and has been investigated elsewhere for RCA (Butera et al. 2015a; Lagerblad, 2005; Marinković et al. 2013).

Creation of Concrete

In October of 1824, Joseph Aspdin patented a hydraulic lime[47] coined "portland cement," because he wanted to draw attention to the color and quality of the cement produced with the Isle of Portland stone (Hewlett and Liska, 2019). Since, the name portland cement has endured to become the prominent type of cement used in modern day concrete and is no longer a "brand name."

Raw materials (e.g., calcareous, argillaceous, ferric materials in addition to others) are heated at high temperatures allowing the process of calcination to remove 44% of the calcium carbonate mass, through the release of carbon dioxide (Hewlett and Liska, 2019). The non-volatile minerals and compounds produced in a kiln is called cement clinker.

The unhydrated cement clinker consists of the following dominant cement phases: alite (C_3S), belite (C_2S), celite (C_3A), and felite (C_4AF). Additional cement phases are produced to a lesser degree (Hewlett and Liska, 2019). The cement clinker is ground to a fine powder (less than 45 um) with a gray color and with the addition of gypsum (and limestone) is considered portland cement.

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[44] The natural aggregates (of good quality) incorporated into the concrete are generally considered a 'filler' and non-reactive with the interacting water.

[45] Rubblization is the process of reducing existing, hardened concrete into rubble.

[46] The Washington State Department of Transportation reported using 1,470 tons of recycled concrete aggregate in 2017 (WSDOT, 2018a).

[47] Hydraulic lime is a cement binder capable of hardening upon chemical interaction with water and able to harden under water (Hewlett and Liska, 2019).

The addition of water to the portland cement produces a chemical reaction yielding cement-paste and hydrate complexes. Concrete typically consists of 7 to 15% portland cement, 14 to 21% water, 60 to 75% natural aggregates, and up to 8% air by volume (Kosmatka and Wilson, 2016). A diagram for the general production of concrete is provided in Figure 5 and described in detail in Kosmatka and Wilson, (2016), Hewlett and Liska (2019), and other literature.

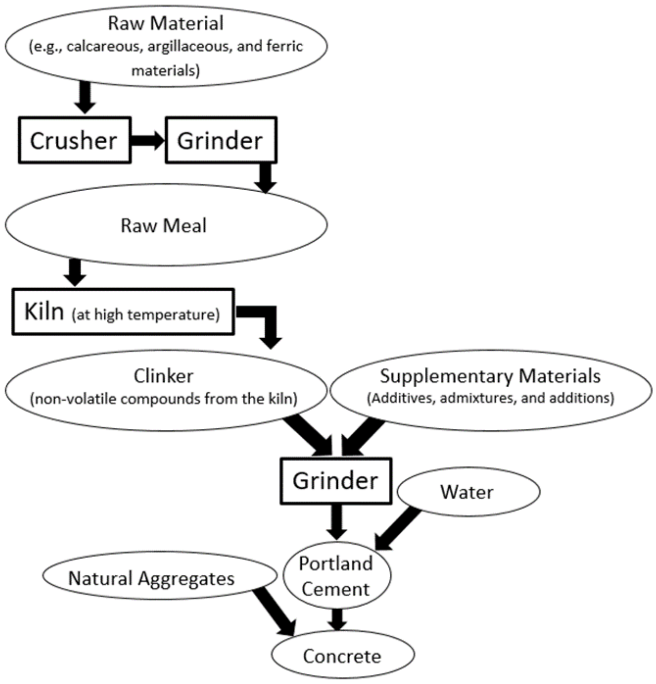

Figure 5: Simplified diagram of concrete production.
The cement is not necessarily portland cement. Modified from Worrell et al. (2001).

Supplementary materials are generally categorized into two classifications: concrete admixtures (e.g., polymers, additions, additives, and superplasticizers) and supplementary cementitious materials. Portland cement may be combined with concrete admixtures to reduce the required water, improve durability and workability, and improve other properties of the concrete mixture. In the United Kingdom, concrete admixture usage increased from around 12% of all concrete made in 1975 to nearly 80% in 2010 (Hewlett and Liska, 2019).

Also, the addition of supplementary cementitious materials to concrete are used to reduce the amount of cement clinker needed. Supplementary cementitious materials include fly ash, silica fume, fibers, and possibly nano-materials.

The water-to-cementitious material (*w/cm*) ratio is defined as the mass of water divided by the mass of cementitious material (including fly ash, silica fume, slag, and others) used to create the concrete. The *w/cm* ratio indicates the mean distance between the cement particles in a cement-paste before it begins to harden. The minimum *w/cm* ratio to completely hydrate ordinary portland cement is 0.42 (Hewlett and Liska, 2019). In concrete, the *w/cm* ratio does not consider the mass of aggregates.

Reducing the *w/cm* ratio added generally increases flexural and compressive strength, lowers permeability, increases water tightness, increases durability, increases the resistance to weathering, creates a better bond between the concrete and the reinforcement, reduces cracking and shrinking due to drying[48], and results in a lower volume change from wetting and drying cycles (Kosmatka and Wilson, 2016).

Aggregate Source

Since around three-quarters of concrete by volume is natural aggregates, the quality of the aggregate used influences the properties of the concrete. The aggregate shape, size, bulk and particle density, strength and toughness, porosity, absorption, composition, thermal expansion and conductivity influence the concretes' properties (Hewlett and Liska, 2019). Aggregates must conform to engineering performance standards to be used in modern day concrete (Kosmatka and Wilson, 2016).

A high quality aggregate is generally considered to be a non-reactive filler or cement replacement in concrete. A concrete with a high quality aggregate source produces a superior-quality concrete and thus a high quality RCA (Dhir et al. 2019).

Washington State's geologic history, including the most recent glaciation during the Pleistocene Epoch, brought numerous ice sheets southward from British Columbia (Ecology, 1981). These ice sheets covered the northern half of the State and allowed for the deposition of glacially derived gravel deposits. The glacially derived gravel deposits provide ideal aggregates for concrete with a superior particle shape, i.e. round, spherical, and flaky.

In 2017, the United States used 46% of construction sand and gravel produced nationally as aggregates in concrete (USGS, 2020a). Washington State's natural aggregates are the most valuable mineral commodity in the State, totaling $277 million in 2017. Also in 2017, the value of sand and gravel aggregates in Washington State was $8.32 per metric ton (USGS, 2020a).

Use of Concrete

The robustness and durability of concrete is demonstrated by the Roman concrete structures surviving today, including the Pantheon and Colosseum in Rome[49]. Modern day use of concrete includes: houses, bridges, dams, concrete roads, road sub-base, and other structural projects (Lagerblad, 2005; USGS, 1998). Concrete is a cost-effective, moldable, and stable material, allowing for its global usage in diverse formats.

The demand for concrete is continuously increasing (Aïtcin and Flatt, 2015; Dhir et al. 2019). Approximately 4 billion tons of cement and over 11 billion tons of concrete are manufactured on a global annual basis (Meyer, 2009; USGS, 2020b). Due to the heterogeneity of the raw materials used to create the portland cement, differing quality of aggregates, unknown water-to-cementitious material ratio, curing time, possible addition of concrete admixtures, additives, and additions, and the initial use of the material, concrete produced should be considered inherently heterogeneous.

The American Society for Testing and Materials (ASTM), Washington State Department of Transportation (WSDOT), and federal agencies have developed standards for concrete and cement that provide guidance to the construction industry. The type of concrete used for a project is typically determined by the compressive strength the material is intended to handle. The *w/cm* ratio is the most significant factor on the compressive strength of concrete, when concrete admixtures, additives, or additions are not added (Hewlett and Liska, 2019).

Concrete durability is often impacted by natural weathering or chemical attacks from brackish, sulfate-rich, or acidic waters (Cabrera et al. 2019). The concrete may also develop cracks from

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[48] Reducing the water-to-cementitious material ratio may increase the risk of shrinkage-induced cracking in addition to thermal cracking.

[49] The cementitious binder used by Romans differs chemically from portland cement.

alkali-silica reactions (ASR)[50] and other alkali aggregate reactions, wetting and drying cycles, and thermal cycles, e.g., freeze and thaw cycles (Hewlett and Liska, 2019). All of these processes degrade the concrete, potentially to the point of deconstruction.

Destruction of Concrete

The worldwide generation of demolition waste has surpassed 3 billion tons per year (Akhtar and Sarmah, 2018). Several articles propose frameworks for the separation of demolition waste and concrete material (Gálvez-Martos et al. 2018; Hjelmar et al. 2016; Ng and Engelsen, 2018; Puthussery et al. 2017; Silva et al. 2014). Internationally, between roughly 41 to 81% of the total input to demolition waste recycling plants is concrete aggregates (Coelho and de Brito, 2013; del Rio Marino et al. 2010; Tam, 2009).

A survey of demolition waste from 19 non-residential projects in the Pacific Northwest revealed an average of 66% concrete (EPA, 1998)[51]. The same survey of residential demolition wastes indicated lower amounts varying from 5 to 24% concrete material (EPA, 1998). The demolition waste is typically separated into discrete materials using air sifters and jigs, magnets, feeders, vibrating screens, spiral separators, manually, and/or other methods (Coelho and de Brito, 2013).

The automatic separation of demolition waste from concrete aggregates can be performed with a 98% efficiency with modern separation devices (Mulder et al. 2007). “Designing for deconstruction” is a concept aiming to manage and separate demolition waste on-site using technology and processing equipment (Hao et al. 2008; Wang et al. 2010).

The size and shape of the aggregate required by the consumer, the required composition of the material, and economic concerns dictate the type of crusher used to create the concrete aggregates (Lee et al. 2010; Ng and Engelsen 2018; Tam, 2008; Youcai and Sheng, 2017). A combination of a jaw crusher and rotary or impact crusher in this order is recommended to produce recycled aggregates with the best shape and size distribution for re-use (Chidiroglou et al. 2007; Marinković et al. 2013; Matias et al. 2013).

Occasionally, an aggregate with less adhered cement-paste will be required by the consumer. Aggregates with less adhered cement-paste are produced after primary crushing using an impact crusher as a secondary crusher (Silva et al. 2014). Processing methods employed by the industry to create aggregate particles with less adhered cement-paste include: acid treatment, heating, mechanical grinding, accelerated carbonation, microwave-based heating, microbial growth of calcium carbonate, and pulsed power discharges (Dhir et al. 2019). There are several treatments capable of strengthening the adhered cement paste (Sharma et al. 2020).

Crushed concrete material and fragmented recycled concrete is also known as recycled concrete aggregate (RCA). In this report, the term “RCA” applies to all concrete material manufactured with portland cement and aggregates or sand and/or containing greater than 90% concrete material (Engelsen et al. 2017; Silva et al. 2014). Figure 6 shows different demolition wastes, with RCA shown in Figure 6a.

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[50] Alkali-silica reactions in concrete refers to a reaction that takes place between certain types of reactive silica in the aggregates and alkali metals (sodium and potassium) in the presence of moisture (Hewlett and Liska, 2019).

[51] The survey of 19 non-residential buildings demolished by R.W. Rhine, Inc., Tacoma, WA and reported by EPA, (1998) contained 167,200 tons of demolition waste.

Figure 6: Different recycled and natural selected aggregates: a) a crushed recycled concrete aggregate (RCA), b) crushed mixed concrete, c) crushed reclaimed asphalt pavement, d) milled reclaimed asphalt pavement, and e) natural crushed limestone.
Adapted from Roque et al. (2016).

Storage of Concrete Material

After separation from other demolition materials, monolithic and crushed concrete material is stockpiled until its re-use, e.g., road sub-base, recycled aggregate concrete (RAC) (Leigh and Patterson, 2004). The RCA stockpile is exposed to environmental conditions, such as wetting and drying cycles, which may enhance the carbonation of the material (Sanger et al. 2020). A stockpile of recycled concrete at a Washington State facility with a general permit[52] and ECY002: Concrete Recycling identified as an activity is shown in Figure 7.

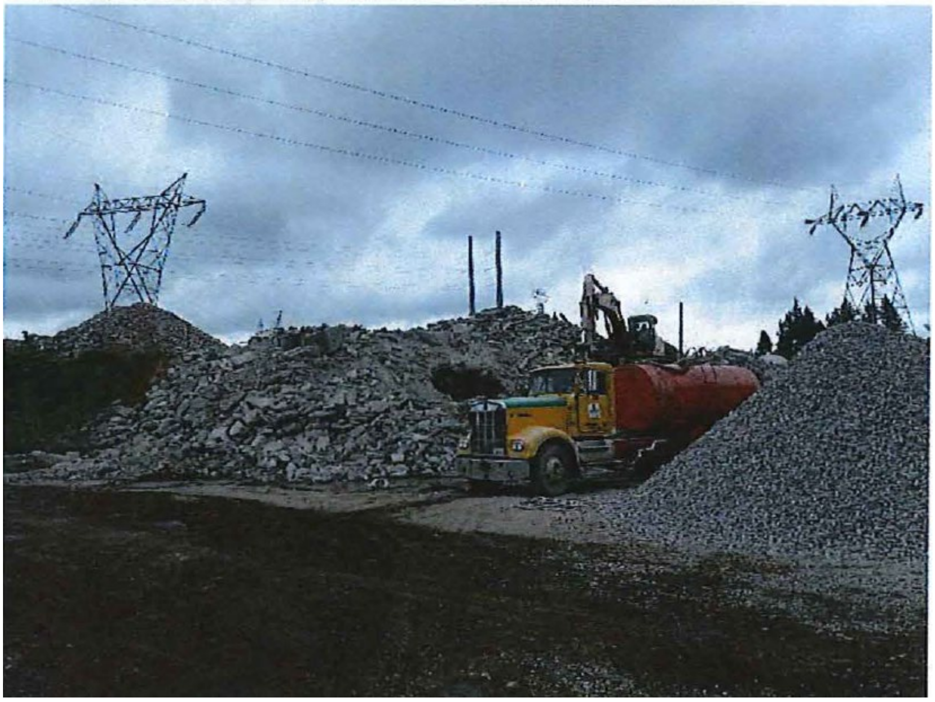

Figure 7: Recycled concrete stockpiles at a permitted[1] facility in Washington State.
Photograph taken at Girard Resources & Recycling LLC in Kent, Washington on August 17, 2017 by Tamara Welty (Ecology) during an unannounced inspection.
[1] Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit 2021. In addition to concrete recycling, other sand and gravel activities may be occurring on the permitted facility.

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[52] The term "general permit" refers to the *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* 2021.

The Ecology Code ECY002: Concrete Recycling is described as:

> "The processing (including, but not limited to, crushing, fracturing, sorting, storing, stockpiling, grading, and washing) of hardened structural concrete to produce a reusable concrete product.
>
> Sites only storing or stockpiling hardened structural concrete, and not otherwise crushing or processing the material are not subject to coverage under this permit unless they conduct additional activities requiring coverage under this permit."
>
> - Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit 2021

Review the Ecology permitting website to help determine which permit may be required: https://ecology.wa.gov/Regulations-Permits?cats=19.

The RCA is typically located in stockpiles at facilities. The geometry and surface area of the stockpile will affect the degree of carbonation the RCA experiences (AzariJafari et al. 2021). Approximately, the stockpiles' outermost foot of RCA is influenced by carbonation. This is because air circulation is restricted by the self-compaction and rehydration, from rainfall, of the RCA material deeper than approximately one foot (Stripple et al. 2018).

Self-cementation is the hydration of the unreacted phases in the fine-grained RCA material. The fine-grained RCA materials are believed to have a larger amount of cement-paste adhered to the particle allowing for self-cementation to an adjacent particle (Bestgen et al. 2016b; Poon et al. 2006).

RCA is heterogeneous in chemical composition and physical properties. The physical properties vary depending on the type of management strategy the facility employs. In addition, the mixing of concrete material received from different sources may also occur at the facility, which compounds the heterogeneity of the material. Facilities permitted by Ecology for concrete recycling under the general permit are not required to log the date and/or source of the concrete received.

Cured concrete material, or RCA, not tainted with chemical, physical, biological, or radiological substances and not presenting a threat to the environment greater than that inherent to the original material is considered an inert waste in Washington State (WAC 173-350-410). The State and industry consider concrete material, or RCA, an inert waste. Inert solid waste is considered an intact waste that cannot undergo significant chemical, biological, or physical transformations (Sharma et al. 2020).

Water Exposure Scenarios

In the scenarios where water is exposed to concrete material, water is considered the main pollution vector for hazardous POC (e.g., chromium, pH, arsenic) released from RCA (Barbudo et al. 2012; Tiruta-Barna and Barna, 2013). Water may be exposed to RCA materials at several stages in the recycling process, including: sorting, stockpiling, and washing. The scenarios where RCA is expected to encounter water include but are not limited to: landfill alternative daily cover, RCA stockpiles, construction fill, and general landfill material (EPA, 2012).

When RCA is exposed to storm, surface, or groundwater the interactions between the water and the material depend on the structure of the stockpile, the receiving plane, motion of water through the stockpile, and whatever may be in contact with the RCA (e.g., soil, immersed in water) (AzariJafari et al. 2021; Tiruta-Barna and Barna, 2013).

Water contact with RCA is expected to occur in different ways. One or more of the water contact mechanisms may be applicable to the expected exposure scenario. Water is exposed to RCA by direct contact, percolation, infiltration, runoff, and/or production of stagnant water layers (Tiruta-Barna and Barna, 2013).

The contact between water and RCA may occur in one or more of the following exposure scenarios identified in the literature:

- **A sloping plane**: the receiving plane of material is steeper than 15 feet per 100 feet; where the primary water contact with the material is runoff of water.
- **A horizontal plane**: rainfall interacts with the horizontal surface of the material and may produce runoff, stagnant layers, percolation through the macroporous material, and infiltration into the natural porous material (i.e. soil) below.
- **A vertical plane**: assuming the material is structurally capable of a "façade," the material may be exposed to precipitation and the main types of water contact possible are runoff and/or infiltration into material if it is indeed porous.
- **Contact with soil**: the material is located in the subsurface; the types of water contact possible are runoff, infiltration, percolation, and stagnation.
- **Completely immersed in water**: The material is completely immersed in water. Encountered when the material is below the ordinary high water mark or located in the subsurface; types of water contact possible include infiltration, direct contact, percolation, and stagnation (Schiopu et al. 2007; Tiruta-Barna and Barna, 2013).

Recycling Concrete Facility Effluent in Washington State

Facilities permitted under the general permit may discharge treated effluent water that does not cause or contribute to a violation of the State water quality criteria, sediment management standards, or 40 CFR 131[53]. For compliance, water discharged from facilities must be at or below the effluent limits for the parameters listed in the general permit for the activity performed at the facility. Regular monitoring is performed on stormwater/process water inputs to groundwater and surface water at permitted facilities for compliance to the general permit[54].

Concrete recycling best management practices (BMPs) must be included in the facility's Stormwater Pollution Prevention Plan (SWPPP). Facilities that did not have permit coverage prior to April 1, 2016 must not place concrete stockpiles:

- Within 100 feet (horizontal distance) of the high water mark of nearby surface water bodies.
- Within 100 feet of a drinking water or irrigation well(s) or a Wellhead Protection Area, unless groundwater is sampled using an Ecology-approved groundwater monitoring program.
- Where there is discharge to ground unless the bottom of the stockpile is separated from the groundwater surface by a minimum of 10 feet[55].

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[53] The *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* 2021 section being cited here is S3.B. Also, 40 Code of Federal Regulations (CFR) 131: Water Quality Standards; Available here.

[54] The *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* 2021 sections being referenced here is S2 Table 2 and S4.

[55] The *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* 2021 section being cited here is S8.F.

Any water discharge to an unlined pond, lagoon, or other type of impoundment is considered a discharge to groundwater. Water ponding at a permitted facility may be considered a discharge to groundwater[56]. In addition, subsurface discharge of wastewater must comply with the Underground Injection Control Program regulations (Chapter 173-218 WAC)[57].

Material acceptance procedures at the facility should ensure that inbound RCA is not a source of dangerous waste (e.g., lead paint, asbestos, polychlorinated biphenyls).

For the Construction Stormwater General Permit, the monitoring of effluent pH must be performed on the stormwater runoff if specific conditions are met (Ecology, 2020).

Re-use of Concrete Material

Concrete material's initial use should always be considered when attempting to re-use the material. The initial use the concrete material (e.g., road, factory, bridge, dam) may expose the material to additional POC not considered in this literature review. Concrete material's second use includes use as a landfill material, bound and unbound aggregates in road projects, and other infrastructure projects (Verian et al. 2013; FHWA, 2018).

The use of RCA has been increasing internationally and domestically. The increase is suggested to be due to numerous studies evaluating the mechanical strength, durability, and environmental effects of recycled aggregate concrete (RAC) and RCA re-use as a base course material: Bestgen et al. 2016b; Bozyurt et al. 2012; Cabrera et al. 2019; Chen et al. 2019; Chidiroglou et al. 2008; Dhir et al. 2019; Junak and Sicakova, 2017; Li et al. 2008, 2009; Malešev et al. 2010; Puthussery et al. 2017; Thomas et al. 2018; WSDOT, 2009, 2014.

A less common application of RCA includes its use to remediate acid mine drainage issues. In column leaching tests[58], the pH of the acid mine drainage increased to above 11 S.U and reduced the concentrations of iron, copper, and zinc in the acid mine drainage leachate (Jones and Cetin, 2017).

The different re-uses of RCA (i.e. back-fill, construction, road-base) may present unique but similar regulatory challenges. For example, the stockpiling concrete recycling facility with properly implemented BMPs will present different environmental risks than stormwater runoff from a construction site.

Ecology encourages the use of RCA to reduce greenhouse gas emissions, lessen the disposal in landfills, and preserve virgin aggregate sources, as well as for economic reasons.

Washington State Department of Transportation's (WSDOT) use of Recycled Concrete Aggregate

Recycled concrete materials are referred to as recycled concrete aggregate (RCA) (WSDOT, 2018b). According to RCW 70A.205.700, Washington State uses the definition of recycled concrete aggregates from the Federal Highway Administration (FHWA). The FHWA defines RCA as purposefully crushed concrete to create aggregates at the desired gradation for re-use (FHWA, 2004, 2015). WSDOT (2020) also defines RCA as coarse and fine aggregates manufactured from hardened concrete mixtures.

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[56] The *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* 2021 section being cited here is S3.H

[57] Chapter 173-218 WAC: Underground Injection Control Program.

[58] A column leaching test is designed to obtain liquid-solid partitioning information on the release of pollutants of concern as a function of the liquid-to-solid (L/S) ratio.

Washington State has superior quality virgin natural aggregates, which contributes to the superior quality of RCA (Dhir et al. 2019; WSDOT, 2014). The State's re-use of RCA in the production of RAC or road sub-base protects dwindling natural aggregates resources, reduce costs and carbon dioxide emissions, and reduces landfill disposal (Bozyurt et al. 2012; Del Ponte et al. 2017; WSDOT, 2014). A 21% economic cost saving was recognized in the re-use of RCA compared to natural aggregates (Lee et al. 2010).

The Washington State Legislature promulgated RCW 70A.205.700 and RCW 70A.205.705[59] in 2015 through Engrossed Substitute House Bill 1695[60]. RCW 70A.205.700 requires the WSDOT to work collaboratively with contractors to propose strategies to increase the re-use and recycling of construction aggregates. This regulation requires WSDOT to use a minimum of 25% RCA on its cumulative transportation infrastructure projects, unless RCA is not readily available and/or cost-effective.

The WSDOT provides a Qualified Products List, QPL, detailing the locations of pre-approved and evaluated sources of RCA. The QPL was created to aid, promote, and encourage the usage of RCA by the construction industry.

The WSDOT requires all recycled materials used in the State to pass the toxicity characteristic leaching procedure, TCLP or Method 1311, for lead. Certification is required indicating the recycled materials are not considered a dangerous waste by the State (Chapter 173-303 WAC). Additionally, WSDOT requires all RCA used on State projects to contain an aggregated weight of less than 1% of adherent fines, vegetable matter, plastics, plaster, paper, gypsum board, metals, fabrics, wood, tile, glass, asphalt (bituminous) materials, brick, porcelain, or other deleterious substance(s) not otherwise noted (WSDOT, 2020). For a full list of requirements for the usage of RCA, see WSDOT Standard Specification Guidelines (WSDOT, 2020).

RCW 70A.205.705, which expired on July 1, 2021, required WSDOT to submit an annual report to the State Legislature on the usage of RCA and implementation of RCW 70A.205.700. According to WSDOT annual usage reports, at least since 2017 the department has not been able to meet the 25% usage requirements outlined by the legislature. In 2017, 2018, and 2019, the cost of transporting the recycled aggregates to the project site was identified as the greatest reason for not meeting the 25% usage requirement. A small fraction of projects identified the use of RCA below the ordinary high water mark as another reason for not meeting the 25% RCA usage (WSDOT, 2018a, 2018b, 2019).

The lack of RCA use is also potentially due to the unfamiliarity with the technical requirements, performance, and durability of the RAC (FHWA, 2018). The WSDOT plans to continue recommending the use of RCA in construction projects through educational outreach to local agencies and the approval of materials on the QPL (WSDOT, 2019). Table 2 displays the maximum percentage of RCA allowable in various applications according to WSDOT Standard Specification Guidelines (WSDOT, 2020).

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[59] Washington State RCW 70A.205.705: Solid Waste Management – Reduction and Recycling: Report to the legislature.

[60] Engrossed Substitute House Bill 1695 – 2015-16: Establishing a priority for the use, reuse, and recycling of construction aggregate and recycled concrete materials in Washington. This bill resulted in RCW 70.95.800/805, which were later renamed RCW 70A.205.700/705, respectively.

Table 2: The maximum allowable replacement amount of recycled concrete aggregate according to the Washington State Department of Transportation Standard Specification Guidelines (WSDOT, 2020).

Purpose End-Use	WSDOT Standard Specification Section[1]	Maximum Allowable Percent (by weight) of Recycled Concrete Aggregate (%)
Coarse Aggregate for Commercial Concrete and Class 3000 Concrete	9-03.1(4)	100
Ballast	9-03.9(1)	100
Permeable Ballast	9-03.9(2)	100
Crushed Surfacing	9-03.9(3)	100
Aggregate for Gravel Base	9-03.10	100
Gravel Backfill for Foundations – Class A	9-03.12(1)A	100
Gravel Backfill for Foundations – Class B	9-03.12(1)B	100
Gravel Backfill for Walls	9-03.12(2)	100
Gravel Backfill for Pipe Zone Bedding	9-03.12(3)	100
Gravel Borrow	9-03.14(1)	100
Select Borrow	9-03.14(2)	100
Select Borrow (greater than 3 feet below Subgrade and side slopes)	9-03.14(2)	100
Common Borrow	9-03.14(3)	100
Common Borrow (greater than 3 feet below Subgrade and side slopes)	9-03.14(3)	100
Foundation Material Class A and Class B	9-03.17	100
Foundation Material Class C	9-03.18	100
Bank Run Gravel for Trench Backfill	9-03.19	100
Fine Aggregate for Concrete	9-03.1(2)	0
Coarse Aggregates for Concrete	9-03.1(4)	0
Aggregates for Hot Mix Asphalt	9-03.8	0
Gravel Backfill for Drains	9-03.12(4)	0
Gravel Backfill for Drywells	9-03.12(5)	0
Backfill for Sand Drains	9-03.13	0
Sand Drainage Blanket	9-03.13(1)	0

[1] See the Washington State Department of Transportation's Standard Specifications for Road, Bridge, and Municipal Construction 2021 (WSDOT, 2020).

Global Carbonation of Cementitious Materials

In 2019, the average carbon dioxide concentration present in ambient air was nearly 410 parts per million (ppm). The atmospheric carbon dioxide concentration climbed at an average of 2.5 ppm per year from 2018 to 2019 (Blunden and Arndt, 2020). An increase in the atmospheric concentration of carbon dioxide results in an increase in the degree of carbonation of the cementitious material (Lagerblad, 2005).

The worldwide manufacturing of cement produces at least 5% of global anthropogenic carbon dioxide emissions (Boden et al. 2017; Rodrigues et al. 2017). Almost 60% of carbon dioxide emissions are sourced from the calcination of the raw materials to produce portland cement

(Hjelmar et al. 2016). The remaining carbon dioxide emissions originates from the fuel burned to produce the heat required for calcination of the raw material (Andrew, 2018; Hewlett and Liska, 2019).

Theoretically over the life-span of the material, the carbon dioxide released during the calcination process reabsorbs to the cementitious matrix, as calcium carbonate (Nielsen and Glavind, 2007; Engelsen et al. 2005). The carbonation rate onto the cementitious matrix of the concrete slows over time, but increases substantially after the concrete is rubbilzed (Engelsen et al. 2005; Hewlett and Liska, 2019; Maia et al. 2018).

Upon reaction with carbon dioxide, between 50 and 90% of the calcium oxide present on the RCA is converted to calcium carbonate over the life-span of the concrete (AzariJafari et al. 2021). The results of a 100-year perspective model for the carbonation of a Danish concrete during original use and after demolition is shown in Figure 8.

Figure 8 is shown for illustrative purposes and displays a chosen set of concrete recycling parameters in the model. See the Carbonation of Cementitious Materials section for important factors affecting RCA carbonation.

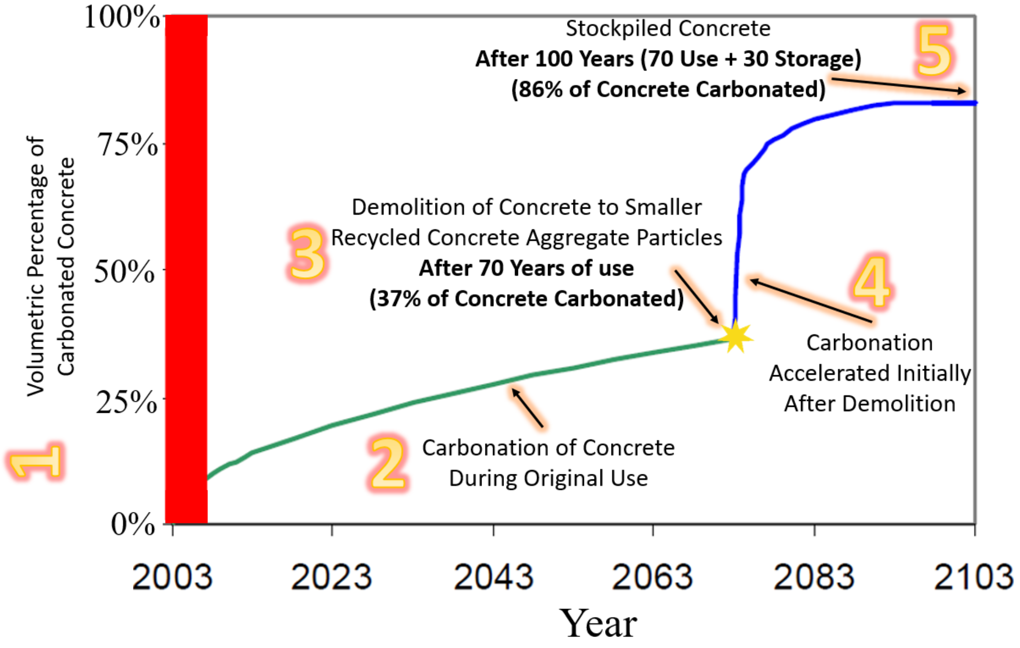

Figure 8: The model results for a 100-year perspective study for the uptake of carbon dioxide by Danish concrete.

Modified from Pade and Guimaraes, (2006).

In Step 1: The volumetric percentage of carbonated concrete, Step 2: Adsorption rate of carbon dioxide during initial use of the concrete, Step 3: Demolition of the concrete to smaller particles increases surface area and allows (Step 4) the rate of adsorption of carbon dioxide to initially increase, Step 5: Carbonation slows with time and the model indicates 86% of the Danish concrete will be carbonated after 100 years (Modified from Pade and Guimaraes, 2006; results depend on model inputs chosen).

A nationwide assessment of the sequestration onto concrete materials in the United States pavement network showed 5.8 million metric tons of carbon dioxide within 30 years. The assessment shows the concrete sequesters 52% of the carbon dioxide after being demolished. If the demolished concrete was stockpiled for an additional 30 years, the concrete could capture up to 11.8 million metric tons (AzariJafari et al. 2021).

The sequestration of carbon dioxide onto cementitious material has been studied extensively in the literature (AzariJafari et al. 2021; Butera et al. 2015a; Engelsen et al. 2005, 2009, 2012, 2017; Galan et al. 2010; Hewlett and Liska, 2019; Houst and Wittmann 2002; Kikuchi and Kuroda, 2011; Kosmatka and Wilson, 2016; Lagerblad, 2005).

Physical Properties of RCA

The physical properties of stockpiled RCA vary significantly at facilities depending on the concrete recycling processes employed. The chemical and physical heterogeneous nature of RCA encountered at Washington State concrete recycling facilities makes it difficult to assign a singular value to the physical properties.

The original water-to-cementitious material (*w/cm*) ratio used when the concrete is manufactured controls several RCA physical properties. The *w/cm* ratio significantly affects the concrete porosity, permeability, and density. In addition, the *w/cm* ratio affects the amount of chemically bound water in the concrete and the amount of unreacted cement present in the RCA (Sanger et al. 2020).

The physical properties of RCA influence the release of pollutants of concern (POC) into the leachate (Bestgen et al. 2016a, 2016b; Coudray et al. 2017; Engelsen et al. 2010; Maia et al. 2018). The RCA particle size has been investigated by numerous authors as a factor for the release of POC from RCA (Bestgen et al. 2016a, 2016b; Chen et al. 2012; Engelsen et al. 2009, 2010; Coudray et al. 2017; Zhang et al. 2018). Other physical properties, i.e. porosity, permeability, of RCA may affect the release of POC, but are seldom reported in the available literature.

This physical properties section of RCA will examine the particle size, shape, porosity, water adsorption, permeability, and different types of densities used to describe RCA in the reviewed literature.

Size, Shape, and Density

Numerous studies indicate there is a direct relationship between the size and shape of the RCA particle to its density (Chidiroglou et al. 2008; de Juan and Gutiérrez, 2009; Galvín et al 2014a). The size and shape of the RCA particles is controlled by the type of crusher used and the maximum crushing diameter determined by the user (Coudray et al. 2017; Matias et al. 2013; Silva et al. 2014).

