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Review

A critical analysis of leaching and environmental risk assessment for reclaimed asphalt pavement management

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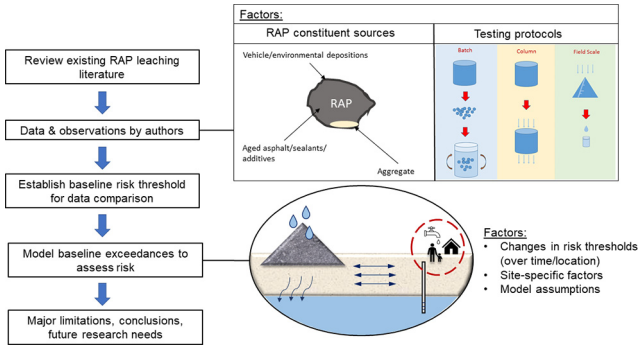
HIGHLIGHTS

- Critical literature analysis of RAP leaching and total concentration data
- Approaches and risk-thresholds are highly variable, and comparison is challenging.
- Direct exposure risk associated with RAP stockpiling is generally low.
- Fate and transport modeling indicate low leaching risk from RAP stockpiles.
- RAP stockpiling unlikely to contaminate underlying or adjacent water supplies

GRAPHICAL ABSTRACT



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ABSTRACT

In the last couple of decades, studies have examined the potential environmental risks associated with reclaimed asphalt pavement (RAP) management through a range of approaches. Variable risk assessment methodologies and uncertainty on RAP behavior in a stockpile or during reuse have led to inconsistent regulatory oversight in many jurisdictions. The objective of this literature review is to provide clarity on the findings pertaining to RAP leaching and assess the potential human and environmental health risks associated with reported constituent mobility from RAP. The reviewed literature focuses on the leaching of metals and organic compounds; direct exposure risk was briefly reviewed and found not to exceed natural soils or aggregates. On occasion, the literature reports elevated leached concentrations of certain metals (e.g., lead) and some polycyclic aromatic hydrocarbons (PAHs) from a few RAP samples. These elevated RAP leachate concentrations are assessed via fate and transport model (US EPA IWEM) to estimate dilution and attenuation of select metals and PAHs under typical environmental conditions and reuse or storage scenarios (e.g., stockpiling). This analysis suggests that most reported leachate concentrations of potential concern would be effectively attenuated at the most conservative conditions simulated (10 m, 100% infiltration); limitations with modeling are acknowledged. Pavement materials and external sources, along with chosen testing protocols, influence RAP leachate concentrations, affecting conclusions for potential environmental impacts of RAP in the literature. Understanding how these variables impact leaching and risk assessment is necessary to maximize and continue beneficial reuse of RAP while safeguarding human and environmental health.

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Abbreviations: AAS, atomic absorption spectroscopy; BaP, benzo(*a*)pyrene; COPC, constituents of potential concern; DAF, dilution attenuation factor; DI, deionized water; GCTL, groundwater cleanup target level; ICP-OES/AES, inductively coupled plasma optical/atomic emission spectroscopy; IWEM, Industrial Waste Management Evaluation Model; LS, liquid to solid ratio; MTs, metric tons; PAH, polycyclic aromatic hydrocarbon; RAP, reclaimed asphalt pavement; RCA, recycled concrete aggregate; REOB, re-refined engine oil bottoms; RSL, regional screening level; SCTL, soil cleanup target level; SPLP, Synthetic Precipitation Leaching Procedure; TCLP, Toxicity Characteristic Leaching Procedure; US EPA, United States Environmental Protection Agency; WTE, waste to energy; WQL, Water Quality Limits.

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1. Introduction

More than 4.3 million km of paved roadways serve the US transportation system, with 94% of these surfaced in asphalt (bituminous) concrete; approximately 360 million metric tons (MTs) (400 million tons) of asphalt pavement is produced in the US annually (NAPA, 2020). Over time, federal, state, and local governments maintain and replace existing asphalt pavements to ensure efficient and safe transportation. Pavement systems can vary depending on the design methodology used and the need of the infrastructure that relies upon it (Nejati et al., 2018; Spreadbury et al., 2020), but a typical asphalt pavement roadway is comprised of multiple structural layers, typically with stabilized subgrade as the foundation, followed by an aggregate base course, and lastly, an asphalt pavement course constructed in lifts as needed. Standard asphalt road maintenance involves milling the pavement (usually the surface course lift but in some cases the full depth of asphalt pavement) and placing a new pavement layer. The removed material, granular pieces of milled asphalt pavement, is referred to as reclaimed asphalt pavement (RAP) (Brantley and Townsend, 1999).

Asphalt concrete manufacturers often incorporate processed RAP as an ingredient in new asphalt pavement to reduce virgin binder demand and provide an additional aggregate source (Roberts et al., 1991; Mehta et al., 2017; Yang et al., 2020). RAP routinely replaces 10–20% of the raw ingredient mass in an asphalt mixture, with replacements upwards of 40% reported (Zaumanis and Mallick, 2014). Testing and field-observations have demonstrated RAP addition to increase an asphalt mixture's resistance to deformation and moisture intrusion, which has resulted in dedicated specifications being developed for its use (Roberts et al., 1991; Aravind and Das, 2006; Zaumanis and Mallick, 2014). The economic savings and environmental benefits, such as greenhouse gas reductions and landfill diversion, associated with RAP reuse are also well documented in the literature (Nassar and Nassar, 2006; Huang et al., 2009; Celauro et al., 2015; Yang et al., 2015).

Most US RAP ends up recycled into new pavement, but since RAP production and demand do not always match with respect to time and location, storing RAP for later use is common practice. In 2017, nearly 73 million MTs of RAP were incorporated into new pavement in the US, with an additional 91 million MTs stockpiled for future use (NAPA, 2019). RAP reuse also expands into other civil engineering applications such as embankment fill and road base or subbase (Taha et al., 2002; Arulrajah et al., 2013; Stolle et al., 2014; Schafer et al., 2019). On rare occasions, excess RAP is landfilled, but this is typically less than 1% of total RAP generated (NAPA, 2019).

If left uncapped, stockpiled RAP will inevitably be infiltrated by rainfall; this has led some to question whether RAP stockpiling and reuse represents a potential pollution source. While RAP's principal component is the mineral aggregate (~95%, by mass), it does contain aged asphalt (~5%, by mass), and over a road's lifetime the surface comes into contact with external deposition, including those from vehicles (e.g., brake pads/tire dust, fluids) and maintenance practices (e.g., sealants, traffic markings). To examine potential RAP environmental concerns, several researchers have reported the results of leaching experiments on RAP over the past three decades (Kriech, 1991; Brantley and Townsend, 1999; Norin and Strömvall, 2004; Legret et al., 2005; Townsend et al., 2013; Aydilek et al., 2017; Mehta et al., 2017; Yang et al., 2020). While most studies have reported pollutant concentrations in RAP leachate at levels below risk-based regulatory thresholds, a few observations of concentrations of potential concern have been reported (Norin and Strömvall, 2004; Yang et al., 2020).

The uncertainty regarding potential environmental emissions has led to inconsistencies among regulatory agencies regarding the appropriate restrictions, if any, that should be placed on RAP storage and unencapsulated reuse. Some government agencies limit or restrict RAP use and stockpiling, but most do not. Recent studies (Herrera, 2019; Murtagh and Vallette, 2017; Niles et al., 2020; Tian et al., 2021) suggest that there may be potential environmental impacts associated with runoff from asphalt roadways and, subsequently, RAP, but the literature lacks an in-depth investigation of how leached concentrations translate to human and environmental health risk. Nonetheless, real or perceived environmental impacts of RAP reuse and storage has major implications on its use in sustainable infrastructure, which may have additional economic and societal impacts (Abbas et al., 2019; Hussain et al., 2019, 2021). A clear understanding of the potential environmental risks associated with RAP reuse/storage would allow for a strategic approach in managing and repurposing this material in a way that best promotes human and environmental health.

Therefore, the goal of this review was to critically assess the existing scientific literature with regard to constituents of potential concern (COPC) in RAP, their potential to leach, and the assessment of human health and environmental risk, as regulatory limits, analytical methods, leach testing methodology, and risk assessment approaches evolve over time and vary depending on regulatory oversight. The objective of this work was to provide sufficient context on the current body of knowledge to provide a clear understanding of the current state of the science and research needs. This review begins with the discussion of potential RAP constituent sources in Section 2, followed by an examination of leaching methodologies/results from studies in Section 3. Section 4 then presents this compiled data in the context of the potential environmental risk, ending in a review of major conclusions derived from this

study and discussion of implications and future research needs based on them in Section 5 (this overview is visually presented in Fig. S1).

2. Asphalt pavement and RAP constituents

Mineral aggregate and asphalt binder represent the two primary components of RAP, and each of these contributes to trace chemical constituents. In addition, small amounts of other chemicals in RAP result from external sources, including road sealants, traffic markings, vehicle emissions, and wear of vehicle components. The two classes of constituents most commonly investigated in asphalt and RAP leaching studies are metals and polycyclic aromatic hydrocarbons (PAHs); chemicals from these classes are the focus of this review. Here we describe the various potential sources of trace chemicals in asphalt pavement and RAP, followed by specific presentations of metal and PAH concentrations reported in the literature.

2.1. Trace constituent sources

Asphalt binder (bitumen) is a petroleum product processed from crude oil and contains an assortment of hydrocarbons, including trace amounts of PAHs (Roberts et al., 1991; Kriech et al., 2002, 2005; Su et al., 2019). The association of asphalt as a petroleum product has motivated multiple investigations into PAH leaching from both asphalt roadways (Kriech et al., 2002; Mangani et al., 2005; Göbel et al., 2007; Mahler et al., 2015; Nielsen et al., 2015; Liu et al., 2017) and RAP (Brantley and Townsend, 1999; Legret et al., 2005; Mehta et al., 2017; Yang et al., 2020). Different asphalt binders exhibit a diverse suite of trace constituents depending on the petroleum source and manufacturing conditions (Roberts et al., 1991; Brandt and de Groot, 2001; Kriech et al., 2002).

Asphalt mix designs may also incorporate additives, including softeners, rejuvenators, and emulsifiers, to provide desired binder and mixture characteristics to meet design climate and traffic conditions for longevity (Hussain et al., 2020; Milad et al., 2020) and for production technologies, such as cold/warm-mix asphalt (Kaseer et al., 2020). Since additives influence the chemical properties of asphalt, it is plausible that their compositions may influence COPC contents in the resulting pavement material (Kriech et al., 2005; Asphalt Institute, 2016) or affect their mobility from an asphalt matrix (Kriech et al., 2002; Červinková et al., 2007). For example, re-refined engine oil bottoms (REOBs) derived from automobile engine oil are used as softening agents in asphalt products. REOBs can have varying formulations and introduce additional sources of metals, including zinc, copper, and molybdenum into asphalt binder (Asphalt Institute, 2016). However, scientific literature regarding RAP leaching as a function of additive addition is scarce.

