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Review

Review of the impact of stormwater and leaching from pavements on the environment

A.J. Kriech, L.V. Osborn *

Heritage Research Group, 6320 Intech Way, Indianapolis, IN 46237, USA

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ABSTRACT

The intensive growth of roadway infrastructure worldwide leads to growing concerns over the health impacts of stormwater runoff and leachate from roadway materials. This comprehensive review combines various sources of information from the last 30 years of research on the impact of pavement stormwater runoff and leaching on the environment. Of the 95 papers found in library searches, 42 papers add significantly to the body of literature around this subject after review of content and quality.

Normally constructed asphalt and concrete pavements were found to release low levels of contaminants during their life. However, deposition from atmospheric pollutants and materials dispersed by vehicles on pavements do have a measurable impact on the quality of stormwater runoff. These tend to be expressed in initial flush from stormwater events. Reuse of old pavements at end of life tend to have little environmental impact when recycled. However, because of deposition of pollutants over their life these materials can have an impact when used in unbound layers of the pavement or in storage before reuse. Water quality can be improved by porous pavements, which allow infiltration of water and drainage to lower layers, thereby filtering many pollutants in stormwater runoff. The challenge is preventing the high initial pavement porosity from plugging over time. Pavement sealers containing coal tar pitch have high levels of polycyclic aromatic compounds and have been shown to impact aquatic life negatively and produce sediment buildup in ponds and streams. Recent studies have investigated photooxidation of pavements and its influence on leaching, but these remain as laboratory-scale studies. Tables outline materials tested, analytical parameters measured, and methodologies to allow readers to easily identify studies most relevant to their focus on impact of stormwater and leaching from pavements on the environment.

1. Introduction

Society's extensive infrastructure developments have significantly impacted natural drainage and water quality, introducing more pollutants into waterways and groundwater during flooding. Structural and nonstructural approaches have been put into place to mitigate flooding from stormwater including the creation of stormwater runoff ponds, separation of sewage from stormwater collection, and underground storage tunnels which relieve the loading on wastewater treatment facilities during heavy rainfall events. Pavement roadways, runways, and parking areas are significant sources of rapid release stormwater events in urban areas because most are relatively impervious to water. Here, we review the last 30 years of research to understand how various pavement materials impact water quantity and quality entering our waterways and groundwater.

Since the 1970's, with the growing recognition of the environmental movement in the United States, the paving industry looked for ways to reduce, reuse, and recycle materials at the end of their life cycle. Today, asphalt pavements are the largest recycled product in North America by source quantity. These end-of-life materials are both recycled and reused in various ways within the pavement structure. Evaluating the environmental impact of pavements at different stages of their life cycle is a focus of this review.

With a strong impetus to develop a circular economy, many industries outside the usual material suppliers such as construction and demolition (C&D) waste, coal fly ash, plastic, and tires see their byproducts and end-of-life wastes as potential viable alternative components for pavements. We discuss the environmental impacts of these byproducts and waste materials included in pavements at the time of construction and when the pavements containing these materials are

* Corresponding author.
E-mail address: linda.osborn@hrglab.com (L.V. Osborn).

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recycled or reused at the end of their life.

Finally, we examine the source of pollutants; those that come from the pavement systems themselves and contaminants that fall and collect on the pavements over time, which are washed away during rainfall.

2. Methods

Peer-reviewed and widely referenced articles within the literature between 1990 and 2021 were assessed for relevance and quality on the topic of *environmental impact of stormwater runoff and leaching from pavements*. Studies were organized into two categories: pavement stormwater runoff, and pavement leachate. These studies were evaluated separately because they are measured under different conditions and give insight into different endpoints of environmental impact. Other reviews have synthesized historical water quality trends related to highways (Kayhanian et al., 2012) and RAP (Spreadbury et al., 2021). Here we itemize a vast array of materials, analytical parameters, methods used and comments such that users can select those publications that best relate to their specific circumstances related to environmental impacts.

2.1. Pavement stormwater runoff studies

Because of water scarcity in many parts of the world, stormwater is often collected and can be used for potable water when properly purified through desalination (Panagopoulos, 2021a, 2021b) and other techniques. It can also be used in industrial applications. Studies that evaluate stormwater runoff events on roads focus on dissolution of adsorbed compounds off the pavement surface in the event of rainfall. Contaminants that build up on pavement surfaces during dry periods are mobilized and carried away with water runoff. Examples include dust and airborne pollutants, materials dropped from vehicles such as lubricants, tire abrasion or spills from trucks carrying chemicals, sealers, and road debris. These studies are important because stormwater events can lead to flooding and can overwhelm the collection and treatment of pollutants before they enter ditches, sewers, streams, lakes, rivers, oceans, and subsurface groundwater.

Porous pavements (sometimes called permeable pavements) are a relatively recent alternative for stormwater management; these studies are included in this review to glean additional information on the benefits and challenges of these systems (Kayhanian et al., 2019). In 2019, Congress enacted the “Water Infrastructure Improvement Act” encouraging practices that catch water where it falls. Porous pavement is a good example of this, allowing water to quickly pass through and get stored below where it is filtered through soil and slowly released to the environment. The rapid drainage of water through porous pavement also enhances contact between the tire and the pavement during rainfall events, which prevents hydroplaning and improves wet friction stopping distance. As the tire rolls over these wet pavements, a suction is created on the back side of the tire that helps to clean particles out of the pores and maintain porosity.

Stormwater runoff studies generally involve runoff simulations from the field, or laboratory, by running water over a material. These studies investigate desorption of materials off pavement surfaces rather than leaching through dissolution mechanisms. Laboratory studies consist of elution of water through a column loosely packed with a sample, then collection and analysis of the eluate, including suspended particulate. Alternatively, field studies involve collection of water runoff from a pavement, or stockpiles of old recovered pavement materials, including construction debris.

2.2. Pavement leachate studies

Leachate studies focus on extractable compounds into water as it passes through materials (e.g., pavement) under various conditions. Landfills were an early area of concern to the Environmental Protection Agency (EPA) because they released significant pollutants back into the environment over time. As a result, the EPA developed tests that simulate and assess the release of compounds through leaching into the environment, one of which is the toxic characteristic leachability procedure (TCLP). These evaluation concepts came from the Resource Conservation Recovery Act (RCRA) of 1976.

Leachate batch studies discussed herein utilized the standardized TCLP EPA method (instrument shown in Fig. 1) or slight variations of this test. Additionally, several studies compared monolithic leachate to standard TCLP leachate. In a standard EPA TCLP, the sample is crushed to less than 9.5 mm to increase the surface area contact, then tumbled in a 20:1 water-to-sample ratio for 18 h. The water is buffered either slightly acidic, neutral, or basic to assess the impact of pH on the leachability of compounds into the water. Conversely, monolithic leachate testing does not involve crushing the sample, therefore only the sample’s outer surface is exposed to water. In this case, if the material tested is impermeable to water, there is a reduced opportunity to extract and solubilize compounds into the water. One important aspect of the TCLP EPA method leachate testing is that the water is filtered to remove particulate, and only compounds that are solubilized into the water are measured.

Lastly, column leachate testing, often measured in bed volumes of water passed through the sample, describes a third type of leachate study. Column leachate testing is employed to analyze water soluble materials within the material, rather than simply removing surface contaminants. Like TCLP, this method also involves water filtration such that only water-soluble compounds are evaluated. Together, these procedures consider dissolution and diffusion processes that contribute to pollution. All three methods of leaching, and the distinctions they raise, are important in understanding potential sources of pollution and mechanisms for entering the water.

2.3. Challenges associated with stormwater runoff and leachate studies

Challenges related to stormwater runoff and leachate studies include *what* to measure and *how* to measure it. Generally, regulatory agencies focus on harmful compounds that may be released into the environment. Quantitative analysis of hazardous substances, such as heavy metals and known toxic compounds, is conducted to determine their concentrations, if present. Comparison to regulatory limits for drinking water, or typical background levels found in residential or industrial settings, determine compliance status. While other materials may be released, only those which are regulated are analyzed. Nonetheless, some studies discussed herein consider non-regulated compounds, and raise questions about their environmental impact. Often, these are emerging pollutants that are found at low levels in the environment, that do not have regulatory limits currently, or known health effects. Examples of such emerging pollutants include antioxidants, such as butylated hydroxytoluene (BHT), and endocrine disruptors (e.g., phthalates). While

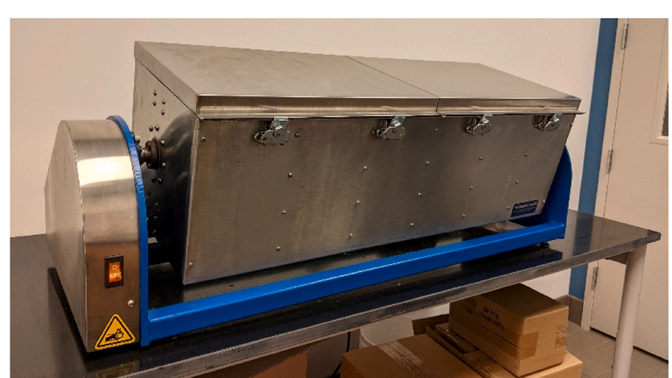

Fig. 1. TCLP instrument used for EPA method.

interpretation of health effects of these non-regulated compounds, if present, may yield more questions than answers, these studies help to direct future research and potential regulations.

Decisions to conduct laboratory versus field testing is another challenge facing researchers. In laboratory settings, variables can be tightly controlled to minimize the impact of confounders present in field studies. However, lack of understanding about potential confounders in the field can lead to misassumptions about the source of the pollutants found. Therefore, well-designed, and well-conducted field studies are invaluable to understanding the impact of confounders on water quality. Validation of laboratory versus field data is ultimately an important and necessary component in understanding water quality issues in the environment.

Over the past 30 years, major strides have been made in analytical instrumentation. Innovation has allowed for better selectivity and sensitivity, therefore improving qualitative and quantitative measurements, greatly advancing our understanding of potential pollutants at low levels in water. Moreover, analysis of complex mixtures, such as asphalt pavements, has been facilitated by advancements in chromatographic separation and mass spectrometry. Altogether, this advancement helps ascertain potential health impacts on plants, animals, and humans. With that in mind, the year these studies were conducted, and the tools used were considered in this review. Studies were evaluated from oldest to most recent within the stormwater runoff and leachate categories.

Strengths and weaknesses in experimental design of the 42 selected studies are critical to determine the importance that individual studies should carry in the literature. Herein, we contemplated the following: Did the study consider confounders or were there validation steps conducted during the research? Are the conclusions reached in agreement with previous published work? If the outcomes run counter to the preexisting body of evidence, were there explanations for the discrepancies? Finally, our conclusions are drawn based on the science presented in these studies, and scientific data gaps identified.

3. Review

3.1. Pavement stormwater studies

Water quality runoff was investigated from stockpiles of salvaged concrete and bituminous paving (Sadecki, 1996). Conducted on behalf of the Minnesota Department of Transportation (DOT) by the Office of Environmental Services, the study investigated stormwater runoff from three stockpiles of reclaimed pavement materials. One was a stockpile of crushed concrete pavement between 4.75 and 19.0 mm. The second contained crushed concrete less than 4.75 mm in size and the third pile was salvaged reclaimed asphalt pavement (RAP). All were placed on a pad with a lined trench collection system for runoff. Metals, total suspended solids (TSS), pH, alkalinity, and chlorides were tested as well as polycyclic aromatic hydrocarbons (PAHs) from the RAP stockpile. The authors conclude that, although there are sediments and leachates emanating from the three stockpiles, the primary concerns were related to TSS, pH, and possible chromium in the concrete runoff. In the RAP stockpiles, no PAHs were detected, alkalinity was neutral, and TSS and metals were lower. The authors recommend that those stockpiles be kept away from environmentally sensitive areas.

