



# Heavy metal removal capacity of individual components of permeable reactive concrete



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## ARTICLE INFO

### Article history:

Received 20 September 2016

Accepted 9 December 2016

Available online 10 December 2016

### Keywords:

Permeable reactive concrete

Permeable reactive barrier

Remediation

Groundwater

Freundlich isotherm

Heavy metal

## ABSTRACT

Permeable reactive barriers (PRBs) are a well-known technique for groundwater remediation using industrialized reactive media such as zero-valent iron and activated carbon. Permeable reactive concrete (PRC) is an alternative reactive medium composed of relatively inexpensive materials such as cement and aggregate. A variety of multimodal, simultaneous processes drive remediation of metals from contaminated groundwater within PRC systems due to the complex heterogeneous matrix formed during cement hydration. This research investigated the influence coarse aggregate, portland cement, fly ash, and various combinations had on the removal of lead, cadmium, and zinc in solution. Adsorption, adsorption, precipitation, co-precipitation, and internal diffusion of the metals are common mechanisms of removal in the hydrated cement matrix and independent of the aggregate. Local aggregates can be used as the permeable structure also possessing high metal removal capabilities, however calcareous sources of aggregate are preferred due to improved removal with low leachability. Individual adsorption isotherms were linear or curvilinear up, indicating a preferred removal process. For PRC samples, metal saturation was not reached over the range of concentrations tested. Results were then used to compare removal against activated carbon and aggregate-based PRBs by estimating material costs for the remediation of an example heavy metal contaminated Superfund site located in the Midwestern United States, Joplin, Missouri.

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## 1. Introduction

The immediate risks of metal-laden waters to the general public were recently shown in the release of mining residuals and heavy metal contaminants near the Animas River, USA and Rio Doce, Brazil (Egler et al., 2014; Gobla et al., 2015; Mariuzzo and Barata, 2016). The severe impact to human life and the high cost of environmental cleanup for related damages has been largely at the expense of the companies and agencies that manage the sites, while the burden of enduring the acute and long-term effects of contamination is placed on the public. Advances in treatment technologies are of high interest especially ones minimizing environmental risks, while remaining cost effective for remediating a majority of the RCRA 8 metals. Cadmium, lead, and zinc were chosen as representative metal contaminants due to their mobility and toxicity especially present in local, Midwestern, groundwater (Carroll et al., 1998; Environmental Protection Agency, 1990). Several methods are available for the remediation of groundwater, however the EPA-preferred method of treatment is with permeable reactive barriers (PRBs), as these typically provide the best cost to benefit ratio (Powell et al., 1998). PRBs are passive systems simply constructed

by cutting a vertical trench downgradient of the contaminant plume, filling the trench with baffles, and installing a reactive medium to intercept and treat the groundwater (Bartzas and Komnitsas, 2010; Collins et al., 2010; Morar et al., 2012; Mulligan et al., 2001; Paul et al., 2003; Wilkin et al., 2008). Typical reactive media components include permanganate, persulfate, activated carbon, or zero-valent iron, all of which remove metals through adsorption or oxidation (ITRC, 2011). Each material is generally toxin specific and costly to produce or regenerate (Bartzas and Komnitsas, 2010; Watts and Teel, 2006). Permeable concrete as a reactive medium, called permeable reactive concrete (PRC), is a potential alternative with benefits to both cost and simplicity of design as compared to traditional PRBs (Knox et al., 2012). Contributions to removal of heavy metals by aggregate or some cement components are known; however, the combined or bulk removal, related to the individual components, has not been previously studied.

