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Leaching Potential of Metals and PAHs of Asphalt Rubber Paving Materials

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Abstract

Asphalt rubber (AR), a waste-based paving binder prepared with waste tyre rubber and asphalt binder, may contain hazardous components that pose environmental risks when applied to the road surface. This study aims to quantify this concern by characterizing the leaching process of AR using a multi-scale experimental method. The effects of various experimental variables on the leaching process were determined, and the leaching of metals and polycyclic aromatic hydrocarbons (PAHs) were characterized at three scales, i.e., binder, mortar, and mixture. The results suggested that AR presents no significant leaching concerns under most conditions, but the leaching of metals from AR might pose risks to the aquatic environments in the areas with acid rain. Furthermore, the leachability of metals increases with service time due to the aging

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of asphalt binder. The findings of this study can contribute to a better understanding of the environmental performance of AR and help conduct AR construction and maintenance in a more sustainable manner.

Keywords: Asphalt rubber, Leaching test, Experimental variables, Metals, PAHs

1. Introduction

The use of crumb rubber (CR) from waste tires in asphalt pavement production has received significant attention in recent decades as it provides an effective solution to the environmental problem caused by the disposal of end-of-life tires (Huang et al., 2007). Two methods, the wet and dry processes, have been employed to recycle waste tires in asphalt pavement (Zhou et al., 2014). In the dry process, crumb rubber replaces the fine aggregate in the asphalt mixture, while in the wet process, crumb rubber is added as a modifier to the hot asphalt binder to produce the modified binder that is then blended with aggregates (Airey et al., 2003). The wet process has been more commonly adopted in field applications due to its ability to produce asphalt mixtures with superior performance as compared to the dry process (López-Moro et al., 2013; Picado-Santos et al., 2020; Yu et al., 2018). The wet process can commonly lead to two types of modified binders: asphalt rubber (AR) and terminal blend (TB) binder. The AR binder contains relatively coarser rubber particles and higher rubber content (>15% by weight of virgin asphalt binder), while the TB binder uses fewer and finer rubber particles (typically <10% by weight of virgin asphalt binder). The AR binder is preferred in field applications due to its good rubber elasticity and proven resistance to rutting and cracking. However, the AR binder also has some limitations such as phase separation issues and relatively poor storage stability. In contrast, the TB binder has improved storage stability by using fewer and smaller rubber particles and increasing the mixing temperature and time of binder preparation, resulting in depolymerization and even dispersion of rubber particles within the binder. Nonetheless, the TB binder faces significant challenges before its widespread use, such as compromised deformation and fatigue resistance due to its lower viscosity and rubber elasticity (Han et al., 2016).

Despite the favorable rheological properties of AR, the recycled crumb rubber derived from waste tires contains impurities that pose a potential threat to the environment when AR is used as a paving material. A typical car tire consists of approximately 60% rubber, 30% carbon black, 6% extending oil, 3% zinc, 2% stearic acid, 1% sulfur, and a small amount of accelerators (Kunioka et al., 2014). Both natural rubber and synthetic rubber can be used in tire manufacturing, and the rubber formulation can differ depending on the source. Toxic pollutants that may be released from tire crumb rubber, including heavy metals such as zinc (Zn) and polycyclic aromatic hydrocarbons (PAHs), have the potential to leach from crumb rubber-modified asphalt pavements, leading to environmental pollution (Fathollahi et al., 2022; Rhodes et al., 2012). Heavy metals are generally non-biodegradable. Therefore, they can bioaccumulate in organisms after being taken into animal bodies. Some heavy metals are toxic when they are present in large amounts, causing biological and physiological complications (Gautam et al., 2016; Tchounwou et al., 2012). Exposure to toxic heavy metals could negatively affect the function of cellular organelles and components, including the mitochondria, nuclei, lysosomes, cell membrane, and enzymes, causing cell damage and cellular function loss. In addition, heavy metals present neurotoxicity risks by interacting with neurons and causing neurotransmitter inhibition. Moreover, metal ions could lead to DNA damage and nuclear protein inactivation, resulting in cell cycle modulation, apoptosis, or carcinogenesis. (Engwa et al., 2019; Tchounwou et al., 2012; Valko et al., 2005). Exposure to PAHs can cause many adverse effects, which may occur via food and water ingestion or dermal contact (Abdel-Shafy and Mansour, 2016; Rengarajan et al., 2015). Some PAHs are ecotoxic and display mutagenic, carcinogenic, teratogenic, or immunotoxic effects on living organisms (Bolden et al., 2017; Burchiel and Gao, 2014). PAHs tend to bioaccumulate in adipose tissues due to their high lipophilicity. Long-term exposure to PAHs may lead to tumor formation in organs such as the bladder, breast, colon, esophagus, lung, pancreas, and skin (Lee, 2010; Rajpara et al., 2017; Yu, 2002). PAHs also have embryotoxicity and are related to reproductive system abnormalities by affecting sperm quality, testicular function, egg viability, and damage nucleic acid (Abdel-Shafy and Mansour, 2016; Bolden et al., 2017; Rengarajan et al., 2015).

However, most of the previous studies have focused on improving the compatibility between crumb rubber and asphalt binder, as well as optimizing engineering performances of the produced AR (Leng et al., 2017; Li et al., 2021; Yu et al., 2017), while limited studies have been conducted to investigate the potential leaching properties of AR. The potential risks associated with the use of AR have not been adequately addressed, and the amount of chemical constituents that can leach out from AR remains unclear. Additionally, previous studies have not examined the influence of inevitable aging on the leaching properties of AR. Therefore, there is an urgent need for an in-depth investigation of the factors that influence the leaching of these chemicals and a fundamental understanding of the potential leaching risks of AR under real-world conditions. Currently, two laboratory test methods established by the United States Environmental Protection Agency (US EPA) are commonly used to investigate the leaching of contaminants from waste road materials, namely the toxicity characteristic leaching procedure (TCLP) and the synthetic precipitation leaching procedure (SPLP). However, these methods were primarily developed to assess the safety of potentially hazardous materials deposited in landfills and have been mainly applied to investigate the leaching risks from polluted soil and demolished concrete (Ai et al., 2019; Hu et al., 2020). To simulate an acidic environment at landfill sites, the pH value of the extraction solution is regulated at 4.2 by SPLP, and it can be as low as 2.88 if TCLP is adopted (US EPA, 1992; 1994). However, as asphalt pavement is rarely constructed in such low pH environments, it is crucial to determine the influence of experimental conditions on the leaching properties of asphalt mixtures to ensure an accurate interpretation of laboratory test results.