Permeable pavements were studied after six years of use (Brattebo and Booth, 2003). Water collected under the permeable pavements showed appreciably improved quality compared to runoff water collected from non-permeable pavements. Concentration of metals, such as copper and zinc, in water were much lower in the filtered underdrain pavements than the nonpermeable pavements. Motor oil was also 89% lower in the permeable sections compared to the impermeable sections in stormwater runoff samples. These results were supported by later studies, which showed the ability of underlying soils to remove pollutants (in porous pavements), through filtration processes or by absorption. These results also suggest that metals released in the stormwater runoff were either in the form of particulate or absorbed onto particulate matter and not water soluble.

Release of organic contaminants from storage of stockpiled RAP in Sweden, were explored both in the field and in the laboratory (Norin and Strömvall, 2004). Drains under the RAP piles allowed water collection at four locations, in concentric rings radiating from the center. Collection closest to the center of the pile had the highest amount of RAP sitting above it, with the lowest liquid-to-solids ratio. Tests were conducted immediately after making the pile, then a year later after collecting water over time. Laboratory studies involved column leaching tests with these same RAP materials. Water was tested for semi-volatile organics, PAHs, and other compounds of interest. Results showed that naphthalene, BHT, and dibutyl phthalate (DBP) an environmentally persistent chemical (Jobling et al., 1995) were the dominant compounds released from the RAP. BHT is a synthetic antioxidant and DBP is a very water-soluble plasticizer; both ubiquitous in the environment, but neither are added to asphalt. Other compounds were detected but could not be identified. Samples taken over time showed larger molecular weight PAHs collected in the water. Laboratory column studies showed lower concentrations of pollutants in the recovered water. The authors conclude that the lower liquid-to-solids ratio for stockpiles collection leads to concentrating the pollutants in the water. Additionally, column studies revealed that the concentration of pollutants decreased significantly after the initial flush, suggesting dissolution rather than diffusion mechanism. The authors conclude that compounds on the surface of the asphalt, including traffic pollution (car and diesel exhaust particulate), crankcase drippings, grease, and spills are the most probable sources of pollutants. Overall, this study emphasizes the importance of conducting more than just laboratory studies and the need for validation with field studies. The best management practices in use today involve constructing RAP piles on impervious paved surfaces with a slope, so that water can be collected and filtered through sand and soil before discharge.

Parking lot sealcoats were studied, and researchers found that the use of coal tar sealants resulted in significant PAH contamination (Mahler et al., 2005). PAH levels in the sediment from stormwater runoff, into ditches and streams, were 65 times higher than unsealed asphalt parking lots. This early study was supported by subsequent studies and has led the ban of coal tar sealants by many municipalities. Coal tar sealers were used widely in military air bases where planes are fueled, primarily to protect asphalt pavements from crankcase drippings and fuel spills. Because coal tar retains its dark color over time, it also was used in parking lots and driveways for aesthetic purposes. Most roads are not sealed with coal tar, so it was limited to special uses and not on streets or highways. One important point is that most of the PAHs are in the form of particulate flaking and have limited solubility in water. However, the particulate PAHs can build up over time in the sediment. Ingestion of these materials cause genotoxic effects to bottom feeding fish and other species, harming them and individuals eating fish taken from these waters.

Stormwater runoff from asphalt, concrete paving bricks (pavers), and crushed stone driveways were studied and evaluated for the amount and quality of runoff through measurement of total particulate, nitrates, and heavy metals (Gilbert and Clausen, 2006). Impermeable asphalt pavement released water the quickest and the source of most pollutants was attributed to materials that collect on their surface during dry periods. Concrete paver bricks collected more water within the lower layers, allowing water to flow between the bricks into the underlying layers of the pavement, instead of quickly shedding the water and pollutants. This water was effectively filtered and was cleaner (less particulates, nitrates, and metals). This study supported the value of using permeable pavers for driveways to improve runoff water quality.

In New Zealand, studies were conducted to better understand stormwater runoff contamination from chip seal asphalt (bitumen) emulsions (Ball et al., 2008). Chip seals are a common and economical pavement type used on rural and low volume roads throughout the world. It involves spraying the asphalt emulsion on the prepared road (Fig. 2) and then quickly dropping stone chips (Fig. 3) over the asphalt emulsion. The emulsion quickly sets through chemical attraction to the aggregate. Evaporation produces an impervious roadway (Fig. 4) with aggregate chips providing friction for traffic. Cationic emulsifiers used to make these asphalt emulsions were determined to be ecotoxic to fish and biodegrade slowly. The study references other works that suggest that many of these emulsifiers are quickly absorbed by soil and do not migrate far from the roadway. Therefore, these emulsions may be safer for rural use than in cities, where there is little soil along the roadway. The study also showed that cured asphalt (after 20 min) was not generally bioavailable and had low water solubility.

An extensive runoff study on Portland cement concrete (PCC) and asphalt pavements, both in laboratory and field settings, was conducted in California (Kayhanian et al., 2009). Ten laboratory-produced pavement types were evaluated that included a variety of compositions used widely in California roads. Additionally, 30 actual pavement sites, in urban and non-urban areas, were monitored for runoff over a three-year period. Both particulate and soluble compounds found in the pavement runoff were tested for metals. The laboratory pavements were artificially aged to simulate 15–18 years in service. Water was passed over the pavement, or through it, as in the case of permeable pavements, to simulate rainfall typical of California. The runoff was collected over an 8-h period, in stainless steel carboys, and tested via inductively coupled plasma-mass spectrometry (ICP-MS). The pH was adjusted at four different pH levels between 4 and 7. The study was also conducted over three different air temperatures of 4, 20 and 45 °C. In laboratory studies, most of the metals originated from corrosion of the metal trays and not from the pavement samples. Nonetheless, chromium was observed in concrete pavements runoff and was determined to come from the Portland cement used, not the aggregate. The authors conclude that chromium in the concrete leaches early in the pavements life and then remains low. Studies like this one are important because the researchers investigated the causes of the anomalies and determined the sources of the metals.

In New Zealand, concentrations in adjacent stream sediments were studied from coal tar and asphalt binders in street pavements to develop a concentration-weighted mixing model to apportion PAHs (Ahrens and Depree, 2010). Based on chemical differences between asphalt and coal tar, the likely sources of PAHs found in aquatics sediments were determined. The challenge is that both asphalt and coal tar look similar physically (viscous black binder) but are very different chemically. Coal tar is produced from pyrolysis tars generated from coal at high temperatures and has significant PAH concentrations. Asphalt binders are produced from the vacuum distillation of crude petroleum and have trace levels of PAHs. Through quantitative analysis of individual PAHs, Ahrens et al. demonstrated that coal tar is the principal source of aquatic PAH pollutants in New Zealand stormwater runoff from pavements.

Fig. 2. Asphalt emulsion sprayed on the prepared road during chip seal process.

Fig. 3. Aggregate stone chips are spread on the freshly sprayed road.

Fig. 4. Newly finished chip seal impervious roadway.

Utilizing stakeholder input and field assessments to select appropriate sampling locations, a study reporting on the sediment of 15 urban stormwater ponds in Minnesota focused on PAHs (Crane, 2014). A suite of 34 PAHs were tested on each sample using US EPA SW-846 method 3550 for an ultrasonic extraction. Forensic analysis was performed to determine the most likely source of PAHs present in the sediment, based on specific ratios of PAHs detected. The study concluded that most of the PAHs present in ponds (67.1%) were the result of particulate from coal tar sealants, used on driveways and parking lots, flaking into runoff. Another 29.5% were attributed to vehicle related sources, and 3.4% from wood combustion particles. Furthermore, differences based on land use, such as industrial versus residential, could not be determined. PAH levels were high enough in three ponds to present risk to benthic invertebrates. Nine ponds exceeded human health risks-based benchmarks that would prompt more expensive disposal of dredging's. The authors recommended banning coal tar sealants.

Permeable pavements' ability to absorb metals and other pollutants was investigated, both in a laboratory setting as well as field pavement, in Xi'an, China (Jiang et al., 2015). The laboratory apparatus was very similar to a column leach study. A porous asphalt pavement (PAP) was used in the study, with a 20% air void mixture to allow water to flow quickly through the permeable pavement. Natural sand was below the PAP to act as a storage and collection zone for water infiltrating into the pavement. Contaminants that buildup on roadways after a 15-day dry spell were collected from three collection sites during a rainfall event. The three sites were selected based on the surrounding businesses that were located along the roadway. All sites were in urban environments where either heavy traffic existed (100,000 vehicles/day), pet and plant markets were along the roadway, or chop houses existed. The runoff water was collected in dust pans and then all three sites were comingled to form a 60-liter sample. Only large debris were filtered out of the samples. Water samples from each site passed through the laboratory produced PAP, then analyzed before entering and after exiting the underlying sand layer. Testing included pH, turbidity, suspended solids (SS), chemical oxygen demand (COD), biological oxygen demand (BOD), ammonia, total nitrogen (TN), hexavalent chromium (Cr(VI)), chloride, zinc, lead, and cadmium. As expected, results showed significant reduction in turbidity and SS due to filtration through the PAP and sand layer. Copper, zinc, and lead were reduced to below limits of detection. Cadmium was reduced some through the filtration and possibly sorption processes. Predictably, pH, COD, BOD, and chlorides were essentially unchanged; this is because there is no mechanism for reducing these organics, except through absorption in the soil layers, which were not evaluated in the study. It was concluded that PAPs can reduce pollutants through filtration and absorption mechanisms in the pavement structure. Nevertheless, long term performance of PAP roadways, from the standpoint of plugging from debris getting carried onto the roadway, was not evaluated. Finally, the actual capacity to absorb heavy metals, and keep them stabilized long-term, was not determined.

Examination of runoff from coal tar sealants determined that particulate from coal tar in runoff induced genotoxicity in fish liver cells and impaired DNA repair capacity a month after placement of the coal tar sealer (Kienzler et al., 2015). Exposure to PAHs and methylated derivatives, N-heterocycles, and other compounds in combination with UVA exposure caused DNA damage even with a 1:100 dilution of runoff. Similar results were found in a follow up study on acute toxicity from coal tar sealed parking lots (Mahler et al., 2015). Ceriodaphnia dubia (cladocerans) and Pimephales promelas (fathead minnows) were used for toxicological tests. This work found a 100% mortality rate from runoff up to 36 days after placement.

Near Gothenburg, Sweden researcher looked at how PAHs partitioned in stormwater runoff collected during a rainfall event from roadway catch basins from two locations (Nielsen et al., 2015). One was on a heavily trafficked roadway and one near a more residential roadway. Samples with the highest TSS contained the highest concentration of PAHs. The filtrated particles (>0.7 μm) contained the highest molecular weight PAHs. Colloidal fractions retained the most PAHs in the 10-nm fraction (smaller particles). The solubilized fraction contained the lowest molecular weight PAHs as hypothesized. This paper conveys the importance of understanding how different PAHs, which have limited solubility in water, can migrate in water and soil. Low molecular weight PAHs are far more mobile than large molecular weight PAHs, which are bound on particles. This study helps determine the most effective mitigation methods for eliminating PAHs in stormwater runoff.