Aggregates used in asphalt pavement production can be sourced directly from mining operations or industrial activity waste products (Modarres and Ayar, 2014; Yoo et al., 2016; Tahmoorian and Samali, 2018; Mikhailenko et al., 2020; Ahmedzade and Sengoz, 2009). Natural aggregates used in asphalt pavement contain various constituents, whose contents are a function of the mineral type and geologic source (Roberts et al., 1991; Yang et al., 2020). Similarly, waste-derived aggregates recycled into asphalt pavement reflect the composition of the parent waste material (Lindgren, 1996; Mijic et al., 2020). Waste-derived aggregate reuse aside from RAP is not commonplace, but examples of previously explored materials for use in asphalt pavement include coal ash (Modarres and Ayar, 2014; Yoo et al., 2016), recycled concrete aggregate (Tahmoorian and Samali, 2018; Mikhailenko et al., 2020), slag (Ahmedzade and Sengoz, 2009), and waste to energy (WTE) ash (Xue et al., 2009; Roessler et al., 2015). Other waste-derived additives, such as crumb rubber from vehicle tires, have also been reported to contain significant quantities of metals (e.g., mercury, aluminum) and PAHs (Azizian et al., 2003).

External sources contributing trace pollutants to asphalt pavement during normal use include those directly attributable to automobiles and other more disperse sources; Table 1 summarizes multiple potential external chemical sources from the literature. Motorized vehicles deposit a variety of fuels, lubricants, and dust particulates under routine traffic loads, which is exacerbated by vehicle accidents and spills. Vehicle components such as brake pads and tires abrade during normal use, depositing particulates containing elements such as zinc, cadmium, and copper (Hewitt and Rashed, 1990; Legret and Pagotto, 1999; National Research Council, 2005; Mummullage et al., 2016). Some vehicle exhaust will be deposited on the pavement surface and can introduce metals (Muschack, 1990) and PAHs (Crane, 2014); elevated PAH concentrations in stormwater runoff and soils near roadways have been associated with automobile emissions (Yunker et al., 2002; Tobiszewski and Namieśnik, 2012; Zgheib et al., 2012; Nielsen et al., 2015).

Nearby commercial and industrial activities can result in the introduction of trace chemicals to the road surface (Lindgren, 1996; Brantley and Townsend, 1999; Legret et al., 2005; Mijic et al., 2020), and atmospheric deposition is a well-established source in urbanized areas (Mangani et al., 2005; Göbel et al., 2007; Liu et al., 2017; Flanagan et al., 2018). Liu et al. (2017) observed unique PAH distributions in runoff from roadways in commercial areas and industrial areas, which were heavily weighted by indeno(1,2,3-c,d)pyrene and dibenzo(*a*)anthracene (characteristic of diesel engines), and acenaphthene, fluoranthene, and phenanthrene (characteristic of coal combustion), respectively.

Road maintenance activities such as applying deicing salts (Legret and Pagotto, 1999), roadway markings (State of Washington, 2015; Hanfi et al., 2020), and sealants (Watts et al., 2010; Mahler et al., 2014, 2015; Baldwin et al., 2017) can also contribute to trace chemicals in RAP. Sealants, particularly coal-tar-based products, are sometimes

Table 1
Constituents reported in asphalt concrete/RAP and their potential sources from the literature.

Sources	Constituents
Asphalt-related	
Asphalt sealants	PAHs[h,i,m]
Asphalt binder	V[l], Ni[l], PAHs[h,i]
Asphalt additives	Cu, Mo, Zn[g]
Aggregate-related	
Recycled concrete aggregate	As[j], Fe[j], Ag[j]
Traffic-related	
Brake pad dust	Pb[a,c], Cu[a,c], Cd[a,c], Zn[a,c]
Tire dust	Zn[a,c,d,e], Cu[a,c,d,e], Cd[a,c,d,e], Pb[a,c,d,e], Cr[a,c,d,e]
Vehicle leakage	Ni[a,c,d,e]Various metals[d], hydrocarbons[d]
Gasoline	Pb[a,c,e], PAHs[a,c,e]
Construction/maintenance related	
Coal-tar sealant	PAHs[h,i]
Traffic paint	Pb[k,n]
Road deicing salts	Zn[b], Cd[b], Pb[b]
Steel roadway construction components	Zn[a,c,d,e]
Roadway markings	Cr[e], Cd[e]
Other	
Pesticide application	Zn[f], Cu[f], Pb[f]

[a] Legret and Pagotto (1999).
[b] Bauske and Goetz (1993).
[c] Hewitt and Rashed (1990).
[d] Muschack (1990).
[e] Hanfi et al. (2020).
[f] Mangani et al. (2005).
[g] Asphalt Institute (2016).
[h] Watts et al. (2010).
[i] Crane (2014).
[j] Mahler et al. (2014, 2015).
[k] Tahmoorian and Samali (2018).
[l] Mehta et al. (2017).
[m] Roberts et al. (1991).
[n] State of Washington (2015).

applied to pavements during maintenance and contain PAHs at concentrations orders of magnitude higher than those present in virgin asphalt and asphalt-based sealants (Watts et al., 2010; Mahler et al., 2014, 2015; Baldwin et al., 2017). Elevated PAH concentrations in stormwater runoff have been specifically linked to coal-tar-based sealants (Mahler et al., 2005, 2010; Yunker et al., 2002; Tobiszewski and Namieśnik, 2012; Biache et al., 2014; Baldwin et al., 2017).

2.2. Metals in asphalt pavement and RAP

Analysis of metal concentrations in a media such as RAP involves a process whereby the samples are size reduced, digested using an acid solution under heat, with the resulting acid solution analyzed for metal content on an appropriate instrument, such as inductively coupled plasma optical/atomic emission spectroscopy (ICP-OES/AES) or atomic absorption spectroscopy (AAS). Results can vary depending on the procedure employed (Kriech et al., 2005); as most studies have been motivated by an environmental assessment, the procedures herein provide the *total environmentally available* concentration (see Table S2 for details). Table 2 summarizes reported metal concentrations for asphalt pavement and RAP samples from five different studies. In a risk assessment, these concentrations are compared to direct exposure thresholds, which will be discussed further later in the review.

Aggregate dominates the mass of asphalt pavement (relative to asphalt binder), and as such, many of the more abundant metals reported (e.g., aluminum, iron) are present in natural aggregates (Lindgren, 1996; Legret et al., 2005; Schafer et al., 2019; Yang et al., 2020). Legret et al. (2005) reported metal concentrations for a RAP stockpile and compared it to a fresh asphalt concrete mixture directly from a plant and a sample of its raw aggregate source. They found that these three materials had similar metals content within an order of magnitude of one another, though some differences were noted and attributed to a lack of material homogeneity. Asphalt binder can also contain measurable quantities of some trace metals such as zinc, nickel, and copper (Kriech et al., 2005; see Table 2).

Lead has been identified as potentially elevated in RAP from older roadways because of historic leaded gas use (Brantley and Townsend, 1999; Legret and Pagotto, 1999; Mehta et al., 2017), but as shown in Table 2, reported lead concentrations range from 6.3 mg/kg-dry (Von Gunten et al., 2020) to 53.8 mg/kg-dry (Rahman et al., 2014), which are comparable to typical aggregates (Schafer et al., 2019) and soils (Smith et al., 2014). Other external sources such as road striping may also be a source of lead in RAP. Depending on the vendor, lead from road striping products on in-service pavements was reported as high as 3000 mg/kg-dry in a 2015 survey of several local roads in Washington, US (State of Washington, 2015).

Arsenic, a trace metal that often poses challenges in waste reuse, was reported in RAP in the range of 1.3 mg/kg-dry (Schafer et al., 2019) to 8.8 mg/kg-dry (Von Gunten et al., 2020), also in the range of natural aggregates and soils (Chirenje et al., 2003; Smith et al., 2014; Welch et al., 2016; Missimer et al., 2018; Schafer et al., 2019). When Von Gunten et al. (2020) examined differences in asphalt pavement as a function of weathering, metal concentrations were higher in weathered samples compared to the unweathered counterparts, but differences in source aggregate and asphalt could not be ruled out as the reason. Based on sequential extraction results (details in Table S2), they concluded that sodium, magnesium, and barium were more mobile from weathered asphalt pavement, while aluminum, chromium, manganese, iron, nickel, arsenic, and molybdenum were less so.

2.3. PAH in asphalt pavement and RAP

The limited number of studies reporting total PAH concentrations (as opposed to leachable concentrations) of asphalt pavement or RAP have utilized methods developed for measuring trace organic contaminants in soil. These procedures utilize an organic solvent to extract organic chemicals away from the matrix and into the solvent, after which the solvent is processed and analyzed. Hundreds of PAHs exist, but most studies report the 16 US EPA priority PAHs (Abdel-Shafy and Mansour, 2016). The challenge with an asphalt-based matrix is that as an organic material itself, the asphalt will dissolve. Since PAH compounds are known to be present in asphalt, their presence is expected when dissolved in an organic solvent.

Table 3 presents reported PAH concentrations for asphalt pavement, binder, and RAP samples from several studies; Table S3 provides the different extraction procedures and parameters such as contact time and particle size reduction. These results are presented as benzo(*a*)pyrene (BaP) equivalents, a toxicity weighted average of seven carcinogenic PAHs (benzo(*a*)pyrene, benzo(*a*)anthracene, benzo(*b*)fluoranthene, benzo(*k*)fluoranthene, chrysene, dibenzo(*a*,*h*)pyrene, and indeno (1,2,3-*cd*)pyrene; see Eq. (1) in the Supplementary material). Individual PAH compound concentrations are presented in Table S1. Tables 3 and S1 present these concentrations alongside their respective risk-based thresholds; however, discussion of potential risk is deferred until later in this review.

The highest BaP equivalent concentration measured for RAP was reported in Su et al. (2019) (15.9 mg/kg-dry), which was an order of magnitude above the other reported studies for RAP and those of reported values from asphalt binders in the literature. The authors identified several factors that likely impacted this measurement, including asphalt source, use of sealants, traffic loading, service life, and stockpile age. PAH compounds are known to transform under various climatic conditions within the environment (Amato et al., 2011; Amato et al., 2014; Liu et al., 2007; Wei et al., 2015). Gbeddy et al. (2020) reviewed such reactions that may occur on the pavement surface, finding photolysis to be the most predominant influence in PAH transformation and degradation. Previous studies (Sadecki et al., 1996; De Lira et al., 2015; Niles et al., 2020) found that as asphalt binder is exposed to weathering (e.g., sunlight, rain), it can oxidize, influencing the diversity and concentration of PAH compounds. RAP binder was found to oxidize faster and to a greater extent at smaller particle sizes (De Lira et al., 2015), suggesting that RAP processing might also influence PAH concentrations in the product.