Therefore, this study aims to quantitatively characterize the leaching process of AR through the experimental methods reasonably representing the field condition. The effects of various experimental variables on the leaching process of AR, including the pH value of the extraction solution, extraction duration, liquid-to-solid ratio, and particle size, were first investigated. Then, based on a compressive analysis of these variables, a multi-scale experimental method was proposed to characterize the leaching behavior of the toxic metals and PAHs from AR at three scales, i.e., AR binder AR mortar, and compacted AR specimens. To better simulate the conditions of asphalt pavement in the field, AR samples were prepared through short-term and long-term aging processes. In addition, artificial acid rain and neutral water were employed as extraction solutions to investigate the leaching risk of AR under diverse climatic conditions.

2. Methodology

2.1. Test materials

In the first part of the study, AR mixtures were prepared using CR modified asphalt binder, natural aggregate, and mineral filler. The modified asphalt binder was produced by integrating 20% of CR (with a particle size of less than 0.25 mm) into a virgin asphalt binder with a penetration grade of 60/70. This mixture was then shear-mixed for one hour at 180 ºC. Subsequently, the modified asphalt binder and aggregates were blended at the temperature of 160-180 ºC. The mixture design implemented in this study was a stone mastic asphalt with a nominal maximum aggregate size of 10 mm (SMA10), which has been widely used as the surface course material for carriageway pavements in Hong Kong since early 2012 (Highways Department, 2016). The binder content was 6.0 wt%. The local granite rocks were employed as coarse and fine aggregates, as well as mineral fillers. The gradation of SMA10 is presented in **Fig. 1**.

In the second part of the study, the AR binder film was produced by subjecting CR modified binder to a heat treatment at 150 ºC inside a rotating bottle for 30 minutes. The loose AR mortar was generated by blending CR modified binder with fine aggregates (d< 2.36mm) at the temperature of 160-180 °C. The compacted AR cylindrical specimens were prepared by mixing CR modified binder, coarse, and fine aggregates at the temperature of 160-180 °C. The mixtures were subsequently compacted using a gyratory compactor to obtain an average air void content of 4±0.5%, which is the designed target air void content of the SMA10 gradation.

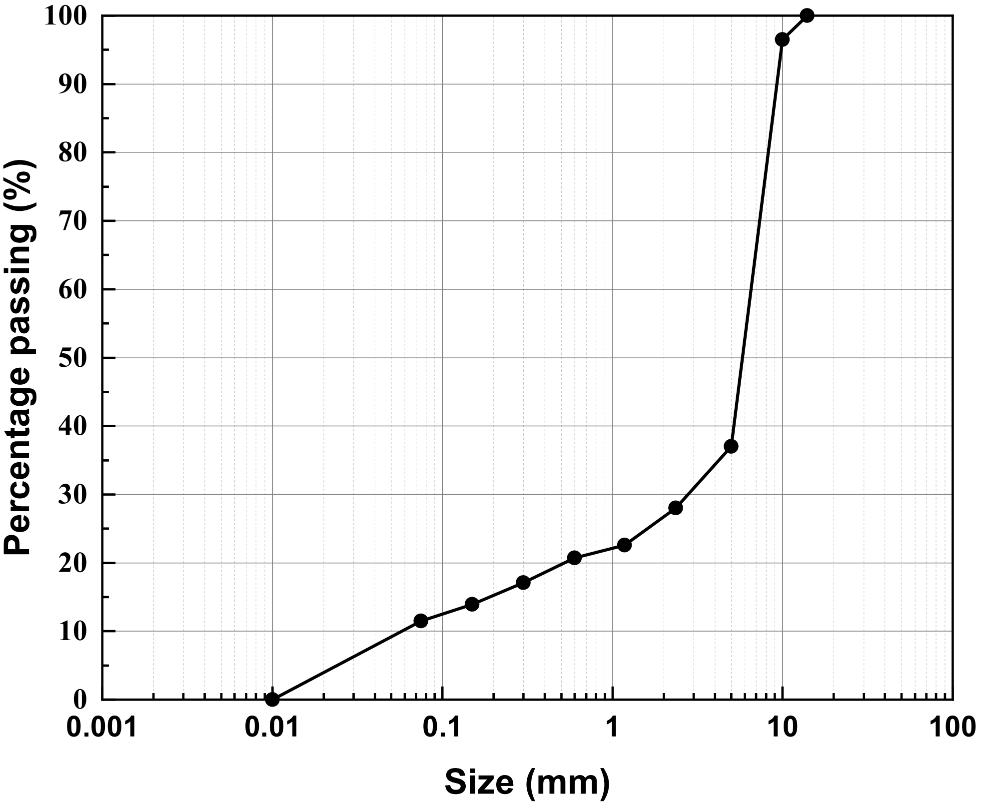

Fig. 1. The aggregate gradation curve of the SMA10 mixture

2.2. Experimental methods

2.2.1. Investigation of the effects of experimental variables

The effects of leaching test variables, including extraction time, liquid-to-solid (L/S) ratio, and extraction solution pH, were investigated in this study. The extraction time primarily determines whether the solid-liquid system has achieved phase equilibrium. During leaching, it is assumed there is sufficient solvent present so that all the solutes in the entering solid can be dissolved into the contacting liquid, and the equilibrium can be attained once the solute is dissolved. Therefore, it is imperative that the extraction time is sufficient to enable complete solute dissolution. The influence of L/S ratios is relatively straightforward, with higher metal concentrations found at lower L/S ratios. However, the L/S ratio should be high enough to ensure a sufficient bulk solution to stabilize the dissolved metals. The pH values of the extraction solution significantly impact the leaching process by altering the metal solubilities. To comprehend the influences of the experimental variables, the Taguchi method was applied (Rao et al., 2008). A subset of variable combinations was selected using the L16 orthogonal array from the Taguchi method.

The experimental design used in this study is presented in **Table 1**, which includes the orthogonal array and the parameters used. The Taguchi method is employed to identify the optimal conditions of the designed variables. This is achieved by evaluating the signal-to-noise (S/N) ratios (Pignatiello Jr, 1988). The S/N ratio is calculated using Equation (1):

$$\frac{S}{N}(x_j) = -10\log\left(\frac{1}{n}\sum_{i=1}^{n}\left({}^{1}/_{y_{ji}}\right)^{2}\right) \tag{1}$$

where x_j denotes the factor x at level j, y_{ji} represents the measured response of samples from the factor x at the j level, and n is the number of experimental runs.

In the situations where the response variable is non-negative, larger values of the S/N ratio are more desirable, and the combination of factors with the highest S/N ratio is considered optimal. The delta values of the S/N ratios are used to determine the significance of each factor. Delta values represent the mathematical range of S/N ratios between all levels for a given factor, which is obtained by calculating the difference between the maximum and minimum S/N ratios for that particular factor.