RCA Particle Size and Shape

The size and shape of the aggregate required by the consumer, the composition of the material, and economic concerns dictate the type of crusher used to create aggregates (Lee et al. 2010; Ng and Engelsen 2018; Tam 2008; Youcai and Sheng 2017). According to Dhir et al. (2019), the type of crusher and number of processing stages the facility employs has a substantial influence on the size and shape of RCA.

Matias et al. (2013) recommends the use of a jaw crusher and rotary crusher, in that order, for the best grain-size distribution and particle shape. The use of an impact crusher as a secondary crusher is recommended when the RCA particles require less adhered cement-paste (Silva et al. 2014). Snyder et al. (2018) reports the use of a jaw crusher reduces the amount of fines produced, as compared to core and impact crushers.

The Federal Highway Administration (FHWA) defines a particle with a diameter greater than 0.187 inches, or 4.75 mm (no. 4 sieve), as a coarse-grained aggregate (RCW 70A.205.700; WSDOT, 2009). Fine-grained aggregates are defined as particles finer than 0.187 inch, or 4.75 mm (no. 4 sieve). In this report, the FHWA size classifications will be used to define fine- and coarse-grained RCA, unless stated otherwise. The coarse- and fine-grained RCA are shown in Figure 9.

Figure 9: Coarse-grained (left) and fine-grained (right) recycled concrete aggregate (FHWA, 2004).

RCA Particle Size

It is well documented that finer-grained RCA particles contain a larger proportion of adhered cement-paste, when compared to their coarse-grained counterparts (Coudray et al. 2017; Engelsen et al. 2005, 2009, 2010; Poon et al. 2006). Coudray et al. (2017) attributed the finer-grained particle size (less than 4 mm) as the primary contributor to the alkaline pH of an RCA leachate. Although, Bestgen et al. (2016a) suggest the pH of the RCA leachate is conditioned more on the composition of the material being assessed.

Chen et al. (2012) and Natarajan et al. (2019) assessed three RCA particle size fractions: gravel- (greater than 4.75 mm), sand- (between 4.75 mm and 0.075 mm), and fine-grained (less than 0.075 mm). The authors generally observed higher pH values for the sand and gravel-sized fractions than the fine-grained RCA. The authors attributed the lower pH in the fine-grained RCA to enhanced carbonation of the finer material, which is supported by Engelsen et al. (2005, 2009) and Zhan et al. (2014).

Coudray et al. (2017) draw a relationship between the maximum chosen crushing particle diameter on the jaw crusher and the percent fines in the crushed aggregate. They state as the chosen crushing diameter decreases the cement-paste adhered to the particle increases (Coudray et al. 2017). The authors also state, regardless of the use of an impact or a jaw crusher the production of fine-grained particles, between 0 and 6 mm, is typically 33 to 52% by weight.

It is important to note that, with decreased particle diameter, the volume-to-surface area ratio declines allowing increased adsorption of carbon dioxide (Abbaspour et al. 2016; Chen et al. 2012; Engelsen et al. 2005, 2009; Law and Evans 2013). Law and Evans (2013) state when the surface area of concrete particles increases, the rate of leaching of specific POC will also increase.

Finer grained RCA particles typically experience increased carbonation, higher water absorption, and have a larger cement-paste content than coarser RCA particles (Coudray et al. 2017; Dhir et al. 2019; Engelsen et al. 2005). Coarser grained RCA particles are typically viewed as favorable for the replacement of virgin natural aggregate in recycled aggregate concrete (Gupta et al. 2009, WSDOT, 2014).

RCA Shape

RCA generally have a rougher surface, higher water absorption, and a lower specific gravity compared to virgin natural aggregates (Gupta et al. 2009). The RCA particle has an irregular shape consisting of angular and rough edges presumably caused by the adhered cement-paste and crushing procedure (del Rio Marino et al. 2010; Gluchowski et al. 2019; WSDOT, 2009). The irregular particle shapes and rough, fractured surface of RCA may contribute to a non-Darcian water flow path[61] through the material (Gluchowski et al. 2019). See the Permeability section for further discussion on non-Darcian flow.

Chidiroglou et al. (2007) characterized the shape of two concrete samples from the United Kingdom employing a jaw crusher according to their elongation and flakiness. The two crushed concrete samples assessed were 64 and 74% equidimensional. The researchers rationalize an increase in the angularity and flakiness of the concrete leads to an increase in the equidimensional percentage post-crushing (Chidiroglou et al. 2007). In addition, an increase in the processing steps of the concrete produces fewer angular/flaky particles (Chidiroglou et al. 2007).

Freeze-thaw cycles may affect both the physical and leachability properties of RCA by increasing the surface area through the expansion and contraction of water (Aydilek, 2015).

RCA Density

The commonest characterization method of an aggregate material is the specific gravity (Silva et al. 2014). There are several other methods employed to measure an aggregates' density (Coutinho, 2006; Table 3). The original water-to-cementitious material (*w/cm*) ratio, curing time, and different stressors influence the concrete's density (Silva et al. 2014; Lagerblad and Trägårdh, 1994).

The original *w/cm* ratio influences the density of the original concrete material (Hewlett and Liska, 2019). The *w/cm* ratio also significantly influences the porosity and permeability of concrete, which has an impact on the density of the material (Hewlett and Liska, 2019, Lagerblad and Trägårdh, 1994). The original strength and type of the concrete as well as its use also has some influence over the density of the RCA (Dhir et al. 2019). All of the densities shown in Table 3 are presented to provide a characterization of the heterogeneity of the RCA encountered in the literature.

Table 3: A description of the types of densities encountered in recycled concrete aggregate (RCA) literature (modified from Dhir et al. 2019).

Type of Density	Description of Density
Specific Gravity (SG)	Or *relative density*, is the ratio of the RCA mass to the mass of an equal volume of water.
Bulk Density (BD)	The ratio of the mass of an aggregate sample to its volume. This density includes the inaccessible and accessible pores of the material and the space between the aggregates.
Particle Density (PD)	The ratio of an oven-dried mass of an aggregate to the volume the aggregate occupies. The volume includes the pores inaccessible to water, but ignores the pores accessible to water.
Oven-Dried Density (ODD)	The ratio of the mass of an oven-dried aggregate to the volume occupied underwater, including all of the pores.
Saturated Surface-Dry Particle Density (SSD)	The ratio between the mass of a water-saturated aggregate and the volume the aggregate occupies, including all pores.

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[61] Darcy's Law is valid in the laminar flow region and the effects due to inertia are neglected. Non-Darcian flow occurs at non-linear laminar and turbulent flows (Freeze and Cherry 1979).

Poon et al. (2006) determined the oven-dried density (ODD) and the saturated surface-dry particle density (SSD) of RCA sourced from a demolition waste facility in Hong Kong. The researchers found the RCA particle size varied the RCA ODD and SSD.

They observed the smallest ODD and SSD when the particle size was less than 5 mm (Poon et al. 2006). This observation is in agreement with other authors, who reported the density of RCA increased with increasing particle size (Dhir et al. 2019; Silva et al. 2014; Zhao et al. 2016). Table 4 reports the RCA densities from the literature reviewed.

Table 4: The densities from reviewed literature including specific gravity (SG), bulk density (BD), particle density (PD), oven-dried density (ODD), and saturated surface-dry particle density (SSD).

Type of Density	Result	Unit	Comment	Reference
Specific Gravity (SG)	2.29 – 2.8	-	RCA	*Abbaspour et al. (2016); Aydilek, (2015); Chen et al. (2012, 2013, 2020); Puthussery et al. (2017); Zhang et al. (2018)*
Bulk Density (BD)	1,210 – 1,245	kg/m^3	RCA from 2 waste piles (aged under a year and less than 1 week)	*Saca et al. (2017)*
Particle Density (PD)	2.2 – 2.76	Mg/m^3	RCA	*Arulrajah et al. (2014); Chidiroglou et al. (2007)*
Oven-dried Density (ODD)	2,065 – 2,088	kg/m^3	Mean fine-grained RCA	*Dhir et al. (2019); Silva et al. (2014)*
	2,325 – 2,327	kg/m^3	Mean coarse-grained RCA	*Dhir et al. (2019); Silva et al. (2014)*
Saturated Surface-dry Particle Density (SDD)	2,295 – 2,300	kg/m^3	Mean fine-grained RCA	*Dhir et al. (2019); Silva et al. (2014)*
	2,442 – 2,438	kg/m^3	Mean coarse-grained RCA	*Dhir et al. (2019); Silva et al. (2014)*
	2.35 – 2.47	g/cm^3	Spanish RCA	*Galvín et al. (2013, 2014b)*
	2.14 – 2.40	g/cm^3	Spanish RCA with particle size between 0.063 mm and 4 mm	*Galvín et al. (2014a)*
	1.67 – 2.41	g/cm^3	Spanish RCA with particle size between 4 mm and 31.5 mm	*Galvín et al. (2014a)*

Natarajan et al. (2019) reported a range of specific gravities for the same Minnesota RCA after eight years of deployment as a sub-base previously reported on by Chen et al. (2013). The authors indicated the SG of the RCA decreased from 2.7 to between 2.13 and 2.50. This change was attributed to the carbonation of the RCA over time.

In contrast to Natarajan et al. (2019), Zhao et al. (2016) indicated a non-carbonated, laboratory created RCA is less dense than its artificially carbonated counterpart. The carbonation of the material caused an increase in density, which was attributed to the conversion of portlandite to ultimately calcite (i.e. calcium hydroxide to calcium carbonate).

Porosity, Water Absorption, and Permeability of RCA

The intrinsic permeability and porosity of RCA is dependent on the original water-to-cementitious material (*w/cm*) ratio used in the manufacturing process (Gluchowski et al. 2019; Houst and Wittmann, 2002; Lagerblad and Trägårdh, 1994).

The porosity, n, is defined as the volume occupied by the pores, V_p, over the total volume, V_t, of the material. The void ratio is defined as the volume of the voids over the volume of the solids. The void ratio is related to the porosity through Equation (1), where e is the void ratio.

$$\text{Porosity } (\boldsymbol{n}) = \frac{\mathbf{V_p}}{\mathbf{V_t}} = \frac{\mathbf{e}}{\mathbf{1+e}} \qquad \text{Equation (1)}$$

According to Freeze and Cherry (1979), the parameter known as intrinsic permeability, k in units of squared length, is converted into the hydraulic conductivity, K in units of length over time, using a well-known equation[62]. However, Freeze and Cherry (1979) remark in non-modernized texts the hydraulic conductivity is sometimes referred to as the coefficient of permeability (Gluchowski et al. 2019).

Although RCA permeability is commonly reported in the literature, the proper terminology used by a few researchers is hydraulic conductivity (Arulrajah et al. 2014; Chen et al. 2012, 2013, 2020; Gluchowski et al. 2019; Zhang et al. 2018). In this text, permeability will be used exclusively and the two terms will be considered synonymous.

Water absorption is the measure of the mass of water absorbed to the material after a 24-hour immersion in a water bath (Dhir et al. 2019). In general, RCAs have increased water absorption compared to natural aggregates (Barbudo et al. 2012).

Porosity and Void Ratio

Gómez-Soberón (2002) reported the porosity of natural aggregates to be less than 3%. Gluchowski et al. (2019) authors attribute elevated porosity observed in the RCA, when compared to natural aggregates, to the presence of adhered cement on the RCA (Gluchowski et al. 2019; Dhir et al. 2019). Further increasing the porosity, RCAs are noted to have an internal pore structure (Deshpande and Hiller, 2012). It is noteworthy that only the atmospherically connected pores contribute to the mass transport in the liquid phase of the RCA, whereas the isolated, disconnected pores do not.

Gómez-Soberón (2002) reported an increase in the porosity of RCA as the particle size decreases. The authors assessed two particle size fractions: 1) 5 mm to 10 mm and 2) 10 mm to 20 mm. The porosity increased from 13.42 to 14.86%, as the particle size decreased.

The RCA porosity will increase as the concrete leaches into the aqueous phase (Abbaspour et al. 2016; Choi and Yang, 2013; Märkl, 2018).

Due to an effect called ‘sealing,’ the formation of carbonate minerals will decrease the RCA porosity (Lagerblad, 2005; Engelsen et al. 2005; Papadakis et al. 1989; van der Sloot and Dijkstra, 2004; Van Gerven et al. 2003; Zhan et al. 2014).

The sealing effect is supported by Zhao et al. (2016) and Zhan et al. (2014), who reported a decrease in the porosity of RCA after carbonation. Zhao et al. (2016) reported a porosity of 14.1% for non-carbonated laboratory created RCA. After artificial accelerated carbonation of the RCA, the authors reported a decrease in porosity to 7.8%. The decrease in porosity was attributed to transformation of calcium hydroxide to calcium carbonate (Zhao et al. 2016).

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[62] See Freeze and Cherry (1979) for a discussion on the conversion between *specific* or *intrinsic permeability* and the hydraulic conductivity.

The void ratio of a material is related to the porosity of the material, through Equation (1), and is unit-less. Chen and Brown (2012) suggest freshly crushed concrete has a larger void ratio, or larger porosity, than carbonated RCA material. They state the void ratio observed in their samples of RCA were between 0.3 and 0.4 (Chen and Brown, 2012).

In Chen et al. (2013), the void ratio varied from 0.28 to 0.36 in RCA sourced from across the United States. Gluchowski et al. (2019) observed a void ratio dependent on the particle size, as shown in Table 5. Although no explanation was provided for the particle size–void ratio results.

Table 5: The void ratios and porosities as a function of particle size of recycled concrete aggregate (Gluchowski et al. 2019).

Particle Size	Void Ratio	Porosity (%)
0 - 8 mm	0.386	27.8
0 - 16 mm	0.543	35.2
0.05 - 16 mm	0.656	39.6

Water Absorption

The higher porosity and the residual, adhered cementitious-paste present on the RCA contributes to an increase in the water absorption of RCA compared to natural aggregates (Coudray et al. 2017; Dhir et al. 2019; Engelsen et al. 2010; Silva et al. 2014). Dhir et al. (2019) and Silva et al. (2014) provide a review of water absorption values encountered in the RCA literature. Dhir et al. (2019) updated a database of water absorption results collected by Silva et al. (2014).

A review of statistical water absorption of RCA, in percentage, is shown in Figure 10. The fine-grain fraction of RCA displays a larger water absorption percentage when compared to coarse-grained RCA, likely due to the increase in adhered cement-paste (Arulrajah et al. 2014; Aydilek, 2015; Dhir et al. 2019; Engelsen et al. 2010).

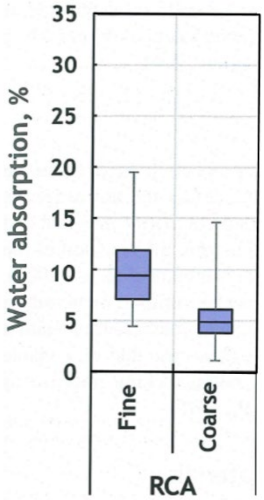

Figure 10: Water absorption of fine- and coarse-grained recycled concrete aggregate particles.
Modified from Dhir et al. (2019).

The water absorption of laboratory created RCA by particle size is observed in Zhao et al. (2016). They studied if the carbonation of the laboratory created RCA influenced the water absorption. They found the water absorption for non-carbonated laboratory created RCA was greater than that of (artificially) carbonated laboratory created RCA for all particle sizes (Zhao et al. 2016).

Permeability

The permeability of water through soils typically obey Darcian, or laminar, flow paths that follow smooth lines called *streamlines* (Freeze and Cherry, 1979). However, not all flow through soils or materials follow a Darcian flow regime. Non-Darcian flow regimes occur when the liquid moving through the material deviates from Darcian streamlines.

The permeability is also influenced by the tortuosity, or a dimensionless ratio of length of the mean actual flow paths to the geometrical length of the flow path. The tortuous flow through concrete material is increased by the internal pore structure of RCA (Deshpande and Hiller, 2012; van der Sloot and Dijkstra, 2004). Additionally, the irregular shapes and rough surface texture creates a non-Darcian flow of water through the RCA (Gluchowski et al. 2019).

The literature reported permeability values of RCA are shown in Table 6. Generally, the coefficient of permeability of RCA ranges several orders of magnitude.

Table 6: The coefficient of permeability values of recycled concrete aggregate, as reported in the reviewed literature.

Permeability Values (m/s)	Reference
3.3 x 10^{-8}	*Arulrajah et al. (2014)*
7.7 x 10^{-6} – 2.6 x 10^{-5}	*Chen et al. (2012, 2013)*
0.8 x 10^{-5} – 1.2 x 10^{-3}	*Chen et al. (2020)*
1.89 x 10^{-5} – 1.02 x 10^{-4}	*Gluchowski et al. (2019)*
2.04 - 2.67 x 10^{-3}	*Poon et al. (2006) and Poon and Chan (2007)*
1.6 x 10^{-5}	*Zhang et al. (2018)*

General Chemical Components in RCA

Major chemical elements in recycled concrete aggregate (RCA) are relatively consistent considering the heterogeneous nature of the RCA. The most abundant elements in RCA are calcium, aluminum, silicon, iron, potassium, sodium, and magnesium. These elements are most likely sourced from the presence of residual, adhered cementitious paste on the RCA (Dhir et al. 2019; Hewlett and Liska, 2019).

Trace elements are observed inconsistently in RCA, i.e. the trace elements vary substantially between RCA samples. The presence of trace elements in RCA may be sourced from exposure to a foreign material, contamination during processing, and/or intrinsic to the raw materials. Trace elements present in RCA include but are not limited to antimony, arsenic, barium, chromium, copper, lead, manganese, molybdenum, nickel, selenium, strontium, and zinc.

This section describes the general consensus for the mineral/chemical components of both non-carbonated and carbonated RCA. The mineral phases containing major and trace elements may transform into carbonate analogues when the RCA is carbonated. Carbon constituents are not discussed in this section. However, the total inorganic carbon content is higher for stockpiled RCA compared to freshly crushed RCA (Dhir et al. 2019).

Cationic-Mineral Components in RCA

The main unreacted calcium-containing cement phases potentially in RCA include, but are not limited to alite (C_3S), belite (C_2S), celite (C_3A), and felite (C_4AF). The calcium-containing mineral phases within RCA are: calcium-silicate-hydrate (*CSH*), portlandite (calcium hydroxide), anhydrite (calcium sulfate), hydroxyapatite ($Ca_5(PO_4)_3(OH)$), calcium carbonate, dolomite ($CaMg(CO_3)_2$, and others (Abbaspour et al. 2016; Bestgen et al. 2016a; Engelsen et al. 2017; Hewlett and Liska, 2019).

The silicon-containing minerals present in/on the RCA include: jennite, tobermorite I, SiO_2, $C_2(A,F)SH_8$, *CSH*, feldspar, and $ZnSiO_3$ (Butera et al. 2015b; Engelsen et al. 2009; Limbachiya et al. 2007). The release of silicon from RCA is controlled by the dissolution of surficial *CSH* and not diffusion from within the concrete (Müllauer et al. 2011).

Aluminum-containing calcium mineral phases control the dissolution of aluminum into the aqueous phase from the RCA depending on the pH of the solution (Engelsen et al. 2009). Aluminum hydroxides and oxides also appear to control the activity of aluminum in alkaline wastes, such as RCA (Abbaspour et al. 2016; Zhang et al. 2018).

The primary mineral phase indicated to be the controlling iron dissolution is hematite, or Fe_2O_3 (Abbaspour et al. 2016). Hydrous ferric oxide (HFO) along with aluminum hydroxides may control the sorption of other POC released from RCA (Ai et al. 2018; Cornelis et al. 2008; Engelsen et al. 2009).

The minerals controlling the release of magnesium from RCA are dolomite ($CaMg(CO_3)_2$) and magnesite ($MgCO_3$), while other minerals containing magnesium may exist: chrysotile, diopside, talc, hydrotalcite, and/or brucite ($Mg(OH)_2$) (Abbaspour et al. 2016; Butera et al. 2015b; Engelsen et al. 2009, 2012). Potassium may exist in cement-paste as potassium oxide, biotite, microcline, illite, muscovite, orthoclase, and syngenite (Abbaspour et al. 2016; Butera et al. 2015b; Engelsen et al. 2009).

The primary barium-containing mineral phases in RCA are barium/calcium/strontium oxymetalates and sulfates, barite ($BaSO_4$), and witherite ($BaCO_3$) (Ai et al. 2019; Butera et al. 2015b; van der Sloot et al. 2000, 2002; Zhang et al. 2018).

Copper may be present in non-carbonated RCA as tenorite (copper oxide), copper hydroxide, and copper ferrite ($CuFe_2O_4$) (Abbaspour et al. 2016; Bestgen et al. 2016; Butera et al. 2015b; Engelsen et al. 2009, 2010). The release of copper from carbonated RCA is predicted to be sourced from malachite ($Cu_2(OH)_2CO_3$), azurite ($Cu_3(OH)_2(CO_3)_2$), and copper carbonate (Abbaspour et al. 2016).

Geochemical results indicate zinc in RCA is mainly controlled by the following zinc-containing minerals: zincite (zinc oxide), zinc hydroxide, willemite (Zn_2SiO_4), calcium zincate ($CaZn_2(OH)_6 \cdot 2H_2O$), and $ZnSiO_3$ (Bestgen et al. 2016a, 2016b; Butera et al. 2015b; Engelsen et al. 2009). It is suggested that the controlling zinc-containing mineral in carbonated RCA may be smithsonite ($ZnCO_3$), with a low solubility in aqueous solutions (Ai et al. 2019).

Nickel hydroxide is considered the dominant mineral controlling the release of nickel from RCA (Butera et al. 2015b; Engelsen et al. 2010).

Both amorphous and crystalline cadmium hydroxide, $Cd(OH)_2$, are potentially formed in RCA (Butera et al. 2015b; Engelsen et al. 2009). The cadmium ions released from RCA will form chloride complexes, $CdCl_2$ and $CdCl^-$, in the presence of excess chlorides in the leachate solution (Engelsen et al. 2012). Manganese may exist as manganite, MnO(OH), in RCA (Butera et al. 2015b; Engelsen et al. 2010).

The release of strontium from concrete appears to leach in a similar manner as barium (Müllauer et al. 2012). That is, the release of strontium appears to be controlled by the dissolution of strontianite, $SrCO_3$ (Müllauer et al. 2012). Other strontium containing minerals potentially existing in RCA include calcium/barium/strontium oxymetalates and sulfates (Butera et al. 2015b; van der Sloot et al. 2000, 2002).

According to geochemical modelling results, the lead-containing complexes potentially found in RCA include lead hydroxides, plattnerite (lead oxide), and lead metalates, such as $Pb_3(VO_4)_2$, $PbCrO_4$, $Pb_3(AsO_4)_2$, and $PbMoO_4$ (Ben Maaouia et al. 2018; Butera et al. 2015b; Cornelis et al. 2008; Engelsen et al. 2010). The formation of lead carbonate, $PbCO_3$, from lead hydroxide, $Pb(OH)_4^{2-}$, is predicted to occur when alkaline material is carbonated (Kosson et al. 2019).

Oliveira et al. (2019) indicate an abundance of spherical titanium oxide (TiO_2) nanoparticles in concrete. The release of titanium from concrete may thus occur as nanoparticles (Oliveira et al. 2019).

Anionic- and Oxyanionic-Mineral Components in RCA

Under elevated salt concentrations, chloride complexes may form with cationic metals in RCA, e.g., cadmium, zinc, copper, and lead (Engelsen et al. 2012, 2017). The source of chlorides in RCA is hypothesized to be from exposure to de-icing salt or salt water.

Sulfur (as sulfate) release from non-carbonated RCA into the aqueous phase is mainly controlled by ettringite and monosulfates. Upon carbonation, gypsum and anhydrite (calcium sulfate) controls the leaching of sulfur (as sulfate). Sulfur (as sulfate) is commonly replaced by oxyanions in several cement-phases, e.g., barium/calcium chromate/sulfate salts (Abbaspour et al. 2016; Barbudo et al. 2012; Butera et al. 2015b; Del Rey et al. 2015).

The oxidation state of the chromium, selenium, vanadium, arsenic, molybdenum, and antimony is dependent on the pH and oxidation-reduction potential conditions of the leachate (Ben Maaouia et al. 2018; Chen et al. 2012; Cornelis et al. 2008; Mulugeta et al. 2011). Generally, these POC leach from RCA predominately in the oxyanionic form for the expected pH range (Mulugeta et al. 2011).

The most stable oxidation states of chromium in RCA is trivalent (Cr^{3+}) and hexavalent (Cr^{6+}) (Ben Maaouia et al. 2018). Since the 2000s, there has been a decline in the use of chromates in construction materials (Del Rey et al. 2015). The addition of reducing agents has aided in the decrease of hexavalent chromium content in concrete (Magistri et al. 2011; Kosson et al. 2014a). Hexavalent chromium is more soluble in aqueous solutions than trivalent chromium (Ben Maaouia et al. 2018; Del Rey et al. 2015).

Hexavalent chromium is generally present in concrete materials as chromate (CrO_4^{2-}) oxyanions (Butera et al. 2015b; Cornelis et al. 2008). Chromate oxyanions may substitute for sulfate anions in several cement-based minerals/phases (Butera et al. 2015b; Cornelis et al 2008; Del Rey et al. 2015; Engelsen et al. 2010).

Geochemical modeling and X-ray diffraction results suggest the chromium-containing minerals in RCA include but are not limited to: $CaCrO_4$, $BaCrO_4$, chromium (III) hydroxide, crocoite (lead chromate), in addition to substitution of chromate into ettringite and *CSH* (Ben Maaouia et al. 2018; Bestgen et al. 2016a; Cornelis et al. 2008; Del Rey et al. 2015; Engelsen et al. 2010).

Pentavalent arsenic (As^{5+}) is the most prevalent speciation in RCA leachate and is generally non-toxic (Sadecki et al. 1996). Pentavalent arsenic is released from RCA as the arsenate (AsO_4^{3-}) oxyanion (Mulugeta et al. 2011).

The substitution of oxyanionic vanadium, primarily vanadate (VO_4^{3-}), for sulfate in ettringite has been well documented (Cornelis et al. 2008; Engelsen et al. 2010; Müllauer et al. 2012; Mulugeta et al. 2011). The hexavalent molybdenum is typically released from RCA in the form of molybdate (MoO_4^{-2}) and may be present in RCA as powellite (calcium molybdate) and lead molybdate (Butera et al. 2015b; Cornelis et al. 2008; Engelsen et al. 2010).

The quadrivalent (Se^{4+}) and hexavalent (Se^{6+}) selenium produce the oxyanionic species selenite, SeO_3^{2-} and selenate, SeO_4^{2-}, respectively, in RCA (Cornelis et al. 2008; Mulugeta et al. 2011). Selenate and selenite may bind with calcium, lead, and barium to produce various hydrated minerals (Cornelis et al. 2008).

The most abundant and mobile oxidation state of antimony in cement-based material is Sb^{5+}, although Sb^{3+} may also be present. The incorporation of antimonite and antimonate, $\mathrm{Sb(OH)}_4^-$ and $\mathrm{Sb(OH)}_6^-$, into cement-hydrate phases such as *CSH*, ettringite, and portlandite (calcium hydroxide) has been reported (Cornelis et al. 2008; Mulugeta et al. 2011). The release of antimony from concrete is related to the decomposition of cement-paste (Magistri et al. 2016).

Phosphorus is often reported and assumed to be from anionic phosphate, PO_4^{3-} (Engelsen et al. 2009). In non-carbonated RCA, geochemical modeling results suggest the mineral hydroxyapatite, $Ca_5(PO_4)_3(OH)$, contains phosphate in concrete (Abbaspour et al. 2016).

Carbonation of Cementitious Materials

Carbonation occurs when carbon dioxide, CO_2, reacts with water to produce carbonic acid, which then interacts with a cementitious material, precipitating carbonates onto accessible cementitious surfaces (Lagerblad, 2005). The carbonation of concrete is a complex exothermic, mainly diffusion-based environmental process, progressing from the exposed surface inwards with time (Ai et al. 2019; Engelsen et al. 2005; Sanger et al. 2020; Walton et al. 1997).

Concrete carbonation occurs during initial use of the material and increases substantially post-demolition (see Figure 8). After demolition, the geometry, shape, and size of the concrete stockpile influences the carbonation experienced by the concrete. Concrete is a finite-sorption media and will absorb a finite amount of carbon dioxide.

Since RCA is considered an alkaline material, the pH of RCA leachate is commonly measured above 10 S.U. with extremes up to 13.2 S.U. (Dhir et al. 2019; Maia et al. 2018; Butera et al. 2014). The observed alkaline pH is primarily due to the dissolution of portlandite, also known as calcium hydroxide, from the cementitious matrix (Engelsen et al. 2012, 2017; Lagerblad, 2005; Sanger et al. 2020). The carbonation of RCA neutralizes the alkaline material, allowing the pH to decrease to near neutral.

The leachate pH affects the aqueous solubility of different mineral phases on the RCA, thus making the pH of the leachant a controlling chemical factor for POC released from RCA. Carbonation of the RCA yields metal carbonates that have different aqueous solubility's than the metal complexes present in the non-carbonated RCA (Ai et al. 2019; Ben Maaouia et al. 2018; Dhir et al. 2019; Engelsen et al. 2010, 2012, 2017; Mulugeta et al. 2011; Sanger et al. 2020; Van Gerven et al. 2003, 2006).

An accurate model for the degree of carbonation RCA has experienced is recommended when conducting a field or laboratory investigation of the RCA pH leachate (Sanger et al. 2020). This is recommended because in early stages (e.g., one to two years) of RCA storage the leachate pH decreases from approximately 13 S.U. to near neutral (Engelsen et al. 2017; van der Sloot et al. 2011).

The chemical and physical parameters of RCA (e.g., particle size, porosity, available calcium (hydr)oxide), and the environmental storage conditions control the degree of carbonation the RCA experiences during initial and end-or-life[63] use (Engelsen et al. 2005, 2009, 2017; Houst and Wittmann, 2002; Natarajan et al. 2019; Sanger et al. 2020; Zhan et al. 2014).

The following section provides background on the carbonation of cementitious materials, such as but not limited to: cement, concrete, recycled concrete aggregate (RCA), and recycled aggregate concrete (RAC)[64]. The chemical and physical heterogeneity of the RCA stored in Washington State must be considered.

The carbonation of cementitious material is described in detail elsewhere, a brief background of the processes affecting the carbonation of cementitious material is provided in this section (AzariJafari et al. 2021; Engelsen et al. 2005, 2009, 2017; Fridh and Lagerblad, 2013; Houst and Wittmann, 2002; Lagerblad, 2005; Papadakis et al. 1989; Sanger et al. 2020).

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[63] "end-of-life", in this report, is the re-use, remanufacturing, recycling, and disposal of recycled concrete aggregate (RCA) materials.

[64] Recycled aggregate concrete (RAC) consists of fresh cement with recycled concrete aggregates as the primary aggregate source; typically requiring additional hydration.

Chemistry of RCA Carbonation

The calcination of limestone produces approximately 60% of carbon dioxide emissions during the production of cement clinker, with the rest of the carbon dioxide emitted from the burning of fossil fuels to heat the kiln (Hjelmar et al. 2016). The cement-phases (e.g., alite and belite) hydrate to form calcium-silicate-hydrate (*CSH*) and calcium hydroxide (as portlandite). These two minerals compose between 70 and 80% of the cement-paste of the RCA (Abbaspour et al. 2016). The original weight percentage of portlandite in a fully hydrated cementitious material is around 15 to 25% (Sanger et al. 2020).

Carbonation leads to surficial chemistry changes to cementitious materials including the degradation of cement phases and transformation to a carbonated counterpart. The surface mineralogy of a non-carbonated cementitious material is largely dominated by calcium hydroxide (as portlandite), calcium-silicate-hydrate (*CSH*), and ettringite in addition to other reacted and unreacted cement phases. Carbonated cementitious surfaces are dominated by calcium, magnesium, and barium carbonates as well as other minor carbonated/non-carbonated cementitious minerals (Ai et al. 2019; Sanger et al. 2020; van der Sloot et al. 2011; Zhao et al. 2016).

One of the most prevalent minerals produced on a carbonated cementitious material is calcium carbonate. The mineral initially forms as aragonite and vaterite but eventually transforms into calcite; the chemical reaction producing calcium carbonate is shown in Equation (5). The aqueous solubility of calcium carbonate is far less than that of portlandite (Sanger et al. 2020). Water acts as a medium for the reaction of carbon dioxide and portlandite (Papadakis et al. 1989). However, if water is present in copious amounts, then the water hinders the transport of carbon dioxide into the cementitious pores (van der Sloot et al. 2011).

$$\mathrm{CO_2}\ (g) + \mathrm{H_2O} \rightarrow \mathrm{CO_2}\ (aq.) \rightarrow \mathrm{H_2CO_3}\ \text{(carbonic acid)} \qquad \text{Equation (2)}$$

$$\mathrm{H_2CO_3} \leftrightarrow \mathrm{HCO_3}^- + \mathrm{H}^+ \qquad \mathrm{p}K_a = 6.35 \qquad \text{Equation (3)}$$

$$\mathrm{HCO_3}^- \leftrightarrow \mathrm{CO_3}^{2-} + \mathrm{H}^+ \qquad \mathrm{p}K_a = 10.33 \qquad \text{Equation (4)}$$

$$\mathrm{CO_3}^{2-}\ (aq.) + \mathrm{Ca}^{2+}(aq.) \rightarrow \mathrm{CaCO_3}(s.) \qquad \text{Equation (5)}$$

The precipitation of calcium carbonate from the consumption of portlandite continues until the available portlandite is consumed (Lagerblad, 2005). Following the consumption of the available portlandite, a pH equilibrium is maintained by the release of calcium cations from *CSH* (Engelsen et al. 2005; Lagerblad, 2005; Sanger et al. 2020).

As the *CSH* phase decalcifies from the release of calcium, the CaO/SiO$_2$ ratio in the *CSH* phase drops (Engelsen et al. 2009, 2017). The elevated silicon percentage in the *CSH* transitions the *CSH* to a highly porous silicon-rich gel. The highly porous silicon-rich *CSH* gel eventually disintegrates during the carbonation process and environmental processes (Abbaspour et al. 2016; Engelsen et al. 2017; Garrabrants et al. 2004).

