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Technical Article

Environmental effects of leachate extracts from reclaimed asphalt pavement: determination of metals, polycyclic aromatic hydrocarbon and acute toxicity to *Daphnia magna*

Efeitos ambientais de extratos lixiviados de pavimento asfáltico fresado: determinação de metais, hidrocarbonetos policíclicos aromáticos e toxicidade aguda para Daphnia magna

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ABSTRACT

Pavimentos asfálticos fresados (PAF) apresentam uma variedade de compostos orgânicos e inorgânicos que podem interagir com o meio ambiente e promover efeitos deletérios à saúde humana. Este estudo investigou o potencial de toxicidade dos PAF por meio da determinação de metais, hidrocarbonetos policíclicos aromáticos (HPA) e testes de toxicidade aguda com *Daphnia magna*. Foram comparados extratos lixiviados e solubilizados de amostras de PAF e de resíduo asfáltico fresco. As análises de Mn apresentaram concentrações acima do critério de aceitação normativo brasileiro para os extratos solubilizados. As amostras resultaram em concentrações de Cd, Cr e Pb, que estão na lista de substâncias que conferem periculosidade aos resíduos. Em relação aos HPA, duas amostras de lixiviado apresentaram concentração de BaP acima do valor máximo permitido e as demais amostras apresentaram valores de BaP próximos ao limite estabelecido. Ademais, foram identificadas nas amostras concentrações de BaA, BbF, IcdP e Chr. Para os ensaios de toxicidade aguda, duas amostras solubilizadas indicaram toxicidade aguda para *Daphnia magna*. Os resultados indicaram que o método de preparo dos lixiviados e do extrato solubilizado influenciaram os valores de metais e a toxicidade aguda. Duas amostras de PAF foram classificadas como resíduos perigosos, sinalizando que tais materiais apresentam potencial para lixiviar substâncias perigosas ao ambiente. Portanto, a disposição em solo desse tipo de resíduo deve ser criteriosa, uma vez que sua composição contém substâncias que podem impactar o meio ambiente e causar efeitos toxicológicos em organismos vivos.

Palavras-chave: rodovias; mistura asfáltica; impacto ambiental; lixiviação; resíduos de construção civil

RESUMO

Reclaimed Asphalt Pavement (RAP) has a variety of organic and inorganic compounds that can interact with the environment and promote deleterious effects on human health. This study investigated the potential toxicity of RAP through metal determination, polycyclic aromatic hydrocarbon (PAH) analysis and acute toxicity tests with *Daphnia magna*. Leached and solubilized extracts of RAP samples and a freshly produced asphalt sample were compared. Regarding metals, Mn was above the Brazilian acceptance criteria in the solubilized extract. The samples showed concentrations of Cd, Cr, and Pb, which are on the list of substances that render waste hazardous. For PAH, two samples of leachate had BaP concentration above the maximum value allowed and the other samples had BaP values close to the established limit. In addition, the samples still presented concentrations of BaA, BbF, IcdP, and Chr. For the acute toxicity tests, two solubilized samples indicated acute toxicity to *Daphnia magna*. The results indicated that the method of preparing the leached and the solubilized extract influenced the values of metals and acute toxicity. Two samples of RAP could be classified as hazardous waste, pointing out that these materials have the potential to leach hazardous substances to the environment. Therefore, the disposal of this type of waste should be carefully evaluated, as its composition contains substances that may degrade the environment and cause toxicological effects on living organisms.

Keywords: road construction; asphalt mixture; environmental impact; leaching; construction and demolition waste.