The extraction solution, which was designed to specific pH values, was prepared using acetic acid, sodium hydroxide, and deionized (DI) water. The test samples were weighed according to the predetermined L/S ratios and mixed with the extraction solution using an agitator at 30 rpm for the designated extraction time. Extraction vessels made of borosilicate glass were used due to their higher thermal conductivity and absence of potential metal contaminants, in comparison to other types of vessels such as high-density polyethylene (HDPE) and stainless steel bottles. Asphalt binder, a temperature-sensitive material, exhibits a decrease in viscosity as temperature increases. Consequently, asphalt binder behaves more like a liquid at higher temperatures and more like a solid at lower temperatures. Wang et al. (2012) reported that a change in temperature of only 15-20 °C can double or halve the viscosity of asphalt binder. Therefore, HDPE bottles, commonly utilized in TCLP and SPLP tests, are not suitable for the leaching tests of AR. The use of HDPE bottles impedes the release of heat generated by friction, which leads to an increase in solution temperature and ultimately, the agglomeration of asphalt mixtures. As a result of agglomeration, the particle size of test samples increases significantly, which largely affects the leaching tests (Ahlberg et al., 2006; Nosrati et al., 2014; Yang et al., 2018).

Moreover, this study quantified the effects of particle size on the leaching of metals from AR. The effective contact surface area between the solid bodies and liquid solutions is dependent on the particle size. The leaching rate of metals from finer particles is expected to be higher due to the larger effective contact surface area with the extraction solution. The test samples used in the TCLP and SPLP are required to have a size of less than 9.5mm. However, AR is composed of a mixture of course aggregate, fine aggregate, mineral filler, and asphalt binder with desired grades and compaction. Therefore, crushing is necessary to reduce the size of AR specimens to regulated levels. However, this crushing process introduces uncertainties as the test samples inevitably have different size distributions. Consequently, it may not be appropriate for AR mixtures to adhere to the size regulations specified in TCLP or SPLP. To evaluate the influence of particle size on the leaching test results, the cylindrical specimens of AR were crushed into loose particles after cooling down to room temperature. The resulting particles were then sieved into four groups based on their sizes: <2.36mm, 2.36-5mm, 5-10mm, and >10mm. The four groups of AR samples were subsequently subjected to a leaching test, using the optimal conditions obtained from the Taguchi analysis.

Table 1 Experimental design: orthogonal array and test parameters

Experiment	Parameters			Sample code
	Extraction time	Solution pH[#]	L/S ratio	
1	12h	12.5	10	S-1
2	12h	9.0	20	S-2
3	12h	5.5	30	S-3
4	12h	2.0	40	S-4
5	24h	12.5	20	S-5
6	24h	9.0	10	S-6
7	24h	5.5	40	S-7
8	24h	2.0	30	S-8
9	48h	12.5	30	S-9
10	48h	9.0	40	S-10
11	48h	5.5	10	S-11
12	48h	2.0	20	S-12
13	96h	12.5	40	S-13
14	96h	9.0	30	S-14
15	96h	5.5	20	S-15
16	96h	2.0	10	S-16

2.2.2. Characterization of the leaching behaviors of AR

In the binder and mortar scale tests, 20 g of AR binder or loose AR mortar was added to the extraction vessel containing 200 ml of extraction solution. The extraction vessel was then agitated at a rate of 30 rpm for 24 hours. For the mixture scale test, a compacted AR cylindrical specimen (diameter = 100 mm, height = 40 mm) weighing 700 g was placed on support inside the extraction vessel, and a total of eight leaching cycles were performed. The duration of the extraction period was up to 64 days (NEN 7345, 1995). To investigate the leaching properties of asphalt mixtures under normal conditions and occasional acid rain events, both neutral and acidic water were used as extraction solutions. The neutral extraction solution was prepared using DI water with a pH of 6.5-7. On the other hand, the acidic solution was made by adding a mixture of sulfuric acid and nitric acid (60/40 wt%) to DI water until the pH reached 4.00 ± 0.05. This pH value is representative of the typical acidity of acid rain (Likens and Bormann, 1974). The use of sulfuric and nitric acids was based on their essential role as components of acid rain. Sulfur dioxide (SO_2) and nitrogen oxides (NO_x) released into the atmosphere react with water, oxygen, and other chemicals to form acid rain (Likens et al., 1972). To simulate real-world conditions, AR samples at the three testing scales were prepared using the short-term and long-term aged binder, representing freshly constructed asphalt pavement and aged asphalt pavement with a service time of 5-10 years (AASHTO, 2013a; 2013b).

2.3. Analytical methods

In this study, the leachates obtained from the leaching tests were subjected to various analytical procedures to determine the concentrations of toxic metals and organic contaminants. The leachates were filtered using syringe filters with a pore size of 0.45 μm, and the resulting filtrate was analyzed using inductively coupled plasma-atomic emission spectrometry (ICP-AES, Spectro Arcos) to measure the concentrations of As, Cd, Cr, Cu, Ni, Pb, Se, and Zn. In addition, the concentrations of PAHs were also determined using gas chromatography-mass spectrometry (GC-MS, Agilent) after liquid-liquid extractions of the leachates. The PAHs were extracted from the leachate solutions using dichloromethane (DCM) through a three-step liquid-liquid extraction procedure at water/DCM ratios of 1:1, 2:1, and 4:1, respectively. The first extraction involved adding 10 ml of leachate solution to a borosilicate glass tube containing 10 ml of DCM, which was then vigorously stirred on a test tube shaker for 90 seconds. The mixture was left to settle for 24 hours, after which the DCM phase was collected, and the extraction process was repeated twice using 5 ml and 2.5 ml of DCM with a rest time of 6h and 3h, respectively. The collected DCM phases were combined, dried with magnesium sulfate (anhydrous granulated for organic trace analysis), and then evaporated under a gentle flow of N2. The extract was dissolved in 1 ml of fresh DCM and was then analyzed immediately. An Agilent J&W Scientific HP-5ms column (5%-phenyl-methylpolysiloxane phase, 30 m × 0.25 mm i.d., 0.25-µm film thickness) with a very low bleed characteristic was used for PAH measurements. The chromatographic conditions for the analysis of PAHs were as follows: the test samples were introduced using a 1.0 µL splitless injection, a heater temperature of 260 ºC, and the solvent delay was 6.5 min. Helium was used as the carrier in a constant flow of 1 ml/min. The oven was initialized at a temperature of 60 ºC for 10 min, then increased to 120 ºC at a rate of 5 ºC /min and further increased to 300 ºC at a rate of 3 ºC /min. The detection was made by mass selective detector source at 300 ºC, quadrupole at 180 ºC, and transfer line at 280 ºC, respectively. Synchronous SIM-SCAN mode (scan range 50 to 550 atomic mass unit) was applied as the acquisition type. All test samples were prepared in triplicates, and the blank samples were used as control groups.