Permeable concrete, also termed pervious concrete, Portland Cement Pervious Concrete (PCPC), no fines concrete, or enhanced porosity concrete, is a best management practice used for stormwater reduction and water quality improvement (American Concrete Institute Committee 522, 2010; Kevern, 2015; Kevern et al., 2008; Sañudo-Fontaneda et al., 2014). Permeable concrete has been shown to remove some metals (copper and zinc), nutrients (nitrogen and phosphorus), organics, and suspended solids from surface waters primarily through physical filtration and adsorption, although stormwater

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volume reduction has been the primary focus of application (Haselbach et al., 2014, 2006; Luck and Workman, 2007; Newman et al., 2002). Permeable concrete typically contains single-sized coarse aggregate bound by sufficient cement paste to coat each aggregate particle and provide load transfer between the cement-coated pieces. Permeable concrete hardens into a highly porous material distinguished from conventional concrete by a high degree of interconnected and tortuous voids (>10% voids by volume) (American Concrete Institute Committee 522, 2010). The two most fundamental components of permeable concrete are cement paste and aggregate, and both have the individual capacity to remove contaminants from solution. Portland cement is well-known as a soil stabilizer or solidifier for use in sequestering heavy metals in soil mixing (Mulligan et al., 2001). Portland cement hydration produces highly alkaline conditions >12.0 S/U and causes metals in solution to precipitate or sorb with the cement hydration products such as calcium-silica-hydrate (CSH) gel (Aziz et al., 2008; Chen et al., 2007). Supplementary cementitious materials (SCMs) such as class C fly ash and natural zeolites have been used in PRBs or water treatment facilities to remove heavy metals through precipitation and adsorption. Removal by fly ash is attributed to the high specific surface area, formation of additional CSH gel, and increased zeta potential as alkalinity increases to improve metal sorption (Bayat, 2002; Czurda and Haus, 2002; Petersen et al., 1996; Purnomo et al., 2008; Wang et al., 2004; Weng and Huang, 1994, 2004).

A wide variety of coarse aggregate materials are used to produce pervious concrete such as crushed limestone, mixed river gravel, high pressure-nonfoliated metamorphics, and cemented sedimentary, well indurated, materials (Kevern et al., 2008, 2010). In general, local-sourced aggregates are used unless chemical, physical, or mechanical properties do not meet prevailing specifications or ASTM C33 (ASTM, 2003). Adsorption by aggregate or soils is also a well-known process generally applied to evaluate the environmental impact risk derived by the natural attenuation of native soils to remove contaminants. Limestone, granite, and silica aggregates have been shown to remove heavy metals from solution in both surface and groundwater applications (Bradl, 2004; Demirkan, 2008; Elliott et al., 1986; Ernst et al., 2016; Lu and Xu, 2009). Calcite contained in powdered limestone, waste sludge, and synthetic sources can adsorb individual metals such as lead, zinc, or cadmium (Ghazy and Ragab, 2007; Merrikhpour and Jalali, 2012; Sibrell et al., 2007). Reaction kinetics and removal rates for calcite with lead or zinc are reported in the literature; however, a combined removal for lead, zinc, and cadmium is not well known (Papadopoulos and Rowell, 1988). This research investigates the removal of lead, zinc, and cadmium from an aqueous solution by three different aggregates (limestone, Kansas River-sourced pea gravel, and inert soda-lime distillation glass beads) used with two cementitious mixtures (100% Portland cement and Portland cement with 25% replaced by Class C fly ash). The results expand the current knowledgebase for PRC, providing the background for design to move the technology into mainstream applications.

## 2. Methods

### 2.1. Materials

Three coarse aggregates were tested (Gravel-G, Limestone-L, Soda-lime glass beads-GB) with two cementitious mixtures: 100% Type I/II ordinary Portland cement (labeled PC) conforming to ASTM C150/C150M-16e1 (2016) or with 25% replacement of Type I/II cement with Class C fly ash (labeled CA) conforming to ASTM C618-15 (2015) with chemical properties measured according to ASTM C311/311M-13 (2013) (ASTM, 2016; ASTM, 2010, 2005). Chemical and physical properties of the materials are summarized in Table 1. Aggregates had a uniform gradation with a maximum nominal size for the pea gravel and limestone of 9.5 mm (3/8 in.) and soda glass beads of 6.0 mm (1/