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INTRODUCTION

The Brazilian road network comprises more than 1.7 million kilometers, of which 12.4% are paved with asphalt concrete (CNT, 2019). This material consists of approximately 95% aggregates (gravel, sand, recycled materials) and the remaining 5% are liquid asphalt or bitumen (ASPHALT INSTITUTE AND EUROBITUME, 2015). Generally, after some decades of use, road surfaces must be recuperated due to traffic wear, material aging, and weathering actions (GARRAÍN; LECHÓN, 2019). For over forty years, the use of recyclable materials for the recovery of asphalt mesh has been common (TXAPA, 2019), among which the reclaimed asphalt pavement (RAP) stands out. RAP is the residue obtained from the milling of damaged pavements and is constituted by aggregates of selected quality, partially covered by aged bitumen that can be reused in the production of bituminous mixtures through cold- or hot-recycling technologies (FARINA *et al.*, 2017; AKBAR *et al.*, 2019; SUN *et al.*, 2019; XIAO *et al.*, 2019).

The use of RAP for the construction of new roads has several environmental and economic benefits, such as the conservation of non-renewable resources, reduction of greenhouse gas emissions, landfill diversity, reduction of maintenance operations, as well as reduction of negative environmental impacts caused by the temporary storage of this material (HUANG; BIRD; HEIDRICH, 2009; CELAURO *et al.*, 2015; YANG *et al.*, 2015; ROMEO *et al.*, 2019; CHHABRA; RANSINCHUNG; ISLAM, 2021; SPREADBURY *et al.*, 2021). Nevertheless, the temporary storage of RAP before its application is an aspect that normally has not been considered. Usually, fixed locations are chosen for the formation of great loading piles of this material. Storage time might be several months or even years in places usually not covered at the base and waterproofed, while during this time RAP remains subject to the weather and percolation of rainwater (XIAO *et al.*, 2017).

Although asphalt concrete is one of the most important building materials in the modern world, the leaching of its compounds into the environment and the potential exposure risks of the population are poorly understood (OLIVEIRA *et al.*, 2019; VON GUNTEN; KONHAUSER; ALESSI, 2020). Concerning RAP, some authors have demonstrated the potential for contamination of its leaching mainly related to Polycyclic Aromatic Hydrocarbons (PAH) and a variety of metals (LEGRET *et al.*, 2005; BUTERA; CHRISTENSEN; ASTRUP, 2014; HOY *et al.*, 2016; MEHTA *et al.*, 2017; MIJIC *et al.*, 2020). Mijic *et al.* (2020), for example, have demonstrated in their studies with RAP leachates that metals such as zinc (Zn) and copper (Cu) were detected above the values recommended by the United States Environmental Protection Agency (USEPA) for water quality limits (WQL). Additionally, Mehta *et al.* (2017), in leaching tests with RAP, detected concentrations of lead (Pb) above the values recommended by the EPA for drinking water. Additionally, they demonstrated that PAH including fluoranthene (Fla), pyrene (Pyr), benz(a)anthracene (BaA), and chrysene (Chr) can leach out of the RAP. Legret *et al.* (2005), in a study with RAP leachates from a French highway, detected BaA, Chr, Benzo[k]fluoranthene (BkF), and Dibenz[a,h]anthracene (DahA).

Experimental leaching studies are normally carried out under controlled laboratory conditions and ideally, when possible, laboratory results are compared with the chemical compounds measured in samples of the same site. Combined results may be used to improve long-term leaching predictions and risk assessment (ENGELSEN *et al.*, 2012). Most of the time this is not possible and the simulation of the leaching in the lab needs to be carefully carried out in a way that best reflects what occurs in the field, once that a few changes in experimental conditions could modify the results of chemical and other analyses (KANG *et al.*, 2011).

In Brazil, these studies are still incipient, which generates the need to propose new research methods on the influence of RAP constitution, toxicity, and interaction with the environment. This paper focused on chemically characterizing the presence and concentration of organic (PAH) and inorganic (metals) contaminants and assessing acute toxicity by daphnia assays.