Conducted on Milwaukee streambeds and parking lots, samples of 30 different streambed sediments and six parking lot dusts were analyzed for PAHs (Baldwin et al., 2017). Most observed PAHs (77%) were attributed to be from coal tar sealants. The study found 78% of the streambed samples would cause adverse effects on benthic organisms.

An extensive laboratory study of permeable pavements, and their role on reducing pollutants in stormwater runoff was conducted in Spain (Hernandez-Crespo et al., 2019). Authors investigated potential clogging of these pavements over time, with dust and dirt building up on pavements, and the resulting impact on permeability and filtration capacity of these pavement systems. Consistent with other studies (Brattebo and Booth, 2003; Jiang et al., 2015), permeable pavements were found reduce the quantity of pollutants and improve the quality of water entering the receiving bodies of water. The study recommends improvement to the underlying layers that store and filter the water before releasing it and stresses the importance of cleaning porous pavements to avoid plugging and to maintain high infiltration (porosity) rates.

In California, asphalt, and concrete permeable pavement systems, as well as underdrain designs, were explored in terms of controlling and maintaining good permeability (Kayhanian et al., 2019). It was found, through study of both actual pavements and laboratory simulations, that there is a great potential for permeable pavements in improving stormwater runoff quality. Maintaining these systems, including clogging prevention, which reduces permeability over time, is the main challenge. Regarding water quality, chromium was the only pollutant observed. Variable levels of chromium were detected in concrete pavements shown to come from the cement itself. The source of other pollutants present in pavement runoffs was determined to be from deposition of materials during use, not from the pavement materials themselves.

A field study was conducted in Madison, Wisconsin, to measure stormwater quality performance on lined permeable pavements and included concrete pavers, pervious concrete, and porous asphalt pavements (Selbig et al., 2019). Water, collected from each pavement type, was analyzed for its quantity and quality. The study found that the infiltrated water contained less particulate but soluble compounds, such as deicing salt, generally moved through these systems quickly. Clogging was a significant problem for all pavements evaluated over the 22-month study.

Within the state of Maryland, seven RAP materials were studies to determine if they could be safely used for highway shoulder edge drop-offs (Mijic et al., 2020). The study looked at constant head and batch leaching for metals. This laboratory study revealed that different metals in the RAP are controlled by different mechanisms within the RAP. Calcium, barium, and magnesium are controlled by carbonate content. Aluminum and iron are controlled by oxide and hydroxides present, as well as the pH of the aggregate. Measurable levels of copper and zinc were released early, after a few bed volumes (first flush) of water, then remained low. Under acidic conditions, arsenic is released in its less toxic, pentavalent form (As(V)). The concentrations of all metals released during the water leach tests were below quality limits, except for copper. An explanation was given for the lack of metal leaching from RAP, however, no account for the sources of copper was presented. Finally, low concentrations of PAHs were detected in stormwater. Although this was a laboratory study exclusively, recommendations were made for additional future studies.

Table 1 summarizes the stormwater runoff studies reviewed.

3.2. Pavement leachate studies

Leachability of a new standard Hot Mix Asphalt (HMA) Surface Mixture, used by the Indiana DOT, was determined in accordance with EPA Method SW846-351 TCLP leaching (Kriech, 1990). Heavy metals, volatiles, semi-volatiles, and PAHs on EPA's list of hazardous compounds were analyzed by a US EPA accredited laboratory. Because the aggregate used was a blast furnace slag byproduct from the iron making process, there were concerns about heavy metal leaching. Low levels of chromium (0.1 mg/L) were observed in the leachate water. No volatile or semi-volatile compounds tested were found above the limit of detection, except for trace levels of naphthalene (est. 0.25 μg/L). At the time, these were below regulatory levels.

In 1991, the Illinois EPA (IEPA) investigated the use of old asphalt pavement materials for use as Clean Fill in highway embankments for the Illinois DOT. The primary goal was to determine the safety of these

Table 1
Summary of storm water runoff studies.

References	Material/Location	Testing	Lab	Field	Batch	Other	Column	Comments	Findings/Conclusions
Sadecki (1996)	Crushed concrete pavement and RAP MN, US	Metals, total suspended solids (TSS), pH, alkalinity, and chlorides, PAHs (RAP)	✓					Extensive data collected and statistical summaries provided. Valence of chromium leaching from crushed concrete would allow better estimate of toxicity	Results lead to best practices for managing recycled pavement stockpiles, which are varied and dynamic
Brattebo and Booth, 2003	Permeable pavements - nine test parking stalls WA, US	Tested surface runoff and subsurface infiltrate for metals and organics, Cu, Zn, Motor oil		✓		✓		PP voids allow stormwater to filter through pavement into soil, which helps reduce contaminants in stormwater	Show favorable value using PP although Zn increased compared to 5-yrs earlier. Limitations: not all climate zones included, and dry conditions not studied
Norin and Strömvall (2004)	RAP from two stockpiles, 4 sections each. Stormwater pond and the groundwater samples Sweden	Sixteen PAH by GC/MS	✓	✓			✓	Naphthalene, butylated hydroxytoluene, and dibutyl phthalate were dominant SVOCs	Results demonstrated the importance in following lab studies with real field measurements. SVOCs emitted likely from traffic, with residues of rubber from tires, unburned fuels, or vehicle exhaust
Mahler et al. (2005)	Sealers; Coal tar-based emulsion, asphalt-based emulsion, unsealed asphalt pavement, and unsealed concrete pavement TX, US	13 PAHs by GC/MS, SVOCs		✓		✓		Best indicator of coal tar is PAH ratios - fluoranthene/pyrene, indeno[1,2,3cd] pyrene/benzo[ghi] perylene, benzo[a] pyrene/benzo[e] pyrene	Evidence provided -parking lot sealcoat could be dominant source of PAHs to watersheds
Gilbert and Clausen (2006)	Asphalt, paver, and crushed stone driveways CT, US	TSS, TKN, nitrate-nitrogen, ammonia–nitrogen, total phosphorus (TP), Zn, Pb, Cu		✓		✓		Runoff from asphalt driveways > paver driveways > crushed stone driveways	Runoff from crushed stone driveways were similar in pollutant concentration to runoff from asphalt driveways. Runoff volume higher in asphalt driveways. Paver driveways = lowest concentrations
Ball et al. (2008)	Cationic asphalt emulsions (4) used for chip sealing New Zealand	Daphnia magna acute studies, Algal growth inhibition, Crustacea. pH and dissolved oxygen of each of the test dilutions	✓		✓			Emulsifier - major contributor to the ecotoxicity. ~ 7-fold variation in ecotoxicity observed in 4 emulsions tested	Most ecotoxic component is the emulsifying agent. Different types have varying degrees of biodegradability. Variation in results suggest that a more complete range of chipseal emulsions needs to be assessed
Kayhanian et al. (2009)	Portland cement and asphalt binder materials - 10 specimens with various mix type and binder materials. 30 field runoff samples California	ICP-MS for chromium	✓	✓		✓		Dissolved Cr in leachate- all pavements; variability of Cr based on time (e.g., first flush); impact of pH on Cr leachability	Based on Cr only, pavement materials are not the source in roadway runoff. Results indicate that sources include road-use, land-use sources, or atmospheric deposition
Ahrens and Depree, 2010	nine residential streets situated within the urbanized catchment New Zealand	28 PAHs by GC/MS		✓		✓		Source identification by ratioing PAH signature source; mixing model used to estimate the fraction of coal tar	PAHs from old coal tar pavements are still ongoing and contribute significantly to aquatic environments. Isomer ratios were used to determine if the source of PAHs was asphalt or coal tar. Parking lot

(*continued on next page*)

Table 1 (*continued*)

References	Material/Location	Testing	Lab	Field	Batch	Other	Column	Comments	Findings/Conclusions
									sealcoats and other products that contain coal tar continue to contribute to waterway PAH contamination
Crane (2014)	Sediment from 15 stormwater ponds in the Twin Cities MN, US	carcinogenic PAHs, TOC, black carbon, sum PAH_{34} by GC/MS, TOC, % Moisture		✓		✓		In MN, a statewide CT-sealant ban was enacted 1/1/2014 due to accumulation of PAHs in many urban stormwater ponds. B[a]P equivalents calculated. CMB modeling performed better than source ratios for source apportionment.	CMB8.2 model was used to determine source based on Σ PAH_{34}. Did not correlate with TOC but log did with black carbon, (sorbs PAHs -making them potentially more bioavailable & toxic). CT-sealants - major source of PAHs (67.1%), followed by vehicle emissions (29.5%) and wood combustion (3.4%). Ban of CT-sealants encouraged based on results of study
Jiang et al. (2015)	Permeable asphalt pavement (PAP), Porous asphalt concrete, Open-graded gravel, natural sand, Geotextiles China	Cu, Zn, Pb, Cd, petroleum pollutants (PP), animal & vegetable oil, BOD, COD, ammonia nitrogen, total phosphorus, chloride, and total nitrogen, pH, SS, Turbidity	✓				✓	Lab simulation materials not fully compacted, but real-world samples with smaller air voids would have better filtration effect Decrease in environmental load based on physisorption & porosity of PAP.	Influent and effluent samples tested. PAP is good at removing Cu, Zn, Pb, and Cd. Not as good on petroleum removal, animal & vegetable oil, BOD, COD and NH_4–N. Marginal reduction on total phosphorus, chloride, and total nitrogen. Increased sampling time
Kienzler et al. (2015)	Coal-tar-based (CTB) sealcoat TX, US/France	ΣPAH16, DNA repair—base excision repair activity		✓		✓		Formamido pyrimidine glycosylase (Fpg)-modified comet assay used to assess genotoxicity, which shows CTB sealcoat to have a greater genotoxic potential when combined with UVA.	Exposure to runoff from CTB-sealed pavement & co-exposure to UVA can damage DNA and impair DNA repair capacity, even 36 days after application. Photo-reactivity of PAHs include oxygenated analogues which likely causes increased genotoxicity
Nielsen et al. (2015)	Multiple stormwater grab sampling at 2 Swedish sedimentation facilities and synthetic colloids and nano-sized particles Sweden	Low, medium, and high-molecular weight PAHs, 14 specific PAHs by GC/MS-SIM, TOC, particle size distribution on the Filtrated and Colloidal fractions		✓		✓		Synthetic suspensions and stormwater samples. Looks at how PAHs partitioned in stormwater runoff Need to complement treatment technologies with techniques that reduce dissolved PAHs.	Stormwater highway runoff contained PAHs that were higher in concentration & MW in Filtrated fraction. TSS was implicated and play an important role in mobility. Risks of secondary pollution from the sedimentation -based systems was highlighted.
Baldwin et al. (2017)	stream bed sediment—composed mostly of silt, but also clay and sand and parking lot dust and particulates WI, US	38 parent PAHs and 25 alkylated PAHs, TOC, toxicity tests	✓			✓		Coal tar–based pavement sealant was the primary source of PAHs. Biological endpoints showed adverse effects Toxicity tests showed that if coal tar-sealants were eliminated, significant improvement of	Ratios of parent and alkylated PAHs were used to differentiate between petrogenic (alkylated & low MW) compounds and pyrogenic (parent & high MW) PAHs. Total PAH_{16} concentrations in streambed sediment

(*continued on next page*)

Table 1 (*continued*)