Birgisdóttir et al. (2007) and Su et al. (2019) (Table 3 and Table S1) reported similar trends of RAP sources from older pavements having elevated concentrations of PAHs relative to the newer pavements. These changes may result from greater exposure to vehicle emissions and deposition, attributed to variability in asphalt source, or a combination of both and other factors. As mentioned previously, coal-tar based sealants, which have been used in Europe and the US, contain PAHs at concentrations orders of magnitude above conventional asphalt-based sealants and unsealed asphalt pavements (Watts et al., 2010; Mahler et al., 2014), and would likely have an impact on RAP PAH concentrations when present.

3. Constituent leaching from asphalt pavement and RAP

The number of scientific studies reporting COPCs in RAP is relatively limited, but the available data does demonstrate that measurable concentrations of metals and PAH compounds are often present; this is the combined result of the aggregate, the asphalt binder, and deposition from external sources during the service life of the road. However, the mere presence of a chemical does not necessarily pose a concern to human health and environmental risk. While the total chemical concentrations summarized in the previous discussion can shed light on some potential risk pathways (discussed later), the primary concern cited for stockpiled or unencapsulated RAP stems from potential contamination of water coming into contact with RAP. Thus, most scientific studies on RAP risks focus on the potential for chemicals to leach from it in water-based extraction procedures.

Scientists have developed various test methods over the past few decades to assess the potential for waste materials to leach COPC, and

Table 2
Total environmentally available metal contents (mg/kg-dry) observed in RAP and asphalt pavement compared to that of conventional aggregates and asphalts. Dashes indicate unmeasured constituents, where BDL is listed designates a lack of detection limit provided.

Study	(Von Gunten et al., 2020)				(Legret et al., 2005)		(Arulrajah et al., 2013)	(Rahman et al., 2014)	(Schafer et al., 2019)	Asphalts	Conventional aggregates			
	Low traffic		High traffic		RAP	Fresh Asphalt Pavement	RAP	RAP	RAP	Six paving asphalts and four roofing asphalts (avg.) (Kriech et al., 2005)	Limerock (Schafer et al., 2019)	Cemented Coquina (Schafer et al., 2019)	RCA (Schafer et al., 2019)	Aggregate (microdiorite) (Legret et al., 2005)
	Fresh	Weathered (25+ years)	Fresh	Weathered (15 years)										
Constituents (mg/kg-dry)														
Aluminum	7,200 ± 1,200	12,600 ± 500	8,100 ± 1400	20,100 ± 1,800	40,380	61,290	–	–	1,580	16.8[a]	1,670	499	4,730	46,960
Arsenic	2.44 ± 0.17	8.8 ± 0.8	2.4 ± 0.6	4.0 ± 0.2	–	–	<5	6.0	1.27 ± 0.586	<0.0250	3.50	1.37	6.38	–
Antimony	–	–	–	–	–	–	–	–	2.15 ± 0.466	0.0325	2.24	1.98	2.27	–
Barium	160 ± 40	390 ± 30	160 ± 50	260 ± 40	–	–	64	453.4	10.2 ± 0.837	0.785	25.6	4.73	39.4	–
Boron	–	–	–	–	–	–	–	–	3.41	–	7.21	2.31	15.8	–
Cadmium	<0.06	<0.2	<0.08	0.25 ± 0.06	0.2	<0.1	<0.1	–	0.218	<0.00500	0.206	0.204	0.396	<0.1
Chromium	14.3 ± 1.0	20.66 ± 0.16	8.5 ± 1.3	18.0 ± 1.6	40	54.0	18	127.0	70.4 ± 9.35	0.905	40.8	79.6	55.1	32
Copper	<2	<2	<2	<4	11	20	–	43.0	6.02 ± 0.619	0.763	2.43	2.26	45.1	14
Iron	9,000 ± 3,000	40,000 ± 10,000	8,000 ± 5,000	22,000 ± 4,000	29,420	51,960	–	–	1,370	–	1,590	1,330	4,290	12,900
Lead	4.8 ± 1.3	6.9 ± 0.2	4.0 ± 0.9	6.3 ± 0.9	7	3	15	53.8	7.90 ± 3.88	0.181	0.826	0.816	12.1	5
Manganese	200 ± 100	700 ± 200	100 ± 50	600 ± 100	–	–	–	–	28.1	0.441	40.2	11.6	63.9	–
Molybdenum	0.96 ± 0.14	2.09 ± 0.16	0.9 ± 0.3	2.2 ± 0.3	–	–	–	4.0	2.15	1.23	3.71	2.09	7.93	–
Nickel	7.7 ± 0.2	17 ± 2	9 ± 2	19 ± 3	23	20	–	106.0	34.1	52.0	16.6	35.1	23.0	14
Selenium	<2	<2	<2	<3	–	–	<3	<3.0	1.23	<0.0250	2.035	1.03	0.998	–
Silver	–	–	–	–	–	–	<5	–	–	0.229	–	–	–	–
Thallium	2.3 ± 0.7	2.6 ± 0.2	1.8 ± 1.0	3.3 ± 1.2	–	–	–	–	–	0.0104	–	–	–	–
Zinc	10.7 ± 0.3	22 ± 2	13 ± 2	31 ± 4	48	86	–	160.0	20.5	1.94	2.24	1.62	127	46

[a] Partly attributed to aluminum foil used in sample handling.

Table 3
BaP equivalents (mg/kg-dry) for several studies on RAP, asphalt pavement, and binder samples. Italicized BaP equivalent values indicate exceedances of residential thresholds while bolded values indicated exceedances of commercial thresholds.

Study and material examined	BaP equivalent (mg/kg-dry)
Legret et al. (2005)	
RAP	*0.21*
Birgisdóttir et al. (2007)	
Base Layer (25 Year)	*0.38*
Wearing Course (25 Year)	*0.22*
Base Course (4 Year)	0.089
Wearing Course (4 Year)	*0.14*
Su et al. (2019)	
Fresh Asphalt Pavement	*0.2*
RAP, S FL, 12–15 Yr	**15.87**
RAP, Central FL, 12–15 Yr	*0.013*
Binder Only	
Asphalt (Legret et al., 2005)	*0.24*
PG 52–28 (Su et al., 2019)	0.012
PG 67–22 (Su et al., 2019)	0.012
Conventional Middle East Penetration (Brandt and de Groot, 2001)	**1.4**
Asphalt (Pinheiro et al., 2013)	0.03
Kriech et al., 2002[a]	
Roofing – A	**1.9**
Roofing - B	**2.3**
Roofing - C	**2.3**
Roofing - D	**2.3**
Paving - 1	**5.1**
Paving - 2	**2.3**
Paving - 3	**2.3**
Paving - 4	**2.4**
Paving - 5	**2.3**
Paving - 6	**1.9**
FL SCTL	
Commercial	0.7
Residential	0.1
US EPA RSL	
Commercial	2.1
Residential	0.11

[a] Elevated BaP equivalents calculated from data in Kriech et al. (2002) are a result of assuming concentrations below detection limit were equal to detection limit.

multiple studies have reported leach test results for asphalt pavement or RAP. Results vary depending on the material source, how samples are collected and processed, and the leaching protocol followed. In addition, several studies have investigated trace chemical concentrations in leachate from actual stockpiled RAP; while not as controlled, these studies most closely represent conditions expected in practice. Here we review the major studies reported to date (both lab studies and field observations), including the methodology employed as well as the metal and PAH concentrations measured. Authors of these studies typically include an assessment of risk based on the leaching results and a comparison to regulatory standards or risk thresholds. In this section, we focus solely on the leaching methodologies and subsequent results presented by the respective authors, with interpretation and discussion of how these measurements relate to environmental risk deferred until later sections.

3.1. Laboratory-based testing

Two major types of leaching protocols have been used to measure chemical leaching from asphalt pavement or RAP: batch and column tests. A batch leach test involves exposing a defined mass of granular solid (e.g., RAP) to a defined volume of leaching solution (e.g., water), often rotating end-over-end for a defined time period and collecting a leachate sample for analysis at the end after separation from the solids. Test variables include the leaching solution employed, liquid to solid ratio (LS), contact time, agitation, and sample preparation. Column tests differ in that the leaching solution is passed through a bed of material (e.g., RAP), allowing leachate samples to be collected over time as more liquid flows through the material. Batch tests are quicker and easier to perform, but column tests are usually considered more representative of true conditions (such as water flowing through stockpiled RAP). Tables 4a and 4b provide a summary of metal and PAH concentrations from studies where batch tests were used to leach asphalt pavement or RAP, while Tables 5a and 5b provide a similar table for studies employing column tests. These studies are described below.

In 1991, a study by Kriech examined six different RAP sources (n = 6) collected from across the state of Illinois, US using a batch leaching procedure and reported metal and PAH concentrations in the leachates. The batch test employed was the Toxicity Characteristic Leaching Procedure (TCLP), a method developed by the US Environmental Protection Agency (EPA) as one means to characterize a solid waste as hazardous. The TCLP uses an acetic acid-based leaching solution designed to simulate conditions in a municipal landfill (Liu et al., 2019a). The majority of the metals and PAH compounds were reported below the study detection limits. The only PAH compounds detected were benzo(*a*)anthracene (1 of 6 sources), benzo(*k*)fluoranthene (1 of 6 sources), naphthalene (2 of 6 sources), and acenaphthene (1 of 6 sources). Elevated concentrations of barium, chromium, and lead measured from one of the sources were attributed to contamination from lubrication fluid additives (barium), crankcase drippings (barium, chromium, lead), and leaded gasoline (lead). Because of the acid used in its leaching solution, the TCLP is well recognized to extract some metals (e.g., lead) to a much greater extent than expected with water (Clavier et al., 2019; Liu et al., 2019a).

Brantley and Townsend (1999) collected six RAP samples (n = 6) from across Florida, US, and conducted both batch and column tests. Several batch leaching procedures were utilized, including the TCLP, a US EPA rainwater leachate protocol called the Synthetic Precipitation Leaching Procedure (SPLP), and one using deionized (DI) water. All of the metals and PAH compounds examined were reported as below detection in the batch test leachates. Stainless steel columns containing 0.9 m of RAP were tested for each RAP source under both saturated and unsaturated conditions. Similar results were observed for most metals and all PAH compounds. An exception was lead, which was detected in several of the RAP samples under both saturated and unsaturated conditions; lead concentrations were attributed to historic deposition from leaded gasoline, and its concentrations dramatically decreased as water passed through the column. The occurrence of lead in the column leachates and not the batch leachates was attributed to the very different LS of each procedure (<1 vs 20). The PAH detection limits in this study, while meeting appropriate targets at the time, are greater than many regulatory PAH risk thresholds today (discussed in more detail later; see Table S4).

Morse et al. (2001) conducted an environmental assessment on several types of construction materials using the SPLP method, one of these being RAP from seven sources (n = 7) across Texas, US. The analysis was limited to leached metals, and elevated concentrations of antimony, barium, and lead were reported from these RAP sources. Another reused construction material, recycled concrete aggregate (RCA), also contained barium and lead concentrations similar to RAP. Several natural aggregates (e.g., gravel, sandstone, limestone) were also observed to have comparable leached concentrations of antimony, barium, and lead to those observed from RAP.