3. Results and Discussion

3.1. Effects of experimental variables

Experimental designs using the Taguchi method were conducted to determine the effects of various experimental variables and obtain the optimal parameters. Sixteen experimental runs were conducted to quantify the concentrations of contaminants in the leachate of AR mixtures. Available metal contents of toxic effects (AMTE), which reflects the leachability of toxic metals/ metalloids, including arsenic, cadmium, chromium, copper, lead, nickel, selenium, and zinc (US EPA, 2005), were selected as the response for analyzing the experimental results. AMTE is defined by Equation (2):

$$AMTE\left(\frac{mg}{kg}\right)=\frac{\sum C_{As}, C_{Cd}, C_{Cr}, C_{Cu}, C_{Pb}, C_{Ni}, C_{Se}, C_{Zn}\ \times L_s}{M_s} \tag{2}$$

where C is the concentration of metals in the leachate (µg/L), Ls is the volume of the extraction solution (ml), and Ms is the dry mass of the sample (g).

The responses of the test samples to different test conditions are illustrated in **Fig. 2**. The measured AMTE values of the AR mixtures varied in the range of 7-34 mg/kg, depending on the test conditions. The results of S/N ratios are presented in **Table 2**. A higher value of the S/N ratio corresponds to a better test condition. The best conditions were found at an extraction duration of 24 h, an extraction solution with a pH of 2, and an L/S ratio of 10. The significance of each factor can be measured using the delta values of the S/N ratios. It can be observed that the effect of the extraction solution pH on the response of the concentrations of the leached metals was more significant, while the L/S ratio had fewer influences compared to the other two factors. **Fig. 3** illustrates the responses of the AMTE values in terms of the extraction durations and the pH values of the extraction solution. It can be observed that significantly more metals were leached from the AR mixtures when the pH of the extraction solution was lower. This finding suggests that the leaching test results of asphalt mixtures are greatly affected by the acidity of the extraction solution, as the mobility of metals varies a lot at different pH values. The previous leaching studies conducted on concrete materials using the extraction solutions with different pH values led to similar findings. It was found that most metals (e.g., Cd, Pb, Zn) in recycled concrete aggregates have a significantly higher leachability at lower pH values (Ai et al., 2019). It can also be observed that the measured AMTE values were first increased with longer extraction time and then decreased, where the peak values were found at the extraction duration of 24 hours, which indicates that the leaching equilibrium of asphalt mixtures can be achieved within 24 hours in the accelerated laboratory leaching test.

The determined equilibrium time is consistent with the recommended extraction durations by TCLP and SPLP (e.g., 18 hours or 24 hours). It can be concluded that the pH values of the extraction solutions are the most significant factors in the leaching test results of asphalt mixtures. The influences of the extraction time and L/S ratio are less important, and an extraction duration of 24 hours and an L/S ratio of 10 were found to be suitable for the leaching test of asphalt mixtures.

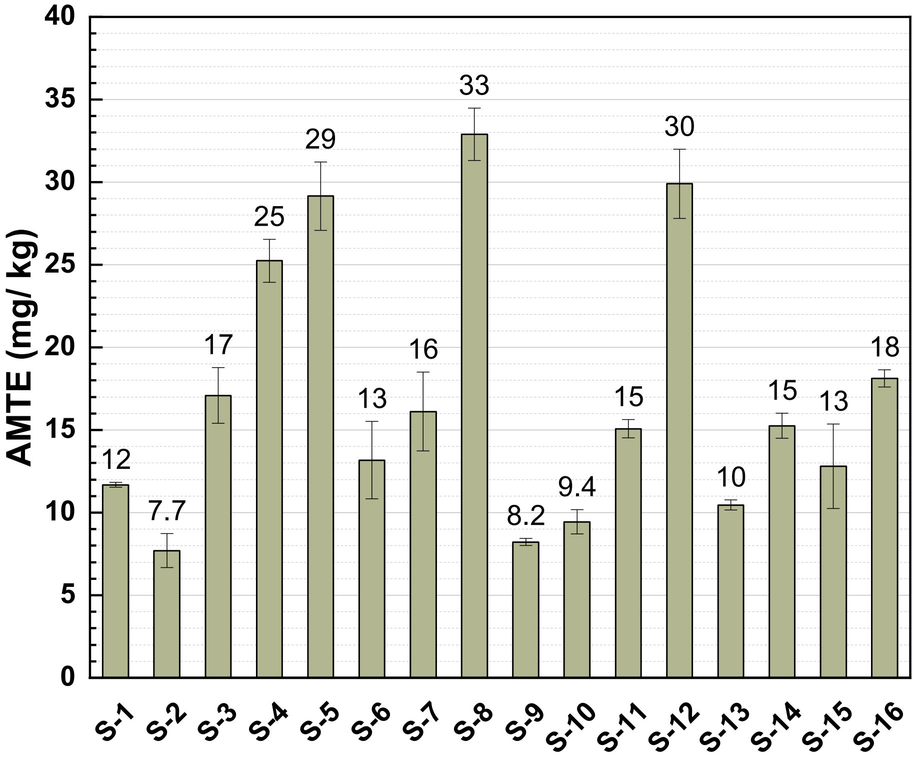

Fig. 2. Responses of the test samples to various test conditions

Table 2 S/N ratio for the response of leaching tests

Extraction time (h)			pH of the extraction solution (-)			L/S ratio (-)		
Level		S/N ratio	Level		S/N ratio	Level		S/N ratio
j=1	12	21.4	j=1	12.5	14.9	j=1	10	16.9
j=2	24	25.3	j=2	9.0	14.2	j=2	20	16.0

j=3	48	21.0	j=3	5.5	17.5	j=3	30	16.3
j=4	96	22.5	j=4	2.0	21.8	j=4	40	15.9
Delta		4.3	Delta		7.6	Delta		1.0

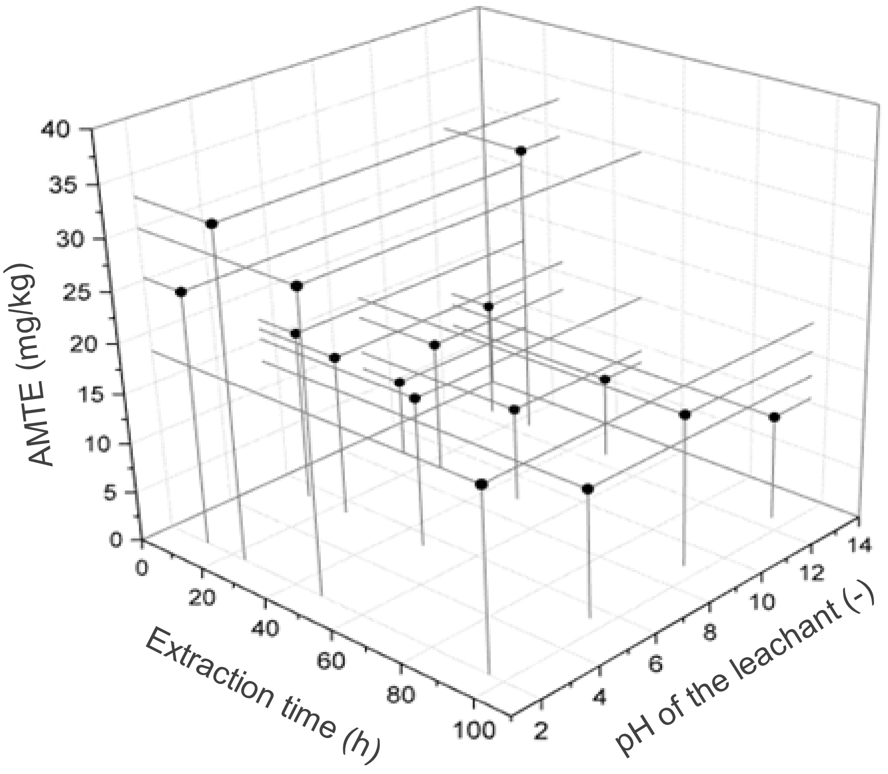

Fig. 3. Responses of the AMTE values in terms of the extraction time and the pH values of the extraction solutions