References	Material/Location	Testing	Lab	Field	Batch	Other	Column	Comments	Findings/Conclusions
								stream health would occur. PAH UV-induced toxicity was also shown in this study	was 55.1 mg/kg (0.6–208 mg/kg).
Hernandez-Crespo et al. (2019)	Permeable Pavements (PP) Spain	Suspended solids, organic matter and nutrients, COD, Total N and Total P, pH, TP, COD, TSS, ammonia, nitrates, nitrite	✓			✓		Adequate cleaning of PP in dry conditions is essential	PP have reduced surface runoff, better infiltration and water quality compared to impermeable pavements. Nitrogen infiltrated the most.
Kayhanian et al. (2019)	PP; Rubberized asphalt concrete (AC) open-graded (OG), rubberized AC gap-graded, OGAC and polymer-modified OGAC, terminal-blend modified binder gap-graded, dense-graded AC, & Portland cement concrete CA, US	pH, conductivity, turbidity, hardness as $CaCO_3$, TSS, TDS, PAHs, O&G, COD, TOC, and toxicity	✓			✓		PP discharged overflow is cleaner than surface runoff generated from impermeable pavement A complete configuration in PP was recommended for this Mediterranean climate to retain larger water volumes, the greater the thickness of the gravel layer, the greater the reduction of effluent volume and the lower the environmental load	Hydrographs of high intensity events (22 mm/10 min) under different pollution build-up levels were determined: 1-month (140 g/m_2) and 6-month (840 g/m_2). Reactive barriers in the gravel layers would improve the quality of infiltrated water even more. Nitrates could be reduced by regulating outflows.
Selbig et al. (2019)	3 permeable pavements: permeable interlocking concrete pavers, pervious concrete, and porous asphalt WI, US	TSS, Total & dissolved Phosphorus, *E. coli*, chloride, and metals		✓		✓		Parking lot runoff from the underdrain toward the monitoring chamber Their configuration allowed direct measurement of the quantity and quality of runoff sent from the parking lot to each permeable pavement.	Part III of a 3-phased 22-month study. 84 of 95 runoff events were sampled for water-quality. Seasonal influx of sediment & high chloride from de-icing agents observed. Elevated levels of pH in PC could have fostered metal precipitation. A PP system with impermeable liner successfully removed sediment (~60% efficiency for TSS) and sediment-bound pollutants from runoff from an asphalt parking lot
Mijic et al. (2020)	RAP from 7 roadways MD, US	pH, EC, hydraulic conductivity, Br tracer tests, Al, As, B, Ba, Co, Cr, Cu, Fe, Mn, Na, Ni, Pb, V, Mg, and Zn	✓		✓		✓	Data modeled to simulate effect of natural formation & distance on trace metal conc. within surface waters Variables included fines content, sand-to-gravel ratio, and coefficient of uniformity. CaO and higher asphalt content showed lower dry unit wt and higher HC. Chapuis equation was better predictor of HC values. pH of the leachates varied between 6.69 and 8.50.	RAP & natural aggregates from shoulders showed similar hydraulic conductivity (HC) (varied between 6.9 × 10-3 - 1.1 × 10-3 cm/s). Cu and Zn in 2 RAPs were above EPA WQLs in first flush but quickly fell below. Transport model showed decreased metal concentrations for all RAP samples (<WQLs) after passing through natural formation and decrease more with horizontal distance

TKN = Total Kjeldahl nitrogen, TSS = Total Suspended Solids, TDS = Total Dissolved Solids, PAHs = Polycyclic Aromatic Hydrocarbons, O&G = Oil and Grease, BOD=Biological Oxygen Demand, COD=Chemical Oxygen Demand, TOC = Total Organic Carbon, ICP-MS=Inductively Coupled Plasma-Mass Spectrometry, GC/MS = Gas Chromatography/MS, RAP = Recycled Asphalt Pavement, SVOCs=Semivolatile Organic Compounds, PAP= Permeable Asphalt Pavements PP= Permeable Pavements, CTB=Coal-tar-based sealcoat. WQL = Water Quality Limit.

materials and their potential impact on groundwater. Collaborative efforts between the Illinois Asphalt Pavement Association (IAPA), the Illinois DOT, and HRG allowed collection and testing of six old pavement millings from around the State of Illinois. Leachability was conducted in accordance with EPA TCLP methods. Kriech (1991) conducted the study and reported the finding to IEPA. Leachates were analyzed for heavy metals, semi-volatiles, PAHs, and polychlorinated biphenyls (PCBs). Metals were determined via ICP-AES. Gas chromatography-mass spectrometry (GC-MS) was employed for analysis of organic compounds. Low levels of barium (<0.5 mg/L) in three samples, and 0.52 mg/L of chromium in one sample, were detected. Low levels of a few PAHs (<1 μg/L) were observed, but no semi-volatiles or PCBs were found above the detection limit.

A larger follow-up study ensued after IEPA reviewed the results. In December 1991, IDOT collected samples from two roads in Illinois. One was a PCC pavement and the other was HMA pavement of similar age (1976) built on the same state highway route, receiving the same traffic. Samples were taken transversely across each pavement, as well as longitudinally. This included outside the wheel path, in the wheel path, between the wheel path and from the shoulder of the road. Per IEPA request, leachates were analyzed for PAH and metals only, as previous results showed no evidence for other compounds of concern. Findings were reported to IEPA (Kriech, 1992a). Low levels of barium (mg/kg range) were detected in the HMA pavements (5 out of 6 samples) taken across the pavement; these were below regulatory limits. Through component source testing, barium was determined to originate from the natural limestone aggregates. Trace levels of a few PAHs were detected, with naphthalene present at the highest concentration, but less than 1 μg/L. Interestingly, the concrete pavements also showed low levels of naphthalene (4 out of 6 samples) taken across the pavement. This suggests other sources of confounding materials, dropped on the pavement from traffic, and not related to asphalt which was originally assumed. After this second study, IEPA determined that asphalt and concrete materials from old pavements could be safely used in Clean Fill applications. The State of Illinois enacted this finding into law.

Research was conducted to study leachability of cold mix asphalt used for patch mix and low volume rural roads in the US (Kriech, 1992b). These types of pavements had never previously been studied. Because these asphalts are often made with water-based asphalt emulsions, and sometimes contain solvents such as kerosene or #2 fuel oil at low concentrations, they were evaluated for metals, volatiles, semi-volatiles, and PAHs using EPA's lists for hazardous materials. No metals, volatiles, and semi-volatiles were observed above the limits of detection of 1 mg/L (metals) and 0.1 mg/L (volatiles/semi-volatiles). Low levels (range μg/L) of a few smaller PAHs, up through pyrene, were measured. It was recommended that these cold mix asphalt stockpiles not be stored near bodies of water.

A study for the Texas DOT was conducted to see if any constituents of environmental concern leached from a total of 33 asphalt stockpiles across the state (Southwest Laboratories, 1993). Materials tested included asphalt patch mix for potholes, RAP (Fig. 5) from milling of old roads, asphalt emulsion precoated aggregate for asphalt chip seals, and reclaimed asphalt millings with a rejuvenator added. Testing included TCLP for metals, semi-volatiles, and volatiles including benzene, toluene, ethyl benzene, xylene, and methyl ethyl ketone. Trace metals were detected, including one sample that contained cadmium just above the standard for drinking water of 0.01 mg/L. One sample contained benzene just over the 5 μg/L limit in drinking water. Compounds for all other samples were below the drinking water standards. The study concluded that asphalt materials used in stockpiles in Texas were not a significant environmental concern.

The University of Florida conducted a comprehensive study of leaching characteristics of asphalt road waste assessing RAP from six stockpiles across Florida (Townsend and Brantley, 1998). Leachate samples were tested for volatiles, PAHs, and metals. Both TCLP and column (lysimeter) leaching were conducted. The TCLP leachate water

Fig. 5. Pile of reclaimed asphalt pavement (RAP).

showed no metals above the detection limit. Additionally, no volatiles or PAHs were found above the detection limit. Only one column test showed lead at concentrations above the groundwater guidance of 15 μg/L. Prior use of leaded gasoline was presumed to be the source of lead, as it was observed only in the oldest RAP sample.

In Europe, aqueous leaching of PAHs was studied from bitumen and asphalt (Brandt and De Groot, 2001). For clarification, in Europe, a bitumen is equivalent to an asphalt binder in North America, whereas asphalt refers to the pavement type used. Nine different commercially available European asphalt binder samples, and one asphalt pavement made from one of the asphalt binder sources were explored. This Dutch study used a standard Dutch Static 30-h static Leachate Test Method, as well as a Dynamic European Union Leachate, using a Zero Headspace Extraction (ZHE) leachate, which is very similar to US EPA TCLP's ZHE leaching. Both total and leachable testing of PAHs were conducted on all nine neat bitumen samples. Asphalt binders and pavement met all criteria for leachate results that were below potable water standards in European Countries. Leachability within the first 3–6 days was predictive of ultimate total leachability of the samples using the Dutch Standard Method. Brandt's study is important because it considered total PAHs in the asphalt binder as well as the leachable PAHs. Knowing both helps to determine the upper limit of what could potentially be released and the propensity to do so over time. This study found that the total PAHs are low and the propensity to leach are orders of magnitude lower.

Similarly, studies of six paving asphalt binders used widely in North America were investigated. Standard EPA TCLP methods were employed to determine total and leachable PAHs (Kriech et al., 2002). Extraction and analysis of PAHs in a complex material, like asphalt binder, is challenging. Authors considered commonly used methods and determined that the best recoveries were obtained using a micro Dimethylsulfoxide (DMSO) extraction. Total PAHs levels were low, and the leachable levels of PAHs were near or below detectable levels.

Complimentary testing was conducted to explore different analytical methods to quantify metals in neat asphalt binders, as well as leachate from ten different asphalt binders (Kriech et al., 2005). Neutron activation was determined to be the most sensitive for evaluation of neat asphalt binders, however, this instrumentation is not readily available. ICP-MS was the most sensitive for testing leachate water from asphalt. Low concentrations (μg/L) of only aluminum, chromium and titanium were detected in the leachate. Wrapping the sample in foil confounded the aluminum concentration.

Leaching was conducted on RAP, as well as RAP recycled back into new pavements at 10% and 20% levels to investigate both PAHs and heavy metals (Legret et al., 2005). Samples were subjected to standard French leaching procedures and column leaching testing for metals and

PAHs. Results showed that small amounts of RAP materials leached early in contact with water, then decreased quickly to non-detectable levels. New pavements that contained RAP showed very low levels of leachate in water. Because samples were crushed for testing, a correlation between particle size (surface area) and leachable compounds was determined. It was suggested that contaminants collected on the RAP over its life tend to release mostly in the first flush when stored in a stockpile.

Four pavement cores of various ages from Denmark were crushed, extracted for total PAHs, and leached using a 64-day column study (Birgisdottir et al., 2007). A model of determining potential cumulative release was built from comparison of total PAHs to available PAHs leached over time. Three of the four pavements met Danish water quality criteria of less than 1.5 mg/kg of total PAHs. The older pavement was just above the limit at 1.7 mg/kg. It was concluded that leaching is diffusion controlled and only a minor portion of PAHs is released from pavements. Samples for this study were ground to <125 μm, which creates a larger surface area. Compared to the standard EPA TCLP procedure, which crushes the material to less than 9.5 mm, this study presents a significantly more severe process. Results suggest that only the smallest, most water-soluble PAHs, such as naphthalene, follow the diffusion model. Confounding pollutants from a fueling station potentially added to the PAHs but contributions were not determined.