Norin and Strömvall (2004) collected milled asphalt from one road project in Sweden (n = 1) and conducted batch (fresh RAP) and column tests (fresh and stored RAP) on size-reduced material (<0.125 mm for batch, <2 mm for column). The batch test consisted of leaching at LS = 100, conducted twice, once at pH = 4 and again at pH = 10 using deionized (DI) water with nitric acid, and sodium hydroxide added as needed to alter pH. This resulted in several elevated concentrations of PAH compounds, such as naphthalene at 3.92 μg/L. PAHs were similarly detected in the column leachates (reported only for one data

Table 4a
Summary of batch tests: results of inorganic metals leaching (μg/L) from RAP samples with a range of detected concentrations reported. Dashes indicate unmeasured constituents, where BDL is listed designates a lack of detection limit provided. Shaded constituents and their corresponding values represent exceedances in US EPA RSLs (2020).

Study	Kriech, 1991	Brantley & Townsend, 1999 (Townsend, 1998)	Morse et al., 2001[b]	Legret et al., 2005	Kang et al., 2011	Shedivy et al., 2012	Hoy et al., 2016	Roque et al., 2016[d]	Aydilek et al., 2017[e]	Yang et al., 2020 (Mehta et al., 2017)	Schafer et al., 2019	Arulrajah et al., 2013	
Extractant Used/Method	TCLP	TCLP, SPLP, DI	SPLP	DI	DI	TCLP, DI	TCLP	DI	ASTM D3987[f]	TCLP	SPLP	ASLP (Acetate)	ASLP (Borate)
Leachate pH	n/a[a]	9.28-9.70	8.76-9.48	7.2-7.8	9.67	5.16-9.58	5.12	9.67, 10.95	8.30-9.34	4.93[h]	7.83 ± 1.10	5[h]	9.2[h]
LS	20:1	20:1	20:1	10:1-30:1[c]	20:1	20:1	20:1	10:1	20:1	20:1	20:1	20:1	20:1
Number of Sources	6	6	7	1	1	5	1	2	7	3	1	1	
Constituents (μg/L)													
Aluminum	-	-	<2000	-	370	6.36-364.55	-	-	<5-272	170-844	240 ± 77	-	-
Arsenic	<5	-	<25.00	-	10	9.32-***95.66***	<10	-	8.90-***<50***[g]	0.05-0.83	<8	<10	<100
Antimony	-	-	<5.00-***6.56***	-	-	5.55-***17.24***	-	-	-	0.12-0.39	<6	-	-
Barium	<200-400	<500	<2000-2016	-	70	21.19-664.81	-	-	<5-29.3	148-1333	270	170	<100
Boron	-	-	-	-	BDL	-	-	-	<5	-	62	-	-
Cadmium	<200	<5	1.02-2.00	<0.1	BDL	BDL	BDL	<0.5	<5	0.21-1.45	<1	<2	<20
Chromium	<50-520	<100	<5.00-6.20	<1	BDL	BDL-1.61	<50	<1.0, 4.6	<25	0.4-3.7	5.00	<10	<100
Copper	-	<500	<100.00	<5	BDL	0.40-14.78	BDL	<1.4, 3.0	<5-28.4	1-619	60 ± 97	-	-
Iron	-	-	-	-	410	BDL-85.33	-	-	<5-10.2	114-6500	399 ± 684	-	-
Lead	<200-***1800***	<10	***20.00-20.44***	<5	BDL	BDL-11.43	BDL	<2.4	<25	0.64-***50.12***	15 ± 12	***20***	<100
Manganese	-	-	<100.00-116.09	-	30	0.65-***1042.09***	-	-	<5	***449-1649***	6.00	-	-
Mercury	<5	-	<2.00	<0.1-0.2	-	-	BDL	-	-	-	-	<1	<10
Molybdenum	-	-	<10.00	<5	BDL	5.28-7.22	-	-	-	0.11-0.48	<6	-	-
Nickel	-	<100	<50.00	<2	BDL	BDL-25.70	<50	<2.6	<5	9.1-20.6	326 ± 562	-	-
Selenium	<25	-	<25.00	-	-	-	-	-	-	0.67-3.84	<2	<10	<100
Silver	<40	-	<100	-	-	0.10-0.92	-	-	-	0.01-0.06	-	<10	<100
Thallium	-	-	<2.00	-	-	-	-	-	-	0.03-0.19	-	-	-
Zinc	-	<500	148-1119	<10-115	BDL	10.04-123.53	1348	<0.6, 0.6	<5-8.90	59-2206	79.0	-	-

[a]Not reported. TCLP fluid (unspecified whether #1 or #2) used as extractant.
[b]pH in parenthesis include range of pHs (n = 7), which is mean value +/− standard deviation. This same method was used to present metals concentrations. If lower concentration range was less than detection limit, the given detection limit was used ("<").
[c]Material was leached thrice, each time at a LS of 10 with extractant tested each leaching cycle. Results present maximum leached during these cycles.
[d]Originally reported in mg/kg-dry. These were converted to μg/L using the provided LS (10). Averages for each source were presented for pH and constituents and are separated by comma.
[e]Some samples in this study were measured with lower detection limits than others. If none were detected between the lower and higher DL, then the higher DL was used ("<").
[f]Method does not prescribe a solution to be used but other operational parameters such as LS and contact time.
[g]Some samples had detection limits of <50 μg/L but some were detected in this range when lower detections were used (39.5 μg/L peak measured).
[h]pH of extraction solution before addition of RAP (not measured after leaching).

point, fresh asphalt at LS = 0.05 and stored asphalt at LS = 0.07) at higher concentrations, which may be attributed to the lower LS; again, naphthalene was the most prevalent at 28.0 μg/L and 9.2 μg/L for the fresh and stored RAP samples, respectively. The authors noted that lighter molecular weight PAHs (i.e., 2–4 aromatic hydrocarbon rings) leached less from columns after storage compared to higher molecular weight PAHs (i.e., 5 ring), which increased; this was attributed to their differing chemical properties (e.g., volatility, solubility). The authors attributed leached naphthalene to several potential sources: vehicle exhaust, asphalt binder, and rubber tire debris.

Legret et al. (2005) conducted both batch and column tests for one RAP source (n = 1) collected from a location in France. Before the batch tests, the RAP was size reduced (<4 mm), and a variety of batch conditions were employed using DI water following a European protocol (AFNOR XP X 31-210). The column tests were conducted under saturated conditions using DI passing through 0.35 m of RAP crushed to <20 mm for LS up to 30. Except for small concentrations of mercury and zinc, metals were not detected in the batch tests or the later stages of the column tests. Most column leachate PAH measurements were below the detection limit, with the highest measured concentrations for copper and zinc observed only during the early phase of the column (days 2, 10); all PAH were below detection limit after day 10 (samples were collected and analyzed at days 25, 50 and 75).

Birgisdóttir et al. (2007) collected asphalt pavement cores from two roads in Denmark (n = 2), one from a service station (built 1980) and another from a highway (built 2001) and examined PAH leaching using a modified column and a tank test procedure. These cores were further divided into wearing (surface) layer and base (structural) layer specimens. Testing similar to batch tests involved placing a crushed asphalt pavement core (95% < 125 μm) into a 10-cm long column and recirculating leachate to achieve an LS of 100. The tank test procedure kept the asphalt pavement cores intact and submerged them for 64 days replacing/sampling the contact water at timed intervals increasing LS over time and its impact on constituent flux. Testing in the modified column showed that PAHs leached to a somewhat greater extent in the surface course compared to the base courses, with the highway core having the highest concentrations. The lower molecular weight PAHs (naphthalene, phenanthrene) were more available to leach than those with an increased molecular weight, such as chrysene, benzo(*bjk*)fluoranthene, and benzo(*a*)pyrene, which were either at very low concentrations or below the detection limit. This trend was also observed for the tank experiments, with the authors reporting

Table 4b
Summary of batch tests: results of PAH leaching (μg/L) from RAP samples with a range of detected concentrations reported. Dashes indicate unmeasured constituents, where BDL is listed designates a lack of detection limit provided. Shaded constituents and their corresponding values represent exceedances in US EPA RSLs (2020).

Study	Kriech, 1991	Brantley & Townsend, 1999 (Townsend, 1998)	Norin & Strömvall, 2004[b]	Legret et al., 2005	Birgisdóttir et al., 2007	Shedivy et al., 2012	Townsend et al., 2013	Mehta et al., 2017
Extractant Used/Method	TCLP	TCLP, SPLP, DI	DI	DI	DI	TCLP, DI	SPLP	TCLP
Measured pH	n/a[a]	9.28-9.70	4, 11[e]	7.2-7.8	n/a	5.16-9.58	4.2[e]	4.93
LS	20:1	20:1	100:1	10:1-30:1[c]	100:1	20:1	20:1	20:1
Number of Sources	6	6	1	1	2[d]	5	5	3
Constituents (μg/L)								
Acenaphthene	<0.13-0.14	<5	0.057	<0.050	BDL-0.05	BDL-0.1342	<0.014	BDL-0.180
Acenaphthylene	<0.20	<5	0.338	<0.050	BDL-0.01	-	<3.5	-
Anthracene	<0.017	<5	<0.018	0.030	BDL-0.05	BDL-3.8343	<0.010	-
Benzo(a)anthracene	<0.017-0.017	<5	-	<0.025	***0.04-0.08***	BDL-***0.0831***	<0.00030-***0.0349***	BDL-***0.27***
Benzo(a)pyrene	<0.240	<0.25	<0.071	<0.010-0.020	BDL-***0.03***	BDL-***0.0544***	<0.00030-0.0233	-
Benzo(b)fluoranthene	<0.023	<1	<0.053	<0.025	*	BDL-0.0677	<0.00010-0.0170	-
Benzo(g,h,i)perylene	<0.110	<5	<0.036	<0.025-0.030	BDL-0.01	BDL-0.0926	<0.0011-0.0442	-
Benzo(k)fluoranthene	<0.017-0.050	<2.5	<0.036	<0.025	*	BDL-0.0555	<0.00020-0.0064	-
Chrysene	<0.033	<5	0.249	<0.025	BDL	BDL-0.1938	<0.00040	BDL-0.28
Dibenzo(a,h)anthracene	<0.068	<2.5	<0.036	<0.025	BDL	BDL-***0.0952***	<0.00010	-
Fluoranthene	<0.017	<5	<0.036	0.050-0.060	0.07-0.20	BDL-2.9548	<0.00070-0.3210	BDL-0.120
Fluorene	<0.015	<1	0.057	0.030-0.040	BDL-0.01	BDL-0.0615	-	BDL-0.260
Indeno(1,2,3-cd)pyrene	<0.022	<1	<0.053	<0.025	BDL	BDL	-	-
Naphthalene	<0.13-***0.49***	<1	***3.92***	<0.100	0.08-***0.50***	BDL-***0.1366***	<0.080	-
Phenanthrene	<0.13	<2.5	0.012	0.250-0.300	0.04-0.50	BDL-0.9697	<0.0016-0.0508	-
Pyrene	<0.060	<0.5	0.062	<0.025	0.06-0.09	BDL-1.1956	<0.00080-0.1358	BDL-0.0960

[a]Not reported. TCLP fluid (unspecified whether #1 or #2) used as extractant.
[b]Adapted from previous work by Larsson (1998).
[c]Material was leached thrice, each time at a LS of 10 with extractant tested each leaching cycle. Results present maximum leached during these cycles.
[d]Four groups of recovered HMA were tested but two of these were from the same road (one for surface course, one for base course).
[e]pH of extraction solution before addition of RAP (not measured after leaching).
*Does not report these values but reports benzo(*bjk*)fluoranthene (BDL-0.04 μg/L).

the highest concentrations of benzo(*a*)anthracene, benzo(*bjk*)fluoranthene, and indeno(1,2,3-*cd*)pyrene in the older (service station) wearing course specimen.