Severe carbonation of RCA results in a pH neutralization of the alkaline material. In long-term field studies, the neutralization of most of the alkaline material in RCA occurs within one (1) to two (2) years after initial construction (Engelsen et al. 2012; Sadecki et al. 1996; Sanger et al. 2020).

A 90 cm thick uncovered RCA road base without traffic had probes measuring pH of the effluent/leachate following interaction with the RCA road base. The pH runoff measured above 8.5 S.U. for at least the first year as shown in Figure 11 (Engelsen et al. 2012; Mulugeta et al. 2011).

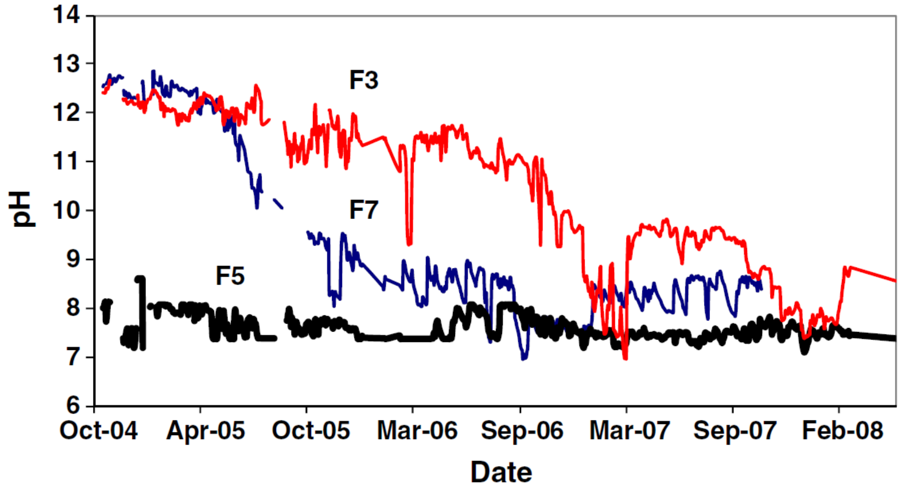

Figure 11: The temporal pH measurements of a pavement test site with uncovered recycled concrete aggregate (F7), asphalt-covered recycled concrete aggregate (F3), and asphalt-covered natural aggregates (F5).
From Engelsen et al. (2012).

The sampling of a roughly 2,000-year-old Roman "cement" aqueduct core was conducted by van der Sloot et al. (2011). The aqueduct core sample is considered fully carbonated and the surface chemistry has changed to predominantly calcium carbonate. Consequently, the leachate pH was near 8 S.U. for all of the samples assessed (van der Sloot et al. 2011).

Depth of Carbonation of Cementitious Material

The carbonation of concrete is described by Fick's first and second law of diffusion (Houst and Wittmann, 2002; Lagerblad 2005; Papadakis et al. 1989). Through the application of Fick's first law, Equation (6) provides the carbonated depth, d_c, in units of length, as a function of exposure time, *t,* in years, and a constant, *k*, in units of length over square root of time (Houst and Wittmann, 2002; Lagerblad 2005).

The constant takes the effective diffusivity coefficient, partial pressure of carbon dioxide in the atmosphere, and the access to reactive compounds in the cementitious material into account (Houst and Wittmann, 2002).

$$\boldsymbol{d_c = k\sqrt{t}} \qquad \text{Equation (6)}$$

Equation (6) was further explored by Lagerblad (2005) for CEM I type concrete without any coating based on the material's strength. In general, the strength of concrete varies inversely with the *w/cm* ratio (Hewlett and Liska, 2019; Kosmatka and Wilson, 2016).

Lagerblad's (2005) suggested *k*-values for concrete by strength are provided in Table 7. The *k*-values indicate a decreasing *k*-value as the strength of the concrete increases and exposure to water saturation increases.

Table 7: The estimated *k*-values for Equation (6) for CEM I type naked concrete surfaces. The *k*-values (in mm/[year$^{1/2}$]) vary depending on the type of exposure to the environment and strength of the material (in megapascal [MPa]).
Adapted from Lagerblad (2005).

	Increasing Strength →			
Type of Storage	**<15 MPa**	**15-20 MPa**	**25-35 MPa**	**>35 MPa**
Wet/Submerged	2 mm/$\sqrt{\text{year}}$	1 mm/$\sqrt{\text{year}}$	0.75 mm/$\sqrt{\text{year}}$	0.5 mm/$\sqrt{\text{year}}$
Buried	3 mm/$\sqrt{\text{year}}$	1.5 mm/$\sqrt{\text{year}}$	1.0 mm/$\sqrt{\text{year}}$	0.75 mm/$\sqrt{\text{year}}$
Exposed	5 mm/$\sqrt{\text{year}}$	2.5 mm/$\sqrt{\text{year}}$	1.5 mm/$\sqrt{\text{year}}$	1 mm/$\sqrt{\text{year}}$
Sheltered	10 mm/$\sqrt{\text{year}}$	6 mm/$\sqrt{\text{year}}$	4 mm/$\sqrt{\text{year}}$	2.5 mm/$\sqrt{\text{year}}$
Indoors	15 mm/$\sqrt{\text{year}}$	9 mm/$\sqrt{\text{year}}$	6 mm/$\sqrt{\text{year}}$	3.5 mm/$\sqrt{\text{year}}$

In addition, other similar models have been reported in the reviewed literature to evaluate the depth and uptake of carbon dioxide into concrete (Andersson et al. 2013; Engelsen and Justnes, 2014).

Factors Impacting Carbonation

Numerous physical and chemical properties and environmental factors influence the rate and depth of carbonation of cementitious composites. These include but are not limited to:

- water-to-cementitious material (*w/cm*) ratio used,
- cementitious material curing conditions,
- type of cementitious materials,
- pre- and post-demolition particle size,
- pre- and post-demolition presence of cracks or damaged zones in the concrete,
- porosity,
- carbon dioxide concentration in the ambient air,
- water content,
- temperature,
- alkali metal content,
- relative humidity,
- environmental storage scenarios, and
- geometry of storage conditions

(ASTM C150/595/1157; Engelsen et al. 2005, 2009, 2017; Hewlett and Liska, 2019; Houst and Wittmann, 2002; Natarajan et al. 2019; Sanger et al. 2020; Stripple et al. 2018; Zhan et al. 2014).

Important Factors of RCA Carbonation

A few major factors influence the uptake of carbon dioxide onto cementitious material. These include the relative humidity and environmental storage conditions, water-to-cementitious material (*w/cm*) ratio used to create the concrete (i.e. effects porosity and permeability), and the particle size and height of the concrete rubble, among others. In addition, the unreacted cement grains carbonate very slowly (Fridh and Lagerblad, 2013).

Crushed RCA in the stockpile self-compacts when exposed to rainfall, preventing the circulation of air into the stockpile. For a RCA stockpile, approximately the outermost foot (~30 cm) of crushed concrete material becomes carbonated (Stripple et al. 2018).

The most commonly reported method for determining the depth of carbonation in a cementitious material is with a colorimetric indicator called phenolphthalein. The indicator is typically sprayed onto a cementitious material and a pink discoloration is observed on the non-carbonated material. The carbonated material does not display a discoloration.

Accelerated artificial carbonation of cementitious materials has been conducted by numerous authors for multiple purposes (Ai et al. 2019; Engelsen et al. 2005; Zhan et al. 2014; Zhao et al. 2016). Artificial carbonation typically occurs in a hermetically sealed vessel with a high-purity carbon dioxide gas constantly or periodically injected. Excess water is commonly avoided when artificially carbonating cementitious materials due to pore-blockage impeding the transport of carbon dioxide (Ai et al. 2019; Zhan et al. 2014).

Relative Humidity

The atmospheric relative humidity[65] where the RCA stockpile is stored is critical to the degree of carbonation the RCA material experiences (Abbaspour et al. 2016; Engelsen et al. 2005; Fridh and Lagerblad, 2013; Hewlett and Liska, 2019; Sanger et al. 2020; Zhan et al. 2014).

The optimum relative humidity for the carbonation of concrete is not agreed upon in the reviewed literature. Generally, the optimum relative humidity is between 40% and 80% (Sanger et al. 2020). The carbonation rate of cementitious material as a function of the relative humidity (in percentage) is shown in Figure 12.

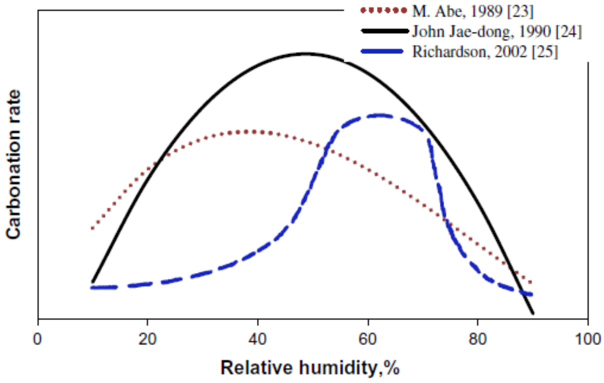

Figure 12: The carbonation rate of cementitious material as a function of the relative humidity (in percentage).
From Zhan et al. (2014).

Environmental Storage Scenarios

The RCA may encounter numerous environmental storage scenarios after demolition. A few of the storage exposure scenarios described in the literature are sheltered or exposed outdoors, indoor (dry), wet/submerged, buried, and intermittent wetting and drying cycles. These storage scenarios will dictate the degree of carbonation the RCA material will experience.

The outdoor or indoor storage of RCA influences the carbonation of the cementitious material. The depth of carbonation increases by a factor between two and four for RCA stored indoors compared to outdoor stored RCA (Hewlett and Liska, 2019).

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[65] Relative humidity is defined as the "dimensionless ratio, expressed in percent, of the amount of atmospheric moisture present relative to the amount that would be present if the air were saturated. Since the latter amount is dependent on temperature, relative humidity is a function of both moisture content and temperature. As such, relative humidity by itself does not directly indicate the actual amount of atmospheric moisture present (NOAA, 2021)"

The rate of carbonation of outdoor stored RCA are subdivided between sheltered and exposed conditions. Exposed storage compared to sheltered storage conditions are expected to be wetter, producing a slower rate of carbonation (Lagerblad, 2005). The carbonation rate of a hardened concrete is decreased from 6.6 to 2.7 mm/year$^{0.5}$ when the concrete surface is exposed to rainfall (AzariJafari et al. 2021).

The rate of carbonation of RCA stored in a dry indoor environment is fast. Although the retardation of RCA stored indoors may be caused by physical contaminants (e.g., paint, carpet, or tiles) on the exposed concrete surface. Indoor storage of RCA may be exposed to higher temperatures than outdoors, allowing for a higher rate of carbonation (Lagerblad, 2005). In addition, the absence for an aqueous phase indoors may 1) allow carbon dioxide access to the RCA pores and 2) not provide a media through which the carbon dioxide can react with the calcium hydroxide (Fridh and Lagerblad, 2013; Papadakis et al. 1989).

If the concrete pores are saturated, submerged, or wet with excess water, the slow transport of carbonate ions to the surface of the RCA limits the degree of carbonation the RCA material experiences. Dry pores are quickly exposed to atmospheric carbon dioxide, but the lack of water slows the formation of calcium carbonate (Papadakis et al. 1989).

When the RCA material is exposed to intermittent wet and drying cycles, the transport of carbonates to the exposed surface of the RCA particle occurs rapidly and in large quantity (Engelsen et al. 2005; Sanger et al. 2020; van der Sloot et al. 2011).

Buried RCA stored underground showed substantial carbonation. The possible explanations for this are the high relative humidity (85%), the organic decomposition occurring, and higher partial pressure of carbon dioxide below ground than normal air (Fridh and Lagerblad, 2013; Lagerblad, 2005).

Water-to-Cementitious Material Ratio

The original water-to-cementitious material (*w/cm*) ratio, defined as the mass of water divided by the mass of cementitious materials, influences several physical, chemical, and structural properties of the hardened concrete (Kamali et al. 2008; Kosmatka and Wilson, 2016). As previously stated, the *w/cm* ratio provides the mean distance between the cement particles in a cement-paste before it begins to harden (Hewlett and Liska, 2019).

A decrease in the original *w/cm* ratio increases the strength and density of the concrete. The *w/cm* ratio controls the concrete porosity and permeability. High *w/cm* ratio concrete have higher porosity than low *w/cm* ratio concrete. Low *w/cm* ratio concrete have more air-accessible porosity than high *w/cm* ratio concrete and contains more non-hydrated cement (Houst and Wittmann, 2002; Sanger et al. 2020).

When the *w/cm* ratio increases the volume of the concrete produced increases, this is shown in Figure 13. The vertical rubber band signifies the same amount of dry cement hydrated by different volumes of water (i.e. different *w/cm* ratio).

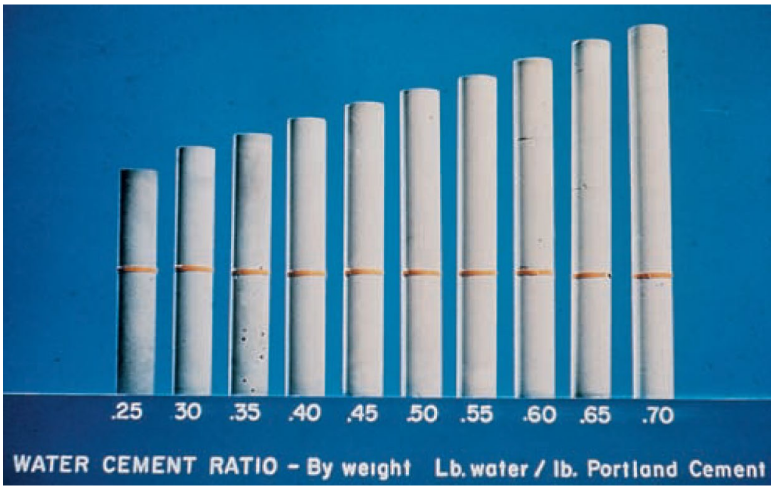

Figure 13: Ten cement-pastes manufactured with same amount of cement with varying water-to-cementitious material ratios from 0.25 to 0.70.
The rubber band indicates the same amount of dry cement added to water at different water-to-cementitious material ratios, thus changing the total volume of the final cement product. From Kosmatka and Wilson, (2016).

Engelsen et al. (2005) suggest a concrete with a low initial *w/cm* ratio yields slow carbonation, while in a high *w/cm* ratio concrete a substantially higher rate of carbonation occurs. As previously mentioned, the sealing effect caused by the formation of carbonate minerals decreases the porosity of the concrete (Engelsen et al. 2005; Lagerblad, 2005; Papadakis et al. 1989; van der Sloot and Dijkstra, 2004; Van Gerven et al. 2003; Zhan et al. 2014; Zhao et al. 2016). However, other authors state that carbonation of concrete material destroys the ettringite crystal bonding between ettringite and *CSH,* allowing for the formation of a less-dense, more-porous concrete (Abbaspour et al. 2016; Majumdar and Stucke, 1981).

Particle Size

The uptake of carbon dioxide is heavily dependent on the particle size of the RCA (Engelsen et al. 2005; Zhan et al. 2014). The rate of carbonation of monolithic cementitious material slows over time, but increases substantially after the cementitious material is crushed (Engelsen et al. 2005; Hewlett and Liska, 2019). Fresh non-carbonated concrete surfaces are exposed by crushing the material during demolition; see Figure 8 (Maia et al. 2018).

The volume-to-surface area ratio is a function of the particle size of the concrete rubble. As the particle size of the concrete rubble decreases the volume-to-surface area ratio decreases, consequently leading to elevated rates of carbonation (Butera et al. 2015a; Law and Evans, 2013). In general, the smaller volume-to-surface area ratio of finer-grained RCA enhances the uptake of carbon dioxide compared to coarser-grained RCA (Engelsen et al. 2005; Nielsen and Glavind, 2007; Zhan et al. 2014).

Additionally, the geometry of a RCA stockpile encountered on a facility will influence the uptake of carbon dioxide by the concrete. A lack of guidance for the shape and size of RCA stockpiles creates greater uncertainty surrounding the uptake of carbon dioxide by concrete stockpiles (AzariJafari et al. 2021).

Other Factors Affecting RCA Carbonation

In 2019, the average global carbon dioxide concentration was greater than 400 parts per million. The carbon dioxide concentration in the atmosphere increased from 2018 to 2019 by 2.5 ppm (Blunden and Arndt, 2020). The partial pressure, or the atmospheric carbon dioxide concentration, has been shown to influence the rate of carbonation of concrete.

If the partial pressure of carbon dioxide increases in the atmosphere, then the rate of diffusion into the cementitious material will also increase (Houst and Wittmann, 2002). If the partial pressure of carbon dioxide in the atmosphere were doubled, the rate of carbonation of cementitious materials would increase by a factor of five (Lagerblad, 2005).

Engelsen et al. (2005) and Lagerblad, (2005) assert the rate of carbonation increases with temperature and vice versa. Sanger et al. (2020) suggests the optimum temperature for the conversion of calcium hydroxide to calcium carbonate is 20°C.

Engelsen et al. (2017) monitored the temperature as a function of depth in the RCA road base for almost a year, shown in Figure 14. The observed thermal gradient from the bottom to top reverses in March to April, around the change of the season.

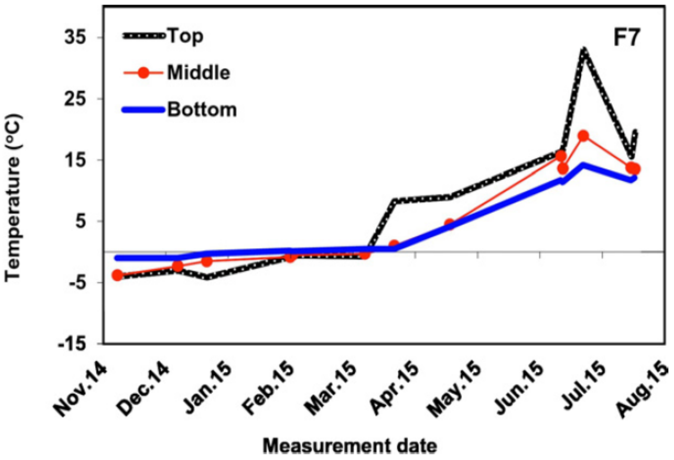

Figure 14: The temperatures (degrees Celsius) measured in the RCA road base not carrying traffic and without asphalt cover (F7) with depth.
Adopted from Engelsen et al. (2017).

Leaching Methodologies

Laboratory leaching methods apply specific leaching conditions that are intended to reflect the leaching scenario the material may encounter in the field and ensure the material field leachate or laboratory eluate[66] meets regulatory compliance (Galvín et al. 2013; Maia et al. 2018; Puthussery et al. 2017). All leaching tests are derived from the two-phase solid-liquid partitioning system, but can incorporate air as the third phase (Chapter 173-340 WAC; Kosson et al. 2014a).

The physical and chemical processes of a leaching test act concurrently on the exposed material to influence the release of pollutants of concern (POC) (Cabrera et al. 2019). Figure 15 shows the chemical and physical processes and the external stresses that influence the field leachate/laboratory eluate POC concentration.

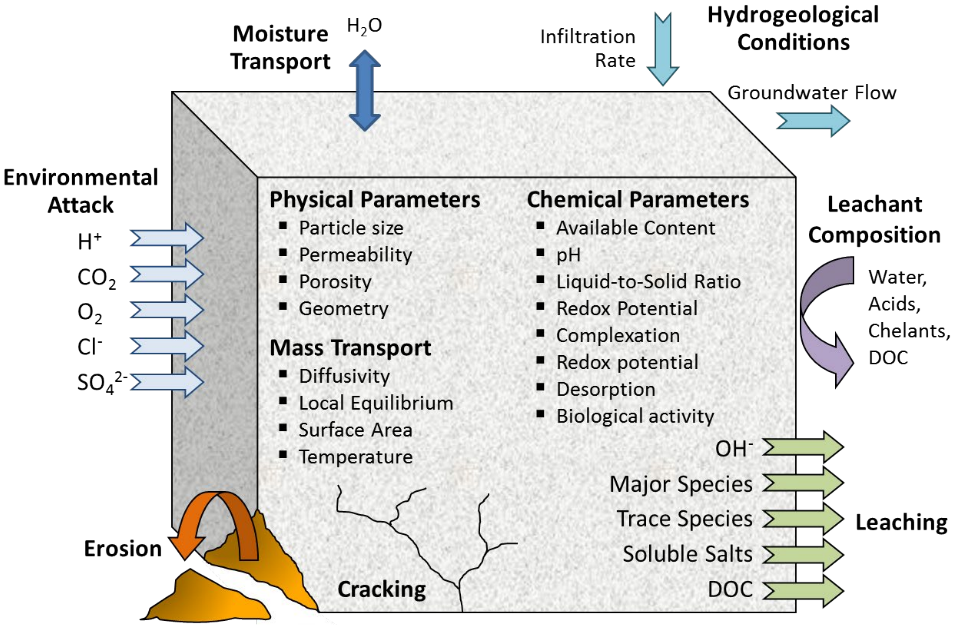

Figure 15: The physical and chemical processes, external stresses, leaching pollutants of concern, and physical degradation a material experiences when leached.
From Vanderbilt, (2020).

Chemical mass transport processes include the dissolution of minerals, degree of carbonation, metal complexation, availability of leachable POC, and sorption onto/off of the RCA matrix. Physical properties of concrete affecting the release of POC include permeability, porosity, particle size, and geometry. Granular and monolithic RCA physical mass transport processes are similar and include advection, surface wash-off ("first flush"), and diffusion.

Diffusion is recognized as the primary mass transport mechanism for specific POC if the RCA is monolithic or coarse-grained (Cabrera et al. 2019). A "first flush" phenomena, or surface wash-off effect, results from the initial infiltration/percolation of liquid, washing off available/soluble materials from the surface (Kosson et al. 2019; Märkl, 2018). The first flush phenomena results in an increase in specific field leachate/laboratory eluate POC concentrations, lasting for a short period after exposure to an aqueous solution (Märkl, 2018;

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[66] From this point forward, this literature review will use the following defined terminology, where "eluate" is used to refer to the solution resulting from a laboratory leaching test and "leachate" is used to refer to the solution collected from/measured in the field following exposure.

Wehrer and Totsche, 2008). An example of a first flush or surface wash-off process is an old-road concrete stockpile releasing elevated concentrations of chloride for around the first month (Sadecki et al. 1996).

The formation of a leached layer is also a possible explanation for the observed decrease in field leachate/laboratory eluate POC concentration as the material ages. A "leached layer" is a layer formed on the RCA particle that is generally depleted of specific available/leachable POC (also called a depletion zone). This generally occurs in the field as the RCA is exposed to rainfall and specific available/leachable POC are released from the material to depletion (Chen et al. 2013; Engelsen et al. 2006, 2012, 2017; Sadecki et al. 1996).

In the reviewed literature, the leaching tests evaluating the POC release from RCA include, but are not limited to: Method 1311, 1312, 1313, 1315, and 1316, as well as U.S. Geological Survey, American Society of Testing Materials, European, Dutch, German, and other non-standard leaching methods.

For more information on leaching methods, see the Previous Leaching Method Literature Reviews section in the Introduction and/or the section Leaching Methods and Ecotoxicity Assessments.

Leachant or Eluent Aggressiveness

The field leachant or laboratory eluent[67] aggressiveness[68] to RCA is an important variable in field and laboratory leaching tests (Kosson et al. 2019). Kamali et al. (2003, 2008) states the aggressiveness of the laboratory eluent(/field leachant) is one of the more important parameters to consider when evaluating the release of POC from the material.

According to the EPA's Leaching Environmental Assessment Framework (LEAF) How-to Guide, the liquid-solid partitioning of POC is influenced by the following factors:

- Field leachant/laboratory eluent pH,
- Liquid-to-solid ratio,
- Oxidation-reduction potential conditions,
- Dissolved organic carbon (DOC) concentration,
- Electrical conductivity and total dissolved solids (TDS), and
- Biological activity (Kosson et al. 2019).

The field leachant/laboratory eluent aggressiveness as well as chemical and physical properties of the concrete influence how the concrete will degrade. A laboratory eluent(/field leachant) with a lower pH and a higher ion concentration gradient between the solid-liquid phases will be more aggressive towards the solid material, in particular more aggressive towards carbonates (Kamali et al. 2008).

It is known that hardened concrete degrades and chemically transforms when exposed to water. The degradation of concrete is accelerated when exposed to an aggressive field leachant/laboratory eluent. Exposure to an aggressive water will cause the cement-hydrate phases present in the concrete to dissolve and possibly precipitate new minerals.

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[67] The term "leachant" refers to the extraction solution prior to a field leaching test of a solid material throughout the rest of this paper. The term "eluent" refers to the extraction solution prior to a laboratory leaching test of a solid material throughout the rest of this paper.

[68] The "aggressiveness" of the field leachant/laboratory eluent to the recycled concrete aggregate is considered high when the chemical gradient between the solid phase material and field leachant/laboratory eluent is also high.

The eluent used in laboratory leaching tests are commonly deionized[69] or pH buffered waters. The initial pH of deionized water is a neutral 7.0 S.U. at 5° C immediately after treatment. Due to the adsorption of atmospheric carbon dioxide into the aqueous phase the pH decreases to as low as 5.8 S.U. (Riché et al. 2006). Although, a study reported the pH of the deionized water used in a laboratory leaching test of RCA to be 8.1 S.U. (Chen et al. 2013).

In personal communication with the authors of Clark et al. (2013), an author stated the pH of the deionized water is easily adjusted through contact with the RCA materials and the low pH of deionized water will not affect the leachability of specific POC (Hee Kang, 2020). Clark et al. (2013) observed a maximum pH of 11.01, 9.57, and 8.30 S.U. for the deionized, low salinity, and high salinity (using artificial sea salt) waters, respectively.

Märkl and Stephan (2016) used deionized and Berlin tap water as the laboratory eluent in a German leaching test, similar to Method 1315, on mortar (fine sand and cement). The Berlin tap water's pH remigrated to a neutral pH after the third elution step, or 11 days of exposure to the mortar (Märkl and Stephan, 2016). The deionized water produced an elevated pH (equal to or greater than 11.5 S.U.) for all elution steps.

The provided explanation is that the elevated ion content of the Berlin tap water allowed for a less aggressive laboratory eluent compared to the deionized water (Märkl and Stephan, 2016).

The presence of chlorides in the laboratory eluent/field leachant will influence the aggressiveness of the eluent/leachant towards the RCA (Bestgen et al. 2016a; Engelsen et al. 2012, 2017). Clark et al. (2013) studied the release of POC from RCA as a function of the laboratory eluent's salinity. The authors show the laboratory eluent salinity will cause varying effects on the release of metals from RCA.

Gascoyne (2002) provides an assessment of the dissolution of cement by groundwater. The authors state the high ionic strength of saline groundwater, particularly if calcium is present in large amounts, will slow down the dissolution rate of the material.

Leachant waters in the field are typically precipitation but may include process and mine dewatering waters. For definitions of stormwater types and wastewaters in Washington State, see the *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* 2021.

Municipal waters and groundwaters commonly have a higher electrical conductivity and potentially a better buffer system than precipitation and deionized waters. Therefore, the municipal waters, groundwaters, and calcium-rich waters may not be as aggressive toward RCA as precipitation and deionized water.

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[69] Deionized water in this paper will refer to deionized, demineralized, distilled, pure, ultrapure, and reagent water as they are processed similarly and will be considered synonymous. The ions in deionized water have been removed by an ion-exchange process; however, this process does not include the removal of dissolved organic compounds. Typically, these waters have a resistivity of at least 18.1 MΩ or an electrical conductivity of less than 0.055 uS/cm (Märkl, 2018).

Leaching of RCA

The leaching results of RCA depends on the type of laboratory leaching method or field collection method[70] used. Laboratory leaching methods are designed to evaluate specific environmental disposal scenarios for the material and compliance to the regulations. Field leaching methods provide a realistic assessment for the release of pollutants of concern (POC) in the materials expected disposal environment.

Batch and parallel batch, column, and lysimeter leaching tests compose the majority of laboratory leaching methods performed on RCA in the literature. Overwhelmingly, batch leaching tests are the most prevalent type of leaching method performed on RCA. The literature seldom investigates the release of POC from RCA in the field or its expected disposal scenario.

Several chemical and physical properties of the RCA and the field leachant/laboratory eluent affect the release of POC from RCA in the reviewed literature. These include the release of the POC from RCA depending on the:

- degree of carbonation or aging in the field or laboratory
- particle size
- curing time

and the release of POC from RCA varying the field leachant/laboratory eluent

- pH
- liquid-to-solid (L/S) ratio
- salinity
- exposure time

The following sections provides a summary of nearly 50 leaching studies of RCA, or alkaline materials. The chemical, inorganic, and organic POC results from laboratory and field RCA leaching studies are presented. When one or more articles disagree with the provided summary, the authors who disagree are noted. The authors may disagree because the RCA material they assessed differed substantially (i.e. particle size, degree of carbonation, intrinsic heterogeneity of the RCA).

Specific leaching POC are removed from the summary tables when the reviewed literature does not provide information on the specific POC. Examples of specific POC removed from the summary tables include but are not limited to silver, thallium, fluoride, germanium, tantalum, yttrium, and beryllium.

Graphical leaching data, not tabulated in the reviewed literature, is not evaluated in this literature review for exceedance of State water quality criteria[71]. Parameters potentially exceeding the State water quality criteria are explained in detail in the following section Environmental Hazards of RCA. The State and/or Federal drinking water standards were not

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[70] A “field collection method” refers to the amount of time between which the leachate sample leaves the recycled concrete aggregate stockpile and is measured or collected for analysis. For example, if a recycled concrete aggregate runoff sample enters a temporary storage container, which is not hermetically sealed and purged of carbon dioxide. Then the RCA leachate (which is no longer in contact with the RCA) will react with carbon dioxide from the atmosphere in the storage container. The effect on the leachate pH and mineralization of pollutants of concern is exacerbated by the number of days the sample is allowed to equilibrate with the atmosphere or the number of days between sampling.

[71] Washington State Chapter 90.48 RCW: Water Pollution Control, Washington State Chapter 173-201A WAC: Water Quality Standards for Surface Waters of the State of Washington, Chapter 173-200 WAC Water Quality Standards for Groundwaters of the State of Washington, and the *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* 2021.

evaluated in comparison to the POC concentrations released from RCA leaching tests in this literature review.

Chemical Eluates/Leachates from RCA

The chemical parameters of the RCA laboratory eluate/field leachate presented in the reviewed literature include the pH, electrical conductivity, oxidation-reduction potential, total solids (dissolved and suspended), hardness, and turbidity.

State water quality criteria for the chemical POC from RCA are shown in Table 8.

Table 8: The chemical Washington State water quality standards for relevant pollutants of concern.

Chemical Parameter	Washington Groundwater Quality Standards[1,2]	Surface Water Quality Standards[1,3]
pH	6.5 – 8.5 S.U.	6.5 – 8.5 S.U.[4,5]
Total Dissolved Solids (TDS)	500 mg/L	Not Applicable
Turbidity	Not Applicable	50 NTU

1. Effluent limits from the *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* 2021.
2. Washington State Groundwater Quality Standards (Chapter 173-200 WAC).
3. Washington State Surface Water Quality Standards (Chapter 173-201A WAC).
4. The pH shall be within the range of 6.5 to 8.5, with a human caused variation within the range of less than 0.2 units for Char spawning and rearing and core summer Salmonid habitat or 0.5 units for Salmonid spawning, rearing, and migration, Salmonid rearing and migration only, non-anadromous interior Redband trout, and indigenous warm water species in freshwaters of the State (WAC 173-201A-200).
5. The aquatic life pH range for {extraordinary, excellent, good, and fair} quality marine waters is between {7.0 to 8.5 with a human-caused variation within the range of less than 0.2 units, 7.0 to 8.5 with a human-caused variation within the range of less than 0.5 units, 7.0 to 8.5 with a human-caused variation within the range of less than 0.5 units, 6.5 to 9.0 with a human-caused variation within the range of less than 0.5 units}, respectively (WAC 173-201A-200).

pH

The pH is a critical factor in the POC release from RCA (Ai et al. 2019). Carbonation, described earlier, occurs during the aging of RCA and causes the pore solution pH of concrete to decrease from around 13 S.U. to near 8 S.U. (Abbaspour et al. 2016; Cabrera et al. 2019; Engelsen et al. 2017; Kosson et al. 2002; Mulugeta et al. 2011). The pH is defined as the negative logarithm of the hydrogen ion concentration (Chapter 173-201A WAC). Typically, the pH ranges from 0 to 14 S.U., although extremes outside of this range have been noted.

The Sand and Gravel General Permit[72] (general permit) prohibits all permittees with concrete recycling identified as an activity from discharging stormwater, process and dewatering water to groundwater and surface water at a pH less than 6.5 and greater than 8.5 S.U. The Washington State dangerous waste criterion for a solid or aqueous waste is a material with a pH greater than 12.5 S.U., as measured by a pH meter using EPA Method 9040C or Method 9045D (WAC 173-303-090(6)).

There is a discrepancy between the RCA laboratory eluate and the field leachate pH results. This difference may originate from the degree of carbonation the RCA has experienced. Alternatively, the pH of the batch leaching methods are generally higher than column and/or field leaching methods due to particle abrasion exposing fresh/non-carbonated RCA surfaces

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[72] *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* 2021.

(Sanger et al. 2020). Column leaching tests provide a more representative pH to field conditions, because the particle abrasion is typically reduced and wetting and drying cycles may be performed to artificially carbonate the material (Sanger et al. 2020).

Attention to environmental processes capable of reducing the field leachate pH (e.g. carbonation, soil acidity) should be considered when evaluating the environmental impacts of the alkaline pH (Engelsen et al. 2012, 2020; Gupta et al. 2018; Sanger et al. 2020). For instance, the number of freeze-thaw cycles the RCA experiences may decrease the field leachate pH observed. The freeze-thaw cycles may cause water to expand and contract, creating new surfaces on the RCA particle (Aydilek et al. 2015).

The range of the RCA laboratory eluate pH was between 7.74 and 13 S.U. for literature using different batch leaching methods (Butera et al. 2014; Galvín et al. 2014a). Three studies evaluated the pH of RCA material using column leaching tests. These three studies observed an RCA laboratory eluate pH between 7.91 and 13.2 S.U. (Butera et al. 2015b; Del Rey et al. 2015).