MATERIALS AND METHODS

Samples

Approximately 30 kg of four RAP samples were obtained of milled asphalt pavement from different locations of the metropolitan area of Curitiba, Paraná, southern Brazil. Samples came from two urban streets (S1: 25°26'44.15"S; 49°11'22.87"W and S5: 25°23'56.95"S; 49°15'32.86"W) and two highways (S2: 25°30'32.14"S; 49°07'59.46"W and S4: 25°27'11.89"S; 49°06'57.72"W). For comparison, a freshly produced asphalt sample was supplied by a local plant (named S3, location coordinate 25°44'53.15"S; 49° 6'17.35"W).

Each sample was homogenized and reduced volume fractions were obtained by the quartering method according to the official Brazilian protocol for solid waste sampling ABNT NBR 10007:2004 (ABNT, 2004). Aliquots of 5 kg were conditioned in inert plastic bags and kept at 4°C until their use in the analyses.

The samples were characterized by granulometric evaluation and bitumen content according to the Brazilian normalized methods (ABNT, 2003; 2013) shown in Table 1.

Table 1 – Values of granulometric and the bitumen content of RAP samples.

Sample	Granulometry (%)			Bitumen content (%)
	filler aggregates (< 0.075 mm)	fine aggregates (0.075-2.0 mm)	coarse aggregates (> 2 mm)	
S1	0.1	84.2	15.7	4.49 (± 0.34)
S2	0.1	50.9	49.0	5.66 (± 0.50)
S3	0.1	75.3	24.6	5,75 (± 0.08)
S4	0.1	58.5	41.4	4.14 (± 0.10)
S5	2.2	80.3	17.5	4.81 (± 0.07)

Source: elaborated by the authors.

Leaching methods

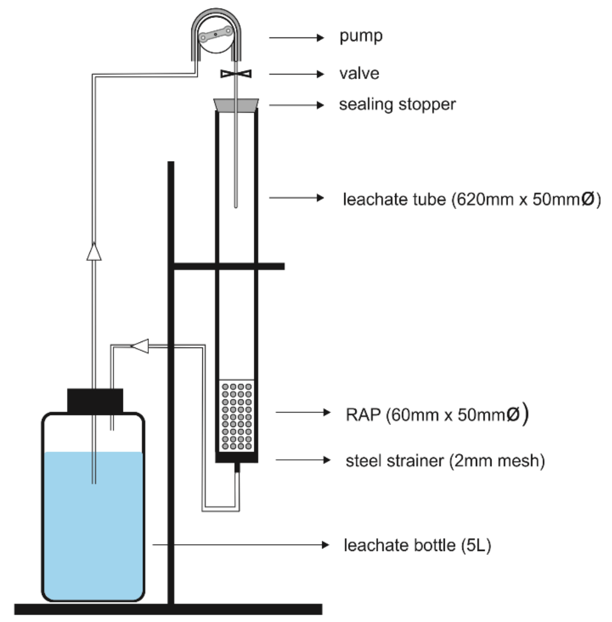
Leachates were prepared based on the Brazilian standardization guide (ABNT, 2004) which is based on the "Toxicity Characteristic Leaching Procedure" (TCLP) method (SW-846 Method 1311; USEPA, 1994). Both experiments used an extraction fluid prepared by mixing 5.7 mL of glacial acetic acid (99.8%, Neon) and 64.3 mL of sodium hydroxide solution 1 mol·L^{-1} (Impex) in ultrapure water (Mega Purity) until complete 1 L; pH was adjusted to 4.90. In leaching experimental methods, the extraction fluid and sample ratio 20:1 were used. All experiments were conducted in triplicate after the sample's granulometry be adjusted to up to 9.5 mm. Reagents were of analytical grade.

Shaker leaching method

The experimental procedure to obtain leachate through orbital agitation (shaker) was based on protocol ABNT NBR 10005:2004 (ABNT, 2004) in which 10 g of sample and 200 mL of extraction fluid were transferred to a 250 mL Erlenmeyer. The flasks were covered and agitated in an orbital shaker device (Ethik) at 30 rpm and 25°C for 18 h. The pH of liquid samples (named "SL") was measured at the end time: 4.95 for SL1, 4.98 for SL2, SL3 and SL4, and 5.02 for SL5.