Water quality was investigated of leachate produced from pavement specimens via a controlled laboratory study (Kayhanian et al., 2010). California DOT aimed to determine if pavement materials were contributing sources of pollution in highway runoff. In collaborative efforts with UC Davis, eight asphalt cement and two PCC specimens were tested. A specially designed apparatus and water distribution tray was employed for evaluation of the water quality of the corresponding leachates. Most organic and inorganic chemical constituents were below or near the reporting limit, except for chromium and vanadium from Portland cement in a single cement sample.

Recycled concrete aggregate (RCA) as granular bases under asphalt pavements was studied in Norway (Engelsen et al., 2012). The pavement sections collected water infiltrating through the roadway for four years. Some sections were not covered with asphalt. The collected water was tested for pH and metals. Results showed that asphalt covered sections had a much slower pH change in the granular recycled concrete base than the uncovered section. This suggests that the asphalt covering over the RCA acts as an impermeable barrier, greatly reducing the rate in which leaching occurs in the granular base. The addition of deicing salts may contribute to increased leaching of metals in the granular layers, due to improved diffusion of salts into the surface of RCA. None of the metals which leached exceeded acceptance criteria with groundwater and surface water in Norway at the time of the study.

Similarly, five different RAP samples from five states used as an unbound granular base were studied (Shedivy et al., 2012). TCLP in both deionized water and buffered solution was performed per method protocol. Leachate was analyzed for metals via ICP-OES and PAHs via high performance liquid chromatography (HPLC). Metals were observed below the maximum contaminant level (MCL) for drinking water, whereas PAHs were measured either below or near the level of detection. One note of caution is that the UV/fluorescence HPLC method, required by EPA Method SW-846-8310, has limitations for analysis of complex materials, like asphalt, due to lack of chromatographic resolution. With advances in sensitivity, GC/MS is now preferred due to increased resolution and mass spectral confirmation.

A study was conducted in Spain using a variety of recycled aggregates from construction and demolition wastes (Del Rey et al., 2015). RAP, RCA (Fig. 6), and ceramic materials for use in aggregate applications were studied. European leachate test, consisting of a two-step leaching procedures, as well as column leaching at different solid-to-liquid ratios were conducted. Concentration of metals was determined via ICP-MS. Additionally, atomic absorption (AA) spectroscopy aided in quantitation of total chromium. Hexavalent chromium was also tested via a standard European method. Many of the samples (14 out of 20) tested were classified as non-hazardous. The RAP from asphalt and two RCA concrete samples were considered inert in the testing. However, three out of five samples of the ceramic materials and some of the RCA had high levels of sulfate and chromium. Additional work on the type of chromium, however, found that all the ceramics were Cr(III) not Cr(VI), which is more toxic. It was revealed that at high pH conditions, Cr(VI) is released, but at pH levels below 5, primarily Cr (III) is released. This speciation of the type of leachable chromium is important in assessing environmental risks from C&D waste materials. Chromium entering the environment in any form should be carefully considered in studies.

The effect of carbonation on leaching of RCA in China was reported (Qin and Yang, 2015). Concrete pavements are alkaline; when crushed and used as aggregate, this alkalinity goes through a two-step repeating process. During rainfall cycles the alkali substances are diffused through leaching. During dry spells, the alkali exposed to air is converted to carbonates from absorption of carbon dioxide from the air. Because rainfall is slightly acidic, this carbonated material is dissolved and leached away. It was concluded that when RCA is used as a granular base, the surface layers above the granular base should be impermeable to rainfall to slow this two-step leaching process. This paper aligns with Engelsen (2020) in Norway, which found that covering the RCA with an impermeable pavement slows the process of pH reduction over time, releasing leachable compounds at a slower rate to the environment.

Dynamic leaching of monolithic surface pavements in France were assessed using European CEN/TC 351 standardized tests (Paulus et al., 2016). An evaluation of diffusion of materials mixed with binders used

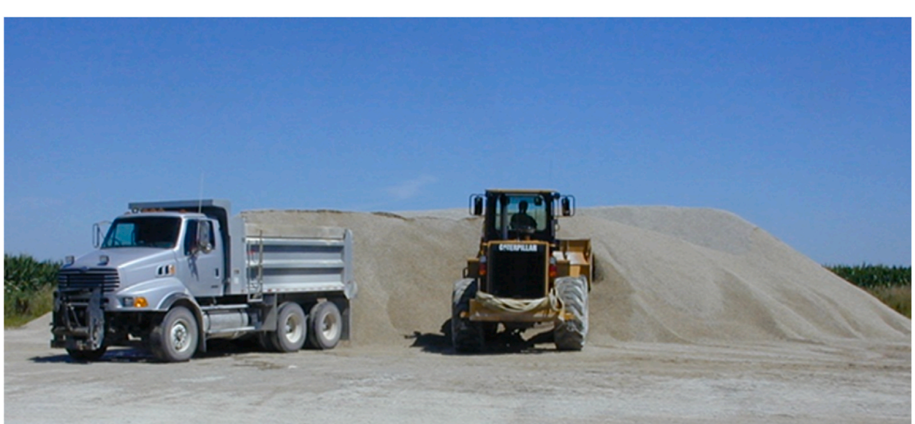

Fig. 6. Pile of recycled concrete aggregate (RCA).

in pavements was presented. Two surface pavements were evaluated; one a hydraulically (cement) bound material, composed of RCA mixed with limestone aggregate, and 7% hydraulic road binder (cementitious), enriched with activated blast furnace slag, and two, an asphalt surface pavement that met French standards. Pavements were evaluated for permeability to water, with average permeability coefficients (K) of 3.68×10^{-6} m/s and 2.12×10^{-7} m/s, for the cement treated surface and asphalt pavement, respectively. Samples were submerged in a vessel of demineralized water and allowed to soak for various times. After a soak period, the water was drained for testing and replaced over the study period. After 64 days of accumulative testing, leachate water was analyzed for pH, conductivity, sulfate, chloride, and fluoride. It was concluded that asphalt surface pavements have little impact on pH and conductivity and released no elements at the level of detection. Conversely, hydraulically bound (cement stabilized) pavements were alkaline, causing the pH of the water to rise to 10.1. Conductivity was between 150 and 220 μS/cm. Sulfate leached continuously throughout the study at ~40 mg/L, except in the last days. Chloride leached slowly over the first 12 days, then stopped. Fluoride was non-detectable in the samples. No trace metals were detected in the sample. It is surmised that the observed pH change is caused by calcium hydroxide leaching from cement over time. Comparable to studies performed on cement-based materials, sulfate and chloride was determined to leach through a pure diffusion mechanism. Hydraulically bound pavement surface was described as designed to carry 50–150 heavy vehicles per day. In contrast to Qin and Yang (2015), intermediate drying of the pavement, which had been shown to form carbonates, then release in rainwater during wet periods, was not investigated in this study. This highlights the importance of simulating field studies to understand real world conditions. In practice, roads used by vehicles are rarely constantly under water, and investigation of the effects of dry periods is essential for full understanding of leaching from roads.

Batch and column leaching of C&D materials in roadways in Portugal were evaluated from an environmental safety standpoint (Roque et al., 2016). Five recycled materials were used, including crushed concrete (CC), crushed mixed concrete (CMC), reclaimed crushed asphalt pavement millings from asphalt pavement, and a control material of crushed limestone aggregate. Batch leaching was tested via European standards (EN 12457-4 (2002)). Column leaching was performed, as described in the paper, by setting columns outside in the natural environment and collecting the leachate from rainfall events. Analyses included metals testing for cadmium, chromium, lead, copper, nickel by ICP. Sulfate and chloride were tested by ion chromatography (IC). Results of batch leaching found that none of the recycled material had metals detected above the Portuguese Standard as inert materials for a landfill. Levels of sulfate, chloride, and dissolved organic carbon (DOC) were within the regulatory limits for inert materials. The pH of the CC and CMC were 11.99 and 11.29 respectively, which are below the standards for hazardous materials. While Portugal had no requirements for pH, the US limits pH at 12.5 for classifying waste materials that would be hazardous by characteristic. Column leaching studies that evaluated different liquid-to-solid ratios had challenges because rainfall was insufficient to reach the last 10 L/kg sample fraction; it was found to be less aggressive than batch leaching. Metal leaching was determined to be low in all cases, but chloride and sulfate levels were higher in the CC and CMC materials than in batch testing. This may have been caused by limited dilution from low rainfall events during the study.

Recycled aggregate concrete (RAC), which is defined as concrete made from waste materials, was investigated in Spain (Cabrera et al., 2019). RACs can include C&D wastes such as concrete, brick, soil, metal, wood, and glass. Other materials include coal fly ash, biomass bottom ash, plastic, tires, volcanic ash (pozzolanic cement), iron and steel slag, or foundry sand. Aiming towards a circular economy, this study looked at the leaching potential of several waste materials. Percolations tests were determined to be most appropriate for granular unbound materials, whereas diffusion testing is best for monolithic materials. In this comprehensive review of C&D waste materials, the authors advise, based on prior studies, as to when and how to assess each potential waste material. The paper suggests both short- and long-term studies for complete evaluation of these various materials. In one study, C&D wastes from Barcelona were explored where a mixed recycled aggregate was used to make a porous concrete pavement. European Standard NEN 7345 method was employed to assess cumulative leaching emissions. The material was determined to be inert by European Standards, therefore they would be safe for use. The authors recommend performing simulation processes that mimic real world situations. Also, diffusion as well as dissolution studies helps to evaluate the effects of waste materials more fully on the environment. Again, total compounds of concern in the Municipal Solid Waste need to be considered in studies like this one.

Dissolved organic carbon on samples of asphalt binder were explored through different aging tests (Xue et al., 2019). This included thin film oven aging (Fig. 7), which simulates oxidation during the manufacturing of HMA materials through a standard Hot Mix facility. The impact of ultraviolet radiation (for 5 and 10 h) was evaluated also. Finally, some samples were subjected to pressure aging vessel (PAV) aging (Fig. 8) for 20 and 48 h. The PAV test was a simulation of long-term aging in the roadway of 8–10 years. Two leaching procedures were performed on the aged asphalt specimens. An 18-h batch leaching test as well as a 30-, 60-, and 90-day long term leaching was conducted. DOC was determined via GC-MS on the dissolved leachate to look for humic, fulvic, and the remaining hydrophilic compounds. Elemental analysis was also performed to determine carbon, nitrogen, sulfur, and hydrogen. Oxygen was determined by difference. Carbonyl index, which is used often to monitor oxidation of asphalt samples, was determined via Fourier transform infrared (FTIR). The results found that between 3.95 and 23.6 mg/L of DOC was released into the water. Leaching was conducted at different pH levels using acid, base, salt, and demineralized water. It was concluded that aging effects appeared to promote leachability of DOC. This laboratory study looked at the asphalt binder (Fig. 9) alone under simulated aging environment with UV radiation. Follow-up investigations to elucidate aging through UV exposure in field settings are necessary, since that is how these materials are used in practice.

Leachate from an asphalt binder and photoproducts were characterized by Fourier Transform ion cyclotron resonance mass spectrometry, revealing a wide array of highly oxygenated water-soluble hydrocarbons (Niles et al., 2020). Thin films of an asphalt binder were prepared by dissolution in a chlorinated solvent, spread on a glass slide and dried under nitrogen. The dried film was submerged under water (25 °C) and subjected to simulated sunlight or darkness. Concluding that asphalt binders can react with UV light in water to generate oxygenated species, the researchers admit that the study was not necessarily simulative of asphalt in actual use.

This and the study by Xue et al. (2019) reported earlier are the first to

Fig. 7. Thin film oven aging.

Fig. 8. Pressure aging vessel (PAV).