For the four studies evaluating the RCA material using a lysimeter leaching method, the laboratory eluate pH ranged from 7.23 to 13.0 S.U. (Butera et al. 2015b; Roque et al. 2016).

Field leaching tests provide the most representative leaching of pH, as these results incorporate the environmental processes the material experiences. The three RCA field leaching studies evaluating the field leachate pH ranged from 7.4 to 12.6 S.U. (Engelsen et al. 2012, 2017; Sadecki et al. 1996). The pH results from the reviewed studies are shown graphically in Figure 16.

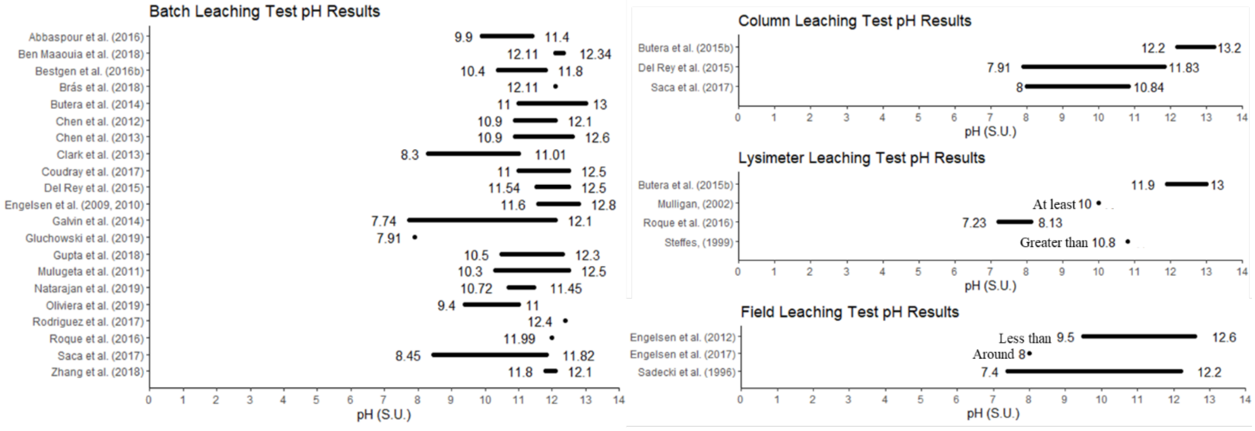

Figure 16: Recycled concrete aggregate pH results, reviewed in this literature review, separated by the leaching method applied.

*The entire reviewed study's pH range in standard units is shown, which **potentially** represents carbonated and non-carbonated RCA samples.*

The acid neutralization capacity of the material is important to know for Method 1313 and other pH-dependent leaching methods. In general, as the RCA's degree of carbonation increases, the amount of acid required to neutralize the alkaline pH decreases (Ai et al. 2019; Ben Maaouia et al. 2018; Bestgen et al. 2016b). In addition, as the particle size decreases the amount of acid required to neutralize the alkaline material per unit mass increases (Chen et al. 2012; Engelsen et al. 2009; Natarajan et al. 2019).

Electrical Conductivity

The electrical conductivity (EC) is a commonly reported parameter. The EC, or specific conductance, is a measure of the solutions' ability to conduct an electrical current at 25°C. This metric reflects the concentration of dissolved ions present in the solution and is correlated to the total dissolved solids (TDS) concentration through various linear/non-linear equations (Rusydi, 2018).

The EC of the laboratory eluate/field leachate can provide important information with respect to the leaching behavior of the cement-based matrix. A moderate relationship between the hexavalent chromium cumulative release ratio and the laboratory eluate EC is observed (Ben Maaouia et al. 2018). The contact time between the laboratory eluent/field leachant and the RCA is suggested to control the EC of the laboratory eluate/field leachate (Abbaspour et al. 2016).

In general, the RCA laboratory eluate/field leachate EC of the batch, column, lysimeter, and field leaching methods were within two orders of magnitude. For all of the leaching methods, the laboratory eluates' EC was between 120 and 11,000 uS/cm for the 15 studies reviewed (Abbaspour et al. 2016; Butera et al. 2015b).

Oxidation-Reduction Potential

Oxidation-reduction potential is not currently regulated as a groundwater or surface water quality criterion. The oxidation-reduction potential is commonly measured in laboratory and field leaching tests, but seldom reported in the literature.

The oxidation-reduction potential, also known as ORP, redox, and Eh, is a measure of the tendency of a chemical species to acquire/lose electrons and thereby become reduced/oxidized, commonly measured in (milli)volts ((m)V). Oxidizing conditions have a positive oxidation-reduction potential, while negative potentials represent reducing conditions. As the oxidation-reduction potential of the field leachant/laboratory eluent changes through exposure to material, the POC (e.g., As, Cr, Se, Mo, Fe, S) speciation may change and affect the solubility and solid-liquid partitioning of the POC (Kosson et al. 2019; van der Sloot and Dijkstra, 2004).

Cement-based materials do not have a substantial amount of compounds capable of being oxidized (Cornelis et al. 2008). Therefore, the oxidation-reduction potential exhibited by cement-based materials during leaching is positive and considered oxidizing (Engelsen et al. 2010). The oxidation-reduction potential of portland cement mortars and concrete from the Netherlands, Norway, and Germany are reported to leach positive, oxidizing conditions (Kosson et al. 2019; Mulugeta et al. 2011).

Abbaspour et al. (2016) reported reducing conditions, or negative oxidation-reduction potential, for laboratory eluates from RCA sourced from northern Virginia. However, through geochemical modeling, the authors suggest the dominant speciation of elements released from the RCA will be in the oxidized form (Abbaspour et al. 2016). The industry may use reducing agents in the concrete mixture to reduce soluble chromates (Magistri et al. 2011).

Butera et al. (2015b) state there is a crucial difference in the oxidation-reduction potential results of field and laboratory RCA experiments. In the field scenarios mild to strong reducing conditions are typically experienced, while laboratory experiments tend to present oxidizing conditions (Butera et al. 2015b).

The oxidation-reduction potential conditions in conjunction with the pH provide an estimate of the valent state of the leaching metal and the mineral present in the RCA (Kosson et al. 2019). The Pourbaix diagram shows the oxidations state of the POC as a function of the oxidation-reduction potential and pH of the field leachate/laboratory eluate (Cornelis et al. 2008). A

Pourbaix diagram is shown in Figure 19, which is discussed in further detail in the Environmental Hazards of RCA section.

Total Solids

The total dissolved solids (TDS) are currently regulated as a secondary groundwater quality contaminant in Washington State (Chapter 173-200 WAC). The groundwater quality criterion for TDS is 500 mg/L. There is no total suspended solid (TSS) or TDS surface water standard in Washington State (Chapter 173-201A WAC).

The general permit requires all permittees with concrete recycling identified as an activity to monitor the effluents of process and mine dewatering water discharging to groundwater for TDS. The limit the general permit allows for the discharge to groundwater of TDS is a concentration of 500 mg/L, measured on a monthly basis. The general permit also requires the effluent of process and mine dewatering water discharging to surface waters to not exceed a TSS concentration of 40 mg/L, measured on a quarterly basis.

The TDS concentration measures the amount of common ions in water and is proportional to the conductivity (Moran, 2018). The TDS concentrations are not typically reported in the reviewed literature.

Rodrigues et al. (2017) reported the TDS release ratio at a L/S ratio of 10 L/kg is 12,000 mg/kg.

Coudray et al (2017) studied the TDS concentration in the laboratory eluate from two leaching tests 1) a standard batch leaching test (with prior particle size reduction to 4 mm) and 2) a non-standard leaching test (without particle size reduction).

The TDS concentration of the RCA laboratory eluate shows a strong correlation to the calcium release ratio regardless of the leaching method (non-standard or standard) and the RCA particle size. Using the non-standard leaching test on RCA, the TDS concentration shows a strong correlation to the sulfate release ratio (Coudray et al. 2017).

The released TDS concentration from the non-standard leaching test decreased with increasing particle size of the RCA. Meanwhile, the standard leaching test suggested the TDS concentration increased with increasing particle size (Coudray et al. 2017).

Sadecki et al. (1996) reported the concentrations of total solids, TSS, total volatile solids, and suspended volatile solids from a coarse-, particles larger than or equal to 4.75 but less than 19 mm, and fine-grained, particles less than 4.75 mm, RCA stockpile field leachate. The total solid concentration decreased with increasing time for both stockpiles. The total solid concentration decreased during the first three months, followed by stabilization of the parameter (Sadecki et al. 1996).

The total solids and TSS concentration from the two stockpiles ranged from 32 to 5,100 mg/L and 1 to 230 mg/L, respectively, for both particle sizes. Sadecki et al. (1996) observed over 97% of total solids leached from the two stockpiles in the dissolved fraction.

Hardness

The water hardness is not currently regulated as a groundwater or surface water quality criterion (Chapter 173-200 WAC; Chapter 173-201A WAC). Although, numerous surface water criteria depend on the water hardness (Ecology, 2019a). The water hardness is a measure of the total calcium and magnesium salts present in the water and is expressed as calcium carbonate, in mg/L of $CaCO_3$ (Chapter 173-201A WAC).

The hardness is not commonly reported in the reviewed literature. Sadecki et al. (1996) reported the summary statistics of water hardness from coarse- and fine-grained RCA stockpile field leachate measured for almost a year. The mean hardness for coarse- and fine-grained concrete was 29 and 31 mg/L of $CaCO_3$, respectively (Sadecki et al. 1996).

Turbidity

Turbidity is not currently regulated as a groundwater quality criterion (Chapter 173-200 WAC). There are surface water criteria for turbidity in fresh and marine waters (Chapter 173-201A WAC). The general permit requires all permittees with concrete recycling identified as an activity to discharge stormwater, process and dewatering water to surface waters at or below 50 nephelometric turbidity units.

Turbidity is defined as the clarity of the water in nephelometric turbidity units (NTU; Chapter 173-201A WAC). Turbidity of RCA field leachate/laboratory eluate is not typically measured or reported in the reviewed literature.

Sadecki et al. (1996) reported the turbidity discharged from a fine-grained, less than 4.75 mm, and coarse-grained, between 4.75 and 19 mm, RCA stockpiles over the course of a year.

The fine-grained RCA stockpile resulted in a broader range, between 1 and 95 NTU, compared to the coarse-grained RCA, between 1 and 28 NTU. However, the average turbidity was 8 and 5 NTU for the fine- and coarse-grained RCA, respectively (Sadecki et al. 1996).

Summary of Chemical Eluates/Leachates from RCA

When reported in the reviewed literature, the effect of the material and/or field leachant/laboratory eluent properties on the chemical RCA field leachates/laboratory eluates are reported in Table 9.

Table 9: A summary of the effect of particle size, liquid-to-solid ratio, curing time, exposure time, salinity, aging, and release with varying pH for the chemical field leachates/laboratory eluates from recycled concrete aggregate.

Physical or Chemical Parameter		pH	Electrical Conductivity (EC)	Oxidation-Reduction Potential (ORP)
Effect of	pH	Not applicable	Increases with increasing acid addition[9]	Increases with increasing acid addition[9]
	Particle Size[1]	Inconclusive	Not reported	Not reported
	Liquid-to-Solid (L/S) Ratio[2]	Decreases with increasing L/S ratio	Decreases with increasing L/S ratio	Not reported
	Curing Time[3]	Decreases with increasing curing time	Not reported	Not reported
	Exposure Time[4]	Decreases with increasing exposure to laboratory eluent	Not reported	Not reported
	Salinity[5]	Decreases with increasing salinity	Not reported	Not reported
	Aging[6, 7]	Decreases with increasing aging	Initial decrease with increasing aging, followed by stabilization	Inconclusive[10]
	Field Aging[8]	Decreases with increasing field aging	Initial decrease with increasing field aging, followed by stabilization	Inconclusive[10]

1. Release of parameter as a function of the particle size; from Coudray et al. (2017); Zhang et al. (2018); Bestgen et al. (2016a, 2016b); and Poon et al. (2006).
2. Only discrete release of the parameter as a function of liquid-to-solid ratio is used; from Gupta et al. (2018); Butera et al. (2015b); and Bestgen et al. (2016a, 2016b).
3. Curing period is the process of hydrating and maintaining the moisture content in freshly cast concrete; from Bestgen et al. (2016b).
4. Exposure time to the laboratory eluent; from Bestgen et al. (2016a).

5. The effect of the laboratory eluent's salinity on the release of the parameter; from Clark et al. (2013); Engelsen et al. (2012, 2017); and Bestgen et al. (2016a).
6. Release of the parameter as a function of the degree of carbonation; field tests are not included in this assessment due to formation of a leached layer, or depletion zone, and other unaccounted for environmental factors.
7. Identified using Ai et al. (2019); Abbaspour et al. (2016); Bestgen et al. (2016a); and Mulugeta et al. (2011).
8. From Engelsen et al. (2012, 2017); Sadecki et al. (1996); and Mulugeta et al. (2011).
9. From Natarajan et al. (2019).
10. In the field scenarios, mild to strong reducing conditions are typically experienced, while laboratory experiments tend to present oxidizing conditions (Butera et al. 2015b).

Inorganic Eluates/Leachates from RCA

Inorganic pollutants of concern (POC) are the most frequently reported constituents released from RCA. Around two dozen inorganic POC released from RCA are reported at least once in the reviewed literature. The chemical and physical parameters affecting the release of inorganic constituents from RCA, if described in the reviewed literature, are provided in the following sections.

The Washington State surface water aquatic life criteria for the relevant inorganic parameters released from RCA are provided in Table 10. The resulting surface water quality criteria determined by the equations provided in WAC 173-201A-240 will not be assessed in this literature review. State surface water quality criteria for human health criteria for consumption of water & organisms as well as the groundwater quality criteria are provided in Table 11 for the inorganic parameters relevant to RCA (Ecology, 2019a; Chapter 173-201A WAC).

Table 10: The inorganic Washington State surface water quality aquatic life criteria for freshwater and marine water for relevant pollutants of concern.

Inorganic Parameter	Aquatic Life Criteria - Freshwater[1]		Aquatic Life Criteria - Marine Water[1]	
	Acute	Chronic	Acute	Chronic
Arsenic	360 ug/L	190 ug/L	69 ug/L	36 ug/L
Cadmium	Equation[2]	Equation[2]	42 ug/L	9.3 ug/L
Chloride (dissolved)[3]	860 ug/L	230 ug/L	None Available	None Available
Chromium (trivalent)	Equation[2]	Equation[2]	None Available	None Available
Chromium (hexavalent)	15 ug/L	10 ug/L	1100 ug/L	50 ug/L
Copper	Equation[2]	Equation[2]	4.8 ug/L	3.1 ug/L
Lead	Equation[2]	Equation[2]	210 ug/L	8.1 ug/L
Mercury	2.1 ug/L	0.012 ug/L	1.8 ug/L	0.025 ug/L
Nickel	Equation[2]	Equation[2]	74 ug/L	8.2 ug/L
Selenium	20 ug/L	5 ug/L	290 ug/L	71 ug/L
Silver	Equation[2]	None Available	1.9 ug/L	None Available
Zinc	Equation[2]	Equation[2]	90 ug/L	81 ug/L

1. Washington State Surface Water Quality Standards; see Ecology (2019a) or WAC 173-201A-240 for details on the criteria.
2. The equation is defined in Ecology (2019a) or WAC 173-201A-240.
3. The criterion based on dissolved chloride in association with sodium. This criterion probably will not be adequately protective when the chloride is associated with potassium, calcium, or magnesium, rather than sodium.

Table 11: The inorganic Washington State groundwater quality standards, surface water quality standards for Human Health Criteria for Consumption of Water & Organisms, and Organisms only for relevant pollutants of concern.

Inorganic Parameter	Washington State Groundwater Quality Standards[1]	Washington State Surface Water Quality Standards for Human Health Criteria for Consumption of Water & Organisms[2]	Washington State Surface Water Quality Standards for Human Health Criteria for Consumption of Organisms only[2]
Antimony	None Available	12 ug/L	180 ug/L
Arsenic	0.05 ug/L[3]	10 ug/L	10 ug/L
Barium	1.0 mg/L[3]	None Available	None Available
Cadmium	0.01 mg/L[3]	None Available	None Available
Chloride	250 mg/L[5]	None Available	None Available
Chromium	0.05 mg/L[3]	None Available	None Available
Copper	1.0 mg/L[3]	1,300 ug/L	None Available
Fluoride	4 mg/L	None Available	None Available
Iron	0.30 mg/L[3, 4]	None Available	None Available
Lead	0.05 mg/L[3]	None Available	None Available
Manganese	0.05 mg/L[3, 4]	None Available	None Available
Mercury	0.002 mg/L[3]	0.14 ug/L	0.15 ug/L
Nickel	None Available	150 ug/L	190 ug/L
Selenium	0.01 mg/L[3]	120 ug/L	480 ug/L
Silver	0.05 mg/L[3]	None Available	None Available
Sulfur (as-Sulfate)	250 mg/L[5]	None Available	None Available
Zinc	5.0 mg/L[3, 4]	2,300 ug/L	2,900 ug/L

1. Washington State Groundwater Quality Standards (WAC 173-200-040).
2. Washington State Surface Water Quality Standards; see Ecology (2019a) or WAC 173-201A-240 for details on the criteria.
3. Metals are measured as total metals.
4. Washington State Secondary Groundwater Quality Standards (WAC 173-200-040).

Dependence on pH

The release of inorganic POC from RCA follows one or more common leaching pattern(s) as a function of pH. When the release concentration/ratio is plotted as a function of the pH, it is possible to identify the pH values where the maximum and minimum release of the POC occurs. The release leaching patterns from RCA are cationic, amphoteric, oxyanionic, and highly soluble or constant and are identified in Figure 17 (Dhir et al. 2019; Kosson et al. 2019).

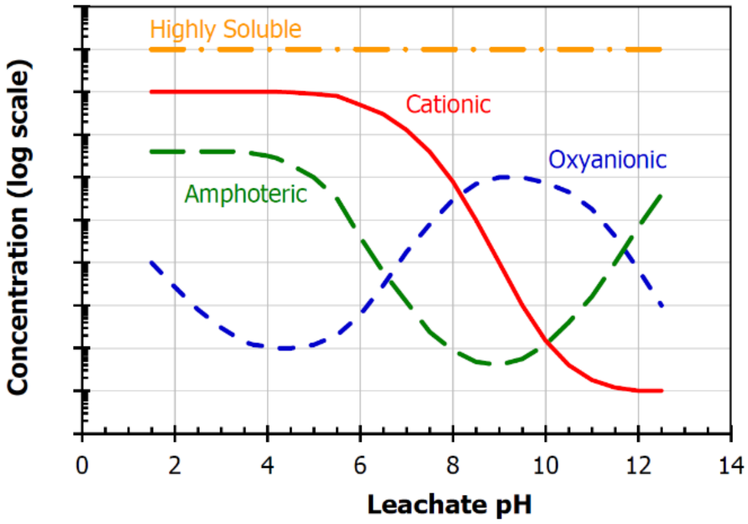

Figure 17: The leaching release patterns dependent on pH for cationic, amphoteric, oxyanionic, and highly soluble pollutants of concern.
Similar to Figure 3 and Figure 4, Figure 17 combines all the possible leaching release patterns for pollutants of concern. Adapted from Kosson et al. (2019).

The highly soluble, or constant, release pattern describes soluble species (e.g., chloride, sodium, selenium) that leach to the point of depletion from the material. The cationic species (e.g., cadmium, nickel, and lead) have a maximum release in the acidic range and a minimum release in the alkaline region. The cationic species behaves as such in RCA due to binding to hydrate phases in RCA.

Amphoteric species (e.g., copper, zinc) have a similar maximum to the cationic species pattern. The amphoteric species' minimum release from the material is located in the mid-alkaline range and increases with increasing pH. In cement systems copper and zinc behave as a cationic species due to binding to the hydrate phases, i.e. they are not controlled by hydroxides at high pH.

Oxyanionic release patterns commonly have a maximum in the neutral to mid-alkaline range and minima in the acidic regime. The oxyanionic species released from RCA include but are not limited to chromate (CrO_4^{2-}), vanadate (VO_4^{3-}), and arsenate (AsO_4^{3-}).

The pH dependent release leaching pattern of inorganic POC from RCA and identified pH ranges, where a maximum and minimum release occurs, is shown in Table 12. Also shown in Table 12 is the dominant oxidation state of the inorganic POC released from and/or present in the RCA.

Table 12: A summary of the leaching pattern as a function of pH, the pH where the maximum and minimum release occurs, and dominant oxidation state for the inorganic pollutants of concern from recycled concrete aggregates.

Inorganic Parameter	Leaching Pattern as a Function of pH[1,2]	pH Where Maximum Release Occurs (S.U.)[1,2]	pH Where Minimum Release Occurs (S.U.)[1,2]	Dominant Oxidation State in Recycled Concrete Aggregate[3]
Aluminum	Amphoteric	2 – 3	5 – 9	Al^{3+}
Antimony	Not reported	Not reported	Not reported	Sb^{5+}
Arsenic	Highly Soluble/Constant/Cationic	1 – 4	6; 10 – 12	As^{5+}
Barium	Cationic	3 – 4	12 – 13	Ba^{2+}
Cadmium	Cationic	1 – 3	Greater than 7	Cd^{2+}
Calcium	Cationic	1 – 6	13 – 14	Ca^{2+}
Chromium	Amphoteric/Oxyanionic	1 – 3; 9 – 13	5 – 6.5	Cr^{6+}; Cr^{3+}
Cobalt	Amphoteric	3 – 4; 11 – 13	8 – 10	Not reported
Copper	Amphoteric/Cationic	1 – 3	7 – 10	Cu^{2+}
Iron	Amphoteric/Cationic	1 – 2	7 – 12	Fe^{3+}
Lead	Amphoteric/Cationic	1 – 3	6 – 12	Pb^{2+}
Lithium	Cationic	4 – 6	13	Not reported
Magnesium	Cationic	1 – 5	12 – 13	Mg^{2+}
Manganese	Cationic	1 – 3	10 – 12	Not reported
Molybdenum	Amphoteric/Oxyanionic	1 – 2; 6 – 11	4	Mo^{6+}
Nickel	Amphoteric/Cationic	1 – 6	10 – 13	Not reported
Phosphorus	Not reported	Not reported	Not reported	PO_4^{3-}
Selenium	Highly Soluble/Constant	Not reported	Not reported	Se^{4+}; Se^{6+}
Silicon	Cationic	1 – 3	11 – 13	Not reported
Strontium	Cationic	4 – 5	11 – 13	Not reported
Sulfur (as-Sulfate)	Not reported	Less than 10	Greater than 10	SO_4^{2-}
Vanadium	Amphoteric/Oxyanionic	1 – 2; 9 – 11	4 – 7	V^{5+}
Zinc	Amphoteric/Cationic	1 – 3	8 – 12	Zn^{2+}

1. Leaching pattern identified in laboratory experiments artificially modifying the pH of the laboratory eluent.
2. From: Abbaspour et al. (2016); Bestgen et al. (2016a, 2016b); Butera et al. (2015b); Chen et al. (2012); Dhir et al. (2019); Engelsen et al. (2009, 2010); Kosson et al. (2014a); and Zhang et al. (2018).
3. Dominant oxidation state for the respective parameter as identified in Abbaspour et al. (2016); Barbudo et al. (2012); Bestgen et al. (2016a); Butera et al. (2015b); Cornelis et al. (2008); Engelsen et al. (2009, 2010, 2012, 2017); Gupta et al. (2018); Mulugeta et al. (2011); and Zhang et al (2018).

Effect of Aging

The effect of aging or carbonation of the material has been noted to have a significant effect on the release of POC from RCA, because it affects the pH of the field leachate or laboratory eluate (Ai et al. 2019; Kosson et al. 2014a, 2019; Mulugeta et al. 2011). The manner in which the RCA is carbonated impacts the chemical, physical, and mechanical properties of the material. The two methods commonly employed in the reviewed literature are accelerated artificial carbonation in the laboratory and natural field exposure to atmospheric carbon dioxide and environmental conditions.

Numerous authors have used accelerated artificial carbonation methods to evaluate the effect of carbonation on the release of POC (Ai et al. 2019; Müllauer et al. 2012; Mulugeta et al. 2011). See the Carbonation of Cementitious Materials section for more detail on artificial carbonation of RCA.

Artificial carbonation results in an increase in density and a decrease in the porosity of RCA as compared to the non-carbonated counterparts (van der Sloot and Dijkstra, 2004; Zhan et al. 2014; Zhao et al. 2016). By comparison, the field carbonation of RCA produced a decrease in the specific density of RCA, which was attributed to the formation of calcium carbonate (Natarajan et al. 2019). The creation of a leached layer, or a depletion zone, on the RCA particle is further attributed to the discrepancies between the laboratory and field release of POC from RCA (Engelsen et al. 2012; Mulugeta et al. 2011).

The difficulty and expense of measuring, incorporating, and reproducing environmental field parameters in the laboratory are recognized as a reason for the discrepancies observed between laboratory and field leaching results (Maia et al. 2018; Sanger et al. 2020; van der Sloot and Dijkstra, 2004).

A few authors have assessed the release of POC from RCA as the material is carbonated by exposure to atmospheric carbon dioxide concentrations on the laboratory bench (Abbaspour et al. 2016; Bestgen et al. 2016a, 2016b). The RCA treated in this manner may be considered "sheltered," as the formation of a leached layer, or depletion zone, does not form.

There are several environmental factors influencing the release of POC from RCA in the field, e.g., thermal and precipitation cycles, partial pressure of carbon dioxide, biological growth, etc. (Aydilek et al. 2015; Engelsen et al. 2006, 2012, 2017). The environmental parameters are seldom studied individually and are commonly applied as a synergistic effect on the release of the POC from RCA.

Table 13 provides a summary for the release of inorganic parameters from RCA as the material ages in the field and in the laboratory.

Table 133: The effect of aging in the field and laboratory on the release of inorganic parameters from recycled concrete aggregate as reported in the reviewed literature.

Inorganic Parameter	Effect of Field Aging[1,2,3]	Effect of Aging in the Laboratory[3,4,5]
Aluminum	Decreases with increasing aging	Inconclusive[7]
Antimony	Increases before decreasing with increasing aging	Increases with increasing aging
Arsenic	Increases before decreasing with increasing aging	Increases with increasing aging
Barium	Not reported	Decreases with increasing aging
Boron	Inconclusive	Not reported
Calcium	Increases with increasing aging	Decreases with increasing aging
Chloride	Decreases with increasing aging	Not reported
Chromium	Increases before decreasing with increasing aging	Increases with increasing aging[8]
Copper	Decreases with increasing aging	Decreases with increasing aging
Iron	Not reported	Undulates with increasing aging
Magnesium	Increases with increasing aging	Increases with increasing aging
Molybdenum	Decreases with increasing aging	Increases with increasing aging[9]
Nickel	Decreases with increasing aging	Not reported
Potassium	Not reported	Generally, decreases with increasing aging
Selenium	Increases before decreasing with increasing aging	Increases with increasing aging[9]
Silicon	Increases with increasing aging[6]	Constant or increasing with increasing aging
Sodium	Decreases with increasing aging	Not reported
Strontium	Not reported	Decreases with increasing aging
Sulfur (as-Sulfate)	Decreases with increasing aging	Increases with increasing aging
Vanadium	Increases before decreasing with increasing aging	Increases with increasing aging
Zinc	Not reported	Decreases with increasing aging

1. Release of the parameter as a function of the time exposed to environmental factors; including but is not limited to the carbonation of the material, formation of a leached layer, natural precipitation cycles, thermal cycles.
2. Identified using Engelsen et al. (2006, 2012, 2017); Mulugeta et al. (2011); and Sadecki et al. (1996).
3. It should be noted that the degree of carbonation, material source, and article-specific objectives differ between the referenced studies.
4. General release of the parameter as a function of the degree of carbonation; field tests are not included in this assessment due to formation of a leached layer, or depletion zone, and other unaccounted for environmental factors.
5. Identified using Abbaspour et al. (2016); Ai et al. (2019); Bestgen et al. (2016a); Engelsen et al. (2009); Müllauer et al. (2012); and Mulugeta et al. (2011).
6. The decalcification of the calcium-silicate-hydrate phase biases the calcium-silicon ratio towards silicon, which results in elevated silicon release at high pH. This is supported by Abbaspour et al. (2016), Chen et al. (2004), Chen et al. (2013), and Engelsen et al. (2009, 2017).
7. Results from Abbaspour et al. (2016) and Ai et al. (2019) suggest a fairly consistent release of aluminum as carbonation increases, while Müllauer et al. (2012) suggests a decreasing aluminum release as carbonation increases.
8. Results from Bestgen et al. (2016a), Müllauer et al. (2012), Mulugeta et al. (2011) differ from Ai et al. (2019), who suggests the release decreases with increasing aging.
9. Mulugeta et al. (2011) differs from Ai et al. (2019), who state the release of the parameter from carbonated recycled concrete aggregate is within the same range as non-carbonated RCA.

Liquid-to-Solid Ratio Dependence

The liquid-to-solid (L/S) ratio is a commonly manipulated variable of a laboratory leaching method. The concentration of inorganic POC released from RCA may increase or decrease as the L/S ratio increases (Gupta et al. 2018; Kosson et al. 2002, 2014a, 2019). Maia et al. (2018) reports a decrease in the heavy metal concentration with increasing L/S ratio is potentially due to the dilution of the laboratory eluate.

The release POC concentration from RCA is measured either discretely or cumulatively as a function of the L/S ratio (Butera et al. 2015b; Gupta et al. 2018). Only discrete POC release trends from laboratory RCA leaching methods are identified as increasing or decreasing with increasing L/S ratio in Table 14. If a POC's trend is marked as inconclusive, a trend could not be established from the provided data in the literature.

In field leaching methods, the L/S ratio is controlled by the volume of precipitation the stockpile receives divided by the total mass of material. The volume of water infiltrating through a 90 cm road base over one year was 6580 L, for an estimated L/S ratio of 0.09 L/kg (Engelsen et al. 2012). At that rate, over the decade the estimated L/S ratio will increase to 1 L/kg (Engelsen et al. 2017).

Table 14: The release of pollutants of concern (as a concentration) from recycled concrete aggregates dependent on the liquid-to-solid ratio of the discrete laboratory leaching method.

Inorganic Parameter	Trend of Liquid-to-Solid (L/S) Ratio[1]
Aluminum	Relatively constant, slight increase with increasing L/S ratio
Antimony	Inconclusive
Arsenic	Inconclusive
Barium	After initial increase, a decrease with increasing L/S ratio
Calcium	Decreases with increasing L/S ratio
Chromium	Decreases with increasing L/S ratio
Copper	Decreases with increasing L/S ratio
Iron	Decreases with increasing L/S ratio
Lithium	Decreases with increasing L/S ratio
Magnesium	Increases with increasing L/S ratio
Molybdenum	Generally, decreases with increasing L/S ratio
Nickel	Generally, decreases with increasing L/S ratio
Phosphorus	Generally, decreases with increasing L/S ratio
Sodium	Decreases with increasing L/S ratio
Strontium	Decreases with increasing L/S ratio
Sulfur (as-Sulfate)	Initial decrease followed by stabilization as L/S ratio increases
Zinc	Inconclusive

[1] Only discrete release of the parameter in a laboratory leaching method as a function of liquid-to-solid ratio is used; from Gupta et al. (2018); Butera et al. (2015b); Chen et al. (2013); and Bestgen et al. (2016a, 2016b).

Particle Size

The effect of the RCA particle size on the release of inorganic POC in laboratory leaching methods has been reported by numerous authors (Bestgen et al. 2016a, 2016b; Chen et al. 2012; Coudray et al. 2017; Zhang et al. 2018;). Several authors suggest the finer-grained, typically less than 4.75 mm, RCA particles have an increased proportion of adhered cement-paste and lower volume-to-surface area ratio (Chen et al. 2012; Coudray et al. 2017; Engelsen et al. 2005, 2010; Natarajan et al. 2019).

The diminution in volume-to-surface area ratio will result in a higher rate of carbonation. The increase in the adhered cement-paste results in an increase in the inorganic POC release for the finer-grained RCA particles compared to the coarser-grained (Bestgen et al. 2016a, 2016b; Chen et al. 2012; Coudray et al. 2017; Sadecki et al. 1996; Zhang et al. 2018).

The shorter distance the contaminant must travel, increased amount of cement-paste, and decrease in the volume-to-surface area ratio are the postulated reasons for the general trend of increasing release of POC from finer RCA particles (Coudray et al. 2017; Galvín et al. 2014a; Law and Evans 2013).

If no other studies exist on how the RCA particle size effects the release of inorganic POC from RCA, summary data collected in a field study is substituted to hypothesize the effect of particle size on the release (Sadecki et al. 1996). The effect of the RCA particle size on the inorganic POC release from RCA is summarized in Table 15.

Table 15: A summary of the effect of the recycled concrete aggregate particle size on the release of the pollutants of concern as shown in the reviewed literature.

Inorganic Parameter	Effect of Particle Size[1]
Aluminum	Decreases with increasing particle size[3]
Arsenic	Inconclusive[5]
Barium	At material pH, increases with increasing particle size
Calcium	Inconclusive[2]
Chloride	Generally, decreases with increasing particle size
Chromium	Generally, decreases with increasing particle size
Copper	Decreases with increasing particle size
Iron	Generally, decreases with increasing particle size[4]
Magnesium	Generally, no difference
Potassium	Generally, decreases with increasing particle size[5]
Silicon	Generally, decreases with increasing particle size
Sodium	Possible increase with increasing particle size[6]
Sulfur (as-Sulfate)	Generally, decreases with increasing particle size
Vanadium	Generally, no difference[6]
Zinc	Decreases with increasing particle size

1. Release of parameter as a function of the particle size; from Bestgen et al. (2016a, 2016b); Chen et al. (2012); Coudray et al. (2017); and Zhang et al. (2018); supplemented with Sadecki et al. (1996).
2. Zhang et al. (2018) did not observe a difference in the amount of calcium leached between the material pH and acidic pH for three grain sizes between 0.075 mm and 75 mm. Coudray et al. (2017) observed mixed results between their two leaching methods. Bestgen et al. (2016a) noted the larger RCA particles leach lower calcium concentrations than the smaller particles, except one. Sadecki et al. (1996) measured higher ranges of calcium from their fine-grained RCA stockpile than their coarse-grained stockpile.
3. Below a pH of 9 S U., the release of aluminum may change (Zhang et al. 2018).
4. Although Bestgen et al. (2016a) did not observe an increase/decrease based on particle size, Zhang et al. (2018) and Bestgen et al. (2016b) suggest an increase in the iron field leachate from finer-grained RCA.
5. The results from Sadecki et al. (1996) disagree with this conclusion.
6. Possible observation from the results of Sadecki et al. (1996).