The influences of particle size on the measured metal concentrations are shown in **Fig. 4**. It can be observed that particle size greatly influences the leaching test results. Finer particles present significantly higher leachabilities than larger particles. The TCLP and SPLP methods that have been principally applied only recommended sample particle sizes of < 9.5 mm. However, the random particle size distribution after crushing and sieving processes will introduce huge uncertainties to the leaching test results. Moreover, the crushing process may introduce impurities to the samples and result in incorrect measurements.

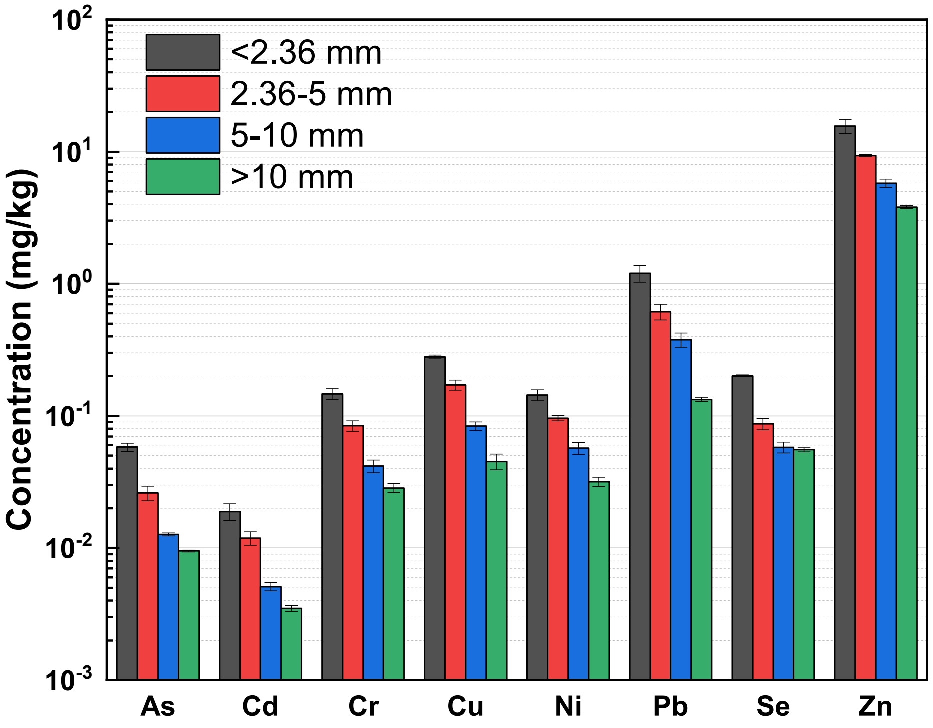

Fig. 4. Measured metal concentrations affected by different particle sizes

To address these issues, a multi-scale experimental method was proposed to achieve a comprehensive and representative evaluation of pollutant leaching from asphalt pavements by incorporating realistic gradations. This method involves utilizing the test samples at three scales: asphalt binder, loose asphalt mortar, and compacted asphalt mixture. Asphalt binder, which is a critical component of the asphalt mixture, is used to create film samples with fixed thickness in the batch leaching test. Asphalt mortar, also known as the fine aggregate matrix (FAM), plays a crucial role in bonding the aggregates together to form the asphalt mixture. It is a composite material consisting of asphalt binder, filler, and fine aggregates with a maximum nominal size of <2.36mm. The small particle size distribution of asphalt mortar makes it suitable for accelerated batch leaching tests. It is also free from contamination that may occur during the crushing and sieving processes. Moreover, asphalt mortar is easy to prepare in a laboratory setting and has been widely used in mechanical property testing, allowing for easy comparison of leaching test results. Compacted asphalt specimens, representative of actual field conditions, were employed in the proposed method to evaluate the long-term leaching properties of asphalt pavements using tank leaching tests. This approach facilitates the examination of both surface wash-off-driven and diffusion-driven leaching processes, while taking into account the realistic gradation and compaction properties of asphalt pavements. The proposed multi-scale experimental method has the potential to provide a more comprehensive and accurate evaluation of pollutant leaching from asphalt pavements.

3.2. Leaching behaviors of AR

3.2.1. Leaching of toxic metals/ metalloids

Fig. 5 shows the overall leaching test results of the AR binder, mortar, and mixture. ST and LT represent short-term and long-term aged samples, while -N and -A indicate whether the extraction solution is neutral or acidic. It can be observed that remarkably more toxic metals were found in the leachates using acidic extraction solutions despite the scales of test samples. It is expected since the majority of metals have higher solubilities in a more acidic environment, and the objective of this investigation is to verify whether the release of toxic metals from AR will be a concern to the local water environment when the pavement is exposed to acid rain. The detailed concentrations of toxic metals in the leachates of AR binder, mortar, and mixture are presented in **Table 3**. Only trace amounts of toxic metals were detected in the leachates when AR samples were exposed to neutral water. Among all the toxic metals, Zn shows the highest concentrations as zinc oxide (ZnO) was used as an activator for the vulcanization of rubber, and it represents approximately 1−2% of tire rubbers by weight (Degaffe and Turner, 2011; Taheri et al., 2011).

Nonetheless, AR materials present little leaching risk to the environment under neutral conditions, as the measured concentrations of toxic metals in the leachates were well below the regulated levels. However, when artificial acid rain was used as the extraction solution, elevated concentrations of toxic metals were found in the leachate. Elevated levels of metals were found in the leachates due to increased mineral dissolutions under acidic environments. Although most of the toxic metals were measured in values below the regulated levels, AR materials present certain risks of leaching Cd and Zn to the aquatic environment with higher concentrations compared to the maximum allowed values in the standard. The results suggest that AR is generally safe to use as a paving material without bringing concerns to the environment under normal circumstances. However, the leachability of toxic metals from AR can be dramatically escalated with the increased acidity of rains, which may lead to the leaching of toxic metals into the runoff that would eventually harm the aquatic environment.