Fig. 9. Samples of asphalt binder.

explore the photooxidation of asphalt on leachability of asphalt.

Samples from three RAP stockpiles in New Jersey were subjected to different conditions with UV radiation and precipitation weathering (Yang et al., 2020). Column leaching tests included a secondary column of soil to evaluate metal attenuation. Trace metal analysis was performed on all the samples and compared to MCL standards for drinking water. One unaged and three different weathering simulations were explored. All samples met MCL standards, however, lead was measurable in two of the samples. The study suggests that the soil type under and around the RAP when used as an unbound granular bases should be considered. Low pH soils (pH < 5) should be avoided beneath unbound RAP that contains trace metals in the leachate such as lead, nickel, and manganese.

In Norway, a review of the leaching performance of recycled aggregates was conducted, including different approaches to evaluating recycled aggregates for various road applications (Engelsen, 2020). This thorough report goes through the appropriate steps in determining the suitability of using recycled aggregates. Starting with leaching properties in relationship to total content chemistry in the neat material, acceptable leachable content, and long-term leaching. By characterizing the recycled material for total chemicals present especially ones that are a concern from a health safety and environmental standpoint a clearer risk determination can be made as to the potential to release compounds over the life of the recycled aggregate.

A review of leaching and total concentration data for RAP was analyzed to estimate the risk (Spreadbury et al., 2021). Trace metals and PAH compounds exist in RAP, from both the primary asphalt pavement binder and aggregate, but external sources appeared to dominate with evidence of leachable contaminants from emissions and wear from vehicles. Except for elevated naphthalene concentrations (Norin and Strömvall, 2004), potential leaching risks cited by authors of the reviewed literature were limited aside from elevated naphthalene concentrations (Norin and Strömvall, 2004). The authors aggregated the existing data as much as possible and modeled these data to estimate the risk. A better understanding of factors that affect RAP leaching (e.g., aggregate/asphalt type, traffic exposure), reflective testing protocols, and robust risk assessment approaches can result in reevaluating best management practices to maximize RAP reuse and ensure the protection of human health and the environment.

4. Discussion

The most often studied compounds of environmental concern by researchers were toxic heavy metals and PAHs due to carcinogenic and genotoxic potential. Findings show that neither metals nor PAHs are released by asphalt or concrete pavements at regulatory levels in stormwater runoff or leachate in numerous studies conducted across four continents. RAP and RCP were frequently studied, both processing these materials once removed from the pavement and then stockpiling them before recycling.

The 18 studies on stormwater runoff explored the impact of different pavement systems and treatments on the quality of stormwater runoff. Sediments near coal tar sealed pavements were shown to have a higher PAH loading. PAHs have very negative impacts on aquatic species. A growing body of evidence has led to the restriction or ban of these coal tar sealers in pavement applications, such as driveways and parking lots. Their aesthetic and fuel resistance values are overshadowed by their potential harm to the environment.

Permeable pavement systems hold great promise for improving stormwater runoff quality and reducing the rate at which stormwater enters receiving bodies of water. This reduces the risk of flooding and improves water quality. However, the ability of these systems to maintain high permeability over time is challenging, since dust and sediments tend to inhibit permeability. Ultimately, clogging of pores negates the benefits of porous pavements. Methods of cleaning these pavements have shown some success in keeping them adequately permeable, but a complete solution is still needed. None of the studies addressed the accumulation of pollutants in the soils under the permeable pavement or the potential for contamination from spills that would be uncontained.

Our assessment of the pavement leachate studies is that leaching both short- and long term are critical in determining the potential to release any materials that could adversely impact water quality. Field studies to validate laboratory studies are recommended to determine realistic levels of leachability as the materials change through their life cycle. These studies help to determine if leaching is diffusion driven or equilibrium driven. The impact of pH and changes in pH over time, depending on the environment and soil that the recycled aggregate is used within, are important to consider. These should all be measured against drinking water and groundwater quality standards; the roadway should not be treated any differently than if these materials were placed in a landfill with limits on leachate. Table 2 summarizes the pavement leaching studies reviewed.

Based on the 42 papers reviewed, the interest in understanding stormwater runoff and leaching of materials from pavements is significant. The studies originated in 10 countries on four different continents and across 8 different US states. Requests for information on pavement

Table 2
Summary of pavement leachate studies.

References	Material	Testing	Lab	Field	Batch	Mono-lith	Column	Comments	Findings/Conclusions
Kriech (1990)	Hot Mix Asphalt (HMA) IN, US	VOCs, SVOCs, PAHs, Metals	✓		✓			AC-20 asphalt cement; aggregate #11 Levi slag, #11 stone, and #24 sand	0.1 mg/L Chromium, no VOCs or SVOCs, trace levels of naphthalene (0.25 μg/L). Below regulatory DW limits.
Kriech (1991)	6 Recycled Asphalt Pavement (RAP) samples IL, US	PCBs, SVOCs, PAHs, and metals	✓		✓			Illinois DOT -concerned about using RAP as clean fill. EPA determined it could be used as a clean fill. One sample high in Ba, Cr, & Pb attributed to surface contamination from crankcase dripping and leaded gasoline.	No PCBs in RAP. Leachate contained no SVOCs, trace levels of naphthalene in 2 of 6 samples and acenaphthene in 1 sample near DL. Other than trace levels of Ba in 4 of the samples, all regulated metals were ND. Sample E, however, contained Cr & Pb.
Kriech (1992a)	Portland Cement Concrete (PCC), Asphalt, and Soil surrounding roadways for use as clean fill IL, US	PAHs, Metals	✓		✓			Performed for Illinois Asphalt Pavement Association. Contiguous sections of Concrete & Asphalt roads – same traffic patterns.	Trace levels of Naphthalene detected in both PCC and asphalt. Barium was detected in all the PCC, but only 2 of the asphalt leachates.
Kriech (1992b)	Cold-Mix Asphalt: Asphalt Emulsion, Cutback Asphalt, Gelled Asphalt IN, US	VOCs, SVOCs, PAHs, Metals	✓		✓			PAH levels detected were similar to HMA & concrete pavements and soils from road shoulders. The aggregate was Indiana Limestone	Barium, naturally found in limestone, leached ~5 ppm, but when coated with asphalt, ND (DL = 2 ppm). No VOC, SVOCs detected other than 2–4 ring PAHs ranging from 0.10 to 8.0 μg/L
Southwestern Lab, 1993	5 types of asphalt, 33 samples, all stockpiled RAP TX, US	Trace metals, VOCs, SVOCs	✓		✓			Synthetic precipitation leaching procedure used by the Texas DOT	Trace levels of metals, VOC, and SVOCs may leach from some asphalt materials, but levels are not present in "environmentally significant amounts"
Townsend and Brantley (1998)	6 different RAP sources FL, US	VOCs, PAHs, Heavy Metals, pH, ORP, DO, Conductivity, TDS, Alkalinity, COD, and NPOC	✓		✓		✓	3 types: TCLP, synthetic precipitation and a deionized water leaching procedures. Batch-scale and leaching columns	Pb in oldest RAP samples at highest conc. (from leaded gasoline and crankcase oil). Batch tests were more dilute than the column test. Overall, RAP would pose minimal leachate risk when used as fill
Brandt and De Groot (2001)	9 asphalt cements, some oxidized, one HMA mix Netherlands	PAHs	✓		✓	✓		Water distribution coefficients calculated for PAHs. Static and dynamic leaching compared. If you know concentration of PAH in the asphalt, you can calculate leachable concentrations from aqueous solubilities	Static leach showed increasing concentrations in first days reaching steady state between 3 and 6 days. Equilibrium concentrations of PAHs stay well below surface water limits
Kriech et al. (2002)	6 paving asphalt cements, 4 roofing asphalt cements IN, US	29 Polycyclic Aromatic Compounds	✓		✓			Paving asphalts selected from the Strategic Highway Research Program Asphalt binder PAC concentrations also reported	Leachates had trace levels of naphthalene and phenanthrene (2 of 29 PACs investigated) in 3 of 10 samples, but < DW levels.
Kriech et al. (2005)	6 paving asphalt cements (same materials used for Kriech et al., 2002) IN, US	Total and leachable metals by neutron activation energy, X-Ray Fluorescence, and ICP-MS	✓		✓			All techniques helpful for results on asphalt cements; ICP-MS best for leachate analysis. Aluminum foil used skewed Al results.	The neat asphalt contains quite a few metals at variable levels based on crude source. All leachable metals (Al, Cr, Ti, Zn) were well below DW standards.
Legret et al. (2005)	RAP vs new conventional asphalt pavement France	PAHs by HPLC, Metals, total hydrocarbons, conductivity, pH, total organic carbon (TOC), chloride & sulfate	✓	✓	✓		✓	Differences between batch leaching tests and column experiments were not very significant. Grain size of the material and the water flow rate influential.	Pollutant leaching is generally weak & below DW standards. RAP sometimes had higher conc. of TH and PAHs but no significant differences. Initial column leaching stages higher but decrease rapidly reaching BDL

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Table 2 (*continued*)

References	Material	Testing	Lab	Field	Batch	Mono-lith	Column	Comments	Findings/Conclusions
									levels. Core samples from field confirmed results.
Birgisdottir et al. (2007)	Hot mix asphalt drilled from two existing roads –2002 Denmark	PAHs, total and leachable by GC/MS	✓		✓	✓	✓	64-day tank leaching test	Leaching of PAHs was diffusion controlled for 7 PAHs. Bases on calculations, only a small % of PAHs in the asphalt is leached out over 25-years.
Kayhanian et al. (2010)	10 asphalt and concrete pavement specimens, aged & unaged, course & dense graded, modified & unmodified CA, US	Conductivity, Harness, pH, TDS, TSS, COD, DOC, TOC, O&G, TPH, Metals, Nutrients, PAHs, Methylene blue active substances	✓			✓		Special system designed - water flow over pavement specimen and water samples. Nearly all detectable metal concentration was obtained during early experimental stage	Nearly all detected Cr and V concentrations were associated with concrete as compared to asphalt. Most organics and other metals detected in leachates for all pavements were near or below DL.
Engelsen et al. (2012)	Field site, foam glass, RCA, natural aggregates, and RCA + RAP Norway	pH, Al, As, Ca, Cd, Cr, Cu, Mg, Mo, Na, Ni, Pb, S, V and Zn, anions Cl−, Br−, SO4 2− and F−, DOC and DIC. XRD on RCA		✓				Extensive field collected leachate study. Release of major elements as function of time and pH. Simplified method for risk assessment. Mg increased over time due to drop in field pH, while Al and Ca decreased with time.	4-years of monitoring release found that field concentrations of Cd, Ni, Pb and Zn were low. 2 of winter seasons, Cr and Mo levels increased. De-icing salt suspect. Temp influenced amount of infiltration water. Exposure conditions also effected carbonation
Shedivy et al. (2012)	RAP (5 States) + one new asphalt WI, US	PAHs by HPLC, pH, Electrical Conductivity, Oxidation/Reduction Potential, and Metals	✓		✓			Column leaching planned but not yet performed HPLC – Fluorescence was used for PAH analysis	Most PAH concentrations of the RAP leachates were near or < DL and below groundwater intervention values. Metals tested were below MCL concentration in DW, except for Mn & As (Mn 1042.09 μg/L and As 95.66 μg/L from new RAP). Cr and Cd were <DL both in leachate from TCLP fluid and DI water.
Del Rey et al. (2015)	Recycled aggregates from Construction and Demolition Waste (CDW) Spain	Chromium and oxidation states and sulfate	✓				✓	Leached sulfate and Cr were mainly released by bricks and tiles Five unused ceramic materials, two old, crushed concretes and one new mortar.	Chromium and sulfate are focus of study of CDW, which were mainly leached from the ceramic materials (bricks and tiles). Total Cr, Cr (III), Cr (VI) were also tested in the leachates. Generally, Cr (III) was released from bricks and tiles, but recycled aggregates mainly released Cr (VI), which is highly leachable and toxic. Authors encouraged legislation that assess the levels of Cr (VI) in the leachate
Qin and Yang (2015)	Recycled concrete aggregate China	Acid intake and pH drop	✓			✓		Theoretical model the acid intake of in-situ RCA layers Authors point out limitation of the column test, which does not represent the in-situ layer.	Although RCA use is encouraged, when used in road layers, intermittent leaching occurs and neutralizes the alkali of the RCA. Leaching intervals fosters carbonation of the RCA's lingering mortar. Dense pavement on top can block CO_2 to promote carbonation in the RCA, which would slow down the drop in pH of the RCA layer, delaying release of toxic metals influenced by pH.
Paulus et al. (2016)	A hydraulically bound material (concrete) and an	DOC, sulfate, pH, conductivity, and inorganic analyses	✓			✓		Dynamic surface leaching test	Here a 64-day diffusion test was performed on asphalt concretes (AC) and