Curing and Exposure Time and Salinity

The RCA's curing time is an important material property affecting both the strength of the material and the release of the inorganic POC from RCA reused in recycled aggregate concrete (Bestgen et al. 2016b). As previously described, the field leachant/laboratory eluent's salinity may have a substantial effect on the degradation of the cement present in the RCA, controlling the release of POC from RCA (Bestgen et al. 2016a; Clark et al. 2013; Engelsen et al. 2012, 2017). The RCA exposure time to the laboratory eluent appears to have a mixed effect on the four POC assessed (Bestgen et al. 2016a).

The effects of these parameters on the release of inorganic POC from RCA are summarized in Table 16.

Table 16: The effect of curing time for a material incorporating recycled concrete aggregates, exposure time to the laboratory eluent, and the field leachant/laboratory eluent salinity on the release of inorganic pollutants of concern from recycled concrete aggregates.

Inorganic Parameter	Effect of		
	Curing Time[1]	Exposure Time[2]	Salinity[3]
Aluminum	Not reported	Not reported	Generally, release decreases from low saline, high saline, to deionized waters.
Arsenic	Not reported	Not reported	Increases with increasing salinity
Barium	Not reported	Not reported	Generally, release decreases from deionized, high saline, to low saline waters.
Cadmium	Not reported	Not reported	Inconclusive
Calcium	Decreases with increasing curing time	Generally, decreases with increasing exposure to eluent	Increases with increasing salinity
Chromium	Decreases with increasing curing time	Increases with increasing exposure to eluent	Generally, release decreases from low saline, high saline, to deionized waters.
Copper	Decreases with increasing curing time	Decreases with increasing exposure to eluent	Increases with increasing salinity
Iron	Undulates with increasing curing time	Generally, after an initial increase there is a slight stabilization, except for freshly crushed samples	Not reported
Lead	Not reported	Not reported	Generally, release decreases from high saline, deionized, to low saline waters
Manganese	Not reported	Not reported	Increases with increasing salinity
Mercury	Not reported	Not reported	Inconclusive
Zinc	Not reported	Not reported	Generally, release decreases from high saline, deionized, to low saline waters

1. Curing period is the process of hydrating and maintaining the moisture content in freshly cast recycled aggregate concrete (or concrete with recycled concrete aggregates); from Bestgen et al. (2016b).
2. Exposure time to the laboratory eluent; from Bestgen et al. (2016a).
3. The effect of laboratory eluent's salinity on the release of the parameter; from Bestgen et al. (2016a); Clark et al. (2013); Engelsen et al. (2012, 2017).

Organic Eluates/Leachates from RCA

The release of organic laboratory eluates/field leachates from RCA are seldom investigated with respect to the chemical and physical parameters of the leached material or laboratory eluent/field leachant (e.g., eluent/leachant pH, L/S ratio, or degree of carbonation). Organic pollutants of concern (POC) consists of a large category of chemical compounds that have one or more carbon atoms covalently bonded to other elements (i.e. hydrogen, oxygen, nitrogen). Although there are several organic POC present in RCA (e.g., polycyclic aromatic hydrocarbons, polychlorinated biphenyls), they are seldom investigated in the field leachate/laboratory eluate.

The State water quality criteria for organic POC potentially leaching from RCA are shown in Table 17.

Table 17: The Washington water quality standards for relevant organic pollutants of concern.

Organic Parameter	Washington State Groundwater Quality Standards[1]	Washington State Surface Water Quality Standards[2,3]
Phenol	Not Applicable	18,000 ug/L
Polychlorinated Biphenyls	0.01 ug/L	0.00017 ug/L
Polycyclic Aromatic Hydrocarbons (PAHs)	0.01 ug/L[4]	Variable[5]

1. Washington State Groundwater Quality Standards (WAC 173-200-040).
2. The Washington State Surface Water Quality Standards for Human Health Criteria for Consumption of Water & Organisms (WAC 173-201A-240).
3. There may be other Washington State Surface Water Quality Standards other than Human Health Criteria for Consumption of Water & Organisms; see Ecology (2019a) or Chapter 173-201A WAC.
4. The Washington State polycyclic aromatic hydrocarbons (PAH) Groundwater Water Quality Standard is 0.01 ug/L for 15 of the 16 EPA PAH; Benzo(a)pyrene has a standard of 0.008 ug/L (Chapter 173-200 WAC).
5. The Washington State Surface Water Quality Standards range from no criterion to 3,100 ug/L depending on the PAH chemical evaluated; see Ecology (2019a) or WAC 173-201A-240 for more information.

Dissolved Organic Carbon (DOC)

The dissolved organic carbon (DOC) concentration is not currently regulated as a groundwater or surface water quality criterion in Washington State (Chapter 173-200 WAC; Chapter 173-201A WAC).

DOC is the organic matter capable of passing through a 0.45 um filter dissolved in the aqueous phase. Particulate organic matter is the organic matter larger than the 0.45 um (EPA Method 415.3). DOC consists of complex organic molecules with a high affinity to bind to heavy metals. When DOC concentrations are increased in the laboratory eluent/field leachant prior to exposure to the RCA, the eluate/leachate concentrations of specific POC may increase by several orders of magnitude in the alkaline regime (van der Sloot and Dijkstra, 2004).

The leaching of DOC is highly dependent on the pH, ORP, and salt strength of the laboratory eluate/field leachate (van der Sloot and Dijkstra, 2004). The leachable DOC from RCA is observed to decrease rapidly after the first few washes, potentially due to the wash-off phenomena (Märkl and Stephan, 2016).

The DOC laboratory eluate concentration from RCA batch leaching methods range from 1.1 to 30 mg/L (Butera et al. 2014; Rodrigues et al. 2017). The DOC laboratory eluate concentration for RCA column and lysimeter leaching tests range from 4.0 to 5.8 mg/L and

2.674 to 7.0 mg/L, respectively (Butera et al. 2015b; Roque et al. 2016). The field DOC concentration ranged from 1 to 15 mg/L (Engelsen et al. 2017).

The DOC concentration leaching from RCA was consistently above 10 mg/L for the entire pH range assessed, between 2 and 13 S.U., in a laboratory leaching method. At the lowest pH assessed, pH less than 2 S.U., the dissolved DOC leached at a maximum from RCA at a concentration between 11 and 12 mg/L (Kosson et al. 2014a).

The DOC release ratio from RCA is provided by Butera et al. (2015b) as a function of the L/S ratio. However, the DOC release ratio is shown as a cumulative release from the RCA. Discrete release of DOC from RCA was not provided as the L/S ratio is increased; as such, the discussion on Butera et al. (2015b) is limited.

Model results of RCA sorption presented suggest humic substances present in the dissolved and particulate organic matter are significant for trivalent chromium (Engelsen et al. 2010). The DOC in the field leachate may cause fluctuations in the measured aluminum concentrations. However, the field leachate DOC concentration does not appear to be correlated to the cadmium concentrations (Engelsen et al. 2012, 2017).

Polycyclic Aromatic Hydrocarbons (PAHs)

Consisting of a group of more than 100 organic aromatic compounds, polycyclic aromatic hydrocarbons (PAHs) are commonly sourced from asphalt production, vehicular emissions, and waste incineration among other sources (Fan and Lin, 2011). In 1976, the EPA developed a list of 16 priority PAHs that has been accepted by many researchers as representative of all PAHs (Achten and Andersson, 2015). The 16 EPA priority PAHs include: acenaphthene, acenaphthylene, anthracene, benzo(a)anthracene, benzo(a)pyrene, benzo(b)fluoranthene, benzo(ghi)perylene, benzo(k)fluoranthene, chrysene, dibenzo(a,h)anthracene, fluoranthene, fluorene, indeno(1,2,3-cd)pyrene, naphthalene, phenanthrene, and pyrene.

PAHs are currently regulated as a carcinogenic groundwater quality contaminant in Washington State. The groundwater quality criterion for PAHs is 0.01 ug/L for 15 of the 16 EPA PAHs; benzo(a)pyrene has a standard of 0.008 ug/L (Chapter 173-200 WAC). The Washington State surface water criteria for the 16 PAHs vary from no criterion to 3,100 ug/L depending on the PAHs evaluated (Chapter 173-201A WAC).

Butera et al. (2014) detected PAHs in trace amounts in the composition of RCA materials. The compositional mean sum of the 16 EPA priority PAHs in 14 RCA samples was 1.36 mg/kg of raw RCA material, although numerous PAHs analyzed were below detection limits (Butera et al. 2014). Within the same order of magnitude, Dhir et al. (2019) assessed a larger database of RCA samples (N=27) and found the PAHs composition to be 4.0 mg/kg of raw RCA material.

Butera et al. (2014) provided the total PAHs composition, but did not analyze the PAHs content of the laboratory eluate. Strufe et al. (2006) studied the release of PAHs from a concrete material sample (or RCA) using a batch leaching method. The results from Strufe et al. (2006) indicate a PAHs concentration in the RCA laboratory eluate of 6 ug/L.

Polychlorinated Biphenyls (PCBs)

Polychlorinated biphenyls (PCBs) are currently regulated as a groundwater quality carcinogen in Washington State. The groundwater quality criterion for PCBs is 0.01 ug/L. (Chapter 173-200 WAC). The State PCB surface water quality standards for the human health criteria for consumption of water & organisms and organisms only is 0.00017 ug/L (Chapter 173-201A WAC; Ecology, 2019a).

PCBs belong to a broad group of synthetic organic chlorinated hydrocarbons. The number of chlorine atoms and their position on the biphenyl molecule determine the physical and chemical properties associated with the PCB congener. There are 209 PCBs congeners (EPA, 2003). The general chemical structure of PCBs is shown in Figure 18.

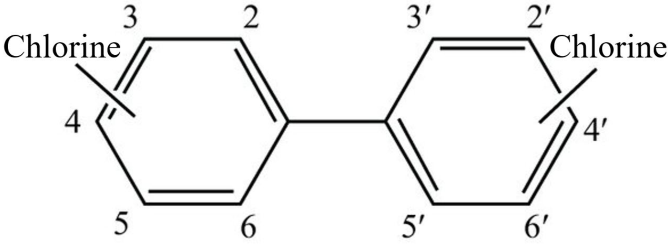

Figure 18: The general chemical structure of polychlorinated biphenyls (Modified from Faroon and Ruiz, 2015).

Butera et al. (2014) assessed the composition of 14 RCA samples for PCBs. Despite the ban on PCB manufacturing in 1979, PCBs were detected in the "new-" and "old-" era RCA samples (Butera et al. 2014). Dissimilar PCB congener profiles were observed between the "old" and "new" RCA, suggesting the background levels of PCBs in the raw materials may be of concern (Butera et al. 2014).

A method to calculate the total PCB content of the RCA sample is to multiply the sum of seven PCBs by a factor between 1 and 10. The seven PCB congeners summed and then multiplied by 5 are PCB28, PCB52, PCB101, PCB118, PCB138, PCB153, and PCB180 (Butera et al. 2014). This method is a common method in determining the total PCB content.

The total PCB composition ranged from 2.3 to 34 ug/kg of RCA material (Butera et al. 2014). These results compare favorably to Dhir et al. (2019), who state the total composition of PCBs in 27 RCA samples is between 1.45 and 400 ug/kg with an average of 18.6 ug/kg.

Strufe et al. (2006) measured the PCB concentration in the laboratory eluate from RCA batch leaching tests. The authors did not observe a PCB concentration in the laboratory eluate above 0.01 ug/L at a L/S ratio of 2 L/kg. In addition, Engelsen et al. (2002) did not detect PCBs in the RCA laboratory eluate above 0.01 ug/L.

Concrete Admixtures

For a detailed review of different types of concrete admixtures (e.g., polymers, additives, additions, and superplasticizers) used in concrete/cement mixtures, see Engelsen et al. (2014), Frigione (2013), and/or Hewlett and Liska (2019). A few studies investigate the release of concrete admixtures from cement-based materials. One concrete admixture that is used and has been studied is naphthalene sulfonate-formaldehyde (NSF), a common water-reducer admixture polymer.

Gu et al. (2020) investigated the release of NSF from cement-based materials. The authors found the behavior of NSF's desorption and adsorption from cement-based materials appears to be related to the hydration process of the cement, water-to-cementitious material ratio, and the amount of NSF added. The authors recommend cautious use of concrete (with NSF added) in distribution pipes and water tower structures.

Märkl and Stephan (2016) investigated the release of NSF and melamine sulfonate-formaldehyde (MSF) from cement-based materials using a German batch leaching method similar to Method 1315. The authors observed a lower leaching potential for NSF and MSF when Berlin tap water was used as the laboratory eluent instead of deionized water. At most, 2 % of the total active component of the NSF and MSF leached after 56 days of exposure to the laboratory eluent. These results suggest the release of MSF and NSF from cement-based materials occurs in low amounts (Märkl and Stephan, 2016).

Onofrei and Gray (1989) studied the release of radioactively labeled NSF and found that NSF was strongly bound and immobilized by the cement hydrate phases, e.g., calcium-silicate-hydrate and calcium-aluminate-hydrate. It was concluded by Gascoyne (2002) that the use of NSF in cement-based material in direct contact with an aquifer would increase the dissolved organic carbon in the groundwater.

In a field study, Ruckstuhl et al. (2003) evaluated the release of NSF from cement-based materials that were in direct contact with groundwater. The concentration of NSF approximately 60 m down-gradient was observed at a maximum of 58 ug/L. The NSF polymer chain was not detected above four units, or oligomers, in the groundwater.

Ruckstuhl et al. (2003) state that roughly 80% of the leached NSF was biodegraded in the aquifer. The properties (e.g., dissolved oxygen, oxidation-reduction potential) of the Zurich aquifer groundwater are not described in the article.

It is important to remember that NSF and MSF are two concrete admixture, polymer materials used in concrete. There are potentially several other concrete admixtures, additives, additions, and/or polymers that may have a similar or different release from concrete material (Engelsen et al. 2014; Hewlett and Liska, 2019). For instance, the use of a fluoropolymer[73] to make cement mortar waterproof has been explored (Krishnan et al. 2013).

Supplementary Cementitious Material

The addition of supplementary cementitious materials (e.g., fly ash, silica fume, and fibers) to concrete reduces the amount of cement clinker required. Concrete materials containing documented supplementary cementitious material substitution were excluded from this assessment.

It is important to note that Kosson et al. (2014b) state the release of antimony, arsenic, chromium, lead, and vanadium were similar between concrete materials with and without replacement of coal combustion fly ash (up to 45%). Although, the authors also state there is ongoing research into the effect the supplementary cementitious materials on the release of POC from concrete.

Silica fume is usually classified as a supplementary cementitious material (FHWA, 2005). The addition of silica fume in the cement manufacturing process may decrease the depth of leaching from cement (Kamali et al. 2003).

Other Types of Organic Eluates/Leachates

There are other lesser reported organic POC from RCA in the reviewed literature. They include but are not limited to hydrocarbon fraction, carbon nanotubes, phenol index, and biological activity.

Hydrocarbon Fraction

Strufe et al. (2006) studied the release of hydrocarbons from RCA using a batch leaching test. The hydrocarbon fraction reported is the summation of the fractions: C6 – C10, C10 – C25, C25 – C35, and benzene. The authors observed the sum of the hydrocarbon fraction release ratio to be 0.19 mg/kg at a L/S ratio of 2 L/kg (Strufe et al. 2006).

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[73] A "fluoropolymer" is a repeating chain of carbon-based chemical units containing multiple or one fluorine-carbon bond(s). The fluoropolymers are conventionally employed for oil and water repellency in the textile industry. Fluoropolymers undergo limited degradation from weathering and Ultraviolet light (Krishnan et al. 2013).

Carbon Nanotubes

According to Oliviera et al. (2019), the presence of elongated and amorphous forms of carbon nanotubes are reported in concrete samples. Carbon nanotubes and other nanoparticles are suspected to be from a range of commercial products the material is exposed to when the concrete is originally used.

Phenol Index

The phenol index is a common and important indicator of water quality. Saca et al. (2017) reported the phenolic compounds in concrete laboratory eluates sourced from batch and column leaching tests. The authors suggest the phenol present may potentially originate from concrete's exposure to steel producing equipment and from exposure to foundry and coke making processes.

The phenol index reported by Saca et al. (2017) are provided in Table 18. The RCA Sample 1 was collected from a stockpile after one week of storage and RCA Sample 2 was collected after under a year of storage[74] (Saca et al. 2017).

Table 18: The discrete phenol index results from the column and batch leaching tests of recycled concrete aggregates.
From Saca et al. (2017).

Liquid-to-Solid Ratio	RCA Sample 1	RCA Sample 2	Leaching Method
0.1 L/kg	1.2 mg/L	Less than 0.1 mg/L	CEN/TS 14405
2 L/kg	1.1 mg/L	0.05 mg/L	(SR) EN 12457-1
10 L/kg	0.37 mg/L	0.1 mg/L	(SR) EN 12457-2

Biodegradation

Biological activity in RCA stockpiles may play a large role in the chemistry of the field leachate produced. The transformation of insoluble metal species into soluble metallic species by biological activity is commonly referred to as bioleaching. However, bioleaching generally occurs in the acidic range followed by the production of exopolysaccharides[75] (Brás et al. 2018).

Sulfate reducing bacteria can reduce sulfate present in gypsum and cement hydrate phases under anaerobic and reduced conditions (Jang and Townsend, 2003). In aerobic and oxidizing conditions, sulfate-producing and nitrifying bacteria may produce sulfuric and nitric acid, respectively (Stroup-Gardiner and Wattenberg-Komas, 2013). The inorganic acids produced by these bacteria act to dissolve silicon-rich calcium-silicate-hydrate gel and calcium hydroxide in the cement/concrete (Gascoyne, 2002).

Fungal microbes may also degrade concrete through the production of organic acids from the degradation of cellulose (George et al. 2013). Depending on the oxidation-reduction potential and the biological activity, production of the inorganic and organic acids may reduce the material pH (Kosson et al. 1996).

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[74] Saca et al. (2017) emphasizes the recycled concrete aggregate sample (aged 1 year) had a near neutral pH (for the batch and column leaching tests) indicating substantial carbonation of the recycled concrete aggregate sample.

[75] "Exopolysaccharides" are primarily composed of polysaccharides and proteins and are macromolecules secreted by microorganisms (Angelin and Kavitha, 2020).

Environmental Hazards of RCA

The production of new concrete requires significant energy consumption, creates carbon dioxide emissions, and uses copious amounts of water (Hewlett and Liska, 2019). In order to preserve limited mineral resources, reduce greenhouse gas emissions, and produce less waste the re-use of RCA material is recommended to reduce the environmental footprint of concrete production (RCW 70A.205.700; Del Ponte et al. 2017; Lee et al. 2010; Tiruta-Barna and Barna, 2013).

Economic benefits associated with the re-use of RCA include the reduction of mining and potential decrease in transportation and material costs (RCW 70A.205.700; Bozyurt et al. 2012; Lee et al. 2010; USGS, 2002). Thus, the beneficial re-use of RCA is apparent, although the process and management of concrete recycling must be monitored and strictly controlled to reduce environmental risks (Barbudo et al. 2012; Tiruta-Barna and Barna, 2013).

The release of pollutants of concern (POC) from RCA into the air, soil, and water occurs through different mechanisms. The interaction of water with the RCA stockpile causes the transport and transfer of the POC into the aqueous phase (Tiruta-Barna and Barna, 2013). Migration of aqueous-phase POC into the soil column beneath a stockpile has the potential to reach underlying aquifers (Engelsen, 2020; Gupta et al. 2018; Oliveira et al. 2019).

The primary transport mechanism of POC from stockpiles of RCA is solid-to-liquid phase partitioning that attempts to reach a chemical equilibrium (Engelsen et al. 2012, 2017; Oliveira et al. 2019; Tiruta-Barna and Barna, 2013). The solid-liquid phase partitioning of POC depends on the chemical gradient difference between the two phases (Kamali et al. 2008). The pH of the field leachate/laboratory eluate generally governs the release and solubility of POC from RCA. Thus, the potential environmental risk does not depend entirely on the composition of the material, but it depends primarily on the availability and solubility of the minerals present on the RCA surface (Barbudo et al. 2012; Del Rey et al. 2015; Galvín et al. 2013).

Complicating the leaching of POC from RCA, the degree of carbonation is critical to the RCA pH and decreases from near 13 S.U to around neutral (Ai et al. 2019; Butera et al. 2014; van der Sloot et al. 2011). The carbonation of the material may also affect the oxidation-reduction potential of the resulting field leachate/laboratory eluate (Mulugeta et al. 2012). Carbonation of the RCA produces metal carbonates (e.g., $CaCO_3$, $NiCO_3$, $BaCO_3$) with different aqueous solubility than minerals present in non-carbonated RCA. This makes the RCA degree of carbonation a critical parameter in the leaching of POC (Ai et al. 2019; Sanger et al. 2020).

One of the intended purposes of this report is to identify the POC potentially leaching from RCA in order to better inform Ecology's Water Quality Program as they develop appropriate requirements for concrete recycling. The identification of potential POC leaching from international and domestic RCA was performed through a comparison of literature batch, column, lysimeter, and field leaching results to State water quality criteria and the effluent limits of the general permit[76].

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[76] Washington State Chapter 90.48 RCW: Water Pollution Control, Washington State Chapter 173-201A WAC: Water Quality Standards for Surface Waters of the State of Washington, Chapter 173-200 WAC Water Quality Standards for Groundwaters of the State of Washington, and the *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* 2021.

This report provides a brief discussion on the applicability of leaching and ecotoxicity methods in relation to compliance with regulatory criteria. The effect of carbonation, soil neutralization, and dilution of the alkaline pH, as well as released POC, from RCA is also discussed. In addition, international studies evaluating the toxicity of concrete laboratory eluate are examined.

Leaching Methods and Ecotoxicity Assessments

For compliance and regulatory reasons, the releases of POC from RCA are frequently measured using laboratory batch, column, and lysimeter leaching methods (Galvín et al. 2013; Butera et al. 2015b). RCA plant managers need an easy way to perform leaching methods that quickly provide them with compliance results for the environmental evaluation of the received materials (Cabrera et al. 2019).

Batch leaching tests provide a quick, reliable, and low cost compliance assessment tool to evaluate the leachable content of the RCA. Although, batch leaching methods may fail to incorporate several important physical and chemical parameters, occasionally rendering the results useless (Galvín et al. 2013).

Toxicity is measured by an organism's response to the synergistic toxic effect of the aqueous sample (e.g., wastewater discharge, RCA stockpile runoff). The synergistic toxic effect is the sum total effect caused by the aqueous sample, rather than by its separate components. Internationally, toxicity tests have been performed on laboratory created concrete aggregate eluate samples (Brás et al. 2017, 2018; Choi et al. 2013; Rodrigues et al. 2017).

Leaching Methods

Due to the reproducibility, ease of performing, and reasonable cost of batch leaching tests, environmental risks and hazards of RCA are commonly established using this leaching method. However, batch leaching method results should be viewed judiciously as the abrasive stirring in the method exposes fresh, non-carbonated, leachable RCA surfaces to the laboratory eluent (Cabrera et al. 2019; Sanger et al. 2020).

Batch leaching tests also may not represent the 'field truth' leaching. The formation of a leached layer on the RCA particle surface in the field will influence the leachate pH and the release of trace-elemental POC (Engelsen et al. 2012; Mulugeta et al. 2012).

Other laboratory leaching methods are generally more labor intensive and intended to produce results more representative of field leaching than batch methods. Examples of other laboratory leaching methods include column (i.e. percolation tests, Method 1314), lysimeter, box method, and monolith tests (i.e. Method 1315).

The parameters influencing the RCA leaching processes in the laboratory and field are provided in Cabrera et al. (2019) and other documents as:

- Physical and chemical reactions (e.g., particle size, temperature, eluent/leachant pH).
- External stresses from the environment (e.g., degree of carbonation, liquid-to-solid ratio).
- Leaching of matrix main constituents (e.g., calcium, aluminum, iron).
- Degradation of RCA matrix (e.g., cracks, erosion).

The laboratory leaching methods used in the reviewed literature are applicable to the RCA encountered in Washington State. Field leaching studies are considered more representative of the POC concentrations leaching from a stockpile of RCA. This is because field leaching incorporates environmental conditions, which the laboratory leaching methods may not.

Ecotoxicity Assays

Ecotoxicity is an estimate of the toxic potential on living organisms. The evaluation of the ecotoxicity of RCA is complicated due to the heterogeneity of the material, exposure to site-specific or physical contaminants during original use, and the chemically complex mixture of the field leachate/laboratory eluate.

Many EPA and Ecology guidance documents are available on potentially applicable ecotoxicity tests. Chapter 173-205 WAC was established to eliminate the discharge of toxics in toxic amounts and for inclusion in the National Pollutant Discharge Elimination System (NPDES) permits. The general permit[77] is an NPDES permit.

The whole effluent toxicity testing (Chapter 173-205 WAC) is designed to protect aquatic life by treating toxic substances and attaining Washington State water quality criteria. If the effluent has "suspected toxicity because of apparent damage to aquatic biota," then the permitted facility may be required to conduct acute and chronic whole effluent toxicity testing (WAC 173-205-040(1)(f)).

Per WAC 173-205-030, the general permit Fact Sheet states the following:

> "The [State] Water Quality Standards for Surface Waters require that the effluent not cause toxic effects in the receiving waters. The commonly available detection methods cannot detect many toxic pollutants. However, one can measure toxicity directly by exposing living organisms to the wastewater in laboratory tests and measuring the response of the organisms. Toxicity tests measure the aggregate toxicity of the whole effluent, and therefore this approach is called whole effluent toxicity (WET) testing.
>
> Ecology does not expect toxicity caused by unidentified pollutants in the discharge as determined by the screening criteria given in Chapter 173-205 WAC. Therefore, the proposed permit does not include whole effluent toxicity testing. Ecology may require effluent toxicity testing in the future, if it receives information that toxicity may be present in this effluent. The permit requires that discharges cause no toxicity."
>
> – Fact Sheet for the Sand and Gravel General Permit, 2021

A few literature articles, described later, applied ecotoxicity tests to concrete laboratory eluates (Brás et al. 2017, 2018; Choi et al. 2013; Rodrigues et al. 2017). These studies use short-term ecotoxicity tests representing a conservative approach to the effects resulting from exposure to RCA laboratory eluate. The ecotoxicity assays used by the studies reviewed include:

- Short-term (30-minute) bacterial bioluminescent test: marine bacterium *Vibrio fischeri* is used to evaluate the toxicity of the concrete laboratory eluate (Rodrigues et al. 2017).
- *Daphnia magna* short-term acute toxicity test: mobility inhibition of micro-crustacean water fleas following 24- and/or 48-hour exposure to the concrete laboratory eluate (Choi et al. 2013; Rodrigues et al. 2017).
- 16-hour microplate susceptibility test: concrete laboratory eluate exposure hinders the growth of microbial eukaryotic yeast *Saccharomyces cerevisiae* (Rodrigues et al. 2017).
- *Lemna gibba* L. (Duckweed) inhibition test: the effect on growth caused by exposure to concrete laboratory eluate, according to standard methods for water and wastewater examination (APHA, 1998; Brás et al. 2017, 2018).

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[77] *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* 2021.

Potential Hazards of RCA

Leaching results presented in the text and tabular format of the aforementioned leaching literature were evaluated for exceedance of the State surface water and groundwater quality criteria as well as the limits established in the general permit[78] (Chapter 173-200 WAC; Chapter 173-201A WAC).

Graphical leaching data, not tabulated in the reviewed literature, are not evaluated for exceedance of the State water quality criteria in this literature review. Exceedance in graphical data was commonly observed, but not confirmed.

If a study conducted a static pH batch leaching test, similar to Method 1313[79], on the RCA, then the leaching POC results for a laboratory eluate pH at or less than 7 S.U. are ignored. This is because the acidic pH laboratory eluate conditions used in these leaching methods are not anticipated in the materials' expected disposal scenario (Bestgen et al. 2016a, 2016b; Butera et al. 2015b; Engelsen et al. 2009, 2010; Kosson et al. 2014a). For more information on the alkaline pH of the RCA laboratory eluate/field leachate, see pH section in Chemical Eluates/Leachate from RCA.

The abstracts of select reviewed literature used to determine if the POC exceeded the State water quality standards are provided in Appendix B.

The laboratory eluates/field leachates released from RCA in exceedance of either the State water quality criteria or effluent limits are presented in Table 19.

The elements calcium, silicon, aluminum, magnesium, potassium, sodium, vanadium, molybdenum, cadmium, cobalt, boron, lithium, phosphorous, strontium, cesium, rubidium, tin, fluoride, titanium, thallium, silver, beryllium, scandium, germanium, yttrium, zirconium, niobium, hafnium, tantalum, bismuth, tungsten, gallium, and uranium did not leach from RCA in exceedance of the State surface water or groundwater criteria, if the criteria exists (Chapter 173-200 WAC; Chapter 173-201A WAC).

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[78] *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* 2021.

[79] Method 1313 evaluates the solid-liquid partitioning of pollutants of concern, in 9-10 parallel batch extractions at static pH values, over a wide range of laboratory eluate pH values.

Table 19: The recycled concrete aggregate field leachate/laboratory eluates of environmental concern identified for exceeding the State water quality criteria or not meeting a requirement of the Sand and Gravel General Permit[80] in at least one leaching method.

Pollutants of Environmental Concern	Above Washington Groundwater Quality Criteria[1,2]	Above Washington Surface Water Quality Criteria[2,3]
Antimony[4]	No criterion	Batch (B), Lysimeter (L)
Arsenic[5]	Field (F)	B, L, F
Barium[6]	B	No criterion
Chloride[7]	F	F
Chromium[8]	B, Column (C), F	B, C, L, F
Copper[9]	Not above criterion	B, C, L, F
Iron[10]	B, F	No criterion
Lead[11]	B	B, C, L
Manganese[12]	B	No criterion
Mercury[13]	Not above criterion	B, C
Nickel[14]	No criterion	B, C, F
pH[15]	B, C, L, F	B, C, L, F
Polycyclic Aromatic Hydrocarbons[16]	B	B
Selenium[17]	B, C, L	B, C, L
Sulfur (as sulfate)[18]	B, C	No criterion
Total Dissolved Solids[19]	B, F	No criterion
Total Suspended Solids[20]	No criterion	F
Turbidity[20]	No criterion	F
Zinc[21]	Not above criterion	B, C

1. Washington State Groundwater Quality Standards (WAC 173-200-040).
2. The effluents limit(s) for the parameter in the Sand and Gravel General Permit.
3. Includes an assessment of all toxics substances criteria listed in Table 240 of the Washington State Surface Water Quality Standards reported in the reviewed literature (WAC 173-201A-240).
4. Butera et al. (2014, 2015b), Galvín et al. (2013), and Gupta et al. (2018). Batch (B), column (C), and lysimeter (L) reported in the literature.
5. Butera et al. (2014, 2015b), Engelsen et al. (2012), Saca et al. (2017), and Sadecki et al. (1996). Field (F), batch (B), column (C), and lysimeter (L) reported in the literature.
6. Butera et al. (2014) and Gupta et al. (2018). Field (F), batch (B), column (C), and lysimeter (L) reported in the literature.
7. Sadecki et al. (1996). Field (F), batch (B), column (C), and lysimeter (L) reported in the literature.
8. Barbudo et al. (2012), Bestgen et al. (2016a), Butera et al. (2014, 2015b), Clark et al. (2013), Del Rey et al. (2015), Engelsen et al. (2012), Galvín et al. (2013, 2014a), Gupta et al. (2018), Limbachiya et al. (2007), Roque et al. (2016), Rodrigues et al. (2017), Saca et al. (2017), Sadecki et al. (1996), and Strufe et al. (2006). Field (F), batch (B), column (C), and lysimeter (L) reported in the literature.
9. Barbudo et al. (2012), Bestgen et al. (2016a), Butera et al. (2014, 2015b), Clark et al. (2013), Del Rey et al. (2015), Engelsen et al. (2012), Galvín et al. (2013, 2014a), Gluchowski et al. (2019), Gupta et al. (2018), Limbachiya et al. (2007), Roque et al. (2016), Saca et al. (2017), and Strufe et al. (2006). Field (F), batch (B), column (C), and lysimeter (L) reported in the literature.
10. Bestgen et al. (2016a) and Sadecki et al. (1996). Field (F), batch (B), column (C), and lysimeter (L) reported in the literature.
11. Butera et al. (2014, 2015b) and Limbachiya et al. (2007). Field (F), batch (B), column (C), and lysimeter (L) reported in the literature.
12. Clark et al. (2013). Batch (B), column (C), and lysimeter (L) reported in the literature.
13. Saca et al. (2017) and Strufe et al. (2006). Batch (B) and lysimeter (L) reported in the literature.
14. Butera et al. (2014), Engelsen et al. (2012), Galvín et al. (2013), Gluchowski et al. (2019), Gupta et al. (2018), Saca et al. (2017), and Strufe et al. (2006). Field (F), batch (B), column (C), and lysimeter (L) reported in the literature.
15. Abbaspour et al. (2016), Ben Maaouia et al. (2018), Bestgen et al. (2016b), Brás et al. (2018), Butera et al. (2014, 2015b), Chen et al. (2012, 2013), Chen et al. (2013), Clark et al. (2013), Coudray et al. (2017), Del

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[80] *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* 2021.

Rey et al. (2015), Engelsen et al. (2009, 2010, 2012), Galvín et al. (2014), Gluchowski et al. (2019), Gupta et al. (2018), Mulligan, (2002), Mulugeta et al. (2011), Natarajan et al. (2019), Oliviera et al. (2019), Rodriguez et al. (2017), Rogue et al. (2016), Saca et al. (2017), Sadecki et al. (1996), Steffes, (1999), Strufe et al. (2006) and Zhang et al. (2017). Field (F), batch (B), column (C), and lysimeter (L) reported in the literature.