Shedivy et al. (2012) collected RAP from five different states (five sources, n = 5) across the US (Ohio, Wisconsin, California, New Jersey, Colorado) in addition to one new asphalt pavement material that was from a local source (Wisconsin). Batch tests were performed on size-reduced material (<19.1 mm) using the TCLP protocol (fluid #1) and deionized (DI) water at an LS of 20. RAP from one source (New Jersey) was also subjected to additional LS ratios (5, 10, 15). The authors reported higher leached metal concentrations using TCLP compared to DI water, noting no differences for leached PAH concentrations, which were slightly above or below the detection limit. PAH leaching did increase with increasing LS for the New Jersey RAP source (the only source where LS was varied) for the following constituents: acenaphthalene, benzo(*a*)anthracene, benzo(*b*)fluoranthene, benzo(*a*)pyrene, benzo(*ghi*)perylene.

Townsend et al. (2013) addressed the need for lower PAH detection limits from their earlier study (Brantley and Townsend, 1999) and conducted batch (SPLP) and column tests (saturated and unsaturated conditions) on five RAP samples (n = 5) collected throughout Florida, US. Out of the five RAP sources included in this study, only one of them leached PAHs during batch testing above detection limits (see Table S5) ranging from 6.4 ng/L (benzo(*k*)fluoranthene) to 321 ng/L (fluoranthene). Column testing under unsaturated conditions showed fluoranthene, pyrene, benzo(*k*)fluoranthene, benzo(*g,h,i*)perylene, benzo(*a*)pyrene, and benzo(*b*)fluoranthene leaching out at measurable concentrations from at least two of the five RAP sources studied with these reduced to below their respective detection limits by LS 1.2. PAHs were detected in 4 of the 5 RAP sources during saturated column testing (see Table S6), which either were below the detection limit by the second leaching event (LS = 0.8) or did not show any meaningful changes in concentration throughout the experiment.

Aydilek et al. (2017) analyzed metals leachability from seven RAP sources (n = 7) collected from highways throughout Maryland, US using both batch and column experiments (the column dataset also published in Mijic et al. (2020)). Batch tests were performed on size-reduced material (<2 mm) using DI water, and leached metals were consistent between individual sources, although elevated concentrations of copper were detected in two sources. Higher leached concentrations were observed in the source with greater *total* copper concentration and lower asphalt binder content. Column specimens were prepared by compacting RAP at 2% dry of their optimum dry density to a height of 0.155 m with solution pH stabilized between 6.0 and 6.5. Boron, copper, manganese, nickel, and zinc concentrations rapidly decreased within the first few pore volumes of flow (LS) for all RAP samples, which was attributed to the release of their water-soluble forms, exhibiting a "wash-off" effect. Like the batch tests, elevated copper and zinc concentrations were observed in two RAP samples initially but decreased with increasing water infiltration. In comparison, the authors reported considerable leaching from control aggregates (natural stone) for arsenic, copper, chromium, and zinc. The authors attributed this to the acidic (pH 5.66) and basic (pH 8.82) effluents generated by these materials that increased the solubility of these trace metals in solution.

Mehta et al. (2017) collected from three RAP sources (n = 3) along with one sample of freshly made hot mix asphalt pavement in New Jersey, US. Batch (TCLP) and column leaching tests (artificial rainwater, pH ~5) were performed to assess the mobility of PAHs and metals, with the metals dataset also published in Yang et al. (2020). For batch tests,

Table 5a
Summary of column tests: results of metals (μg/L) leaching from RAP samples with a range of detected concentrations reported. Dashes indicate unmeasured constituents, where BDL is listed designates a lack of detection limit provided. Shaded constituents and their corresponding values represent exceedances in US EPA RSLs (2020).

Study	Brantley & Townsend, 1999 (Townsend, 1998)	Legret et al., 2005	Roque et al., 2016[d]	Yang et al., 2020 (Mehta et al., 2017)	Mijic et al., 2020 (Aydilek et al., 2017)
Extractant Used/Method	SPLP	DI	n/a	Artificial Rainwater	n/a
pH	4.2[a]	7.1-8.4	7.35-7.66	~5[a]	7.53-8.50
LS	Varied[b]	0.5-30:1[c]	5:1	n/a	Varied[g]
Number of Sources	6	1	1[e]	1[f]	7
Constituents (μg/L)					
Aluminum	-	-	-	6.22-128.35	<25-320
Arsenic	-	-	-	0.11-5.18	<25
Antimony	-	-	-	0.03-0.56	-
Barium	<500	-	-	33.65-233.87	14.2-182
Boron	-	-	-	-	31.6-608
Cadmium	<5	-	0.6	0.01-0.04	<2
Chromium	<100	-	6	0.14-0.71	<25
Copper	<500	<5-13	1.8	0.49-62.70	<5-16.1
Iron	-	-	-	0.35-36.51	<25-224
Lead	<10-***38.3***	<5	1	0.01-0.12	<25
Manganese	-	-	-	0.09-1.84	<5-426
Mercury	-	<0.1-0.3	-	-	-
Molybdenum	-	<5	-	0.67-6.70	-
Nickel	<100	-	3	0.06-1.90	<5-108
Selenium	-	-	-	0.10-0.82	-
Silver	-	-	-	0.01-0.04	-
Thallium	-	-	-	0.00	-
Zinc	<500	<10-71	2	0.92-11.64	23-213

[a]Reported before adding RAP material (not presented after leaching).
[b]Leachate samples were collected intervals based on days (40–42, maximum).
[c]Leachate samples were collected at intervals and ranges observed during these are reported.
[d]Originally reported in mg/kg-dry at a LS of 5, which was used to convert to μg/L. These reflect total cumulative constituents leached at this LS.
[e]Compared to this study's batch testing, only one source (crushed RAP) was presented.
[f]One source was tested (NORTHRAP) but two treatments from this group were subjected to columns (untreated and UV-treated).
[g]Measured in pore volumes.

one of the RAP sources exhibited elevated lead concentrations, which the authors attributed to the historical use of tetraethyl lead and white roadway paint. This source also leached the highest concentrations for water-soluble PAHs (these were near or below detection in other RAP sources). Elevated concentrations of other metals (calcium, aluminum, iron, and manganese) were attributed by the authors to increased mineral dissolution under the acidic TCLP solution. The freshly mixed asphalt concrete exhibited lower concentrations for most metals and PAHs than RAP, which the authors suggest is due to exposure to road materials, vehicle emissions, and dust deposition. Column tests on these RAP sources showed that metals and PAHs were detected at much lower concentrations than their batch counterparts, which was attributed to the differences in extraction solution, contact time, and LS.

Other studies not focused on RAP, but using RAP as a comparison or control, provide some additional RAP leaching data; such studies include those evaluating coal fly ash (Kang et al., 2011; Hoy et al., 2016), natural and other construction/demolition debris-derived aggregates (Arulrajah et al., 2013; Roque et al., 2016), and WTE ash (Schafer et al., 2019). An in-depth discussion on RAP leaching in these studies is limited, but they generally conclude that RAP does not release metals at elevated concentrations compared to the other materials studied. Relevant RAP data from these studies are presented in Tables 4a, 4b and 5a, 5b.

3.2. Field measurements from RAP stockpiles

Three studies report trace constituent concentrations of leachate generated from stockpiled RAP under field conditions. One study (Sadecki et al., 1996) measured concentrations of both stormwater runoff and leachate while the other two (Norin and Strömvall, 2004; Licbinsky et al., 2012) examined just the leachate (see Table S7 for these concentrations).

Sadecki et al. (1996) examined a RAP stockpile in Minnesota, US, placed on a graded, impermeable membrane using a sampling and flow monitoring system with dataloggers and automatic sequence samplers. Two other stockpiles consisting of RCA (<19 mm, <4.75 mm) were also included in this study (PAHs were not examined in the leachate from the RCA stockpiles). Metals were occasionally observed from the RAP leachate above detection limits but were within the range of concentrations measured from the two RCA stockpiles; the trend for metals concentrations was to decrease over time. PAHs leached from the RAP stockpile were reported close to or below detection limits (for the time).

In Sweden, Norin and Strömvall (2004) assembled field-scale RAP stockpiles and sampled from them at a dedicated test site (in comparison to their lab-based experiments, see Tables 4a, 4b and 5a, 5b). Their analysis was limited to organic constituents, including PAHs. Leachate samples were collected from areas where LS was lowest (center of stockpile) and from areas where LS was highest (near outside edges of stockpile). Two stockpiles representing different methods of processing RAP were also studied: "scarified" RAP from the top 3 cm of a highway and "dug" RAP, which represented the top 10 cm of the same highway (i.e., same source, n = 1). Leachate samples from the stockpiles identified naphthalene in 8 of 27 samples, two of these with concentrations exceeding 10 μg/L (concentrations below this threshold were not reported by the authors).

In the Czech Republic, Licbinsky et al. (2012) portioned off one part of an active stockpile and buried two samplers within to collect rainwater infiltrate for two sampling events (after 60, 135 days) to measure

Table 5b
Summary of column tests: results of PAH leaching (μg/L) from RAP samples with a range of detected concentrations reported. Dashes indicate unmeasured constituents, where BDL is listed designates a lack of detection limit provided.