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Table 2 (*continued*)

References	Material	Testing	Lab	Field	Batch	Mono-lith	Column	Comments	Findings/Conclusions
	asphalt concrete. France								hydraulically bound monoliths. Limited release of toxic constituents in the AC is shown. For the monoliths, ~20,000 mg/ m_2 of sulfate was leached (1 order of magnitude > than chloride diffusion & 2 orders of magnitude > than fluoride.
Roque et al. (2016)	Recycled CDW, crushed (cr) concrete (C), cr mixed C, cr reclaimed (rcld) asphalt pavement (pvt), milled rcld asphalt pvt, natural cr limestone Portugal	Heavy metals, Cl-, SO_4, pH	✓		✓		✓	Batch and lysimeter tests	CDW in civil engineering works required evaluation of leachability. Batch and lysimeter tests were conducted with Cl and SO4 lower in the batch test than lysimeter test and dissolved organic carbon flipped. Batch test results were compared with Portuguese limit values, with the studied CDW samples releasing substances far below these limits.
Cabrera et al. (2019)	Recycled aggregate concrete, and CDW Spain	pH, heavy metals, and other elements, carbonation	✓		✓	✓	✓	Review paper Review includes environmental risks of RAC, the various reactions in CDW (i.e., pH variations on release), the various universal leaching methods, and legal regulations including EPA, WHO, and EU leachate limits. Specific applications should be tested and not just in the laboratory.	With the boom in use of recycled materials, many wastes like fly ash, bottom ash, glass waste, steel slag, tires, and plastics can be used as new raw materials in the manufacture of concrete. Concrete degrades primarily due to decalcinization, attack by sulphates, alkali-carbonate and alkali-silica reactions, attack by sea water or water with chlorides or acid attack; degraded concretes tend to produce more leachate contaminants. Assessment of potential environmental risk, concepts and mechanisms which control release, and leaching characteristics of unbound recycled materials were also discussed.
Xue et al. (2019)	60/80 pen grade asphalt, and SBS modified China	Dissolved Organic Compounds (DOC), fulvic & humic acids, FTIR, XPS, pH	✓		✓			30, 60 and 90 days, pH of leachants significant Thermal (TFOT and PAV aged asphalt binders) aging showed higher depletion rates for long-term leaching periods while UV aging showed higher leachability of DOC during short-term leaching process.	Leachability of DOC from aged asphalt related to aging process (thermal oxidation and UV) and leachants. DOC was time and pH dependent in both short and long-term leaching studies and was significantly influenced by the chemical composition. Aging effects the chemical composition of the asphalt binder and the subsequent leachate. For short-term aged samples, leaching of DOC appeared to be pH-dependent while leaching time was more influential for long-term aging samples. FTIR showed leachates mainly consisted of hydrocarbon and oxygen containing compounds. Fulvic acids and hydrophilic organic carbon fractions were high while a

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Table 2 (*continued*)

References	Material	Testing	Lab	Field	Batch	Mono-lith	Column	Comments	Findings/Conclusions
									small % of humic acids was detected.
Niles et al. (2020)	PG 76-22 Asphalt, polymer modified FL, US	FT-ICR MS Analysis of neat binder and leachate for composition & NPOC	✓		✓			Atypical leaching process and photooxidation HRMS revealed two structural motifs in asphaltenes: an aromatic core with alkyl side chains (island) or several covalently linked aromatic cores (archipelagos). Asphaltenes with high concentration of archipelago motifs were shown to “crack” to produce small water-soluble polyaromatic hydrocarbons (PAHs) through photofragmentation, whereas samples enriched in single-core island motifs were not.	In the study, the 168-h irradiated samples lost 6.56% of the asphalt as water soluble compounds while the water insoluble asphalt fraction increased by 4.1%. Analysis of the leachable compounds using ultrahigh-resolution Fourier transform ion cyclotron resonance mass spectrometry (FT-ICR MS) high resolution mass spectrometry confirmed the presence of large, oxygenated hydrocarbons. This study concluded that asphalt binders can react with ultraviolet light (UV) in water to generate oxygenated species. The researchers found oxygenated species of Polycyclic Aromatic Compounds (PAC) present in the water.
Yang et al. (2020)	RAP, HMA, and soil NJ, US	Metals		✓	✓		✓	pH important Time-series concentrations of select elements in 6 sets of column experiments showed that Ca, K, Mg, and S leached out fast, i.e., hours to <1 day, in both fresh-HMA and north-RAP samples, and in both RAPs and soil columns showed slower leaching during the 4 days of experiment due to drop in EC of column solutions.	Leachate from a RAP & a soil column were tested for potential release of trace elements. Mn and Ni found in RAP were largely attenuated in the soil. Conclusion: RAP can be used as unbound material except for in acidic (pH < 5) environments. Suspended particulates not included and may add to the environmental load. Unfiltered samples frozen for future study; also plan to include time release and rate studies of trace metals in strong acidic environments.
Engelsen (2020)	Recycled aggregates Norway	pH, Metals,	✓	✓	✓		✓	Globally, C&D waste is one of largest solid waste streams Inorganics in RCA were focus, mainly concrete, bricks, and natural aggregates. External field conditions will influence leachate results due to differences in pH, buffer capacity, water contact, and saturation.	In addition to in-depth chemistry & reaction kinetics of PC, plasticizers, air entrainers, superplasticizers, retarders, accelerators, spray concrete accelerators, & external sources during primary service life were also discussed. RCA with low leachable content is critical to facilitate high material recycling. Simplified dilution models by using the net infiltration were recommended as well as consideration of new formulations of today and in future - accurately determining direct environmental impacts.
Spreadbury et al. (2021)	RAP FL, US	Metals & PAHs	✓	✓	✓		✓	Review with Risk modeling Comparing RAP data is challenging due to variety of approaches. Risk models use different assumptions and calculations, estimates should be followed with field	Factors influencing RAP leachates include pavement materials, external sources, and testing procedures. Review suggests that contamination of underlying or nearby water

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Table 2 (*continued*)

References	Material	Testing	Lab	Field	Batch	Mono-lith	Column	Comments	Findings/Conclusions
								monitoring to ensure human and environmental health	supplies is unlikely from stockpiles of RAP, although on occasion, elevated leached concentrations of certain metals and some PAHs. These elevated RAP leachate data were assessed using US EPA IWEN fate and transport model to estimate dilution and attenuation of select metals and PAHs under typical environmental conditions, reuse, or stockpiling

CDW=Construction and Demolition Waste, HMA=Hot Mix Asphalt EC = Electrical Conductivity.

leaching are frequent, and come from Environmental Regulatory agencies, DOT, Non-Governmental Organizations, as well as private citizens. People want to know if it is safe to pave boat ramps, line fish hatcheries and drinking water ponds with paving materials. Is RAP or RCA safe to use for specific applications and store in stockpiles near water or over drinking water wells? Do pavements diminish water quality near lakes and rivers? These papers help to answer many of these questions. An overall trend shows that both concrete and asphalt pavements, absent of use deposition, appear to have minimal leachate concerns. They tend to not release heavy metals or toxic pollutants when built and constructed using standard road construction materials. New areas of study on the impact of UV (sunlight) will need further research including field validation (Xue et al., 2019; Niles et al., 2020).

Because roads eventually wear out from traffic, aging, and weathering, they eventually reach the end of their life. When these are removed through milling machines, as in the case of asphalt pavements, or demolished in the case of concrete, they should be appropriately managed. Since the 1970's, with the advent of the milling machine, asphalt pavements have been evaluated for recycling into new pavements. Greater than 98% of all pavements are reused or recycled in the US today (Williams et al., 2017). During the process, RAP is stockpiled, crushed, and screened to size, then reincorporated into new paving materials. RAP and reclaimed concrete from pavements were evaluated for alternative reuses in several studies including use of these materials in unbound granular bases underneath new pavements. Results showed that these generally have minimal impact on stormwater runoff and leachate. Old concrete pavements go through a similar process of crushing and separation and removal of reinforcing steel before being made into RCA. In Europe more so than in the US, when buildings are demolished, they are processed to sort concrete, metal, brick, wood, and glass for reuse or recycling. Managing these materials and finding appropriate uses were a focus of some of the papers reviewed. Are they safe and what is an appropriate reuse or recycling use for these materials? Until they are reused or recycled are, they safe for storage in stockpiles? Results show that it depends on what and how they will be used. Typically, if they can go back into their original use there are fewer issues of concern. However, when they are used in new ways such as unbound granular base materials, they need additional study. Understanding their potential to leach in the environment in which they will be placed helps to determine the safety of their new use.

Nontraditional materials from other industries can find their way into widespread use in the construction industry. Millions of tons of fly ash from coal fired power plants have been used for decades safely to replace 25–40% of Portland cement in concrete pavements. These pozzolanic materials have similar performance characteristics to Portland cement concrete over the lifetime of the pavements. Extensive laboratory and field studies have been conducted to fully understand these materials and qualify them through specifications as replacement for Portland cement. There are many benefits for their use in reducing greenhouse gas emissions from cement manufacturing and improved long-term performance in concrete. As other alternative waste materials are considered for pavement, they will need to be properly assessed for their long-term impact on water quality and recyclability.

5. Conclusions

Results of this review can assist state, county, city managers, engineers, and scientists in understanding and managing the impacts of pavement stormwater runoff and leachate by providing easy identification of studies most relevant to specific circumstances based on materials, testing parameters, and methodologies. In general, stormwater runoff and leachate from asphalt pavements and concrete that are normally constructed show low concentrations of pollutants. When adding waste or new additives to asphalt, robust leaching studies should be conducted to ensure encapsulation. Environmental pollutants that fall on or deposit on the pavement during its life can release low levels of PAHs and other pollutants in storage during rainfall events, especially during "first flush" rainfall events.

Concrete pavements that are reclaimed have similar profiles because they also collect similar contaminants over their life. Since concrete pavements have been found to release low levels of chromium from the cement depending on source and pH, it is important to monitor this element. In all cases, reclaimed pavement materials should be managed considering their surroundings to prevent these first flush pollutants from entering the waterways. Best practices include building these stockpiles on impervious bases with slope control to drain first flush through an absorptive media. However, in environmentally sensitive areas where groundwater is impacted easily, there should be proper evaluations when using these materials in place of conventional natural aggregates.