16. Strufe et al. (2006). Batch (B) reported in the literature.
17. Butera et al. (2014, 2015b), Clark et al. (2013), Del Rey et al. (2015), Galvín et al. (2014), Gupta et al. (2018), and Saca et al. (2017). Batch (B), column (C), and lysimeter (L) reported in the literature.
18. Del Rey et al. (2015), Galvín et al. (2014a), and Saca et al. (2017). Batch (B), column (C), and lysimeter (L) are reported in the literature.
19. Rodrigues et al. (2017) and Sadecki et al. (1996). Field (F) and batch (B) are reported in the literature.
20. Sadecki et al. (1996). Field (F) is reported in the literature.
21. Bestgen et al. (2016a), Butera et al. (2014), Clark et al. (2013), and Saca et al. (2017). Field (F), batch (B), column (C), and lysimeter (L) reported in the literature.

Potential Chemical Hazards of RCA

The chemical laboratory eluates/field leachates of RCA are described in the Chemical Eluates/Leachates from RCA section. The chemical hazards released from the RCA potentially exceeding the State water quality criteria or general permit effluent limits[81] include pH, total dissolved solids (TDS), total suspended solids (TSS), and turbidity.

The RCA degree of carbonation affects the pH of the field leachate/laboratory eluate produced. The solubility of POC-containing minerals present on the RCA surface depends on the pH and salinity of the field leachant/laboratory eluent.

It is asserted that the rate of carbonation increases with environmental temperature, and vice versa (Engelsen et al. 2005; Lagerblad, 2005). An increase in the environmental temperature where the RCA is stored may increase the leached depth into the cement matrix (Kamali et al. 2003).

pH

The State dangerous waste criterion for pH is a solid or aqueous waste with a pH equal to or greater than 12.5 S.U or less than 2 S.U., as measured using EPA Method 9040C or Method 9045D (WAC 173-303-090(6)). The general permit requires a permitted facility, with concrete recycling identified as an activity, to discharge storm and process water between a pH of 6.5 and 8.5 S.U.

The primary contributor to the alkaline pH is the dissolution of portlandite, or calcium hydroxide, from the cementitious matrix of RCA (Engelsen et al. 2017; Sanger et al. 2020). Other metal hydroxide complexes are reported to dissolve and precipitate, which may contribute to the alkaline pH to a certain degree (Abbaspour et al. 2016; Butera et al. 2015b; Cornelis et al. 2008; Engelsen et al. 2009, 2010, 2012; Gupta et al. 2018; Kosson et al. 2019; Mulugeta et al. 2011).

The release of alkali hydroxides from freshly cured and crushed RCA raises the pH to between 13 and 14 S.U (Cabrera et al. 2019; Engelsen et al. 2017). As the RCA is carbonated, the pH of the field leachate/laboratory eluate decreases to a near-neutral pH, between 8 and 8.5 S.U. (Abbaspour et al. 2016; Engelsen et al. 2012, 2017; Kosson et al. 2002; Mulugeta et al. 2011).

The pH of the RCA laboratory leaching tests ranged from 7.23 to 13.2 S.U. (Butera et al. 2015b; Roque et al. 2016). The RCA field leachate pH ranged from 7.4 to 12.6 S.U. (Engelsen et al. 2012; Sadecki et al. 1996). These wide ranges suggest the field leachate pH of RCA is heavily influenced by the RCA degree of carbonation.

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The carbonation of RCA is a slow process that is dependent on numerous factors, previously described in the Carbonation of Cementitious Materials Section. In field studies, the RCA field leachate approaches a near-neutral pH within a year or two (Sanger et al. 2020).

An exceedance of the State dangerous waste pH criterion, 12.5 S.U., may occur if the RCA field leachate/laboratory eluate is sourced from a freshly crushed RCA or an RCA with a low degree of carbonation.

In addition, stockpiles of RCA may discharge water in exceedance of the State pH water quality criterion at least for the first year. Best management practices (BMPs) to reduce the pH of the field leachate sourced from RCA stockpiles are provided in the Best Environmental Management Practices Section. These BMPs along with treatment technologies should be implemented to control and neutralize the release of alkaline field leachate.

The calcium concentration in the laboratory eluate/field leachate is suggested to be associated with the alkaline pH, as a result of the portlandite dissolution (Bestgen et al. 2016a). The RCA calcium oxide content may provide an estimate of the degree of carbonation or pH produced by the material (Bestgen et al. 2016b).

Total Solids (Dissolved and Suspended)

The general permit requires a permitted facility with concrete recycling identified as an activity to discharge process and mine dewatering water to groundwater at or below a total dissolved solid (TDS) concentration of 500 mg/L. The general permit also requires the discharge of process and mine dewatering water to surface water to have a total suspended solid (TSS) concentration of 40 mg/L or less.

Sadecki et al. (1996) states over 97% of the total solids released from the two RCA stockpiles were in the dissolved fraction. The total solid concentration decreased during the first three months of the study, before stabilizing. The maximum TSS and TDS concentration measured in the Sadecki et al. (1996) was 236 mg/L and 5,100 mg/L, respectively. The authors indicate sediment falling into the sample collector at the beginning of the study may have contributed to these extreme values.

The TDS concentration of the RCA laboratory eluate shows a strong correlation to the calcium release ratio regardless of the leaching method (non-standard or standard laboratory leaching test) and the RCA particle size. The non-standard laboratory leaching test, without particle size reduction of the RCA, indicated the TDS concentration shows a strong correlation to the sulfate release ratio (Coudray et al. 2017).

The released TDS concentration from a non-standard laboratory leaching test, without particle size reduction, decreased with increasing particle size of the RCA. Meanwhile, the standard leaching test suggested the TDS concentration increased with increasing particle size (Coudray et al. 2017).

Rodrigues et al. (2017) also observed an exceedance of the TDS concentration limit of 500 mg/L. The authors report a TDS concentration of 1,200 mg/L in their RCA laboratory eluate.

The literature suggests RCA field leachate/laboratory eluate from the field/laboratory may exceed the effluent limits for TDS and TSS in the general permit. Although, the RCA field leachate TDS and TSS concentrations decrease with time as long as freshly crushed RCA is not added to the stockpile.

Turbidity

The general permit requires all permitted facilities with concrete recycling identified as an activity to discharge effluent waters at or below 50 nephelometric turbidity units (NTU).

The fine-grained RCA stockpile studied by Sadecki et al. (1996) produced a maximum turbidity of 95 NTU. The mean of the 25 samples collected during the yearlong study was 8 NTU (Sadecki et al. 1996). Additionally, the coarse-grained RCA stockpile produced an average turbidity of 5 NTU, with a maximum of 28 NTU (Sadecki et al. 1996). These results suggest the fine-grained RCA has a low likelihood of exceeding the 50 NTU limit required by the general permit.

Potential Inorganic Hazards of RCA

The release of inorganic POC from RCA in exceedance of the State surface water or groundwater criteria depends on the solubility of the inorganic POC-containing minerals. Following the dissolution of available/soluble minerals, diffusion becomes the primary mechanism releasing POC from RCA. To date, the general permit[82] does not require testing of inorganic POC for compliance.

The speciation of the constituents released from RCA depends on the pH, oxidation-reduction potential conditions, and the content of humic substances of the field leachant/laboratory eluent. The pH of RCA in laboratory and field leaching tests may be between near neutral and extreme alkaline, depending on the material's degree of carbonation.

The oxidation-reduction potential of RCA materials has been reported as either highly oxidizing or mild to strongly reducing (Abbaspour et al. 2016; Cornelis et al. 2008; Engelsen et al. 2010; Mulugeta et al. 2011). The discrepancy in the reported oxidation-reduction potential is due to the mild to strong reducing conditions experienced in the field, while laboratory leaching tests generally present oxidizing conditions (Butera et al. 2015b).

The alkaline pH and generally oxidative conditions of cement-based materials allow specific metals to form oxyanions (Cornelis et al. 2008). The primary charge on arsenic, chromium, selenium, molybdenum, and vanadium is oxyanionic in field leachates from a pH of 8.4 to 12.6 S.U. This is also true of the laboratory eluates (Mulugeta et al. 2011).

In addition to the inorganic constituents with the potential to exceed the State water quality criteria listed in Table 19, inorganic POC of potential environmental concern are molybdenum, lithium, strontium, and vanadium (Butera et al. 2015b; Engelsen et al. 2012, 2017; Gupta et al. 2018; Oliviera et al. 2019). They are of potential environmental concern due to the possibility of elevated release concentrations from RCA. However, these potential environmental POC either are below the regulatory criteria or not regulated by the State.

Several inorganic analytes have been shown to leach from RCA that did not exceed the State water quality standards, if the criteria exists. These inorganic contaminants are aluminum, calcium, silicon, magnesium, potassium, sodium, cobalt, boron, cadmium, phosphorous, tin, molybdenum, lithium, vanadium, fluoride, cesium, rubidium, titanium, tungsten, strontium, and gallium (Butera et al. 2015b; Clark et al. 2013; Engelsen et al. 2012, 2017; Galvín et al. 2013; Gupta et al. 2018; Oliveira et al. 2019; Poon et al. 2006; Saca et al. 2017; Sadecki et al. 1996; Chapter 173-200 WAC; Chapter 173-201A WAC).

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[82] *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* 2021.

Several inorganic analytes have been shown to leach from RCA below analytical detection limits. These include silver, thallium, germanium, beryllium, scandium, yttrium, zirconium, niobium, hafnium, tantalum, bismuth, and uranium (Clark et al. 2013; Gupta et al. 2018; Oliveira et al. 2019).

Cationic/Amphoteric Hazards

Inorganic cationic POC are generally leached from RCA in a cationic or cationic/amphoteric release pattern. These patterns leach a maximum POC concentration in the acidic range with a minimum observed in the alkaline range. The POC presenting an environmental hazard that leach in a cationic or cationic/amphoteric release pattern from RCA include barium, copper, iron, lead, manganese, nickel, and zinc.

Barium

The barium[83] laboratory eluate concentration released from RCA in two studies were above the State groundwater quality criterion of 1.0 mg/L in a batch leaching method (Butera et al. 2014; Gupta et al. 2018; Chapter 173-200 WAC). There are no State surface water quality criteria for barium (Chapter 173-201A WAC).

Several leaching studies using batch, column, lysimeter, and field leaching methods did not have concentrations exceeding the States' groundwater quality criterion for barium (Butera et al. 2015b; Clark et al. 2013; Del Rey et al. 2015; Galvín et al. 2013, 2014a; Poon et al. 2006; Rodrigues et al. 2017; Saca et al. 2017; Sadecki et al. 1996; Strufe et al. 2006). The heterogeneity of the RCA composition may affect the availability of barium (Dhir et al. 2019).

The release of barium from RCA appears to decrease as the material becomes carbonated in a laboratory setting (Ai et al. 2019; Müllauer et al. 2012). Two out of the eight RCA samples assessed by Gupta et al. (2018) were above the groundwater quality criterion for barium at a L/S ratio of less than 2 L/kg. As the L/S ratio was increased to 10 L/kg, the release of barium decreased to below the criteria for all RCA samples evaluated (Gupta et al. 2018).

The finest particle size, less than 0.075 mm, released more barium up to a pH of 9 S.U. Above a pH of 10 S.U. the finest particle size released less than the coarser size fractions (Zhang et al. 2018). In addition, the release of barium from RCA generally increased from low salinity, to high salinity, to deionized water (Clark et al. 2012).

Copper

The copper[84] laboratory eluate/field leachate concentration did not exceed the State groundwater criterion of 1.0 mg/L in the RCA batch, lysimeter, column, and field leaching tests (Chapter 173-200 WAC). The State marine chronic and acute surface water quality criteria for copper is 3.1 ug/L[85] and 4.8 ug/L[86]. The State's copper surface water quality criterion for human health consumption of water & organisms, 1,300 ug/L, was not exceeded in the batch, leaching, column, and field leaching tests (WAC 173-201A-240).

The State copper aquatic life surface water quality criteria for marine waters were exceeded in the reviewed literature. Batch, column, lysimeter, and field leaching results of RCA exceeded the copper acute and chronic aquatic life criteria for marine water (Barbudo et al. 2012; Bestgen et al. 2016a; Butera et al. 2014, 2015b; Clark et al. 2013; Del Rey et al. 2015; Engelsen et al. 2012; Galvín et al. 2013, 2014a; Gluchowski et al. 2019; Gupta et al. 2018; Limbachiya et al. 2007; Roque et al. 2016; Saca et al. 2017; and Strufe et al. 2006). The State's

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[83] The metal is measured as total metal in Chapter 173-200 WAC.
[84] The metal is measured as total metal in Chapter 173-200 WAC.
[85] A 4-day average concentration not to be exceeded more than once every three years on average.
[86] A 1-hour average concentration not to be exceeded more than once every three years on average.

aquatic life surface water quality criteria for freshwater were calculated using an equation; therefore, there are no definitive values for comparison of the reviewed literature results.

Although initially high values of copper were detected in the field leachate, Engelsen et al. (2012) indicate the concentrations decrease as the RCA ages. Organic matter from the soil may have intermixed with the RCA during processing in Engelsen et al. (2012); this may enhance the leachability of copper.

Copper laboratory eluate concentrations have also shown to decrease as the RCA material ages (Abbaspour et al. 2016; Bestgen et al. 2016a, 2016b). The leaching results indicate a declining copper release from RCA as the particle size increases (Bestgen et al. 2016a, 2016b; Coudray et al. 2017).

Iron

The iron[87] released from RCA exceeded the State secondary groundwater quality criterion of 0.30 mg/L in one batch and field leaching study (Bestgen et al. 2016a; Sadecki et al. 1996; Chapter 173-200 WAC). There is not a State surface water quality criterion for iron (Chapter 173-201A WAC).

Three of the four studies using batch leaching tests, and the only study employing column and lysimeter leaching tests, on RCA did not exceed the States' criterion for iron. A field study of RCA leachate reported the iron concentration to be 1.2 mg/L. The release of iron from RCA is not expected to result in an environmental concern (Bestgen et al. 2016a; Sadecki et al. 1996).

Lead

The lead[88] laboratory eluate/field leachate concentration from RCA exceeded the State groundwater criterion of 0.05 mg/L in one batch leaching study (Limbachiya et al. 2007). The State aquatic life surface water quality criterion for chronic exposure to lead in marine waters was exceeded in four studies using a batch, column, and lysimeter leaching methods (Butera et al. 2014, 2015b; Limbachiya et al. 2007). The State aquatic life surface water quality criterion for acute exposure to lead in marine waters was not exceeded using the batch, column, lysimeter, or field leaching methods.

There are no State lead surface water quality criteria for human health consumption of water & organisms or organisms only. Additionally, the State's lead aquatic life surface water quality criteria for freshwater are calculated using an equation, thus there are no definitive values for comparison of the reviewed literature results.

The RCA field leaching study (Engelsen et al. 2012, 2017) reported the lead field leachate concentration was below the detection limit for the duration of the study. This may relate to the availability of lead in the composition of the RCA, as the standard deviation of lead ranged two orders of magnitude (Dhir et al. 2019). As such, the leachable lead concentration is typically below the detection limit and referred to as negligible in several articles (Barbudo et al. 2012; Galvín et al. 2013; Gluchowski et al. 2019; Engelsen et al. 2012, 2017; Rodrigues et al. 2017; Strufe et al. 2006).

Manganese

The manganese[89] laboratory eluate concentration exceeded the State groundwater quality criterion of 0.050 mg/L once in the reviewed literature using Method 1311 (Clark et al. 2013; Chapter 173-200 WAC). There are no State surface water quality criteria for manganese (Chapter 173-201A WAC). Other studies evaluating the release of manganese from RCA

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[87] The metal is measured as total metal in Chapter 173-200 WAC.

[88] The metal is measured as total metal in Chapter 173-200 WAC.

[89] The metal is measured as total metal in Chapter 173-200 WAC.

suggest the environmental risk from manganese is low (Butera et al. 2014, 2015b; Gupta et al. 2018; Limbachiya et al. 2007; Strufe et al. 2006).

The high standard deviation associated with the RCA manganese composition may affect the availability of manganese (Dhir et al. 2019). The release of manganese from RCA follows a cationic leaching pattern. An acidic laboratory eluent, such as that used in Method 1311, may not represent a typical RCA disposal scenario field leachant. Additionally, the release of manganese from RCA follows a direct relationship with salinity (i.e. increasing salinity results in an increase in the release of manganese from RCA; Clark et al. 2013).

Mercury

The State groundwater quality criterion for mercury[90] is 0.002 mg/L and was not exceeded in the reviewed literature (Chapter 173-200 WAC). The State surface water quality criteria for chronic exposure to mercury for fresh and marine water were exceeded in two RCA batch and a column leaching test (Saca et al. 2017; Strufe et al. 2006). The State human health criteria for the consumption of water & organisms and organisms only were both exceeded in a column leaching test (Saca et al. 2017).

However, five batch leaching studies were not able to detect mercury in the RCA laboratory eluate (Barbudo et al. 2012; Clark et al. 2013; Galvín et al. 2014a; Rodrigues et al. 2017).

Nickel

There is no State groundwater quality criterion for nickel. The State has established nickel concentration standards for marine surface water quality criteria for aquatic life based on acute and chronic exposure, 74 ug/L[91] and 8.2 ug/L[92] respectively. The nickel standards for human health criteria for the consumption of water & organisms and organisms only are 150 ug/L and 190 ug/L. An equation is used to calculate the nickel threshold for the acute and chronic aquatic life criteria for freshwater and will not be assessed (Chapter 173-201A).

The nickel concentration thresholds for marine surface water quality criteria for aquatic life based on acute and chronic exposure were exceeded in RCA batch and column leaching tests (Butera et al. 2014; Galvín et al. 2013; Gluchowski et al. 2019; Gupta et al. 2018; Limbachiya et al. 2007; Saca et al. 2017; Strufe et al. 2006). The chronic nickel marine surface water quality criterion was also exceeded in a field leaching test (Engelsen et al. (2012). The State human health criteria for the consumption of water & organisms and organisms only were both exceeded in two batch leaching studies (Butera et al. 2014; Gupta et al. 2018).

Several studies report the nickel release from RCA is below the chronic aquatic life criterion for marine water, or 8.2 ug/L (Barbudo et al. 2012; Del Rey et al. 2015; Engelsen et al. 2017; Galvín et al. 2014; Butera et al. 2015b; Rodrigues et al. 2017; Roque et al. 2016). The field release of nickel from RCA is initially high. However, the nickel release decreases with age due to the formation of a carbonation layer and a depletion zone (Engelsen et al. 2012).

Zinc

The State groundwater water quality criterion for zinc[93] is 5 mg/L and was not exceeded in the reviewed batch, column, lysimeter, and field leaching tests. The State water quality criteria for acute and chronic exposure aquatic life criteria for marine water is 90 ug/L[94] and 81 ug/L[95],

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[90] The metal is measured as total metal in Chapter 173-200 WAC.
[91] A 1-hour average concentration not to be exceeded more than once every three years on average.
[92] A 4-day average concentration not to be exceeded more than once every three years on average.
[93] The metal is measured as total metal in Chapter 173-200 WAC.
[94] A 1-hour average concentration not to be exceeded more than once every three years on average.
[95] A 4-day average concentration not to be exceeded more than once every three years on average.

respectively. An equation is used to calculate the zinc threshold for the acute and chronic aquatic life criteria for freshwater and will not be assessed (Chapter 173-201A).

The marine surface water standard for acute and chronic exposure to zinc was exceeded in one column and three batch leaching studies (Bestgen et al. 2016a; Butera et al. 2014; Clark et al. 2013; Saca et al. 2017).

The heterogeneity of zinc in the RCA may affect the availability for leaching (Dhir et al. 2019). The zinc concentration in the RCA laboratory eluate/field leachate is often reported below the analytical detection limit using batch and field leaching methods (Barbudo et al. 2012; Butera et al. 2014; Engelsen et al. 2012, 2017; Galvín et al. 2013, 2014a; Gluchowski et al. 2019; Gupta et al. 2018; Rodrigues et al. 2017; Strufe et al. 2006).

Ai et al. (2019) reported that fresh RCA generally leached higher zinc concentrations than the carbonated counterpart. The release of zinc from RCA decreased with increasing particle size (Chen et al. 2012). An increase in the chloride composition in the laboratory eluent/field leachant may enhance the leaching of zinc (Engelsen et al. 2012; Clark et al. 2013).

Anionic Hazards

Specific anionic hazards are generally highly soluble species that follow a ‘constant’ release pattern from the RCA. The soluble anionic species leach to the point of depletion from the RCA. This implies that the anionic hazards may leach an elevated concentration for a short duration before the RCA particle forms a leached layer or depletion zone (Engelsen et al. 2012; Sadecki et al. 1996).

Sulfur (as Sulfate)

The State’s secondary groundwater quality criterion for sulfate is 250 mg/L (Chapter 173-200 WAC). The sulfur (as sulfate)[96] released from RCA exceeded the groundwater quality criterion in one column and two batch leaching studies (Del Rey et al. 2015; Galvín et al. 2014a; Saca et al. 2017). The sulfate laboratory eluate concentrations exceeded the criterion by a factor between almost 2.7 and 23 times.

Although, eight, two, and one leaching study(ies) did not exceed the State sulfate groundwater criterion in batch, lysimeter, and column leaching tests, respectively (Barbudo et al. 2012; Butera et al. 2014; Butera et al. 2015b; Galvín et al. 2013, 2014a; Gluchowski et al. 2019; Limbachiya et al. 2007; Rodrigues et al. 2017; Roque et al. 2016; Strufe et al. 2006).

The release of sulfate from RCA is controlled by ettringite and monosulfates, if calcium carbonate was added to the cement. The source of the elevated release of sulfates may be from the addition of gypsum to the cement clinker during production (Butera et al. 2014).

Upon concretes’ carbonation, calcium carbonate and gypsum form with the latter, controlling the release of sulfate from RCA. Using lysimeters and column leaching methods, sulfate released from RCA showed an initial decrease followed by a stabilization as the L/S ratio was increased from between 1 or 2 L/kg to 10 L/kg (Butera et al. 2015b). Butera et al. (2015b) suggests less than 3% of the total sulfate present leached from the RCA.

There is an observed discrepancy between the laboratory and field sulfate release concentration from RCA in the alkaline pH range as the material ages (or as the degree of carbonation increases). In the laboratory, the sulfate[97] released from RCA increases as the depth of carbonation increases (Butera et al. 2015b; Engelsen et al. 2009; Müllauer et al. 2012).

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[96] Sulfur (as sulfate) is hereby referred to as sulfate. The leachate ratio of sulfate to sulfide is between 2.7 and 3.2, indicating the primary speciation of sulfur is sulfate in RCA leachate (Engelsen et al. 2012).

[97] Sulfate (as sulfur) is reported in Müllauer et al. 2012).

However, in the field the leachate sulfate[98] concentration declined after about 100 days of leaching (Engelsen et al. 2012).

It was suspected that the formation of a leached layer, or a depletion zone, on the surface of the RCA occurred. After the formation of a leached layer, the primary release mechanism for sulfate from RCA is diffusion (Engelsen et al. 2012, 2017). Generally, as the RCA particle size increased, the sulfate concentration decreased (Coudray et al. 2017).

Chloride

The State secondary groundwater quality criterion for chloride is 250 mg/L (Chapter 173-200 WAC). The State freshwater quality criteria for aquatic life for chronic and acute exposure to chlorides is 230 mg/L[99] and 860 mg/L[100], respectively (Chapter 173-201A WAC).

The only study to exceed the State groundwater and surface water quality criteria is a field study of two RCA stockpiles (Sadecki et al. 1996). The fine-grained and coarse-grained RCA[101] stockpile leached a maximum chloride concentration of 750 and 910 mg/L over the year, respectively.

Many other studies using laboratory and field leaching methods suggest the release of chlorides from RCA is in general below State groundwater and surface water quality criteria (Butera et al. 2014, 2015b; Del Rey et al. 2015; Engelsen et al. 2017; Galvín et al. 2013, 2014a; Gluchowski et al. 2019; Rodrigues et al. 2017; Roque et al. 2016; Saca et al. 2017; Strufe et al. 2006).

Chloride content in construction materials, such as RCA, is typically limited as chloride may lead to the corrosion of steel reinforcement (Saca et al. 2017). The chloride present on the RCA is suspected of being sourced from exposure to de-icing salt (Engelsen et al. 2012, 2017; Sadecki et al. 1996). The release of chlorides from RCA showed a decreasing trend over time due to its high aqueous solubility (Sadecki et al. 1996).

Chlorides are expected to form metal complexes, which may enhance the release of specific metals from RCA. The salinity, directly related to the chloride concentration, of the laboratory eluent/field leachant has been shown to affect the release of barium, copper, zinc, chromium, arsenic, manganese, and lead from RCA (Clark et al. 2013; Engelsen et al. 2012, 2017). The releases of chlorides from RCA are not expected to present an environmental risk because the chloride concentration should quickly drop below the regulatory criteria (Sadecki et al. 1996).

Oxyanionic Hazards

Oxyanionic hazards released from RCA are oxymetalates or a metal bonded to one or more oxygen atoms with a net negative charge. Chromium, arsenic, selenium, and antimony are released from RCA and are capable of forming oxyanions depending on the pH and oxidation-reduction potential conditions of the field leachate or laboratory eluate. The oxyanionic species released from RCA include, but are not limited to, chromate (CrO_4^{2-}), vanadate (VO_4^{3-}), and arsenate (AsO_4^{3-}).

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[98] Sulfur concentrations are reported in Engelsen et al. (2012).

[99] A 4-day average concentration not to be exceeded more than once every three years on average.

[100] A 1-hour average concentration not to be exceeded more than once every three years on average.

[101] Sadecki et al (1996) evaluated the release of contaminants from two stockpiles. The two stockpiles were a fine-grained recycled concrete aggregate stockpile consisting of particles finer than 4.75 mm and a coarse-grained recycled concrete aggregate with particles greater than 4.75 mm but less than 19 mm.

As an example, the Pourbaix diagram (Figure 19) shows the speciation and form of chromium as a function of the oxidation-reduction potential and pH of the field leachate/laboratory eluate. See Cornelis et al. (2008) for a detailed discussion on Pourbaix diagrams and oxyanions present in cement-based materials.

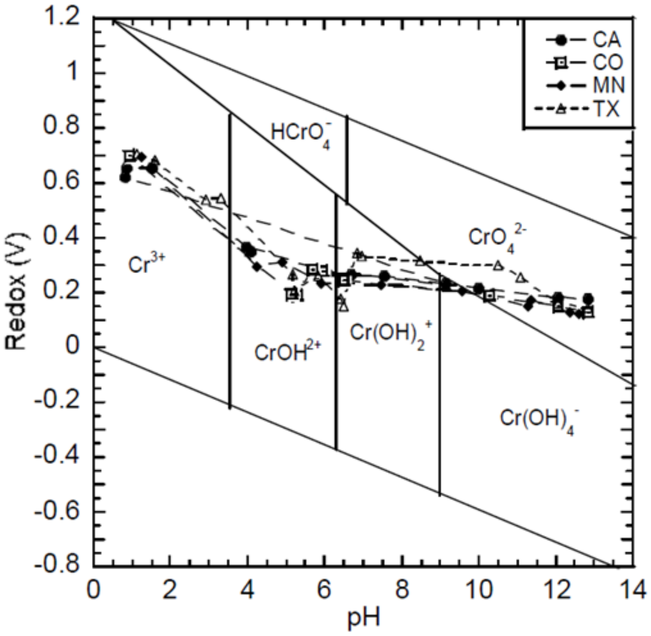

Figure 19: The chromium speciation and form as a function of pH and oxidation-reduction potential (Redox [in volts]), or a Pourbaix diagram, joined with RCA laboratory eluate chromium results from a batch leaching method similar to EPA Method 1313.

CA is recycled concrete aggregate from California, CO is recycled concrete aggregate from Colorado, MN is recycled concrete aggregate from Minnesota, TX is recycled concrete aggregate from Texas; modified from Chen et al. (2012).

Chromium

The release of chromium from RCA occurs primarily as hexavalent chromium with minor amounts of trivalent chromium (Butera et al. 2015b; Del Rey et al. 2015). For the alkaline pH and positive-oxidizing conditions of the RCA field leachate/laboratory eluate, the form of hexavalent chromium leached from RCA occurs as the oxyanionic chromate ion (CrO_4^{2-}) (Ben Maaouia et al. 2018; Chen et al. 2012; Cornelis et al. 2008; Mulugeta et al. 2011).

The State groundwater quality criterion for chromium[102] is 0.05 mg/L (Chapter 173-200 WAC). The State freshwater chronic and acute surface water quality criteria for hexavalent chromium is 10 ug/L[103] and 15 ug/L[104], while the marine chronic and acute surface water criteria for hexavalent chromium is 50 ug/L[105] and 1,100 ug/L[106], respectively.

The State surface water quality standards for trivalent chromium depend on an equation and will not be evaluated. There are no State trivalent or hexavalent chromium surface water quality criteria for human health consumption of water & organisms or organisms only.

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[102] The metal is measured as total metal in Chapter 173-200 WAC.

[103] A 4-day average concentration not to be exceeded more than once every three years on average.

[104] A 1-hour average concentration not to be exceeded more than once every three years on average.

[105] A 4-day average concentration not to be exceeded more than once every three years on average.

[106] A 1-hour average concentration not to be exceeded more than once every three years on average.

Chromium Exceedance

Despite the decline in the use of chromium in construction materials, the leaching of chromium from RCA remains an environmental concern (Engelsen et al. 2012; Maia et al. 2018). The field leachate/laboratory eluate chromium concentration from RCA in the field and laboratory has been shown to exceed the State groundwater chromium criterion in numerous studies (Ben Maaouia et al. 2018; Bestgen et al. 2016a; Butera et al. 2014, 2015b; Chen et al. 2012; Del Rey et al. 2015; Engelsen et al. 2010, 2012; Galvín et al. 2014a; Gupta et al. 2018; Limbachiya et al. 2007; Rodrigues et al. 2017; Saca et al. 2017; Sadecki et al. 1996).

The chronic and acute freshwater aquatic life criteria for hexavalent chromium was exceeded in the two batch leaching studies distinguishing the speciation of chromium in the RCA leachate (Del Rey et al. 2015; Butera et al. 2014). The State marine waters aquatic life criteria for acute and chronic exposure to hexavalent chromium was exceeded in one batch leaching test (Del Rey et al. 2015).

The majority of the leaching studies report the total chromium released from RCA without description of the speciation. However, other leaching studies report an average of at least 80% of total chromium released from RCA is in the oxyanionic form [i.e. chromate (CrO_4^{2-})] as hexavalent chromium (Butera et al. 2014; Del Rey et al. 2015; Mulugeta et al. 2011). A comparison between 80% of the total chromium leaching results and the State surface water quality hexavalent chromium criteria may provide insight into the environmental concern of hexavalent chromium released from RCA.

These comparisons include a substantial degree of uncertainty, as the actual amount of hexavalent chromium released from RCA remains unclear. Using 80% of the total chromium leaching result values indicates that:

- The chronic and acute freshwater aquatic life criteria for hexavalent chromium were exceeded in the majority of reviewed RCA leaching studies, both in the field and laboratory (Barbudo et al. 2012; Bestgen et al. 2016a; Butera et al. 2014, 2015b; Del Rey et al. 2015; Engelsen et al. 2012; Galvín et al. 2013, 2014a; Gupta et al. 2018; Limbachiya et al. 2007; Rodrigues et al. 2017; Roque et al. 2016; Saca et al. 2017; Sadecki et al. 1996; Strufe et al. 2006).
- The aquatic life chronic exposure to hexavalent chromium in marine waters was exceeded in both the field and laboratory leaching tests (Ben Maaouia et al. 2018; Bestgen et al. 2016a; Butera et al. 2014, 2015b; Chen et al. 2012; Del Rey et al. 2015; Engelsen et al. 2012; Galvín et al. 2014a; Gupta et al. 2018; Limbachiya et al. 2007; Rodrigues et al. 2017; Saca et al. 2017; Sadecki et al. 1996).
- The aquatic life acute exposure to hexavalent chromium in marine waters was exceeded in one batch leaching test (Del Rey et al. 2015).

Chromium Release Trends

The solubility of chromium is heavily dependent on the pH and oxidation-reduction potential of the field leachate/laboratory eluate (Cornelis et al. 2008). The leaching pattern of chromium is identified as both amphoteric and oxyanionic. This means that the maximum release concentration of chromium from RCA occurs at a pH of 1 to 3 S.U. and 9 to 13 S.U. and a minimum release concentration occurs at a pH of 5 to 6.5 S.U. (Dhir et al. 2019).

There is a discrepancy in the reviewed literature regarding the release of chromium from artificially carbonated RCA[107]. In the majority of reports, as the RCA becomes carbonated the

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[107] Artificially carbonation in this context means either allowing the material to be exposed to the atmosphere for a set amount of time in a sheltered environment or artificial accelerated carbonation. The degree of carbonation the material may have experienced may be different between the RCA assessed in the different studies.

leaching of chromium from RCA increases substantially in comparison to non-carbonated RCA (Bestgen et al. 2016a; Müllauer et al. 2012; Mulugeta et al. 2011). However, another author reports a decrease in the amount of chromium released from carbonated RCA as compared to non-carbonated RCA (Ai et al. 2019).

Engelsen et al. (2012) conducted a field study of the chromium leachate concentration. The decline in the chromium concentration after 100 days of exposure to the environment was attributed to the formation of a leached layer, or a depletion zone, on the RCA particles. The decrease was almost two orders of magnitude.

The chromium concentration released from RCA is shown to decrease as the liquid-to-solid (L/S) ratio increases (Bestgen et al. 2016a, 2016b; Gupta et al. 2018). As the finer-grained RCA particles are noted to have an increase in the amount of adhered cement-paste, the release of chromium from RCA increases as the particle size decreases (Bestgen et al. 2016a; Coudray et al. 2017; Sadecki et al. 1996).

An increase in the reaction time between the RCA and the laboratory eluent as well as the salinity, enhances the amount of chromium released from the RCA (Bestgen et al. 2016a; Clark et al. 2013). In addition, a rise in the amount of curing time will decrease the release of chromium from RCA (Bestgen et al. 2016b).