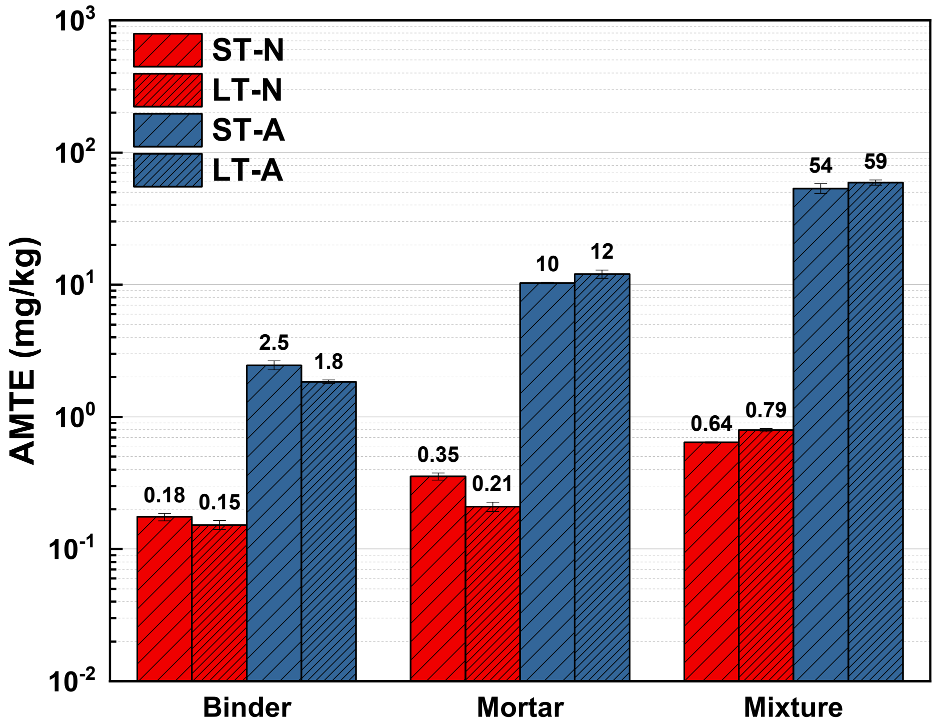

Fig. 5. Measured AMTE values of the leachates from AR binder (24h), mortar (24h), and mixture (64d)

Table 3 Concentrations of toxic metals (μg/L) in the leachates of AR samples

Constituents (μg/L)	Binder		Mortar		Mixture		GCTLs
	pH=7	pH=4	pH=7	pH=4	pH=7	pH=4	
As	-	0.5-1.5	-	0.8-1.6	-	1.7-1.8	10
Cd	0.7-1.3	0.7-1.2	0.6-0.8	2.6-3.0	0.7-1.6	5.9-7.0	5
Cr	0.5-1.3	4.7-7.4	-	6.2-6.7	-	7.1-16.2	100
Cu	5.6-6.4	8.7-14.1	4.4-4.7	7.8-9.7	4.9-7.0	21.5-28.5	1000
Ni	0.1-0.2	6.1-6.5	0.5-0.6	4.3-5.3	0.8-0.9	15.2-17.3	100
Pb	0.9-1.5	10.6-11.1	1.1-1.8	3.2-3.7	0.3-0.4	6.0-10.4	15
Se	-	2.1-5.5	-	5.7-12.2	-	13.6-17.7	50
Zn	6.6-7.6	145.8-204.0	13.4-28.4	987.0-1170.2	55.0-71.8	5274.1-5834.1	5000

Note: Shaded values indicate exceedances in regulation levels; GCTLs: groundwater and surface water cleanup target levels (DEP, 2005); - : Not detected (detection limit = 0.1 μg/L).

Regarding the influence of aging, the results suggest that aging appears to exert divergent effects on the measured metal concentrations across samples of varying scales (**Fig. 5**). This observation is primarily drawn from the test results conducted under acidic conditions (represented by ST-A and LT-A), given that metal leaching from AR consistently remained at remarkably low levels (i.e., less than 1 ppm) under neutral environments, rendering them inconsequential. It is noteworthy that the long-term aged samples exhibited reduced metal leaching in the binder-scale test while manifesting increased metal leaching in the mortar and mixture-scale tests. During the aging process, oxidative reactions occurred, leading to the transformation of light hydrocarbons in the asphalt binder into heavier compounds. This transformation resulted in an increase in the viscosity and stiffness of the aged asphalt binder. As the leaching of metals from AR binder is essentially a diffusion-controlled process, the long-term aged AR binder became much more rigid, which impedes the diffusion of metals from the asphalt binder to the extraction solution. However, unlike the binder scale leaching test, mortar and mixture scale leaching tests are influenced by both diffusion and advection effects. The advective leaching process occurs as a liquid physically flows through the sample. It is impossible to happen in the binder scale test as asphalt binder is highly viscous and considered impermeable to water. However, the advection effect plays a crucial role in mortar and mixture scale tests. The advective leaching dominates the leaching process in the mortar scale test since the water rinses through loose AR mortar without restrictions. Because the long-term aged AR binder is more rigid and brittle, the coating between binder and aggregate became inferior, leading to lower resistance to water flush. The flush strips the AR mortar, generating more finer particles and even stronger advection effects. Therefore, although the diffusion of toxic metals from long-term aged AR binders was slow, more metals were transported and leached out from the AR mortar by the advection effect. In the mixture scale test, the compaction process was expected to stabilize the AR mixture and restrict the migration of contaminants by decreasing the effective surface area and permeability of toxic metals. As a result, the leaching speed was much lower in compacted AR mixtures, and the measured leaching concentrations of toxic metals from the compacted AR mixtures through a 64-day test duration was comparably higher than that of the AR mortar, which had a test duration of only 24 hours. The aging process caused the loss of bonding strength and weakened coating in long-term aged AR mixtures, which deteriorates the stabilization effect and makes the material more vulnerable to water penetration. Therefore, long-term aged AR mixtures also presented a higher leachability than short-term aged AR mixtures. These findings suggest that the leachability of toxic metals from AR mixtures is likely to increase with longer service life due to the inevitable aging of asphalt binder, generating more environmental concerns.