Porous pavements made from asphalt and concrete conclude that simple filtration of the water through the pavement and underlying drainage layers greatly reduces pollutants entering waterways. One challenge is how to keep these pavements from plugging over time. Also, highly water-soluble compounds such as deicing salts tend to pass through these pavements unabated.

Multiple studies found that coal tar pitch sealed pavements have negative impacts on stormwater and leachate. Particulate flakes from these pavement surfaces over time can get carried in stormwater runoff to ponds, lakes and rivers and negatively impact aquatic fish and invertebrates. Evidence suggests that these materials should be restricted from use on pavements.

Laboratory studies have looked at the impact of sunlight and water on formation of photooxidation products in leachate of neat asphalt binders. Further studies in this area are necessary to understand the impacts on the environment under real-world conditions.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Anthony J. Kriech reports financial support was provided by Asphalt Institute.

Heritage Research Group was hired by the Asphalt Institute to perform a comprehensive literature review to determine if compounds that leach from asphalt pavements pose an environmental risk or hazard under typical conditions.

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References

Ahrens, M.J., Depree, C.V., 2010. A source mixing model to apportion PAHs from coal tar and asphalt binders in street pavements and urban aquatic sediments. Chemosphere 81, 1526–1535.
Baldwin, A.K., Corsi, S.R., Lutz, M.A., Ingersoll, C.G., Dorman, R., Magruder, C., Magruder, M., 2017. Primary sources and toxicity of PAHs in milwaukee-area streambed sediment. Environ. Toxicol. Chem. 36, 1622–1635.
Ball, G.F.A., Herrington, P.R., Patrick, J.E., 2008. Environmental Effects of Emulsions. Opus International Consultants Ltd [Unpublished material].
Birgisdottir, H., Gamst, J., Christensen, T.H., 2007. Leaching of PAHs from Hot mix asphalt pavements. Environ. Eng. Sci. 24. Number 10.
Brandt, H.C.A., De Groot, P.C., 2001. Aqueous leaching of polycyclic aromatic hydrocarbons from bitumen and asphalt. Water Res. 35, 4200–4207.
Brattebo, B.O., Booth, D.B., 2003. Long-term stormwater quantity and quality performance of permeable pavement systems. Water Res. 37, 4369–4367.
Cabrera, M., Galvin, A.P., Agrela, F., 2019. Leaching issues in recycling concrete. Civil Struct. Eng. 329–356.
Crane, J., 2014. Source apportionment and distribution of polycyclic aromatic hydrocarbons, risk considerations, and management implications for urban stormwater pond, sediments in Minnesota, USA. Arch. Environ. Contam. Toxicol. 66, 176–200.
Del Rey, I., Ayuso, J., Galvin, A.P., Jimenez, J.R., Lopez, M., Garcia-Garrido, M.L., 2015. Analysis of chromium and sulphate origins in construction recycled materials based on leaching test results. Waste Manag. 46, 278–286.
Engelsen, C.J., 2020. Leaching performance of recycled aggregates. Adv. Construct. Demolit. Waste Recycl. 439–451.
Engelsen, C.J., Wibeote, G., van der Sloot, H.A., Lund, W., Perkovic, G., 2012. Field site leaching from recycled concrete aggregates applied as sub-base material in road construction. Sci. Total Environ. 427–428, 86–97.
Gilbert, J.K., Clausen, J.C., 2006. Stormwater runoff quality and quantity from asphalt, paver, and crushed stone driveways in Connecticut. Water Res. 40, 826–832.
Hernandez-Crespo, C., Fernandez-Gonzalvo, M., Martin, M., Andres-Domenech, I., 2019. Influence of rainfall intensity and pollution build-up levels on water quality and quantity response of permeable pavements. Sci. Total Environ. 684, 303–313.
Jiang, W., Sha, A., Xiao, J., Li, Y., Huang, Y., 2015. Experimental study on filtration effect and mechanism of pavement runoff in permeable asphalt pavement. Construct. Build. Mater. 100, 102–110.
Jobling, S., Reynolds, T., White, R., Parker, M.G., Sumpter, J.P., 1995. A variety of environmentally persistent chemicals, including some phthalate plasticizers, are weakly estrogenic. Environ. Health Perspect. 103, 582–587, 1995 Jun.
Kayhanian, M., Vichare, A., Green, P.G., Harvey, J., 2009. Leachability of dissolved chromium in asphalt and concrete surfacing materials. J. Environ. Manag. 90, 3574–3580.
Kayhanian, M., Vichare, A., Green, P.G., Alaimo, C., Hwang, H.M., Signore, J., Troxler, M., Jones, D., Harvey, J., 2010. Water quality evaluation of leachate produced from pavement specimens under controlled laboratory conditions. Road Mater. Pavement Des. 11 (1), 9–28.
Kayhanian, Masoud, Fruchtman, B.D., Gulliver, J.S., Montanaro, C., Ranieri, E., Wuertz, S., 2012. Review of highway runoff characteristics: Comparative analysis and universal implications,. Water Res. 46 (1), 6609–6624. https://doi.org/10.1016/j.watres.2012.07.026. In press.
Kayhanian, M., Li, H., Harvey, J.T., Liang, X., 2019. Application of permeable pavements in highways for stormwater runoff management and pollution prevention: California research experiences. Int. J. Transportat. Sci. Technol. 8, 358–372.
Kienzler, A., Mahler, B.J., Van Metre, P.C., Schweigert, N., Devaux, A., Bony, S., 2015. Exposure to runoff from coal-tar-sealed pavement induces genotoxicity and impairment of DNA repair capacity in the RTL-W1 fish liver cell line. Sci. Total Environ. 520, 73–80.
Kriech, A.J., 1990. Evaluation of Hot Mix Asphalt for Leachability. Heritage Research Group [Unpublished manuscript].
Kriech, A.J., 1991. Evaluation of RAP for Use as a Clean Fill. Heritage Research Group [Unpublished manuscript].
Kriech, A.J., 1992a. Leachability of Asphalt and Concrete Pavements. Heritage Research Group [Unpublished manuscript].
Kriech, A.J., 1992b. Leachability of Cold Mix Asphalts. Heritage Research Group [Unpublished manuscript].
Kriech, A.J., Kurek, J.T., Osborn, L.V., Wissel, H.L., Sweeney, B.J., 2002. Determination of Polycyclic aromatic compounds in asphalt and in corresponding leachate water. Polycycl. Aromat. Comp. 22, 517–535.
Kriech, A.J., Osborn, L.V., Kurek, J.T., Moberly, A.C., Stockburger, A., Kovar, L., 2005. Trace elements in asphalt cement (bitumen) and asphalt cement (bitumen) leachate: results and comparison of analytical techniques. Assoc. Asphalt Paving Technol. 1, 1–17.
Legret, M., Odie, L., Demare, D., Jullien, A., 2005. Leaching of heavy metals and polycyclic aromatic hydrocarbons from reclaimed asphalt pavement. Water Res. 39, 3675–3685.
Mahler, B.J., Van Metre, P.C., Bashara, T.J., Wilson, J.T., Johns, D.A., 2005. Parking lot sealcoat: an unrecognized source of urban polycyclic aromatic hydrocarbons. Environ. Sci. Technol. 39, 5560–5566.
Mahler, B.J., Ingersoll, C.G., Van Metre, P.C., Junz, J.L., Litte, E.E., 2015. Acute toxicity of runoff from sealcoated pavement to Ceriodaphnia dubia and Pimephales promelas. Environ. Sci. Technol. 49, 5060–5069.
Mijic, Z., Dayioglu, A., Hatipoglu, M., Aydilek, A., 2020. Hydraulic and environmental impacts of using recycled asphalt pavement on highway shoulders. Construct. Build. Mater. 234.
Nielsen, K., Kalmykova, Y., Strömvall, A., Baun, A., Eriksson, E., 2015. Particle phase distribution of polycyclic aromatic hydrocarbons in stormwater — using humic acid and iron nano-sized colloids as test particles. Sci. Total Environ. 532, 103–111.
Niles, S.F., Chacón Patiño, M.L., Puman, S.P., Rodgers, R.P., Marchall, A.G., 2020. Characterization of an asphalt binder and photoproducts by fourier transform ion cyclotron resonance mass spectrometry reveals abundant water-soluble hydrocarbons. Environ. Sci. Technol. 54, 8830–8836.
Norin, M., Strömvall, A.-M., 2004. Leaching of organic contaminants from storage of reclaimed asphalt pavement. Environ. Technol. 25, 323–340.
Panagopoulos, A., 2021a. Energetic, Economic and Environmental Assessment of Zero Liquid Discharge (ZLD) Brackish Water and Seawater Desalination Systems. Energy Conversion and Management, p. 235.
Panagopoulos, A., 2021b. Study and evaluation of the characteristics of saline wastewater (brine) produced by desalination and industrial plants. Environ. Sci. Pollut. Control Ser. 1–14.
Paulus, H., Schick, J., Poirier, J.E., 2016. Assessment of dynamic surface leaching of monolithic surface road materials. J. Environ. Manag. 176, 79–85.
Qin, Y., Yang, H., 2015. Carbonation dominates the acid intake of recycled concrete aggregate subjected to intermittent leaching. Construct. Build. Mater. 89, 110–114.
Roque, A.J., Martins, I.M., Freire, A.C., Neves, J.M., Antunes, M.L., 2016. Assessment of environmental hazardous of construction and demolition recycled materials (C&DRM) from laboratory and field leaching tests application in road pavement layers. Procedia Eng. 143, 204–211.
Sadecki, R.W., 1996. An Investigation of Water Quality in Runoff from Stockpiles of Salvaged Concrete and Bituminous Paving. Texas Department of Transportation.
Selbig, W.R., Buer, N., Danz, M.E., 2019. Stormwater-quality performance of lined permeable pavement systems. J. Environ. Manag. 251, 109510.
Shedivy, R., Meier, A., Edil, T.B., Tinjum, J.M., Benson, C.H., 2012. Leaching Characteristics of Recycled Asphalt Pavement Used as Unbound Road Base. Department of Geological Engineering, University of Wisconsin-Madison [Unpublished manuscript].
Southwestern Laboratories, 1993. Final Report Asphalt Stockpile Testing Project. Environmental Services, a division of SWL for the Texas Department of Transportation.
Spreadbury, C.J., Clavier, K.A., Lin, A.M., Townsend, T.G., 2021. A critical analysis of leaching and environmental risk assessment for reclaimed asphalt pavement management. Sci. Total Environ. 775, 1–16.
Townsend, T.G., Brantley, A., 1998. Leaching Characteristics of Asphalt Road Waste. Department of Environmental Engineering Sciences, University of Florida [Unpublished manuscript].
Williams, B., Copeland, A., Ross, T.C., 2017. National Asphalt Pavement Association (NAPA), Asphalt Pavement Industry Survey on Recycled Materials and Warm-Mix Asphalt Usage, vol. 138. Federal Highway Administration Information Series.
Xue, Y., Hu, Z., Wang, C., Xiao, Y., 2019. Evaluation of dissolved organic carbon released from aged asphalt binder in aqueous solution. Construct. Build. Mater. 218, 465–476.
Yang, Q., Yin, H., He, X., Chen, F., Ali, A., Mehta, Y., Yan, B., 2020. Environmental impacts of reclaimed asphalt pavement on leaching of metals into groundwater. Transport. Res. Part D 85, 102415.