Column leaching method

For the column method, a leaching device was developed based on previous studies (CÓRDOBA, 2014). The system (Figure 1) consisted of a borosilicate glass column with an internal volume of 1.6 L and, in output, a stainless-steel mesh (2 mm) was placed to prevent the loss of sample particles. A peristaltic metering pump (Provitec) was used to recirculate the extraction fluid between leacher tube and reservoir bottle. The sample (100 g) was mixed with portions of the extraction fluid and transferred to the leacher, maintaining a volume of liquid sufficient to maintain a height of 40 cm; the remainder of the extractor liquid was transferred to the reservoir (named leachate bottle in Figure 1) and the flow of the peristaltic pump was set (30 L·h^{-1}) to keep a constant volume inside the leacher column. Recirculation was maintained for 18 h at 25°C. The pH of liquid samples (named "CL") was measured at the end time: 5.07 CL1 and CL5; 5.06 CL2 and CL4; 4.88 CL3.



Source: elaborated by the authors.

Figure 1 – Column leacher sketch developed for leachate preparation of RAP samples.

Solubilized extraction method

To obtain the solubilized extracts, the standardized method ABNT NBR 10006:2004 (ABNT, 2004) was strictly followed. 250 g of sample and 1 L of ultrapure water were transferred to a 2 L Erlenmeyer flask that was covered and remained for 7 days at 25°C. After this period, the samples were filtered through a nylon membrane (0.45 µm). The pH of solubilized extracts (named "SE") was checked at the end: 6.91 SE1; 7.12 SE2; 7.24 SE3; 7.13 for sample SE4 and 7.52 for sample SE5.

Metals analyses

The samples preparation method was conducted following the protocol of Standard Methods for the Examination of Water and Wastewater guide (APHA, 2005). The analyses were performed by ICP-OES (Optima 8300, PerkinElmer) and the concentration determined by linear regressions obtained with standards solutions of high purity (Qhemis) at the following concentrations: 0.05, 0.10, 1, 3, 5, 8, and 10 mg·L^{-1}. For leaching and solubilized extracts, analyses of cadmium (Cd), chromium (Cr), Iron (Fe), manganese (Mn), lead (Pb), and zinc (Zn) were performed.

Polycyclic aromatic hydrocarbons analysis

Sample preparation and PAH analysis were carried out following the NIOSH method 5515 (NMAM, 1994). Liquid samples were prepared by solid-phase extraction (SPE) with octadecyl silica (ODS) cartridges (1,000 mg, Macherey-Nagel) conditioned with 10 mL of methanol (grade HPLC, Vetec) and 5 mL of ultrapure water; 200 mL of sample were applied and the cartridge dried by vacuum for 120 seconds. Elution was performed with 10 mL of acetonitrile (grade HPLC, Honeywell). The eluate was evaporated until dried and the residual was redissolved in 1 mL with ethyl acetate (HPLC grade, Honeywell), filtered into vials using a Polytetrafluoroethylene (PTFE) syringe filter (0.22 µm, Allcrom). Considering the procedure above, the samples were concentrated 200 times before the analysis.

The analyses were performed by gas chromatography with flame ionization detection (CG-2010 plus, Shimadzu), using a sample injection of 1 µL in splitless mode at 290°C. Separation was conducted on a DB-5 capillary column (Agilent, 30 m long, 0.25 mm i.d. and 0.10 µm film thickness) with helium as carrier gas at 35 cm·s^{-1} (White Martins). The analysis started with a column at 60°C for 1 min followed by heating until 200°C (rate of 5°C·min^{-1}) and after until 280°C (rate of 2°C·min^{-1}). The detector was kept at 300°C. After adjusting the chromatographic method, the retention time of each compound was performed by analyses of individual PAH standard solution diluted in ethyl acetate (1 mg·L^{-1}).