3.2.2. Leaching of PAHs

The concentrations of 16 US EPA priority pollutant PAHs that are potential human carcinogens were measured in this study (NTP, 2005), namely acenaphthene (Ace), acenaphthylene (Acy), anthracene (Ant), fluoranthene (Flt), fluorene (Flu), naphthalene (Nap), phenanthrene (Phe), pyrene (Pyr), benz[a]anthracene (BaA), benzo[b] fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[ghi]perylene (BghiP), benzo[a]pyrene (Bap), chrysene (Chr), dibenz[a,h]anthracene (Dba), and indeno[1,2,3-cd]pyrene (Ip). Only eight lightweight PAHs were detected at measurable concentrations in the leachates of AR materials, including Ace, Acy, Ant, Flt, Flu, Nap, Phe, and Pyr. **Fig. 6** illustrates the overall PAH leaching results. It was found that concentrations of light molecular weight PAHs (e.g., Nap, Acy, Ace, and Flu) are higher than those with relatively heavier molecular weights (e.g., Phe, Ant, Flt, and Pyr). It was attributed to the low solubilities of PAHs in water, while the lower molecular weight PAHs have relatively higher solubilities than those with increased molecular weights. A slight increase in PAH concentrations was found when acidic water was used. Similar findings were reported in a previous study which suggested that lightweight PAHs were more soluble in acidic water (Batchamen Mougnol et al., 2022).

A possible explanation for this phenomenon is the cation− π interaction between lightweight PAHs and protons in acidic water, which is a stabilizing electrostatic interaction between the polarizable pi electron cloud of an aromatic ring and a cation. The delocalized conjugated π system in benzene rings generates polarity, which is more condensed in lightweight PAHs, providing them slight solubilities in a polar liquid such as water. Also, this electron-rich π system can have a noncovalent molecular interaction with an adjacent cation (i.e., H^+), which is abundant in acidic water, to form a stable binding that is much stronger than hydrogen bonds

(Frontera et al., 2011; Hunter and Sanders, 1990; Mahadevi and Sastry, 2013). As a result, lightweight PAHs form more stable structures and present higher solubilities in acidic water.

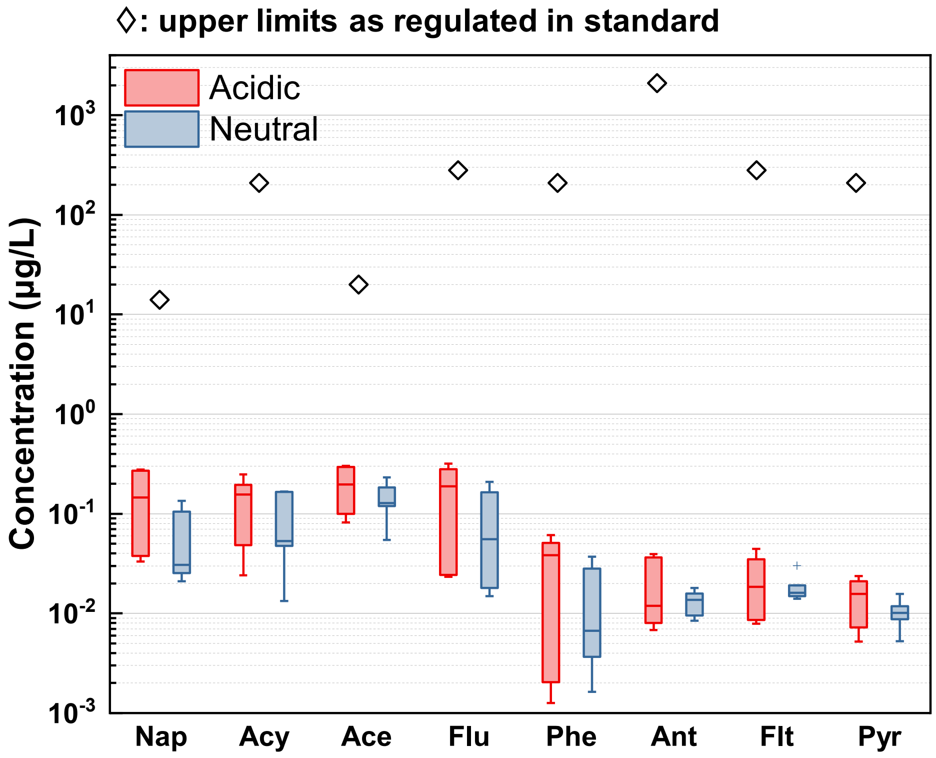

Fig. 6. The overall PAH concentrations in AR leachate and the regulatory maximum levels (DEP, 2005)

The measured concentrations of PAHs in the leachates of AR are presented in **Table 4**. The influence of aging on the leaching of PAHs was insignificant, although long-term aged samples showed marginally decreased concentrations of PAHs than that of short-term aged samples. It was mostly due to the evaporation of low molecular weight PAHs during the laboratory aging process, especially naphthalene, as it has the highest volatility. The test results indicated that only lighter molecular weight PAHs (i.e., less than five hydrocarbon rings) could be found in the leachates of AR, and their concentrations were very low. The impact of PAHs on health can be evaluated by the toxic equivalency factor (TEF), based on the BaP equivalent concentration (BaP_{eq}) that assumes that all carcinogenic PAHs are as potent as BaP (Nisbet and Lagoy, 1992). The BaP_{eq}-TEF values were calculated by Equation (3):

$$\begin{gathered}\left(BaP_{eq} - TEF\right)_{\Sigma PAH} = \\ [Nap] \times 0.001 + [Acy] \times 0.001 + [Ace] \times 0.001 + [Flt] \times 0.001 + \\ [Phe] \times 0.001 + [Ant] \times 0.01 + [Flu] \times 0.001 + [Pyr] \times 0.001 + \\ [BaA] \times 0.1 + [Chr] \times 0.01 + [BbF] \times 0.1 + [BkF] \times 0.1 + \\ [BaP] \times 1 + [Ip] \times 0.1 + [DbA] \times 1 + [BghiP] \times 0.01\end{gathered} \tag{3}$$

Table 4 Concentrations of PAHs (μg/L) in the leachates of AR samples

Constituents (μg/L)	Binder		Mortar		Mixture		GCTLs
	ST	LT	ST	LT	ST	LT	
Acenaphthene	0.233-0.254	0.139-0.183	0.093-0.134	0.083-0.120	0.082-0.122	0.054-0.101	20
Acenaphthylene	0.165-0.190	0.121-0.167	0.050-0.149	0.047-0.096	0.049-0.056	0.013-0.024	210
Anthracene	0.008-0.009	0.008-0.010	0.013-0.016	0.009-0.018	0.013-0.017	0.014-0.015	2100
Benzo(a)anthracene	-	-	-	-	-	-	0.05
Benzo(a)pyrene	-	-	-	-	-	-	0.05
Benzo(b)fluoranthene	-	-	-	-	-	-	0.5
Benzo(g,h,i)perylene	-	-	-	-	-	-	0.2
Benzo(k)fluoranthene	-	-	-	-	-	-	210
Chrysene	-	-	-	-	-	-	4.8
Dibenzo(a,h)anthracene	-	-	-	-	-	-	0.005
Fluoranthene	0.016-0.018	0.135-0.209	0.015-0.035	0.016-0.045	0.008-0.019	0.009-0.030	280
Fluorene	0.164-0.279	0.135-0.209	0.057-0.140	0.054-0.119	0.015-0.024	0.018-0.023	280
Indeno(1,2,3-cd)pyrene	-	-	-	-	-	-	0.05
Naphthalene	0.135-0.195	0.097-0.105	0.026-0.177	0.021-0.107	0.036-0.038	0.025-0.033	14

Phenanthrene	0.028-0.051	0.026-0.037	0.008-0.051	0.005-0.031	0.002-0.004	0.001-0.004	210
Pyrene	0.012-0.020	0.012-0.016	0.009-0.024	0.011-0.021	0.005-0.010	0.005-0.007	210

Note: GCTLs: groundwater and surface water cleanup target levels (DEP, 2005); - : Not detected (detection limit = 0.1 μg/L).