Arsenic

The State groundwater quality criterion for arsenic[108] is 0.05 ug/L (Chapter 173-200 WAC). The State surface water quality human health criteria for consumption of water & organisms and organisms only for arsenic is 10 ug/L. The freshwater surface water quality criteria for acute and chronic exposure to arsenic is 360 ug/L[109] and 190 ug/L[110], respectively. The arsenic marine surface water quality criteria for acute and chronic exposure is 69 ug/L[111] and 36 ug/L[112], respectively (Chapter 173-201A WAC). The State classifies arsenic as a carcinogen.

Arsenic occurs naturally in some aggregates used in concrete, but this is not believed to be the sole source of arsenic in RCA (Sadecki et al. 1996). Naturally occurring arsenic is well documented in limestone with a concentration of less than 1 to 20 mg of arsenic per kg of limestone, potentially suggesting the cement-paste is one of the sources of the arsenic in RCA (DECOS, 2012; Pichler and Mozaffari, 2015). Thus, arsenic is present in all commercial cements in minor amounts.

Arsenic Exceedance

The concentration of arsenic exceeded the State groundwater quality criterion in every field and laboratory leaching study evaluating the release of arsenic from RCA. These leaching studies include batch, column, lysimeter, and field leaching methods (Barbudo et al. 2012; Butera et al. 2014; Butera et al. 2015b; Clark et al. 2013; Del Rey et al. 2015; Engelsen et al. 2012; Engelsen et al. 2017; Galvín et al. 2013, 2014a; Gupta et al. 2018; Poon et al. 2006; Rodrigues et al. 2017; Saca et al. 2017; Sadecki et al. 1996; Strufe et al. 2006).

The arsenic State surface water human health criteria for consumption of water & organisms and organisms only is exceeded in batch, lysimeter, and field leaching tests (Butera et al. 2014, 2015b; Engelsen et al. 2012; Saca et al. 2017; Sadecki et al. 1996). The arsenic concentration thresholds for marine and fresh surface water quality criteria for aquatic life based on acute and chronic exposure were also exceeded by the field leachate in Sadecki et al. (1996). The State

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[108] The metal is measured as total metal in Chapter 173-200 WAC.

[109] A 1-hour average concentration not to be exceeded more than once every three years on average.

[110] A 4-day average concentration not to be exceeded more than once every three years on average.

[111] A 1-hour average concentration not to be exceeded more than once every three years on average.

[112] A 4-day average concentration not to be exceeded more than once every three years on average.

marine surface water quality criterion for chronic exposure to arsenic for aquatic life was also exceeded in one batch leaching test (Butera et al. 2014).

Arsenic Release Trends

The most common speciation of arsenic in RCA field leachate is pentavalent arsenic (As^{5+}), which is less toxic than trivalent arsenic (As^{3+}) (Sadecki et al. 1996). Although rarely studied independently in the literature, trivalent oxidized arsenic produces arsenite (AsO_3^{3-}) and oxoarsinite (AsO_2^{-}), while pentavalent oxidized arsenic yields arsenate (AsO_4^{3-}) compounds (Cornelis et al. 2008, Mulugeta et al. 2011).

The release of arsenic from RCA as a function of pH follows the highly soluble, or constant, leaching release pattern (Dhir et al. 2019). Although another author identified the release pattern of arsenic from RCA to be cationic, stating the release of arsenic decreases with increasing pH (Zhang et al. 2018).

Sadecki et al. (1996) conducted a field leaching study of RCA stockpiles. In this study, the arsenic concentrations ranged from 13 to 760 ug/L and 6 to 430 ug/L in the coarse- and fine-grained RCA stockpiles, respectively. As the study progressed, the arsenic field leachate concentration decreased. The observed decrease was potentially attributed to a first flush phenomenon, the formation of a leached layer or a depletion zone (Engelsen et al. 2012). Mulugeta et al. (2011) indicate the release of arsenic from RCA was higher in a field leaching study compared to the same material in a laboratory leaching test.

The arsenic release from RCA increased after the material was artificially carbonated for all RCA samples assessed (Mulugeta et al. 2011). According to the charge-based analysis performed on the resulting laboratory eluate, the primary charge of the arsenic released from both the carbonated and non-carbonated RCA was anionic. In addition, the primary charge on the released oxy-arsenic species from RCA in both the laboratory eluate/field leachate was anionic for the entire range of pH assessed (8.4 to 12.6 S.U.) (Mulugeta et al. 2011).

There was not a substantial effect to the arsenic laboratory eluate concentration when the particle size was varied from 75 mm to less than 0.075 mm (Zhang et al. 2018). The arsenic laboratory eluate concentration increases with increasing laboratory eluent salinity (i.e. arsenic concentration increased from deionized, to low salinity, to high salinity waters; Clark et al. 2013).

Selenium

The leaching of selenium from RCA has been identified to pose a potential environmental risk (Maia et al. 2018). The State groundwater quality criterion for selenium[113] is 0.01 mg/L (Chapter 173-200 WAC). The State surface water quality aquatic life criteria for freshwater chronic and acute exposure to selenium is 5.0 ug/L[114] and 20.0 ug/L[115], respectively. The marine State surface water quality criteria, or the human health criteria for consumption of water & organisms and organisms only, were not exceeded. See Table 10 and Table 11 for the other selenium surface water quality criteria.

The release of selenium from RCA may exceed the State groundwater quality criterion in batch, column, and lysimeter leaching studies (Butera et al. 2014, 2015b; Clark et al. 2013; Del Rey et al. 2015; Gupta et al. 2018). Selenium released from RCA also may exceed the State freshwater aquatic life criterion for chronic exposure in batch, column, and lysimeter leaching studies (Butera et al. 2014, 2015b; Clark et al. 2013; Del Rey et al. 2015; Gupta et al. 2018; Saca et al. 2017). The State surface water quality criterion for freshwater acute exposure to

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[113] The metal is measured as total metal in Chapter 173-200 WAC.
[114] A 4-day average concentration not to be exceeded more than once every three years on average.
[115] A 1-hour average concentration not to be exceeded more than once every three years on average.

selenium was exceeded in three batch leaching studies (Butera et al. 2014; Clark et al. 2013; Del Rey et al. 2015).

The release of selenium from RCA follows a constant, or highly soluble, leaching pattern (Dhir et al. 2019). Although seldom studied independently, the formation of the oxyanionic selenium species (e.g., selenite, selenate) exist in RCA field leachate/laboratory eluate. The dominant charge of the oxy-selenium species released from RCA in the field and laboratory is anionic (Mulugeta et al. 2011).

The selenium release pattern as a function of pH differed between the field and laboratory. The explanation provided is the formation of a leached layer, or depletion zone, on the RCA in the field. The selenium releases from carbonated RCA samples were 3 to 7 times higher than the non-carbonated RCA samples (Mulugeta et al. 2011). Another author noted the selenium releases from carbonated and non-carbonated RCA generally fall in the same range (Ai et al. 2019).

Antimony

Antimony is not a regulated groundwater contaminant in Washington State. The State antimony surface water criteria for human health criteria for consumption of water & organisms and organisms only is 12 ug/L and 180 ug/L, respectively (Chapter 173-201A WAC).

The antimony released from RCA exceeded the water & organism criterion in three batch studies and one lysimeter leaching study (Butera et al. 2014, 2015b; Galvín et al. 2013; Gupta et al. 2018).

The presence of antimony in RCA appears to be quite variable. The European Union has made it common practice to add tin, iron, or antimony salts as reducing agent additives to cement clinker. The purpose of adding a reducing agent is to decrease the release of soluble chromates (i.e. hexavalent chromium; Magistri et al. 2011).

The primary charge associated with oxy-antimony species released from RCA in the field and laboratory was anionic. The concentration of antimony was observed to increase by a factor of 1.5 to 8 for carbonated RCA when compared to the non-carbonated RCA counterparts (Mulugeta et al. 2011).

Gupta et al. (2018) assessed the release of antimony from RCA as a function of the liquid-to-solid ratio. The antimony exhibited a similar laboratory eluate concentration over the entire range of L/S ratios assessed (0.5 to 10 L/kg). The mechanism reportedly controlling the leaching of antimony from RCA was solubility (Gupta et al. 2018).

Potential Organic Hazards of RCA

The Washington State groundwater criterion for polycyclic aromatic hydrocarbons (PAHs) is 0.01 ug/L[116] (Chapter 173-200 WAC). The State surface water quality human health criteria for consumption of water & organisms and organisms only for PAHs vary depending on the PAHs[117] (Chapter 173-201A WAC).

The total PAHs concentration from RCA laboratory eluate was measured to be 6 ug/L from concrete material using a batch leaching test. The RCA field leachate may exceed the State

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[116] The Washington State polycyclic aromatic hydrocarbons (PAH) Groundwater Water Quality Standard is 0.01 ug/L for 15 of the 16 U.S. EPA PAH; Benzo(a)pyrene has a standard of 0.008 ug/L (Chapter 173-200 WAC).

[117] The Washington State surface water quality standards range for polycyclic aromatic hydrocarbons (PAH) from no criterion to 3,100 ug/L depending on the PAH chemical evaluated; see Ecology (2019a) or WAC 173-201A-240 for more information.

groundwater criterion for PAHs and the surface water quality criteria for benzo(a)anthracene, benzo(a)pyrene, and benzo(b+j+k)fluoranthene (Strufe et al. 2006).

The State groundwater quality criterion for polychlorinated biphenyls (PCBs) is 0.01 ug/L (Chapter 173-200 WAC). The State PCB surface water quality standards for the human health criteria for consumption of water & organisms and organisms only is 0.00017 ug/L. The marine surface water standards for acute and chronic exposure to PCBs are 2 ug/L[118] and 0.014 ug/L[119], respectively. The freshwater surface water standards for acute and chronic exposure to PCBs are 10 ug/L[120] and 0.030 ug/L[121], respectively.

The presence of PCBs was confirmed in the composition of the newer and older RCA samples. The compositional concentration of PCBs in RCA is reported to be between 1.45 and 400 ug/kg, with a mean concentration of 18.6 ug/kg (Dhir et al. 2019). The PCB congener profile was different between the “old-” and “new-” era RCA samples (Butera et al. 2015b).

Strufe et al. (2006) and Engelsen et al. (2002) indicated the leaching of PCBs from RCA is less than 0.01 ug/L. However, the State surface water standard for human health criteria for consumption of water & organisms and organisms only is orders of magnitude below 0.01 ug/L. Although there is no evidence of environmental risk of PCBs sourced from RCA, the leaching of PCBs from RCA remains a potential environmental concern.

pH Neutralization and Dilution of Released Contaminants

The carbonation process produces physical, chemical, and mineralogical changes to the RCA that influence the release of POC and the material pH (Abbaspour et al. 2016; Ai et al. 2019; Lagerblad, 2005; Tiruta-Barna and Barna, 2013). Carbonated RCA results in a lower field leachate/laboratory eluate pH as compared to non-carbonated RCA (Engelsen et al. 2017). An accurate model of the degree of carbonation the RCA has experienced is recommended prior to an investigation into the release of POC from RCA (Sanger et al. 2020).

As previously described[122], the water contact mechanisms between the field leachate and the RCA differ depending on the structure of the stockpile and motion of water through the stockpile. The RCA-impacted field leachate may discharge to waters of the State, where similar and different pH neutralization processes occur in surface water and groundwater discharge.

Dilution of the RCA impacted field leachate is expected to occur when the field leachate intercepts a water of the State. Dilution because of discharge into a surface water body is seldom discussed in the literature. The focus centers around the field leachate discharge to a subsurface aquifer and subsequent dilution followed by further dilution as the impacted groundwater intercepts a surface water body.

pH Neutralization

The pH of the field leachate/laboratory eluate depends on the availability of leachable calcium and other metal (hydr)oxides (Abbaspour et al. 2016; Bestgen et al. 2016a, 2016b; Butera et al. 2015b; Cornelis et al. 2008; Engelsen et al. 2009, 2010, 2012, 2017; Gupta et al. 2018). In a field leaching scenario, the highest leachate pH is observed immediately after the leachate leaves the RCA material (Sanger et al. 2020). The RCA alkaline pH may be neutralized and

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[118] A 24-hour average not to be exceeded.

[119] A 24-hour average not to be exceeded.

[120] A 24-hour average not to be exceeded.

[121] A 24-hour average not to be exceeded.

[122] See the Water Exposure Scenarios section for more information on expected scenarios of RCA exposure to water.

potential risks minimized depending on the specific conditions existing on-site (i.e. infiltration rate, organic matter content of soils) (Oliveira et al. 2020).

See BMP C252: Treating and Disposing of High pH Water in the Stormwater Management Manual for Western Washington (Ecology, 2019b).

Surface Water pH Neutralization

The primary pH neutralization mechanism for atmospherically exposed surface water is the diffusion of carbon dioxide (Sanger et al. 2020). Again, the carbonation reduces the field leachate pH. The acidity present in surface waters is from the presence of excess hydrogen ions and the carbonate system (Gupta et al. 2018).

Subsurface pH Neutralization

If the water exposed to RCA percolates into the subsurface soils, then components in the soil may react with the field leachate to neutralize the alkaline pH. Neutralization of the alkaline pH in the subsurface occurs via interaction of the field leachate with soil and groundwater acidity, soil vapor carbon dioxide, and carbonation (Sanger et al. 2020; Gupta et al. 2018; Oliveira et al. 2020).

The groundwater systems' neutralization is sourced from inorganic carbon species (e.g., bicarbonate, carbonic acid). Since groundwater systems are site-specific, RCA research seldom discusses groundwater chemistry; while, the soil neutralization capacity depends on the biological activity, soil type, and soil acidity (Gupta et al. 2018).

The soil acidity may be divided into two classes: (1) cation exchange capacity, known as "exchangeable" acidity, and (2) "hydrolytic" acidity. The exchangeable acidity consists of soil cations (i.e. alumino-hydroxo complexes) that act to release hydrogen ions (increasing acidity). The released hydrogen ions, along with the soils' natural hydrolytic acidity, will act to neutralize a portion of the alkaline RCA leachate until exhausted (Oliveira et al. 2020).

Gupta et al. (2018) used a geochemical model to show the soil neutralization capacity of a 1-meter soil column with variable soil acidity. The soil acidity ranged from 0.5 to 9 milliequivalents (meq) per 100 grams (g) of soil.

Gupta et al. (2018) determined a 1-meter soil column with a soil acidity of 0.5 meq/100 g of soil could neutralize the alkaline pH produced from exposure to RCA for 20-years at a liquid-to-solid (L/S) ratio of 0.5 L/kg. Their model results indicate that an increase in the soil acidity increases the potential to neutralize the alkaline pH. Gupta et al. (2018) results also indicate that the soil acidity cannot be relied on for long-term pH control, since at high L/S ratios, the soils' acidity is expended.

However, the modeling approach used a constant alkaline pH, where in reality carbonation would lower the pH of the field leachate infiltrating through the subsurface over time. By not accounting for this and other pH neutralization mechanisms decreasing the alkaline pH, Gupta et al. (2018) results may underestimate the neutralization of the alkaline pH.

Chen et al. (2020) evaluated the transport of alkaline pH through different subgrade soils. They evaluated four types of clayey Unified Soil Classification System soils: ML14, CL25, CH38, and SC10. The authors extrapolated the soil column experimental results using a numerical model to determine the amount of time the alkaline front takes to travel through five meters of soil.

The study indicates silt (ML14) and clay (CL25 and CH38) classified soils retard the movement of the alkaline pH front, compared to clayey sand (SC10). The chemical properties (i.e. cation exchange capacity) of clayey sand allow the alkaline front to migrate far quicker than through silt or clay classified soils. This implies the propagation of the alkaline pH front though the soil is directly proportional to the mineralogy of the soil (Chen et al. 2020).

As previously mentioned, RCA alkaline pH may be neutralized by inorganic carbon species in the subsurface. Oliveira et al. (2020) indicate organic matter content, capable of being transformed to carbon dioxide, present in the soil promotes carbonation and neutralization of the RCA alkaline leachate. The results suggest once the soil acidity is depleted, the carbonation could provide sufficient buffering against the alkaline RCA leachate (Oliveira et al. 2020).

Dilution of Released Pollutants

Dilution of the alkaline field leachate occurs when the leachate reaches a surficial water body or a subsurface aquifer (Engelsen et al. 2012, 2020). This dilution of the field leachate is expected to further neutralize the alkaline pH (Gupta et al. 2018; Townsend et al. 2016). The mixing of the RCA field leachate with groundwater and surface water is expected to dilute the concentrations of the released POC (Engelsen et al. 2012; Engelsen, 2020).

Townsend et al. (2016) investigated the dilution of pH-impacted water using a mass and charge balance approach. The authors determined an appreciable pH decrease at high dilution factors. They also observed a greater neutralization effect of the alkaline pH if the groundwater had a higher alkalinity[123].

Equations are provided in Engelsen et al. (2012) to calculate the site specific groundwater (DF_{gw}) and “hydrologic connectivity[124]” surface water (DF_{sw}[125]) dilution factors of the RCA leachate, shown in Equation (7) and (8) respectively (Engelsen et al. 2020). The infiltration rate (I; L/year) shown in Equation (9) can be calculated; using the infiltration constant (I_f; year/L) and annual precipitation (P; L/year) (Engelsen et al. 2012).

$$\mathrm{DF_{gw}} = (\mathrm{L_{gw}} \cdot \mathrm{I})/\left[(\mathrm{k} \cdot \mathrm{i} \cdot \mathrm{d_{mix}}) + (\mathrm{L_{gw}} \cdot \mathrm{I})\right] \qquad \text{Equation (7)}$$

$$\mathrm{DF_{sw}} = (\mathrm{k} \cdot \mathrm{i} \cdot \mathrm{d_{mix}} \cdot \mathrm{L_{sw}})/\mathrm{Q}_{sw} \qquad \text{Equation (8)}$$

$$\mathrm{I} = \mathrm{I_f} \cdot \mathrm{P}^2 \qquad \text{Equation (9)}$$

where:

- DF_{gw}: The groundwater dilution factor from mixing of the field leachant with the groundwater,
- DF_{sw}: The surface water dilution factor from mixing of the field leachant with the groundwater followed by connection to a surface water body,
- L_{gw}: The length of the stockpile in the groundwater flow direction,
- L_{sw}: The width (L) of the stockpile area perpendicular to the groundwater flow direction,
- k: The hydraulic conductivity (L/year),
- i: The hydraulic gradient (L/L),
- d_{mix}: The mixing zone thickness (L) of the aquifer, and
- Q_{sw}: The surface water flow rate (L^3/year).

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[123] Alkalinity refers to the ability of a solution to neutralize acids and bases to maintain a stable pH value (typically measured in mg/L of $CaCO_3$).

[124] Hydrologic connectivity refers to the hydrogeologic connection between surface water bodies and groundwater bodies. The hydrologic connection between surface water bodies and groundwater may be seasonal and/or depend on other environmental factors.

[125] The surface water dilution factor provided assumes the RCA field leachate is first diluted by groundwater and then enters the surface water body through hydrologic connectivity.

The released POC leachate concentrations from the stockpile are multiplied by the DF_{gw} and DF_{sw} to obtain the resulting surface water POC concentration (accounting for hydrogeologic connectivity).

Using a standard set of Norwegian parameters, the dilution factor for concrete leachate into groundwater is 7.1%, with hydrologic connectivity into a small stream with another 5% dilution factor (for a small stream). The dilution factor for exclusive discharge to a surface water body from a RCA stockpile is not discussed (Engelsen et al. 2012).

Numerous mass transport parameters are not accounted for in Equations (7) and (8). For instance, the previous mentioned equations do not account for the chemical sorption to the sediments, lateral dispersion, and/or other technical aspects involved in the fate and transport of the leachate in the aquifer (Engelsen, 2020; Fetter, 2017).

If the ambient groundwater or surface water concentrations of POC are detectable, then these equations may need to be adjusted to account for the upgradient concentration (Chapter 173-340 WAC).

Ecotoxicity Assays

The synergistic, antagonistic, and bioavailability of the chemically complex RCA field leachate/laboratory eluate (e.g., alkaline pH, heavy metals, organics) may contribute to the toxic response observed and make the results difficult to comprehend (Rodrigues et al. 2017; Tiruta-Barna and Barna, 2013). International studies evaluating the toxicity of RCA laboratory eluate typically use short-term ecotoxicity tests representing a conservative approach. For a description of the reviewed ecotoxicity tests from the literature, see the section on Leaching Methods and Ecotoxicity Assessments.

Lemna gibba L.

Brás et al. (2018) evaluates the effect of concrete eluate, produced using laboratory batch leaching method EN 12457-4 (2002), on the inhibition of growth of the duckweed fronds. Duckweed, or *Lemna gibba L.*, is able to grow across a pH range of 3.5 to 10.5 S.U., but the optimal pH range is 4.5 to 8.3 S.U.

The constraining factor for plant growth may be the high degree of salinity of the concrete eluate. The laboratory created concrete eluate had an electrical conductivity of 4,200 uS/cm, which exceeds the upper limit of tolerability by medium sensitivity plants (4,000 uS/cm) (Brás et al. 2018).

Compared to the control, consisting of deionized water, and other materials assessed, the concrete eluate had the greatest impact on the growth of duckweed fronds. The concrete eluate generated a mean of twelve (12) fronds, while the average control eluate yielded 22 fronds. However, the higher mineral content of the concrete laboratory eluate produced a positive effect on the photosynthetic pigment development (Brás et al. 2018).

Daphnia magna and Other Biota

The *Daphnia magna* survival pH is generally between 7 and 8.5 S.U. The alkaline pH produced from water's exposure to RCA was between 10 and 12.4 S.U. (Choi et al. 2013; Rodrigues et al. 2017). The pH may be the cause of adverse effects to the test species (Choi et al. 2013).

Choi et al. (2013) evaluated the ecotoxicity of an alkaline material composed of ordinary portland cement and Pozzolan ash supplementary cementitious material, the authors considered the specimen concrete. They evaluated the ecotoxicity of the laboratory created concrete eluate produced after allowing the specimen to interact with water for 30 days. The concentrations tested on *Daphnia magna* were 100, 50, 25, 12.5 and 6.25% of concrete laboratory eluate diluted with deionized water.

The effective concentration (EC_{50})[126] of the concrete laboratory eluate could not be determined because the results showed 100% immobilization of the *Daphnia magna* after 24-hr exposure to the lowest concentration tested, 6.25% of the concrete eluate. It is also noted in Choi et al. (2013) that the eluate produced by concrete created at a 0.4 water-to-cementitious material ratio was highly toxic to *Daphnia magna* and that incorporation of loess into a concrete may potentially reduce the toxicity (Choi et al. 2013).

Rodrigues et al. (2017) developed a process to assess the ecotoxicity potential of construction materials. The ecotoxicity of a laboratory fortified concrete, hereby referred to as RCA, eluate was characterized using the proposed framework. They estimated the ecotoxicity indices using three acute toxicity tests, previously described in the Ecotoxicity Assays section. The ecotoxicity indices of RCA laboratory eluate reported in Rodrigues et al. (2017) are provided in Table 20.

Table 20: The ecotoxicological indices (shown as a percentage [%] of concrete eluate) of the ecotoxicity tests performed with laboratory fortified concrete eluate.
From Rodrigues et al. (2017).

		Vibrio fischeri	***Daphnia magna***		***Saccharomyces cerevisiae***
Material	**pH**	**EC_{50} (30 min Exposure)**	**EC_{50} (24 hr Exposure)**	**EC_{50} (48 hr Exposure)**	**EC_{50} (16 hr Exposure)**
Concrete Eluate Ecotoxicity Indices (% of laboratory eluate)	12.4 S.U.	Greater than 100% 'No Effect'	6.8%	5.5%	30.2%

The exposure to the RCA eluate caused adverse effects to two living organisms, the *Saccharomyces cerevisiae* yeast and *Daphnia magna* water fleas. The EC_{50} for *Daphnia magna* occurred at a lower percentage of the RCA laboratory eluate than the yeast (Rodrigues et al. 2017).

In ecotoxicology if an EC_{50} is greater than 100%, the results indicate the antagonist is of 'no effect' or the effect is negligible to the test organism (Viegas, 2020). The exposure to 100% laboratory eluate concentration induced 'no effect' to the *Vibrio fischeri*. It is important to remember that an EC_{50} greater than 100% implies the response in the test organism [*Vibrio fischeri*] could not be detected, not that the antagonist [RCA laboratory eluate] is non-toxic (Kennedy et al. 2000).

Rodrigues et al. (2017) recommend the use of the *Saccharomyces cerevisiae* yeast as a screening tool for ecotoxicity of demolition waste, due to its ease of performing and low-cost. They suggest components other than the specific metals evaluated in the study (i.e. metals not sampled in the study) may have influenced the test organism's response to the RCA eluate.

Despite the lack of *Vibrio fischeri* bacteria inhibition, the toxicity to *Daphnia magna* allowed the authors to qualify the RCA laboratory eluate as having evidence of ecotoxicity under the French proposal document Criteria on the Evaluation Methods of Waste Ecotoxicity (ADEME, 1998; Rodrigues et al. 2017).

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[126] A half maximal effective concentration (EC_{50}) refers to the empirical/statistical estimate of an antagonist's (i.e. toxicant, field leachate, laboratory eluate) concentration to induce a particular response in half (or 50%) of the test population under a particular set of conditions.

Environmental Best Management Practices

The re-use of recycled concrete aggregate (RCA) is typically either disposal in a landfill or recycling (EPA, 2018). Recycling of the material commonly includes crushing, transporting, and storage in a stockpile until its potential re-use – for example, as recycled aggregate concrete (RAC) or as road (sub-)base (Leigh and Patterson, 2004). The use of sprinklers for dust suppression is common to reduce the air emissions/pollutants from concrete recycling (del Rio Marino et al. 2010).

Similar to other forms of construction activities, the recycling of concrete has raised environmental concerns on water quality, air quality, noise, waste generation, and other local impacts (Snyder et al. 2018). The application of best management practices (BMPs) and proactive construction and design controls at facilities storing RCA stockpiles have generally been able to mitigate these environmental concerns.

A general permit[127] violation, in the form of a discharge of water in exceedance of water quality regulations, may occur at facilities (1) when the BMPs are not implemented correctly or (2) due to user error. In order to protect the waters of Washington State (State), there are several BMPs identified in the general permit that attempt to mitigate the negative environmental effects of storing stockpiles of RCA.

Employee training and education along with proper housekeeping are repeatedly stated as one of the more critical BMPs to reduce environmental impacts of concrete recycling and production (CalCIMA, 2012; EPA, 2018; Raju and Kameswari, 2015). The BMPs for the production and storage of cementitious materials are sourced in part from California, EPA, Ecology's Stormwater Management Manual for Eastern and Western Washington, and Snyder et al. (2018).

The different re-uses of RCA (i.e. back-fill, construction, road-base) may present unique but similar regulatory challenges. For example, the stockpiling concrete recycling facility with properly implemented BMPs will present different environmental risks than stormwater runoff from a construction site.

Sand and Gravel General Permit Requirements

The general permit requires that all permitted facilities develop a Site Management Plan (SMP). This plan is a dynamic site-specific document described in detail in the general permit. The following is a brief description of the SMP, which includes a site map and four main parts:

1. Erosion and Sediment Control Plan (ESCP)

The ESCP contains the sediment and erosion control BMPs the permittee will implement at a facility and a schedule for implementation.

2. Monitoring Plan

A Monitoring Plan must be developed for compliance with water quality discharge limits established in the general permit. The Monitoring Plan must identify monitoring points providing representative sampling of all point source discharges to waters of the State.

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[127] *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* 2021.

3. Stormwater Pollution Prevention Plan (SWPPP)

The State Stormwater Management Manual for Western/Eastern Washington provides guidance in the development of an SWPPP (Ecology, 2019b, 2019c). In addition, EPA's Developing Your Stormwater Pollution Prevention Plan can be used to assist with developing the SWPPP (EPA, 2007). The SWPPP contains several sections as described:

- Measures to Prevent Commingling (of stormwater and process water).
- Runoff Conveyance and Treatment BMPs
  - A list of runoff and treatment BMPs are provided, although it is noted the BMPs applied on-site are not limited to those listed.
- Innovative BMP
  - BMPs reducing energy consumption or providing better operational management practices beyond Ecology's Stormwater Management Manual (Eastern/Western Washington) are encouraged if they achieve compliance with the general permit.
- Inventory of Material and Pollutant Sources
  - An inventory is needed to catalogue the types of materials handled at the site that are exposed to runoff or precipitation, as well as a potential pollutant and pollutant sources.
- Source Control BMPs
  - This section describes the BMPs implemented to achieve all known, available, and reasonable methods of prevention, control, and treatment for compliance to the stormwater discharge water quality limits identified in the general permit.
- Concrete Recycling BMPs
  - If the permittee received coverage under the general permit for the first time on or after April 1, 2016 for their facility, then the facility must not create a new RCA stockpile in specific locations (See general permit in Appendix A).
  - This section establishes material acceptance procedures to ensure inbound RCA is not a source of dangerous waste.

4. Spill Control Plan

The Spill Control Plan must include the material(s) of concern, spill control plan strategies, and response to a spill. All spills or unplanned discharge of an oil or hazardous material must be reported by calling the National Response Center and the Washington Emergency Management Division.

Water Quality BMPs

Water quality BMPs are implemented at the planning and design stage to mitigate specific RCA contaminant(s) from entering a water body of the State. The majority of BMPs described in the literature are to prevent the migration of pollutants of concern (POC) from the manufacturing or re-use of concrete materials. Specific BMPs related to the manufacturing and production of cement/concrete are provided because similar mitigation strategies are necessary.

The Stormwater Management Manual for Western and Eastern Washington provides several applicable BMPs (Ecology, 2019b, 2019c). In addition to others, these include:

- BMP C252: Treating and Disposing of High pH Water (Western Washington Manual)
- BMP C154 (E): Concrete Washout Area (Western/Eastern Washington Manual)
- S429 BMPs for Storage or Transfer (Outside) of Solid Raw Material, Byproducts, or Finished Products (Western Washington Manual)
- BMP C151E: Concrete Handling (Eastern Washington Manual)

In addition, the EPA provides a Stormwater BMP on Concrete Washout (EPA, 2012b).

California Construction & Industrial Materials Association (CalCIMA) published a draft Ready Mixed Concrete Process Water BMPs manual in conjunction with the California State Water Resources Control Board. This manual describes in detail the applicability, target constituents, and estimated costs (capital and maintenance) of implementing the recommended BMPs. A draft manual is available (CalCIMA, 2012).

Snyder et al. (2018) presents water BMP mitigation strategies for the usage of RCA as unbound road (sub-)base, fill, and use in RAC. They emphasize knowledge of the original use of the concrete is critical in determining the type of contamination to expect.

The placement of the RCA in a stockpile should be designed with water protection in mind (i.e. distance from the waterway and redundant BMPs). Snyder et al. (2018) suggest diverting RCA-impacted stormwater runoff using conventional BMPs (e.g., straw bales, grass/filter channels, and berms around stockpiles; maintaining a perimeter around and covering stockpiles; treatment of the water as needed).

Mitigation strategies for the reduction of the total suspended and dissolved solids in the stormwater runoff from RCA stockpiles include the use of bioswales, hardy vegetation, and “floc” logs. The use of pH “shock” logs or pH logs, pH adjustment technology, carbon dioxide bubblers, and chemical addition (according to Ecology-specific guidance) are also suggested to help neutralize the alkaline pH leachate (Snyder et al. 2018). The structural BMPs implemented on-site need to be maintained and inspected frequently to ensure proper operation.

Depending on the soil acidity, the soil may be able to neutralize the alkaline leachate. This is not a pragmatic solution as the soil may ultimately reach a finite neutralization capacity (Chen et al. 2020; Gupta et al. 2018; Snyder et al. 2018). The propagation of the alkaline front through a soil is dependent on the chemical constituents of the soil (Chen et al. 2020).

The Washington State Department of Transportation (WSDOT) limits the height of all aggregate stockpiles to 24 feet, except those in excess of 200 cubic yards, which cannot exceed 4 feet in height (WSDOT, 2020). The limitation of the height of the stockpile decreases the loss to air, which may decrease the amount of fines settling into nearby waterways (Oliveira et al. 2019).

Future Studies

The general permit[128] requires a Discharge Monitoring Report (DMR) be submitted to the Department of Ecology (Ecology) on a quarterly basis. The DMRs provide details about the water quality discharged from actively permitted facilities, These include the type of wastewater discharged (i.e. stormwater, process water) and the type of receiving waters (i.e. surface water, groundwater). Facilities designated as inactive are only required to submit DMRs in certain circumstances.

Other than basic leachate properties provided by the DMRs, there is a lack of recycled concrete aggregate (RCA) leachate data generated by stockpiles of RCA in Washington State. In order to establish a correlation between RCA presented in the reviewed literature to Washington State-specific RCA, a leaching study is recommended.

Options for future studies of RCA leachate include:

- Investigate the RCA leachate from concrete stockpiles at the monitoring point (defined in the facilities general permit) at numerous permitted RCA facilities,
- Conduct a laboratory leaching investigation of RCA samples collected from stockpiles across the state, and/or
- Conduct a field leaching investigation of RCA samples at a site with an established baseline.
- Investigate the effectiveness of best management practices (BMPs) on reducing the RCA leachate to levels below effluent limits in the general permit.

If a field and/or laboratory leaching investigation is selected, certain parameters will need to be considered:

- **Type of RCA material to be tested**
  - Type of cementitious material to be assessed (e.g. CEM I, CEM II, fly ash, silica fume, slag).
  - Source of the aggregates (possibly a control stockpile).
  - Water-to-cementitious material (*w/cm*) ratio, porosity, permeability, and density of the original concrete.
  - Type of crusher used to create the RCA.
  - Amount and thickness of the residual, adhered cementitious material on the RCA.
  - Shape and the particle size of the RCA.
  - Degree of carbonation or the age of the RCA.
  - Source and original use of the RCA.
  - Addition of new RCA over time to the concrete stockpile.
- **Environmental factors impacting the RCA leachate**
  - The relative humidity, environmental conditions, temperature, particle size, and partial pressure of carbon dioxide will influence the carbonation rate of the RCA.
  - The amount and quality of precipitation will influence the carbonation rate and the leaching of pollutants of concern (POC) from RCA.
  - The above environmental factors change with time and space and therefore must be measured throughout the study due to their potential effect on the leachate.