The quantification of 16 PAH was performed applying the linear regressions obtained by external calibration. Analytical solutions were prepared with analytical standards (PAH Mix, 10 ppm in acetonitrile, Supelco) diluted in ethyl acetate (grade HPLC, J.T.Baker) to obtain the following concentrations: 5, 2.5, 1.0, 0.5, and 0.25 mg·L^{-1} of each analyte. The abbreviations adopted for the compounds were: Naphthalene (Nap), Acenaphthylene (Acy), Acenaphthene (Ace), Fluorene (Flu), Phenanthrene (Phe), Anthracene (Ant), Fluoranthene (Fla), Pyrene (Pyr), Benz[a]anthracene (BaA), Chrysene (Chr), Benzo[b]fluoranthene (BbF), Benzo[k]fluoranthene (BkF), Benzo[a]Pyrene (BaP), Indeno[1,2,3-cd] pyrene (IcdP), Dibenz[a,h]anthracene (DahA), and Benzo[g,h,i]perylene (BghiP).

Acute toxicity

Bioassays with *Daphnia magna* were carried out following the NBR 12713 guide (ABNT, 2016). The organisms were cultivated according to ABNT NBR 12.713:2016 and DIN 38.412:1989 protocols (DIN, 1989; ABNT, 2016). Before the toxicity tests, the pH of the samples was verified and adjusted to 7 using NaOH (0.01 mol·L^{-1}) for samples LEA3, LSA1, LSA2, LSA3, and LSA4. Test solutions were prepared by dilution of samples in culture medium to get 50, 25, 12.5, 6.25, and 3.125% (v/v). Ten neonates were exposed to 25 mL of the test solutions and maintained for 48 h at 20°C in the dark, in three replicates. The control test was performed with a culture medium and the sensitivity of the organisms was verified with potassium chloride.

The results were expressed in terms of the toxicity factor (TF) that corresponds to the highest sample dilution in which no immobility greater than 10% of organisms is observed. In case of no toxic effect is observed, the TF value is 1 (without dilution) and the other dilutions are the TF number: 2, 4, 8, 16, and 32 (highest dilution) (ABNT, 2016). After 48 h of exposure, the number of immobile organisms was recorded to calculate the percentage of immobility and TF.

RESULTS AND DISCUSSION

Metals analyses

Brazilian acceptance criteria (ABNT, 2004) were used to evaluate the relevance of metals concentrations (Table 2). Leach test limits (LTL) were used to compare the concentration of the metals in the leachates by column (CL) and by shaker (SL) while solubilization test limits (STL) were applied to SE. Regarding the results for the leachates, the concentration of the metals was below the Brazilian acceptance criteria in all the analyzed samples. For SE, only the concentration of Mn in samples S1 and S5 were above Brazilian criteria.

In the leaching samples, Zn was the metal with the highest concentration, with sample S2 being the one with the highest value in both tests (CL2 [41.65 mg·L^{-1}] and SL2 [40.49 mg·L^{-1}]). In the solubilized extract, only in sample S1 it was possible to quantify the element Zn. The occurrence of this metal in RAP can occur naturally, through the composition of the rocks used in the "mineral skeleton" of the pavements (SAXBY, 1969), or by anthropogenic sources, such as, for example, the presence of Zn in brake pads and tires, which can reach the pavement and consequently the waste. The occurrence of Zn can be due to the friction of the tire with the road surface, or by the abrasion of brake pads with metallic items, as pointed out by the study of Hjortenkrans, Bergbäck and Aggerud (2007) and Hong *et al.* (2020).