The mean values of the BaP$_{eq}$-TEF results of AR are illustrated in **Fig. 7**. It can be observed that the AR binder has the highest BaP$_{eq}$-TEF values, followed by AR mortar and mixture. This is because PAHs are a class of chemicals that occur naturally in crude oil and are believed to be presented in asphalt binder, although the concentrations of PAHs in asphalt binder are often very low (i.e., ppm levels). Therefore, the potential leaching of PAHs from AR is attributed to the asphalt binder content, which was approximately 80% in the AR binder, 20% in the AR mortar, and 5% in the AR mixture, respectively. Also, the calculated BaP$_{eq}$-TEF results were found at low levels (i.e., less than 0.01 μg/L), which indicated that the leachates of asphalt paving materials were at little or no risk for harmful health effects (Chepelev et al., 2015; Moffat et al., 2015). It was mainly because PAHs are found less frequently in water due to their low solubilities compared to air and soil. Exposure to PAHs can potentially increase the risk of certain types of cancer. However, the primary exposure route is through the diet (food/ drinking water exposures), which further reduces the cancer risks through dermal exposure to the leachate of AR. It should be noticed that the BaP$_{eq}$-TEF results in this study only demonstrated that the leachates of AR have few harmful health effects on humans. However, there are still health risks in contact with asphalt pavement as the PAHs can be emitted from asphalt mixtures into the air, especially during the construction of asphalt mixtures at high temperatures. A complete investigation of PAH exposure from contact with asphalt pavement is beyond the scope of this study. More details can be found in previous studies on asphalt emissions (Autelitano et al., 2017; Cui et al., 2020; Khare et al., 2020).

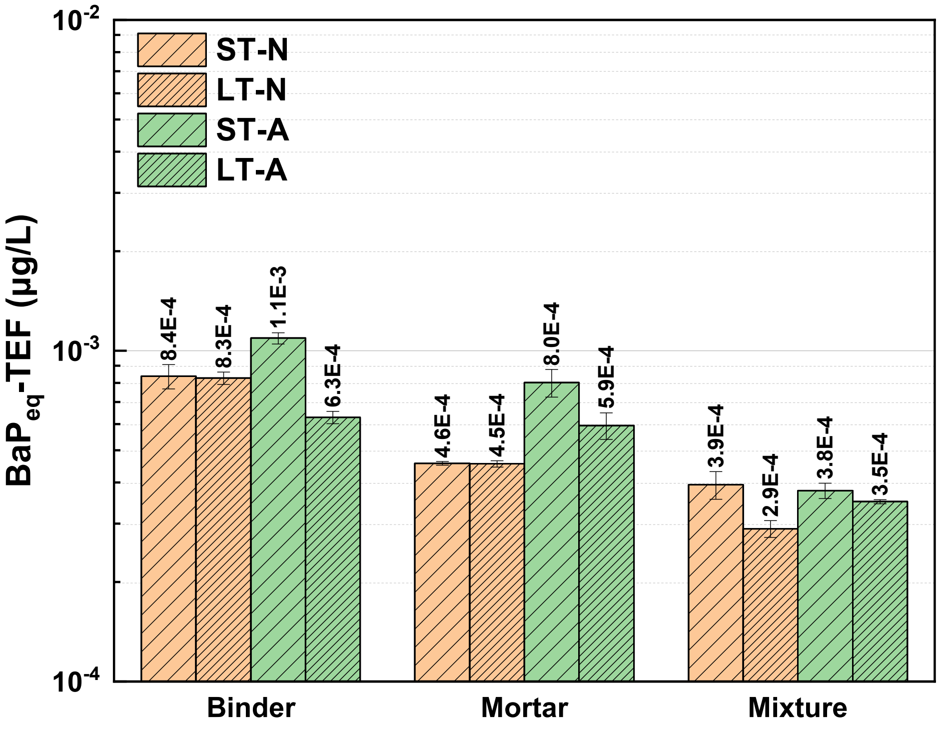

Fig. 7. The BaP$_{eq}$-TEF results of AR

4. Conclusions

This study was conducted to comprehensively evaluate the leaching properties of AR, a widely used waste-recycled asphalt paving material in field applications. The first part of the study focused on the investigation of the effects of experimental variables on the leaching process of AR, including the pH of the extraction solution, sample particle size, extraction duration, and L/S ratio. Following this, a novel method was developed and employed to comprehensively investigate the leaching behaviors of AR at three scales: binder, mortar, and mixture. Based on the experimental findings, the following conclusions can be drawn:

- The particle size of samples and the pH of the leachant are the predominant factors in the leaching test of AR. An L/S ratio of 10 and a test duration of 24 h are generally suitable for batch leaching tests, whereas longer durations are required for evaluating the long-term leaching property using tank leaching tests.

- The proposed multi-scale leaching method can generate a more comprehensive and accurate evaluation of pollutant leaching from AR. Test results exhibited good repeatability, and the proposed method facilitated easy comparison of results. The findings reveal that the leachability rank follows the order of binder < mortar < mixture across three scales.
- In order to reflect the true conditions of asphalt pavements in the field, it is important to use a leachant that simulates rainfall. The use of neutral and acidic leachant in this study has provided insights into the behavior of pollutant leaching under different conditions.
- The test results revealed that asphalt binder is the dominant source of PAHs in asphalt pavements. The leaching of PAHs generally displays low environmental risks due to their low solubility in water. Only eight lightweight molecular PAHs (i.e., Ace, Acy, Ant, Flt, Flu, Nap, Phe, and Pyr) were detected at measurable levels.
- AR is generally safe as a recycled waste material for surface pavement, but under certain conditions such as exposure to acidic runoff or stormwater, the leaching of toxic metals from AR can exceed water regulation limits and pose a risk to the environment.
- The aging of asphalt pavements may lead to increased leachability of toxic metals, while reduced releases of PAHs are expected.

In future studies, the effects of other types of aging (e.g., thermo-oxidative aging, photo-oxidative aging, and water aging) on the leaching behaviors of AR will be investigated in addition to oxidative aging, as they may also affect leaching test results. Moreover, the ecotoxicological effects of these pollutants on aquatic organisms should be studied in the future to better understand the influences of leachates from AR on the aquatic environment.

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