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[128] *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* 2021.

- **Type of leaching methods to be employed**
    - Laboratory leaching methods, such as the four new EPA leaching methods described in Kosson et al. (2014a, 2019), can be used to describe the RCA eluate characteristics.
    - A field RCA leachate investigation may be conducted to assess the potential leaching of chemicals; common, heavy, and trace metals; and organic leachates as a function of time.
        - The formation of a leached layer, or depletion zone, and/or a first flush on the RCA will affect the release of POC over the course of the investigation (Engelsen et al. 2012, 2017; Märkl, 2018).
- **Type of monitoring to be conducted in the field leaching investigation**
    - Measure the chemical parameters of the leachate immediately following contact with the RCA in order to prevent influence from atmospheric conditions.
    - Commonly, field leaching studies of RCA collect the leachate using a synthetic membrane or liner following percolation through the RCA. This collection method will determine the potential environmental risk to surface waters following the leachates' contact with RCA.
    - Groundwater monitoring of RCA leachate is seldom reported in the literature and may require application of standard groundwater approaches (i.e. upgradient and downgradient piezometers and/or wells for characterization of the subsurface soil and groundwater).
- **Type of chemical analyses to be conducted in the field and/or laboratory**
    - Chemical parameters of leachates (e.g., pH, electrical conductivity, oxidation-reduction potential) should be measured in the field, if possible, and before laboratory analysis.
    - Common, heavy, and trace metal analyses should be performed in the laboratory with an appropriate approved method.
    - Organic analyses (e.g., PCBs, PAHs, and possibly supplementary mixtures) should be performed in the laboratory with an appropriate approved method.
- **Application of ecotoxicity assays to the RCA leachate**
    - Develop an ecotoxicity testing strategy to evaluate the toxicity of the chemically complex RCA leachate to Washington State aquatic test organisms.
    - Evaluate the applicability of Chapter 173-205 WAC to effluents from facilities permitted with concrete recycling identified as an activity.
- **Implementation and effect of best management practices (BMPs) applied to RCA**
    - Study the implementation of BMPs applied to the RCA leachate (i.e. floc logs, pH logs, straw bales, bioswales, carbon dioxide bubblers, berms, chemical addition, and height of stockpile).
    - Investigate the effect of BMPs on the leachate's pH; total dissolved solid; total suspended solid; turbidity; common, heavy, and trace metals; and organic pollutants of concern (POC).

The use of these proposed future study techniques may help us understand the fate and transport of the alkaline pH as well as POC concentrations released from RCA.

Conclusions from the Literature

The intended purpose of this report is to provide the Washington State Department of Ecology (Ecology) Water Quality Program with the current science of pollutants of concern (POC) released from recycled concrete aggregate (RCA) that may potentially exceed (not meet) effluent Sand and Gravel General Permit (general permit) limits or Washington State (State) water quality criteria[129]. This report should be used to identify potential POC released from RCA, with the understanding that the alkaline pH may be neutralized and other POC concentrations decreased below levels of concern by best management practices (BMPs), treatment technologies, and site-specific conditions.

According to Snyder et al. (2018), when properly maintained conventional and innovative BMPs as well as construction controls are employed in replicate at concrete recycling facilities negative environmental impacts may be mitigated. It should be noted that the BMPs reviewed by Snyder et al. (2018) might not exemplify BMPs currently employed at facilities in the State. The literature on BMP efficiency was not researched as this extends beyond the scope of this literature review.

The POC representing a potential environmental risk to the waters of the State are shown to depend primarily on the solubility and availability of the minerals present on the RCA surface (Barbudo et al. 2012; Del Rey et al. 2015; Galvín et al. 2013). The pH of the laboratory eluent/field leachant is one of the primary controlling factors influencing the solubility of POC from RCA. The diminution of the leachate pH as the RCA carbonates is combined with the production of a leached layer, or mineral depletion zone, in the field leaching scenarios (Engelsen et al. 2006, 2012, 2017; Sadecki et al. 1996).

The following section contains conclusions of the RCA literature review. The results of this literature review should be viewed judiciously with the knowledge the RCA material has a wide degree of heterogeneity. Further study is required to establish a correlation between international and domestic RCA and Washington State-specific RCA. Adding another degree of uncertainty, the leaching method parameters vary significantly between the reviewed studies.

This report is not a review of the concrete or concrete recycling industry, but rather a literature review of the characteristics of RCA and RCA leachate, as well as potential environmental concerns when the material is not properly managed. The sensible management of stockpiles and implementation of BMPs will reduce the negative environmental impacts while retaining the net benefits of recycling concrete.

Leaching Methods and Environmental Pollutants of Concern

It is apparent the leaching method and parameters of the test (i.e. pH, liquid-to-solid ratio, and particle size) should reflect the intended purpose for evaluating the RCA. If the proper leaching test method is not selected, the leaching results provided to the regulator may not be able to verify if the POC leachate concentrations are below regulatory criteria.

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[129] "State water quality criteria" refers to Washington State Chapter 90.48 RCW: Water Pollution Control, Chapter 173-201A WAC: Water Quality Standards for Surface Waters, Chapter 173-200 WAC Water Quality Standards for Groundwaters, and Chapter 173-204 WAC Sediment Management Standards of the State of Washington. *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* 2021.

The leaching methods are not designed to characterize field behavior, due to expense and complexity. Rather, the leaching tests verify if the POC laboratory eluate concentrations are below regulatory levels under the provided leaching test conditions (Maia et al. 2018).

The availability and solubility of the POC minerals on the RCA into the field leachate/ laboratory eluate may reflect the:

- Inherent heterogeneity of the recycled concrete State-wide.
- Sorption of a contaminant onto the concrete from the initial use.
- Formation of a leached layer or depletion zone on the concrete surface.
- First flush or surface wash-off effect.
- Degree of carbonation experienced by the concrete.

The general permit sets effluent limits from facilities on the discharge pH, total dissolved solids, total suspended solids, and turbidity to waters of the State. In addition, the general permit considers the discharge of an oil sheen or petroleum products to waters of the State a violation.

Water discharging from permitted facilities must not cause or contribute to a violation of the State's water quality criteria or sediment management standards and 40 CFR 131[130] (Chapter 173-200 WAC; Chapter 173-201A WAC; Chapter 173-204 WAC)[131].

Leaching Methods

Laboratory batch leaching results provide a simple tool for researchers, regulators, and facility site managers to evaluate the RCA environmental risk (i.e. Method 1311). However, the use of batch leaching tests may provide erroneous results due to the failure to incorporate specific leaching parameters encountered in the materials disposal scenario. Parallel batch leaching methods provide a representative assessment of the materials leaching behavior under varying leaching parameters.

Batch laboratory leaching methods, such as Method 1311, 1312, 1313, and 1316, fail to provide "field truth" leaching from RCA because the abrasive stirring employed by the tests expose fresh, non-carbonated, and leachable RCA surfaces to the laboratory eluent. Regardless, batch leaching methods have been proven consistent, inexpensive, and potentially valuable.

Column laboratory leaching methods, such as Method 1314, provide more representative leaching results for evaluation of environmental risk than batch leaching methods. Especially if the column leaching method incorporates wetting and drying cycles, which will enhance the carbonation of the RCA emulating the changes in the RCA due to long-term storage.

Environmental field parameters are expensive and difficult to measure, reproduce, and incorporate into laboratory leaching methods. Field leaching studies performed on domestic and international concrete material are generally considered the most representative leaching results due to incorporation of environmental conditions.

The amount of time the leachate is no longer in contact with the RCA after the leaching is also important. Longer atmospheric exposure of the RCA leachate will influence the leachates chemical properties (i.e. pH, precipitation of minerals). Thus, the collection of field measurements (e.g., pH, oxidation-reduction potential) should be performed on-site to be representative of RCA leachate discharge.

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[130] 40 Code of Federal Regulations 131: Water Quality Standards.

[131] *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* 2021. The general permit section being cited here is S3.B.

Environmental Pollutants of Concern from RCA

The alkaline pH released from non-carbonated or slightly carbonated RCA poses a risk of exceeding (not meeting) the effluent pH limits in the general permit as well as State water quality criteria. Neutralization of the RCA alkaline leachate occurs via different natural mechanisms in surface waters and subsurface waters. In addition, implementation of best management practices (BMPs) help to neutralize the alkaline pH leachate for compliance to the general permit.

The degree of carbonation of the RCA influences the pH of the leachate and thus the solubility of mineral phases present on the RCA. In a field leaching scenario, the release of metals from RCA generally decreases with increasing aging due to the formation of a leached layer, or depletion zone. Whereas in the laboratory, the release of metal POC from RCA with varying degrees of carbonation appears to be dependent on the laboratory eluate metal being assessed.

The calcium and sulfate release ratios appear to have a strong correlation to the total dissolved solid concentration in the RCA laboratory eluate.

In general, the release of inorganic POC from RCA as a function of the liquid-to-solid (L/S) ratio decreases as the L/S ratio increases in laboratory leaching tests. This is potentially due to the dilution of the laboratory eluate.

The reviewed literature suggests there is increased adhered cement-paste present on the finer-grained RCA particles. The laboratory leaching methods suggest a generally higher release of inorganic POC from the finer-grained RCA particles.

When the concrete is not designed for exposure to sulfates, chlorides, and acidic liquids, subsequent exposure may lead to the degradation of the cement material. Generally, the partitioning of inorganic constituents from the RCA into the field leachate/laboratory eluate is enhanced by increased salinity of the field leachant/laboratory eluent.

The predominant form of chromium (primarily as hexavalent chromium), arsenic, selenium, and usually antimony released from RCA in the field and laboratory leaching methods is in the (oxy)anionic state.

The results of this literature review acknowledge that POC are released from RCA with the potential to adversely affect water quality in waters of the State when proper BMPs and treatment technologies are not utilized. The POC released from RCA posing an environmental concern to State groundwater and surface water bodies are different due to differing water quality criteria.

It should be noted that the POC identified in this literature review do not account for diminution by BMPs employed by the concrete recycling facilities or site-specific conditions (i.e. acidic soils, organic matter). Therefore, the identified POC should be viewed judiciously with the inherent uncertainty associated with the leaching method and the wide degree of heterogeneity of RCA material.

Surface Water Pollutants of Concern from RCA

Possible POC released from RCA with the potential to exceed State surface water quality (Chapter 173-201A WAC) may include, but are not limited to:

- The alkaline pH, sourced primarily from the dissolution of calcium hydroxide, of freshly crushed or slightly carbonated RCA may exceed the surface water quality criterion upper limit of 8.5 S.U. There is a large range of pH reported in the literature from above 13 S.U. for freshly crushed RCA to below 8 S.U. for a fully carbonated RCA.

- Total suspended solid concentrations observed in the RCA leachate were above the effluent limits listed in the general permit.
- Turbidity may exceed the effluent limits specified in the general permit for finer-grained RCA. However, the average turbidity measured is far below the effluent limits of the general permit.
- The release of chloride from RCA may exceed the State surface water quality criterion for a short period of time. As chloride is highly soluble, the release concentration should quickly decrease.
- Release of antimony, arsenic, chromium (VI), copper, nickel, and selenium from RCA frequently exceed at least one of the State surface water quality criteria.
- The release of lead, mercury, and zinc from RCA may exceed at least one of the State surface water quality criteria, albeit these metals do not commonly exceed the State surface water quality criteria.
- Leaching of specific polycyclic aromatic hydrocarbons (PAH) from RCA above the State surface water quality criteria was observed for benzo(a)anthracene, benzo(a)pyrene, and benzo(b+j+k)fluoranthene.
- The RCA polychlorinated biphenyl (PCB) laboratory eluate concentration was investigated over one and a half decades ago. None of the RCA laboratory eluate samples assessed for PCB exceeded 0.01 ug/L. The State's surface water quality criterion for PCBs is orders of magnitude lower than 0.01 ug/L.

Numerous POC (barium, iron, manganese, sulfur (as sulfate), and total dissolved solid) do not have state surface water quality criteria, but did exceed the State groundwater quality criteria.

Groundwater Pollutants of Concern from RCA

Possible POC released from recycled concrete aggregate with potential to exceed State groundwater quality (Chapter 173-200 WAC) may include, but are not limited to:

- The alkaline pH, sourced primarily from the dissolution of calcium hydroxide, of freshly crushed or slightly carbonated RCA may exceed the State groundwater quality criterion upper limit of 8.5 S.U. There is a large range of pH reported in the literature from above 13 S.U. for freshly crushed RCA to below 8 S.U. for a fully carbonated RCA.
- Total dissolved solid concentrations (TDS) observed in the RCA field leachate/laboratory eluate were above the effluent limits listed in the general permit. The TDS concentration appears to be related to the calcium and sulfate concentration in the leachate.
- Sulfur (as sulfate) released from RCA exceeds the State groundwater quality criterion by a factor between 2.7 and 23.
- The chloride release concentration from RCA may exceed the State groundwater quality criterion. However, chloride is highly soluble and expected to rapidly decrease below the groundwater quality criterion.
- Release of arsenic[132], chromium[133], and selenium[134] from RCA exceed the State groundwater quality criteria frequently in the field and laboratory leaching methods.

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[132] The metal is measured as total metal in Chapter 173-200 WAC.

[133] The metal is measured as total metal in Chapter 173-200 WAC.

[134] The metal is measured as total metal in Chapter 173-200 WAC.

- The release of barium[135], iron[136], lead[137], and manganese[138] from RCA seldom exceed the State groundwater quality criteria in the laboratory and field leaching methods.
- Leaching of PAH from RCA above the State groundwater criteria is possible.

Antimony, nickel, total suspended solids, and turbidity do not have groundwater quality criteria, but did exceed the State surface water quality criteria. Zinc[139], copper[140], and mercury[141] were not released from RCA above the State groundwater quality criteria, although these metals were released from RCA above State surface water quality criteria. PCBs do not leach from RCA at concentrations above 0.01 ug/L, the State groundwater quality criterion, and thus are not expected to be a concern to State groundwater bodies.

pH Neutralization and Dilution

The carbonation process produces physical, chemical, and mineralogical changes to the RCA that influences both the material pH and the release of POC, identified in the previous section. In a field leaching scenario, the highest leachate pH is observed immediately after the leachate leaves the RCA material. The RCA alkaline pH may be neutralized and potential risks minimized depending on the specific conditions existing on-site (i.e. infiltration rate, organic matter content of soils, geometry and motion of water through an RCA stockpile).

Surface water pH neutralization of the alkaline leachate occurs primarily from the presence of excess hydrogen ions, known as “hydrolytic acidity,” and the carbonate system. Whereas subsurface pH neutralization mechanisms occur via interaction of the leachate with soil and groundwater acidity, soil vapor carbon dioxide, and carbonation.

The propagation of the alkaline pH front though the soil is directly proportional to the mineralogy of the soil (Chen et al. 2020). Silt (ML14) or clay (CL25 and CH38) classified soils[142] are shown to retard the propagation of the alkaline pH front compared to clayey sand (SC10). The soil acidity may be divided into two classes: cation exchange capacity, known as “exchangeable” acidity, and “hydrolytic” acidity.

The use of RCA as road-base results in a short-term alkaline pH discharge as opposed to a longer-term discharge associated with RCA stockpiles at processing facilities. When subsurface discharge of the field RCA leachate occurs, the soil acidity will become exhausted at a high liquid-to-solid ratio. After the soil acidity is expended and depending on site-specific conditions (i.e. soil organic matter), the soil may use carbonation to sufficiently buffer against the alkaline pH (Oliveira et al. 2020).

There is potential for RCA stockpile water to runoff into a nearby water of the State, if minimum stockpile setbacks from State water bodies are not maintained (Snyder et al. 2018). Dilution of the RCA effluent discharge results in a pollutant of concern concentration that are substantially lower than the non-diluted effluent concentration. The impacted groundwater could intercept a surface water body, hydrologic connectivity, and be further diluted by the ambient surface water (Engelsen et al. 2020).

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[135] The metal is measured as total metal in Chapter 173-200 WAC.
[136] The metal is measured as total metal in Chapter 173-200 WAC.
[137] The metal is measured as total metal in Chapter 173-200 WAC.
[138] The metal is measured as total metal in Chapter 173-200 WAC.
[139] The metal is measured as total metal in Chapter 173-200 WAC.
[140] The metal is measured as total metal in Chapter 173-200 WAC.
[141] The metal is measured as total metal in Chapter 173-200 WAC.
[142] Classified by the Unified Soil Classification System.

Using a standard set of Norwegian parameters, the dilution factor for concrete leachate into groundwater is 7.1%, with hydrologic connectivity into a small stream with a dilution factor of 5%. The dilution factor for exclusive discharge to a surface water body from a RCA stockpile is not discussed. Numerous mass transport parameters (i.e. chemical sorption, lateral dispersion) are not accounted for in the equations used to calculate these dilution factors (Engelsen et al. 2012, 2020).

Application of Whole Effluent Toxicity Testing and Limits

The purpose of the whole effluent toxicity testing in Washington State is to define toxicity limits for inclusion into the National Pollutant Discharge Elimination System (NPDES) permit (Chapter 173-205 WAC). The goal of the whole effluent toxicity testing is to eventually eliminate the discharge of toxics in toxic amounts. For the application of Chapter 173-205 WAC to facilities with concrete recycling identified as an activity, there must be a "suspected toxicity because of apparent damage to aquatic biota" (WAC 173-205-040(1)(f)).

There have been at least three studies published investigating the ecotoxicity of concrete laboratory eluate on aquatic organisms. The biota assessed for their toxicity to concrete laboratory eluate were *Daphnia magna* water fleas, *Vibrio fischeri* bacteria, *Saccharomyces cerevisiae* yeast, and *Lemna gibba* L., also known as Duckweed. Adverse effects were observed following exposure to the concrete laboratory eluate for *Daphnia magna*, *Saccharomyces cerevisiae*, and *Lemna gibba L.* (Brás et al. 2018; Choi et al. 2013; Rodrigues et al. 2017).

The half maximal effective concentration for *Daphnia magna* is 6.8% and 5.5% at a 24- and 48-hour exposure, respectively (Rodrigues et al. 2017). The ecotoxicity of the concrete leachate may produce adverse effects to aquatic biota.

The applicability of Chapter 173-205 WAC to facilities with concrete recycling identified as an activity in the general permit needs to be assessed. This will help determine if State-specific concrete is toxic to State aquatic test organisms.

Best Management Practices (BMPs)

Similar to other forms of construction activities, the recycling of concrete has raised environmental concerns about water quality, air quality, noise, waste generation, and other local impacts. A general permit[143] violation, in the form of a discharge of water in exceedance of State water quality regulations, may occur at facilities (1) where the BMPs are not correctly implemented or (2) due to employee error.

Several BMPs are listed in the general permit, Eastern/Western Washington State Stormwater Management Manuals, and other sources to reduce the negative environmental impact while recycling concrete (Ecology 2019b, 2019c; Snyder et al. 2018; CalCIMA, 2012).

The application of BMPs (i.e. proactive construction and design controls, setbacks from waters of the State, application of conventional and innovative BMPs) at facilities storing RCA stockpiles may be capable of mitigating environmental concerns. Proper employee training and maintenance of BMPs employed are required to maintain the effectiveness of the BMPs (Snyder et al. 2018).

In addition, there is an environmental risk if proper dust control is not implemented at the concrete recycling facility. RCA has been shown to produce titanium and iron oxide

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[143] *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* 2021.

nanoparticles, carbon nanotubes, and other fine particulates. These particulates may present an inhalation hazard to nearby residents and animals and have the potential to reach nearby waters of the State affecting water quality (del Rio Marino et al. 2010; Oliveira et al. 2019).

The proper use of BMPs may provide adequate environmental protection from the POC released from RCA identified in this literature review. Ecology encourages the recycling of concrete material to preserve virgin aggregate sources, reduce greenhouse gas emissions, and lessen the disposal in landfills, as well as for economic reasons. Concrete recycling process controls must be monitored to ensure environmental risks are minimized, while retaining the net benefits of concrete recycling.

Future Work

One of the intended purposes of this report is to identify the pollutants of environmental concern potentially leaching from RCA to better inform Ecology's Water Quality Program for the development of appropriate requirements for concrete recycling. This report acknowledges the uncertainties associated with using international and domestic RCA material as well as a diverse set of leaching methods to identify the environmental hazards. It is recommended that Ecology establish a correlation between RCA presented in the reviewed literature to Washington State-specific RCA.

Options for future study into this material include:

- Investigate the RCA leachate from concrete stockpiles at the monitoring point (defined in the facilities general permit) at numerous permitted RCA facilities,
- Conduct a laboratory leaching investigation of RCA samples collected from stockpiles across the state, and/or
- Conduct a field leaching investigation of RCA samples at a site with an established baseline.

Additionally, Ecology may investigate the effectiveness and utilization of BMPs on reducing the RCA leachates' environmental hazards to below effluent limits in the general permit and State water quality standards. The use of the proposed future study techniques may be able to highlight the fate and transport of the RCA leachate in the environment.

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Ecology documents prepared after June 12, 2014 must identify information sources by the following 11 categories:

1. Peer review is overseen by an independent third party.
2. Review is by staff internal to Department of Ecology.
3. Review is by persons that are external to and selected by the Department of Ecology.
4. Documented open public review process that is not limited to invited organizations or individuals.
5. Federal and state statutes.
6. Court and hearings board decisions.
7. Federal and state administrative rules and regulations.
8. Policy and regulatory documents adopted by local governments.
9. Data from primary research, monitoring activities, or other sources, but that has not been incorporated as part of documents reviewed under other processes.
10. Records of best professional judgment of Department of Ecology employees or other individuals.
11. Sources of information that do not fit into one of the other categories listed.

The information source for each of the following documents is shown in brackets "[#]" at the end of the citation.

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Glossary, Acronyms, and Abbreviations

Glossary

Additives: See Admixture.

Additions: See Admixture.

Admixtures: Concrete admixtures are additives, additions, superplasticizers, and polymers that are added to portland cement to reduce the required water, improve durability and workability, and improve other properties of the concrete mixture. There are several types of admixtures commercially available.

Alkalinity: Refers to the ability of a solution to neutralize acids and bases to maintain a stable pH value (typically measured in mg/L of $CaCO_3$).

Amphoteric: A compound, especially metal (hydr)oxide capable of reacting as both an acid and a base.

Anion: An ion with a negative charge.

Anthropogenic: Human-caused.

Argillaceous: General term applied to material consisting of mainly silica, alumina, and oxides of iron; a common argillaceous material is clay and/or shale.

Best Management Practices (BMPs): General definition means schedules of activities, prohibitions of practices, maintenance procedures, and other physical, structural and/or managerial practices to prevent or reduce the pollution of waters of the state. BMPs include treatment systems, operating procedures, and practices used to control plant site runoff, spillage or leaks, sludge or waste disposal, and drainage from raw material storage. In this permit, BMPs are further categorized as operational, source control, erosion and sediment control, and treatment.

Calcareous: Contains calcium (magnesium) carbonate.

Cation: An ion with a positive charge.

Cement clinker: The non-volatile remains of the calcareous, argillaceous, and ferric materials travelling through the kiln. The clinker is cooled and crushed into fine particles. The cement clinker is not necessarily Portland cement clinker.

Cement: A hardened mixture of water and a powdery substance consisting of calcined lime and clay clinker without aggregates. The cement is not necessarily Portland cement.

Cementitious: See Cement.

Char: Fish (of the genus *Salvelinus*) that are distinguished from trout and salmon by the absence of teeth in the roof of the mouth, presence of light colored spots on a dark background, absence of spots on the dorsal fin, small scales, and differences in the structure of their skeleton.

Clean Water Act: A federal act passed in 1972 that contains provisions to restore and maintain the quality of the nation's waters. Section 303(d) of the Clean Water Act establishes the TMDL program.

Concrete: The combination of dry cement, natural aggregates (e.g., sand, gravel), and water. It generally consists of between 7-15% hardened, hydrated cement, 14-21% water, 60-75% aggregates (coarse and/or fine), and up to 8% air by absolute volume. The cement is not necessarily Portland cement.

Concrete Recycling: The processing (including, but not limited to, crushing, fracturing, sorting, storing, stockpiling, grading, and washing) of hardened structural concrete to produce a reusable concrete product.

Conductivity: A measure of a materials ability to conduct an electrical current. In water, conductivity is related to the concentration and charge of dissolved ions.

Cured concrete: See Concrete and Curing time.

Curing time: The amount of time the cementitious material is allowed to harden. Concrete typically reaches full strength following 28 days after placement (curing time).

Dissolved oxygen (DO): A measure of the amount of oxygen dissolved in water.

Effective Concentration (EC_{50}): See half maximal effective concentration.

Effluent: An outflowing of water from a man-made structure. For example, the treated outflow from a wastewater treatment plant.

Eluate: The extraction solution resulting from a laboratory leaching test of a solid. It is the result of the solid-liquid partitioning between the solid material and the aqueous phase.

Eluent: Used to describe the extraction solution prior to a laboratory leaching test of a solid.

Exopolysaccharides: Primarily composed of polysaccharides and proteins and are macromolecules secreted by microorganisms.

Exposure time: The amount of time the laboratory eluate/field leachate is exposed to the material to be leached.

Facility (or facilities): Refers to an "actively permitted facilities with concrete recycling identified as a Sand and Gravel General Permit activity," located in Washington State.

Fractured concrete: See Concrete.

General permit: The term "general permit" in this text refers to the *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* 2021.

Half maximal effective concentration (EC_{50}): An empirical/statistical estimate of the concentration of an antagonist (i.e. toxicant, field leachate, laboratory eluate) to induce a particular response in half (or 50%) of the test population under a particular set of conditions.

Hardened concrete: See Concrete.

Hydrologic connectivity: The hydrogeological connection between surface water bodies and groundwater bodies. The hydrologic connection between surface water bodies and groundwater may be seasonal or depend on other environmental factors.

Hyporheic: The area beneath and adjacent to a stream where surface water and groundwater intermix.

Leachant: Used to describe the extraction solution to a leaching test of a solid material. The latter half of this report uses this term to refer to the extraction solution prior to a field leaching test of a solid material.

Leachate: A solution obtained by leaching a solid, it is the result of the solid-liquid partitioning between the solid material and the aqueous phase. The latter half of this report uses this term to refer to the leaching results obtained or measured in the field.

Liquid-to-solid ratio: The ratio of the volume of water (in units of volume L^3) to the mass of cement. This ratio is commonly varied in laboratory leaching methods. In the field, the ratio represents the amount of water that has come into contact with the material (assuming the mass of the material is known).

National Pollutant Discharge Elimination System (NPDES): National program developed under the Clean Water Act for issuing, modifying, revoking and reissuing, terminating, monitoring, and enforcing permits, and imposing and enforcing pretreatment requirements. The NPDES program regulates discharges from wastewater treatment plants, large factories, and other facilities that use, process, and discharge water back into lakes, streams, rivers, bays, and oceans.

Nonpoint source: Generally, any unconfined and diffuse source of contamination. Pollution that enters any waters of the state from any dispersed land-based or water-based activity, including but not limited to atmospheric deposition, surface-water runoff from agricultural lands, urban areas, or forest lands, subsurface or underground sources, or discharges not otherwise regulated under the NPDES program. Legally, any source of water pollution that does not meet the legal definition of “point source” in section 502(14) of the Clean Water Act.

Oligomers: A polymer that consists of a “few” repeating chemical units.

Oxyanion: An element bonded to one or more oxygen atoms with a net negative charge (e.g., chromate [CrO_4^{2-}], sulfate [SO_4^{2-}], and carbonate ions [CO_3^{2-}, HCO_3^-]).

Parameter: Water quality constituent being measured (analyte). A physical, chemical, or biological property whose values determine environmental characteristics or behavior.

pH: A measure of the acidity or alkalinity of water. A low pH value (0 to less than 7) indicates that an acidic condition is present, while a high pH (greater than 7 to 14) indicates a basic or alkaline condition. A pH of 7 is considered neutral. Since the pH scale is logarithmic, a water sample with a pH of 8 is ten times more basic than one with a pH of 7.

Point source: Sources of pollution that discharge at a specific location from pipes, outfalls, and conveyance channels to a surface water. Examples of point source discharges include municipal wastewater treatment plants, municipal stormwater systems, industrial waste treatment facilities, and construction sites where more than 5 acres of land have been cleared.

Pollution: Contamination or other alteration of the physical, chemical, or biological properties of any waters of the state. This includes change in temperature, taste, color, turbidity, or odor of the waters. It also includes discharge of any liquid, gaseous, solid, radioactive, or other substance into any waters of the state. This definition assumes that these changes will, or are likely to, create a nuisance or render such waters harmful, detrimental, or injurious to (1) public health, safety, or welfare; (2) domestic, commercial, industrial, agricultural, recreational, or other legitimate beneficial uses; or (3) livestock, wild animals, birds, fish, or other aquatic life.

Polychlorinated biphenyls (PCB): A group of 209 man-made compounds that generally occur as complex mixtures. PCBs are very persistent, lasting for decades in the environment. Like other persistent, bioaccumulative, and toxic chemicals, PCBs move easily between air, water, and land.

Polycyclic aromatic hydrocarbons (PAH): A group of more than 100 different chemicals and generally occur as complex mixtures. They can come from natural sources or from human activity.

Polymers: A substance that is made of a chain of chemical units, typically in repeating subunits. Admixtures are commonly polymers.

Portland lime (cement): A generic term for cement used in virtually all concrete and owes its origin to Joseph Aspdin.

Recycled concrete aggregate (RCA): Defined by United States Department of Transportation Federal Highway Administration as a granular old portland cement concrete that has been removed from service and purposefully crushed to produce concrete aggregates for re-use.

Salmonid: Fish that belong to the family *Salmonidae*. Species of salmon, trout, or char.

Sand and Gravel General Permit: The term "Sand and Gravel General Permit" in this text refers to the *Sand and Gravel General Permit - A National Pollutant Discharge Elimination System (NPDES) and State Waste Discharge General Permit* 2021.

Stormwater: The portion of precipitation that does not naturally percolate into the ground or evaporate but instead runs off roads, pavement, and roofs during rainfall or snowmelt. Stormwater can also come from hard or saturated grass surfaces such as lawns, pastures, playfields, and from gravel roads and parking lots.

Superplasticizers: See Admixture.

Supplementary cementitious material: These materials are used in conjunction with cement in concrete to reduce the amount of portland cement needed.

Supplementary materials: These are supplementary cementitious materials and concrete admixtures.

Surface waters of the state: Lakes, rivers, ponds, streams, inland waters, salt waters, wetlands, underground waters, and all other surface waters and watercourses within the jurisdiction of Washington State.

Total Dissolved Solids (TDS): Those solids that are capable of passing through a glass fiber filter (1.0 – 1.5 um) and dried to a constant weight at 180 degrees centigrade.

Total Suspended Solids (TSS): The particulate material in a liquid sample that does not pass through a glass fiber filter. Large quantities of TSS discharged to a receiving water may have a toxic effect on aquatic life or result in solids accumulation.

Turbidity: A measure of the clarity of water as expressed by nephelometric turbidity units (NTU) and measured with a calibrated turbidimeter.

Waters of the State: Those waters as defined as "waters of the United States" in 40 CFR Subpart 122.2 within the geographic boundaries of Washington State and *"*waters of the state*"* as defined in Chapter 90.48 RCW. This includes groundwater*,* lakes, rivers, ponds, streams, wetlands, inland waters, salt waters and all other surface waters and watercourses within the jurisdiction of the State of Washington.

Water-to-cementitious material ratio (*w/cm*): The mass of water used to hydrate a specified mass of cementitious material.

Water-to-cement ratio (*w/c*): The recommended water-to-cement (*w/c*) ratio needed to completely hydrate a cement is 0.42. The *w/c* ratio indicates the mean distance between the cement particles in a cement-paste before it begins to harden.

Acronyms and Abbreviations

BMP	Best management practice
CalCIMA	California Construction & Industrial Materials Association
CCN	Cement Chemist Notation
EC	Electrical conductivity
Ecology	Washington State Department of Ecology
EPA	U.S. Environmental Protection Agency
GIS	Geographic Information System
L/S	Liquid-to-solid
LCA	Life cycle assessment
NPDES	National Pollutant Discharge Elimination System (see glossary)
PAH	Polycyclic aromatic hydrocarbons
PARIS	Permitting and Reporting Information System
PCB	Polychlorinated biphenyls
POC	Pollutants of concern
QPL	Qualified Products List
RAC	Recycled aggregate concrete
RCA	Recycled concrete aggregate
RCW	Revised Code of Washington
S.U.	Standard units
State	Washington State
SWPPP	Stormwater pollution protection plan
TDS	Total dissolved solids
TSS	Total suspended solids
USGS	U.S. Geological Survey
w/c	Water-to-cement
w/cm	Water-to-cementitious material
WAC	Washington Administrative Code
WET	Whole effluent toxicity

Units of Measurement

°C	degrees centigrade
ft	feet
g	gram, a unit of mass
km	kilometer, a unit of length equal to 1,000 meters
L/s	liters per second (0.03531 cubic foot per second)
m	meter
mg	milligram
mg/d	milligrams per day
mg/kg	milligrams per kilogram (parts per million)
mg/L	milligrams per liter (parts per million)
mg/L/hr	milligrams per liter per hour
Mg/m^3	megagrams per cubic meter
mL	milliliters

mm	millimeters
mmol	millimole or one-thousandth of a mole
MPa	megapascal
mole	an International System of Units (IS) unit of matter
ng/g	nanograms per gram (parts per billion)
ng/kg	nanograms per kilogram (parts per trillion)
ng/L	nanograms per liter (parts per trillion)
NTU	nephelometric turbidity units
S.U.	standard units
ug/g	micrograms per gram (parts per million)
ug/kg	micrograms per kilogram (parts per billion)
ug/L	micrograms per liter (parts per billion)
umhos/cm	micromhos per centimeter
uS/cm	microsiemens per centimeter, a unit of conductivity

Appendices

These appendices are available only on the internet, linked to this report at: https://apps.ecology.wa.gov/publications/SummaryPages/2203003.html.

Appendix A: Sand and Gravel General Permit - A *National Pollutant Discharge Elimination System* (NPDES) and State Waste Discharge General Permit 2021

Appendix B: Select Abstracts of Leaching Literature Used Throughout the *Recycled Concrete Aggregate Leaching: A Literature Review*