The high concentration of Zn obtained in the leachate extracts can be explained by the fact that it was relatively mobile and is readily leached in acidic media due to their amphoteric leaching pattern (BERTHELSEN; OLSEN; STEINES, 1995; LIM *et al.*, 2004; CETIN *et al.*, 2014; GUNTEN *et al.*, 2019). It is worth mentioning that the pH of the leached extracts was between 4.95 and 5.07 due to the acidic characteristics of the extraction solution, which was around 4.90. Similar to this study, Legret *et al.* (2005) and Mijic *et al.* (2020) found the highest Zn values in leached from RAP when the pH was more acidic.

Cd was another metal found in all samples from the leaching tests. The properties of Cd are similar to those of Zn, having greater mobility in acidic media (BERTHELSEN; OLSEN; STEINES, 1995). The occurrence of Cd in RAP can be due to the burning of oils and greases used as lubricants in vehicles, by the abrasion of brake pads or because the Cd can be part of the bitumen used as a binder in pavements (AKPOVETA; OSAKWE, 2014; JIANG *et al.*, 2015; GUNTEN *et al.*, 2019).

Pb was another metal responsible for the hazardous characteristics of the leachates. The highest concentration of Pb was verified at sample S4 for both column and shaker preparation methods: 0.506 mg·L^{-1} for CL4 and 0.108 mg·L^{-1} to SL4. The leachates of samples S3 and S5 presented significant levels of Pb: 0.064 and 0.038 mg·L^{-1} to CL3 and CL5; 0.115 and 0.017 mg·L^{-1} to SL3 and SL5, respectively. The Brazilian drinking water quality standard determines that the maximum value allowed for Pb is 0.01 mg·L^{-1} (BRASIL, 2021), similar to countries like the USA.

Similarly, Brantley and Townsend (1999) and Mehta *et al.* (2017), in studies with leachate from RAP in Florida and New Jersey (USA) respectively, detected concentrations of Pb higher than recommended by the USEPA for WQL, which is 0.015 mg·L^{-1}. The studies also indicated that RAP contamination was due to exposure to traffic emissions and dust deposition.

Table 2 – Metal concentrations (mg L^{-1}) in leachates and solubilized extracts.

Metal	STL	LTL	Column Leaching (CL)					Shaker Leaching (SL)					Solubilized Extract (SE)				
			CL1	CL2	CL3	CL4	CL5	SL1	SL2	SL3	SL4	SL5	S1	S2	S3	S4	S5
Cd	0.005	0.5	0.028	0.035	0.023	0.032	0.051	0.023	0.024	0.024	0.024	0.045	ND	ND	ND	ND	ND
Cr	0.05	5.0	ND	0.093	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Fe	0.30	U	0.709	3.911	0.553	0.637	1.090	0.394	0.231	0.211	0.227	0.339	ND	ND	ND	ND	ND
Mn	0.10	U	0.471	1.733	0.635	2.186	2.089	0.710	1.145	0.709	0.579	1.678	**0.398**	**0.059**	ND	ND	**0.326**
Pb	0.01	1.0	0.005	0.026	0.064	0.506	0.038	ND	ND	0.115	0.108	0.017	ND	ND	ND	ND	ND
Zn	5.0	U	0.963	41.65	0.755	1.337	0.725	10.86	40.49	0.931	0.554	0.509	1.813	ND	ND	ND	ND

Source: elaborated by the authors.

SLT: solubilization test limit; LTL: leaching test limits; ND: not detected; U: undefined.

The comparative study between the leaching and solubilization tests found that the concentrations of metals in the acid leaching tests were higher compared to the solubilized extracts. This fact may be related to the physical properties of some metals, which have greater mobility and are easily leached in acidic media (BERTHELSEN; OLSEN; STEINES, 1995; MEHTA *et al.*, 2017). In the leaching tests, the contact was dynamic, whereas for solubilization the contact was static. The type of agitation also interferes with the extraction process, since the dynamic contact produces great agitation between the particles of the waste, increasing the mechanical action of the extraction liquid on the sample (CAUDURO, 2003).