**Table 1**  
Composition of components.

Chemical	Portland cement (%)	Class C fly ash (%)	Limestone (%)	Gravel (%)	Glass beads (%)
SiO <sub>2</sub>	20.49	40.71	3.75	34.90	74.00
Al <sub>2</sub> O <sub>3</sub>	4.26	18.99	0.50	3.32	1.30
Fe <sub>2</sub> O <sub>3</sub>	3.14	6.05	0.18	0.79	0.04
CaO	63.48	20.10	51.88	0.75	10.50
MgO	2.11	4.82	1.33	0.70	0.20
SO <sub>3</sub>	2.90	0.83	0.00	0.00	0.20
Na <sub>2</sub> O	0.18	1.46	0.00	1.98	13.00
K <sub>2</sub> O	0.47	0.65	0.00	1.17	0.30
CO <sub>2</sub>	1.48	0.00	0.00	0.00	0.00
CaCO <sub>3</sub>	3.41	0.00	92.60	56.39	0.00
LOI	2.20	0.08	0.00	0.00	0.00

4 in.). Absorption of the gravel and limestone was approximately equal at 1% as determined using ASTM C127-14 (2014) (ASTM, 2014). Pea gravel consisted of subrounded to subangular grains while crushed limestone consisted of angular grains. The specific gravities of Portland cement, class C fly ash, limestone, pea gravel, and glass beads are 3.15, 2.69, 2.65, 2.62, and 2.50 respectively. Soda-lime glass beads were used as an assumed nonreactive scaffold for the cement paste to identify the cement's role in removal. The beads were required to provide a similar macro-pore distribution and paste thickness to adequately compare paste with limestone or pea gravel concrete. Soda-lime glass beads are commonly used in distillation columns and were assumed to be nonreactive.

Limestone aggregate was sourced from the lower ledge of the Callaway-Cooper-Mineola facies of Cedar Valley Limestone. The Callaway-Cooper-Mineola is a Devonian calcitic-magnesia limestone which covers about 58% of Missouri, U.S., with about 30% surficially exposed and 28% below grade. The pea gravel aggregate was sourced from the Kansas River drainage basin and procured from aggregate dredging operations located in Bonner Springs, Kansas. The Kansas River flows about 275 km (170 miles) from the confluence of the Smoky Hill and Republican rivers, at Junction City, eastward to its mouth in Kansas City, Kansas. The drainage basin covers just over 155,000 km<sup>2</sup> (60,000 miles<sup>2</sup>). Sediment within the Kansas River reflects the geologic composition of its drainage basin with approximately 160 geologic bedrock units contributing to the sediment of the Kansas River. Most of the coarse materials obtained are of various limestones, dolomites, chert, sand or mudstone, or crystalline calcite (Scott and Hambleton, 1965). A potassium feldspar bearing metamorphic rock also appears in sufficient mass within the sample used for this study. Some gravel contains both potassium feldspar and dolomite, effervescing very weakly in dilute hydrochloric acid. Metamorphic rocks exposed at the surface in Kansas are associated with the Riley and Woodson county igneous intrusives. Granite or granitoid igneous rocks are found in Woodson County and are medium to coarse-grained with intrusions into Pennsylvanian shales (Franks, 1965, 1966). Lower Permian limestones and shales have been slightly altered by contact metamorphism in Riley County which explains the dolomite encased potassium feldspar. Quartzite is apparent within lesser percentages, is more than likely "Silver City" in origin, and derived from Woodson and Wilson counties. Limestone and dolomite fragments are most likely Late Pennsylvanian or Early Permian (270–300 million years ago) in origin but not distinguished from each other as they tend to react similarly in the presence of dilute hydrochloric acid which indicates similar calcium response and therefore a similar effective reactivity with metals. Chert or flint, as known regionally, is a hydrate amorphous silica that occurs along east facing cuestas that form the dominate physical landform of the region, locally termed "The Flint Hills" (Fowler et al., 1935). The Kansas River cuts through the cuestas to transport the chert, sandstone, and

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