Study	Brantley & Townsend, 1999 (Townsend, 1998)	Legret et al., 2005	Norin & Strömvall, 2004[d]		Birgisdóttir et al., 2007	Townsend et al., 2013	Yang et al., 2020 (Mehta et al., 2017)
			Not Stored	Stored			
Extractant Used/Method	SPLP	DI	Acidified Water		DI	SPLP	Artificial Rainwater
pH	4.2[a]	7.1-8.4	4.5	4	8[a]	4.2[a]	~5[a]
LS ratio	Varied[b]	0.5:1-30:1[c]	0.05	0.07		0.4:1[h]	n/a
Number of Sources	6	1	1		2[e]	5	1[g]
Constituents (μg/L)							
Acenaphthene	<5	<0.05	3.0	0.7	<0.007-0.070	<0.014	BDL-0.0878
Acenaphthylene	<5	<0.05	0.5	0.4	<0.030	<3.5	-
Anthracene	<5	<0.025	0.5	0.1	<0.007	<0.010	-
Benzo(a)anthracene	<5	<0.025	<0.01	<0.01	<0.007-***0.180***	<0.0030-0.0086	BDL-0.00103
Benzo(a)pyrene	<0.25	<0.010-0.020	<0.01	<0.01	<0.050	<0.00030	-
Benzo(b)fluoranthene	<1	<0.025-0.025	<0.01	<0.01	*	<0.00010-0.0091	-
Benzo(g,h,i)perylene	<5	<0.025-0.080	<0.01	<0.01	<0.050	<0.0011-0.0910	-
Benzo(k)fluoranthene	<2.5	<0.025	<0.01	<0.01	*	<0.00020-0.0013	-
Chrysene	<5	<0.025-0.045	<0.01	<0.01	0	<0.00040	BDL-0.00251
Dibenzo(a,h)anthracene	<2.5	<0.025-***0.055***	***0.04***	***0.20***	<0.024-***<0.050***[f]	<0.00010	-
Fluoranthene	<5	<0.025	0.1	0.1	0.007-0.078	<0.00070	BDL-0.0162
Fluorene	<1	<0.025	2.1	0.5	<0.030	-	BDL-0.0282
Indeno(1,2,3-cd)pyrene	<1	<0.025-0.050	0.02	0.04	<0.011-0.200	-	-
Naphthalene	<1	<0.100	***28***	***9.2***	0.013-***0.320***	<0.080	-
Phenanthrene	<2.5	<0.025	1.8	0.7	0.012-0.120	<0.0016	-
Pyrene	<0.5	<0.025	0.1	0.1	<0.007-0.054	<0.00080-0.0382	BDL-0.0194

[a]Reported before adding RAP material.
[b]Leachate samples were collected intervals based on days (40–42, maximum).
[c]Leachate samples were collected at intervals and ranges observed during these LS are reported.
[d]Column studies were performed on same stockpiled material at start of study ("not stored") and after a two-year storing period ("stored").
[e]Four groups of recovered HMA were tested but two of these were from the same road (one for surface course, one for base course).
[f]This range includes a value of 0.043 μg/L that was observed, which exceeds the US EPA RSL (2020) for this constituent.
[g]One source was tested (NORTHRAP) but two treatments from this group were subjected to columns (untreated, and UV-treated).
[h]Only LS tabulated.
*Does not report these values but reports benzo(*bjk*)fluoranthene (0.007–0.830 μg/L).

leached metals and PAHs. The corresponding leachate LS was estimated as <1 due to the low volume of leachate collected (<1 L, per event) and the estimated volume of RAP above the collection vessels (0.10048 m^3). The low volume of leachate was attributed to the shape of the stockpile (conical) and low water retention due to low hydraulic conductivity and high evaporation rate. Concentrations of certain metals (copper, molybdenum, lead, vanadium, cadmium) were similar between the first and second sampling events, while others were higher in the first sampling event (barium, cobalt, chromium, zinc), suggesting that these were present on the surface of the RAP particles and "washed out" during this initial contact with rainwater; this trend was also observed for PAHs (except for benzo(*a*)pyrene which was approximately constant). However, some metals (manganese, nickel, antimony) were detected higher in the second sampling event compared to the first.

4. Risk assessment analysis

The literature provides evidence that trace chemical constituents such as metals and PAH compounds exist in RAP, both from the primary asphalt pavement ingredients (aggregate, asphalt binder) and from external sources such as vehicle emissions and wear, and that upon exposure to water, some of these chemicals may leach. Most authors reported that external contributions were likely the dominant source of these trace chemicals, as asphalt binder or newly prepared asphalt pavement has been observed to leach less than some reported RAP samples studied (Brandt and de Groot, 2001; Kriech et al., 2002, 2005; Shedivy et al., 2012; Mehta et al., 2017). RAP from older pavements has leached constituents to a greater extent than newer RAP (Brantley and Townsend, 1999; Birgisdóttir et al., 2007). The presence of trace chemicals in RAP, or the leaching of these chemicals into water, does not by itself indicate that these chemicals pose a risk to human health and the environment. An assessment of risk takes into consideration both the toxicity of the chemical to the receptor of concern (e.g., humans) and the dose of that chemical to which the receptor is exposed (see Fig. S2 for a visual illustration). In this section, we examine the risk posed by stockpiled or unencapsulated RAP by examining the conclusions of various studies and by assessing reported data through the lens of a typical risk assessment approach used for solid wastes similar to RAP.

4.1. Direct exposure risk

A common approach to assessing the potential risk posed by a waste or construction byproduct such as RAP involves examining two pathways: direct human exposure and contamination of groundwater. The direct exposure pathway typically involves scenarios where a material is used in a manner similar to soil and has the potential for human contact through dermal exposure (transfer of chemicals from the material through the skin), inhalation (exposure through particles entering the respiratory tract), and ingestion (hand to mouth contact). In the context of RAP reuse/storage, the direct exposure pathway was not cited as a primary concern in any of the reviewed studies. A plausible explanation for this observation is that direct exposure to RAP is relatively limited to

human receptors (aside from occupational workers) as a risk pathway under normal storage (e.g., aggregate stockyards) and reuse (e.g., bound in the pavement, under pavement/cover soil) conditions.

While the use profile of RAP merits against direct exposure as a significant pathway, for context, we conducted a screening level direct exposure assessment by comparing the total trace chemical concentrations (mg/kg-dry) available to several different risk-based direct exposure clean soil thresholds (see Tables S1 and S8 for direct exposure risk thresholds developed by Florida, US and US EPA for PAHs and metals, respectively). Differences between direct exposure thresholds provided by states or other agencies such as the US EPA are due to the different assumptions used to derive these values, such as exposure over time (e.g., residential vs. workplace), a risk to groundwater, and exposure to vulnerable populations (e.g., children, elderly) (see Table S4 for comparisons by state/agency).

Table S8 compares the metal concentrations from RAP to US EPA and Florida direct exposure risk thresholds showing occasional exceedances in Florida soil cleanup target levels (SCTLs). Compiling the data from various studies showed some RAP to exceed residential SCTLs for arsenic, barium, and manganese with exceedances for commercial soil RSL for arsenic. It is necessary to put these values in context to what may be encountered in the surrounding environment, such as soils and aggregates where metal contents vary based on local geology (Chirenje et al., 2003; Smith et al., 2014; Welch et al., 2016; Missimer et al., 2018; Schafer et al., 2019) (see Table 2 for typical aggregate metal contents). In the case of arsenic, soils throughout the US commonly have average concentrations that exceed residential and commercial US EPA RSLs (Chirenje et al., 2003; Smith et al., 2014; Welch et al., 2016; Missimer et al., 2018), while limerock and RCA, two commonly used aggregates, have exceeded the US EPA commercial RSL (3.0 mg/kg-dry) for As and the residential FL SCTL (2.1 mg/kg-dry) (see Table 2).

Table 3 (and Table S1) compare BaP equivalent concentrations for RAP to US EPA and Florida SCTLs and show exceedances in residential thresholds (aside for one specimen from Birgisdóttir et al. (2007)) while being below commercial thresholds aside for one source observed in Su et al. (2019) (15.87 mg/kg-dry). RAP samples occasionally exceeded residential soil-based risk levels for individual PAHs too: benzo(*a*)anthracene, benzo(*a*)pyrene, benzo(*b*)fluoranthene, and dibenzo(*a*,*h*)anthracene (Table S1). For comparison, these RAP concentrations are within ranges previously reported for soils surrounding nearby high-volume traffic roadways and commercial zones in cities. BaP equivalents in urban soils have been reported as high as 9.4 mg/kg-dry (Liu et al., 2019b), exceeding residential and commercial FL SCTLs. Individually, benzo(*a*)anthracene, benzo(*a*)pyrene, benzo(*b*)fluoranthene, and dibenzo(*a*,*h*)anthracene have been reported in urban soils as high as 1.5 mg/kg-dry, 0.5 mg/kg-dry, 1.19 mg/kg-dry, and 0.6 mg/kg-dry, respectively, surpassing their FL SCTLs (Agarwal, 2009; Peng et al., 2011; Wang et al., 2013).

4.2. RAP leachate concentrations compared to risk-based thresholds in literature

Authors of the different RAP leaching studies through various means use leached concentrations to assess the risk of stockpiled or unencapsulated RAP on water supplies; the typical approach used is to compare concentrations in the leachates to regulatory or risk thresholds. Therefore, the subsequent conclusions drawn from each study are a function of the thresholds by which the researchers chose to compare.

Kriech (1991) concluded that RAP could be used as clean fill based on TCLP results from 6 RAP samples; this decision was based, however, on US hazardous waste criteria not typically used for beneficial use of waste materials. Brantley and Townsend (1999) utilized water-based leaching protocols on 6 FL RAP samples and found trace chemical to not leach at concentrations greater than typical groundwater standard (FL groundwater cleanup target levels (GCTLs)); the leached lead measured in three samples was not considered of notable risk because it followed a "wash-off" leaching pattern, where soluble lead sharply decreased to below detection in subsequent LS sampling events. PAH compounds were not measured above their detection limits, but these detection limits were higher in the 1999 study compared to current (2020) FL GCTL risk-thresholds (see Table S4). Townsend et al. (2013) repeated this methodology on 5 new FL RAP sources using much lower PAH detection limits. They found that only one of these sources leached PAH above the detection limit in batch tests (slightly exceeding RSL for benzo(*a*)anthracene, 0.0349 μg/L); however, all samples leached PAHs above the detection limit during column testing, but measurements remained below FL GCTLs.

In Europe, Norin and Strömvall (2004) reported that total PAH concentrations in the leachates from both the column and field-scale RAP stockpiles exceeded Swedish standards for groundwater in polluted soils at gas stations; no direct comparison to individual PAH thresholds was made. Legret et al. (2005) concluded that leached PAHs from their RAP source were below limit values for drinking water and posed a limited risk, although Dutch target values for groundwater (50 μg/L) were exceeded for total leached hydrocarbons (120 μg/L). Birgisdóttir et al. (2007) did not make any direct comparisons of leached concentrations from RAP to groundwater or drinking water standards but estimated the impact of its PAH flux (leaching) on nearby soil contents over 25 years of simulated leaching. They concluded that the leached concentrations of PAHs from RAP would be unlikely to cause nearby soils to exceed Danish soil quality criteria.

Shedivy et al. (2012) concluded that leached PAHs from the 5 RAP sources did not pose environmental risks based on comparison to Wisconsin groundwater limits (see Table S4). Aydilek et al. (2017) reported that besides some initial elevated copper and zinc concentrations in column tests for two RAP sources, metals were well below US EPA Water Quality Limits (WQL). Mehta et al. (2017) suggest that leached metals from unbound RAP would be largely attenuated in the surrounding soil permitting its use in most environments besides those with acidic conditions (pH < 5).

The different risk-based thresholds, leaching tests, and risk assessment approach employed in the various studies make a comparison of conclusions a challenge; thus, we performed a preliminary screening approach by taking the maximum concentration measured for every RAP sample (regardless of methodology) and compared it to the US EPA RSLs for drinking water (2020) shading concentrations in Tables 4a, 4b and 5a, 5b where those values are greater than their RSL. Out of 41 sources studied, RAP leachate exceeded metal risk-based thresholds for arsenic (7 sources), lead (13 sources), antimony (8 sources), and manganese (5 sources). PAHs, out of 29 RAP sources examined, were reported above RSLs for naphthalene (6 sources), dibenz(*a*,*h*)anthracene (6 sources), benzo(*a*)pyrene (4 sources), and benzo(*a*)anthracene (5 sources).

Several of these observed exceedances (Kriech, 1991; Shedivy et al., 2012; Arulrajah et al., 2013; Mehta et al., 2017) were reported from samples using TCLP and not solutions reflective of typical rainwater infiltration that most stockpiles and reuse scenarios would experience. Neglecting these values would result in manganese no longer exceeding its RSL and arsenic and lead exceeding for 5 (down from 7) and 10 (down from 13) sources, respectively (antimony would be unchanged). For PAHs, this would result in naphthalene exceeding for 3 sources (down from 6), dibenz(*a*,*h*)anthracene exceeding for 5 sources (down from 6), and benzo(*a*)anthracene exceeding for 4 sources (down from 5) (benzo(*a*)pyrene would be unchanged). Several studies (Norin and Strömvall, 2004; Birgisdóttir et al., 2007; Mehta et al., 2017) also greatly size reduced the material beyond that required for testing such that it is not in a form that would exist under stockpile/reuse conditions (i.e., <2 mm). Neglecting results produced from methods that are not nonrepresentative of typical reuse/stockpiling conditions reduce exceedances for naphthalene to 1 source (as reported by Birgisdóttir et al., 2007) and benzo(*a*)pyrene to 3 sources (all from Shedivy et al., 2012) (number of sources exceeding for dibenz(*a*,*h*)anthracene and benzo(*a*)anthracene would remain unchanged). In total, this would

result in 18 sources being reported exceeding an RSL from 30 sources originally. It is important to note that this count does not consider if individual sources may exceed for more than one constituent.

Conventionally used aggregates and binders have been reported to leach some metals and PAHs in concentrations that may exceed certain risk-based thresholds and are within the range or within an order of magnitude of those measured from RAP. The maximum antimony concentration reported from RAP was 17.2 μg/L (see Tables 4a, 4b), and Morse et al. (2001) observed antimony concentrations within the same order of magnitude ranging from 6.26 μg/L (sandstone) to 13.0 μg/L (siliceous sand). Morse et al. (2001) and Schafer et al. (2019) both reported lead concentrations of 15.9 μg/L (limerock) and 15 μg/L (RCA), respectively, when leached using SPLP solution. These leached concentrations are within the range of values observed from RAP under SPLP solution (0.64–38 μg/L; see Tables 4a, 4b and 5a, 5b). Kriech et al. (2005) observed antimony and lead leaching from asphalt binders occasionally above their detection limit (5.0 μg/L), among other metals (Table S9). For naphthalene, leachate from asphalt binder has been reported as high as 0.37 μg/L (Brandt and de Groot, 2001), which is in range or similar order of magnitude of several maximum reported values measured from RAP leachates from Kriech (1991) (0.49 μg/L), Birgisdóttir et al. (2007) (0.50 μg/L), and Shedivy et al. (2012) (0.14 μg/L) (see Tables 4a, 4b and 5a, 5b). Reported concentrations of leached benz(*a*)anthracene and dibenz(*a*,*h*)anthracene from binder have ranged from 0.0001–0.0005 μg/L and 0.0001–0.0003 μg/L, respectively (Brandt and de Groot, 2001; see Table S10). These concentrations are less than maximum concentrations reported for benz(*a*)anthracene and dibenz(*a*,*h*)anthracene from RAP in individual studies (0.03–0.180 μg/L and 0.050–0.20 μg/L, respectively; see Tables 4a, 4b and 5a, 5b), suggesting that these PAHs leach to a lesser extent from asphalt binder than from RAP.

One deficiency in this screening level approach is that leachate concentrations are compared directly to risk-thresholds based on ingesting, or drinking, the leachate. Robust risk assessments use leaching data in addition to site-specific data (assumed and/or provided) to predict concentrations of chemicals at actual points of exposure, and these concentrations are then compared to the risk thresholds (Blaisi et al., 2019).

4.3. IWEM risk assessment modeling

A direct comparison of a constituent concentration measured using a leach test to a risk-based standard or threshold allows one to screen for chemicals of possible concern, but this approach does not take into account the potential dilution and or attenuation a trace chemical will undergo between the point of release (e.g., the base of a RAP stockpile) and the point of exposure (e.g., potable water well). The expected concentration occurring at the location of concern can be predicted using fate and transport models, with model inputs including the concentration at the source and characteristics of the underlying soil and aquifer. Fig. S2 illustrates the modeled RAP exposure pathway and various parameters by which this exposure is dependent upon.

For this study, we utilized the US EPA's Industrial Waste Management Evaluation Model (IWEM), a tool often used in US beneficial use determinations (Li et al., 2010; Park et al., 2012), to estimate the concentration of a leached constituent at distances (10, 50, and 100 m) from a RAP stockpile. Site-specific parameters such as hydrogeologic conditions (e.g., soil/aquifer characteristics) along with material-specific properties (e.g., stockpile shape, hydraulic conductivity, constituent concentrations) were used in the model to calculate predicted constituent concentrations (the 90th percentile concentration) (see Table S11 for parameters, values, and assumptions). For this exercise, US national averages for subsurface conditions were chosen by defaults in the software, and coarse soil was selected as the soil type to provide a conservative approach to risk assessment due to its higher hydraulic conductivity (US EPA, 2015a, 2015b). The US national rainfall average was chosen as the infiltration input at a rate of 0.767 m/yr (NOAA, 2020). IWEM assumes that this entire infiltration rate will penetrate through the stockpile, which is not necessarily accurate under real-world scenarios due to different field conditions (e.g., evapotranspiration rates) and properties of the RAP stockpile itself (e.g., hydraulic conductivity). To estimate fate and transport as a function of rainwater infiltration, this rate was modeled as 10, 25, 50, and 100% of its original value. Modeled stockpile parameters (i.e., size dimensions, bulk density, hydraulic conductivity) were chosen based on those reported previously in the literature (Kandhal and Mallick, 1998; Lavin, 2003; Chesner et al., 1998; Copeland, 2011; Nassar and Nassar, 2006; Zhou et al., 2010; Aydilek et al., 2017).

Figs. 1 and 2 present the IWEM modeling results (numerical values presented in Table S12) for the chemical constituents in Tables 4a, 4b and 5a, 5b that exceed their respective US EPA RSL (metals: arsenic, lead, antimony, manganese; PAHs: naphthalene, dibenz(*a*,*h*)anthracene, benz(*a*)anthracene, benzo(*a*)pyrene). For studies with multiple reported concentrations exceeding RSLs, the maximum value was plotted. The figures present the modeled dilution attenuation factor (DAF) as a function of stockpile infiltration (%) (the DAF represents the ratio of source concentration to receptor concentration) and, as appropriate, include curves for multiple receptor distances (10, 50, 100 m). The horizontal lines added to the figures represent the DAF necessary for the maximum concentration of constituent measured in any of the historic leaching studies below which the US EPA RSL is met.

As expected, as the distance to the receptor increases and the infiltration rate decreases, higher DAFs are achieved. For metals, all reported concentrations are diluted/attenuated to below their respective US EPA RSLs at the most conservative modeled compliance point (10 m) and at all infiltration rates (10–100%) except for the Kriech (1991) lead concentration of 1800 μg/L. This concentration requires a DAF of 120 to reach beneath the US EPA RSL (15 μg/L). As mentioned earlier, this represents one source from one study, which used a TCLP solution to extract the leachate. Acidic landfill leachate conditions, as simulated with TCLP, are unlikely in stockpiled conditions where exposure to rainwater is the likely source of infiltration.

Similarly, all reported concentrations for the PAHs dibenzo(*a*,*h*)anthracene, benzo(*a*)anthracene, and benzo(*a*)pyrene were diluted/attenuated below their US EPA RSLs at all modeled conditions. Naphthalene concentrations also achieved sufficient DAFs at the 10 m simulation at all infiltration rates except for the maximum value reported in Norin and Strömvall (2004) of 28 μg/L. Sufficient DAFs were only achieved at lower infiltration rates (<40% for 100 m) and with increasing distances (≥50 m) from the compliance point. This finding is likely due to naphthalene's relatively high solubility in water in comparison to other PAHs (Abdel-Shafy and Mansour, 2016). Generally, PAHs with lower molecular weight are more mobile in the aqueous phase compared to heavier species (Simon and Sobieraj, 2006; Haritash and Kaushik, 2009; Abdel-Shafy and Mansour, 2016; Baldwin et al., 2017). Heavier PAHs, such as dibenzo(*a*,*h*)anthracene, benzo(*a*)anthracene, and benzo(*a*)pyrene, are more prone to becoming bound in soil or particles than a light PAH such as naphthalene (Simon and Sobieraj, 2006).

Under simulated parameters that reflect typical environmental conditions, the IWEM exercise presented in this section suggests that most reported constituent concentrations from RAP that exceeded US EPA RSLs for drinking water (2020) could achieve sufficient DAFs. Most constituents from these studies needed DAFs <10, which was attainable under the most conservative IWEM scenario modeled (10 m distance, 100% infiltration rate). The elevated naphthalene concentration in Norin and Strömvall (2004) was partly attributed to surface deposition (i.e., gasoline), as its leaching from virgin asphalt binders in the literature was observed at orders of magnitude below this concentration (Brandt and de Groot, 2001; Kriech et al., 2002). This source would likely be quickly depleted under percolation conditions exhibiting a "wash-off" phenomenon.

One limitation of IWEM is that the leachate concentrations cannot be simulated to change over time, meaning that "wash-off" and continuous leaching cannot be modeled simultaneously. Using the "wash-off"

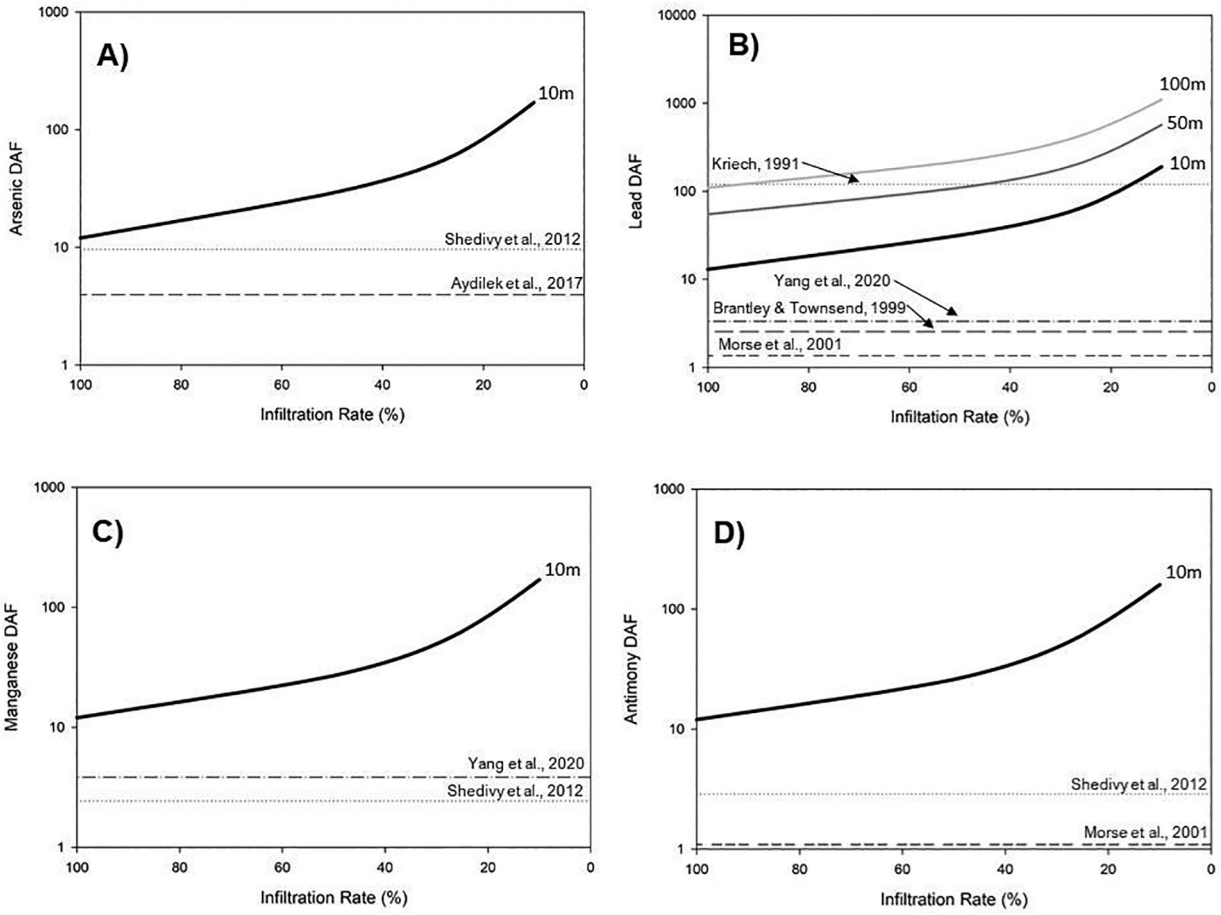

Fig. 1. IWEM modeling DAF attained compared to those needed for identified leached metals of potential concern from each study's concentration to be diluted/attenuated below US EPA RSL values for drinking water for A) arsenic, B) lead, C) manganese, D) antimony as a function of infiltration rate (%) and distance to the receptor (m).

concentration may not provide an appropriate estimate of risk over long-term conditions. A more accurate model would allow leachate concentration inputs to vary as a function of infiltration (or LS). Models are also limited by the availability of material and site-specific data. As shown in this exercise, parameters such as stockpile infiltration rate can influence potential environmental risks. RAP stockpiles can vary in particle size distribution and form a crust-like layer during storage, two factors that change its hydraulic conductivity, and hence rainwater infiltration, over time (Copeland, 2011). Modeling software packages such as IWEM can also contain built-in assumptions and nested models for predicting certain material properties (or changes within them over time) which can vary conclusions depending on the program used. For example, results highly dependent on material-specific inputs such as permeability can be affected by the theoretical framework or assumptions used to model the parameter (Fazelabdolabadi and Golestan, 2020).

5. Summary and future needs

This review examined 17 studies (41 RAP sources total) reporting leaching from RAP (14 of these for metals, 9 of these for PAHs). The leaching pathway was extensively focused on in this study as direct exposure was not deemed a significant pathway of concern in the literature, which the authors attributed to limited physical contact with RAP with human receptors (although these were briefly reviewed and were not found to exceed beyond roadside/urban soils and other conventionally used aggregates). Potential leaching risks cited by authors of the reviewed literature were limited aside from elevated naphthalene concentrations reported by Norin and Strömvall (2004). However, the different risk-based thresholds and leaching approaches (i.e., material preparation, leaching solutions) used in these studies made comparisons challenging. To evaluate potential environmental impacts, the authors compared reported leachate concentrations to US EPA RSLs for tapwater. The following constituents exceeded these limits: for metals, arsenic (7 RAP sources), lead (13 RAP sources), antimony (8 RAP sources), and manganese (5 RAP sources); for PAHs, naphthalene (6 RAP sources), dibenz(*a*,*h*)anthracene (6 RAP sources), benzo(*a*)anthracene (5 RAP sources), benzo(*a*)pyrene (4 RAP sources). Based on the reviewed literature and our risk assessment analysis, the stockpiling and reuse of RAP is unlikely to contaminate underlying or adjacent water supplies under conditions reflective of the modeling exercise performed in this study. However, data regarding the direct measurement of leachate from RAP stockpiles is limited (our review found three studies, of which one only reported the number of naphthalene exceedances above 10 μg/L in terms of PAH analysis). The use of long-term, in-situ groundwater monitoring near RAP stockpiles, embankments, or other structures (e.g., road bases) could be used to confirm similar trends and may be used to overcome challenges associated with modeling real-world scenarios.

Discussions from authors in the literature and direct comparison of leached concentrations from fresh asphalt mixtures and asphalt binder suggests that some elevated concentrations detected in RAP may be the result of external sources (e.g., traffic, sealants); however, material variability (e.g., aggregate and binder sources) can also be a plausible factor for some reported concentrations. A higher level of understanding of the relative impacts of each is necessary, as it may have lasting impacts on aggregate/asphalt products and road debris management, the latter

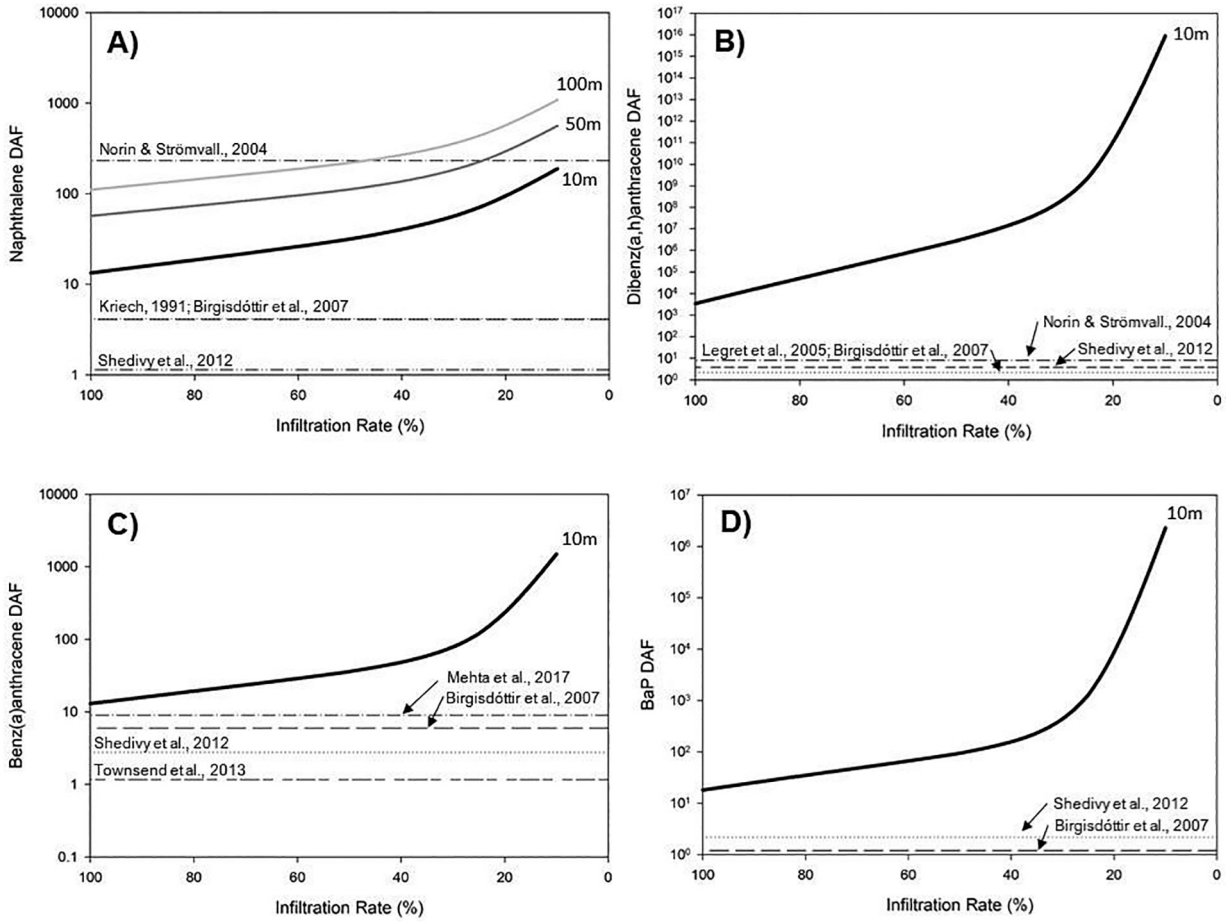

Fig. 2. IWEM modeling DAF attained compared to those needed for identified leached PAHs of potential concern from each study's concentration to be diluted/attenuated below US EPA RSL values for drinking water for A) naphthalene, B) dibenzo(*a*,*h*)anthracene, C) benzo(*a*)anthracene, D) benzo(*a*)pyrene (BaP) as a function of infiltration rate (%) and distance to the receptor (m).

being potentially relevant to other paving materials (e.g., portland cement concrete). Test method variables, such as particle size reduction and leaching solution, can also potentially influence results and interpretations of potential environmental risks. Drawing conclusions on the potential environmental risks from RAP should use results derived from methods that best reflect its state of storage/reuse (e.g., stockpile) and local environmental conditions (e.g., rainwater). A focus on collecting samples on a multi-incremental (e.g., per 1000 km milled/MTs processed) basis may also provide a better sense of chemical variability versus the discrete approach (e.g., singular stockpiles) used in the literature to date.

Risk quantification varies from study to study, with different risk-based thresholds used based on location and guidelines available at the time. Risk evaluation tools, like IWEM, can be used to provide estimates and evaluate what types of institutional and engineering controls may be necessary to control potential environmental risks to a reasonable level. It is important to understand that these models are developed using different assumptions and calculations, and provided estimates should be followed with field monitoring to make definitive conclusions. The use of IWEM in this study identified some limitations with simulating highly soluble ("wash-off") constituents that leach in the short-term and quickly deplete from RAP in the long-term, which could lead to overestimates of potential 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.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.145741.

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