Similar to this research, Córdoba and Schalch (2013) reported that metal concentrations were higher in leaching tests than in solubility tests, except chromium. The authors compared the methods of leaching and solubilization of recycled aggregates from RAP samples. When comparing leaching tests, it seems that leaching by column was more efficient than leaching by shaker to extract metals. Except for Mn in samples SL1 and SL3 and Pb in sample SL3, all other metals showed higher concentrations for column extractions. This fact can be explained by the fact that the column has a downward flow of extracting fluid, which could favor the leaching of metals.

Polycyclic aromatic hydrocarbons analysis

Of the sixteen PAH monitored in this study, nine were detected among the samples. The concentrations of the following compounds were above the quantification limit of the analytical method: Pyrene (Pyr), Anthracene (Ant), Phenanthrene (Phe), Fluorene (Flu), Benzo[a]Pyrene (BaP), Benzo[b]fluoranthene (BbF), Indeno[1,2,3-cd]pyrene (IcdP), Chrysene (Chr), and Benz[a]anthracene (BaA). The other PAH compounds were not detected in the samples. PAH concentrations for all samples and their respective extraction types are summarized in Table 3. The limits of quantification and detection of the nine compounds quantified in this study were determined in accordance with the Brazilian guidelines for analytical validation and their values calculated considering the factor of concentration were: 0.95 and 0.29 μg·L^{-1} for Pyr; 0.85 and 0.26 μg·L^{-1} for Ant; 0.8 and 0.024 μg·L^{-1} for Phe; 0.75 and 0.022 μg·L^{-1} for Flu; 1.25 and 0.7 μg·L^{-1} for BaP; 0.9 and 0.26 μg·L^{-1} for BbF; 0.8 and 0.23 for μg·L^{-1} for IcdP, 0.95 and 0.28 μg·L^{-1} for Chr; 1.1 and 0.33 μg·L^{-1} for BaA.

As metals, the occurrence of PAH in RAP can occur naturally, due to their presence in the bitumen composition (BRANDT; GROOT, 2001; MEHTA *et al.*, 2017), and intensified by anthropogenic sources, through the burning of gasoline and diesel, abrasion of tires, wear of brake linings, and road dust deposition (LEE; DONG, 2010; PERRONE *et al.*, 2014; AZAH; KIM; TOWNSEND, 2015).

Five potentially carcinogenic PAH were detected in the samples (Figure 2). BaP and BaA, for example, were detected in all leachate samples, whereas for the solubilized extract, PHA were not detected only in sample SES1. Chr was not found only in samples S2 and S3 of the leached and solubilized extracts. Sample S2 presented BbF in all methods of extractions. IcdP was detected only in the column leaching sample (CLS2). The presence of BkF and DahA was not detected. The detected concentrations of the seven potentially carcinogenic PAH in the samples are shown in Figure 2.

Similarly, Legret *et al.* (2005), in a study with RAP from a French highway, found four of the seven PAH with the greatest carcinogenic potential. They were

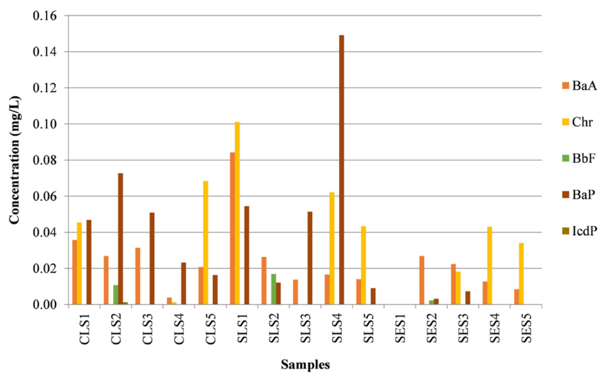
Table 3 – PAH measurement results in mg L^{-1}.

PAH	CLS1	CLS2	CLS3	CLS4	CLS5	SLS1	SLS2	SLS3	SLS4	SLS5	SES1	SES2	SES3	SES4	SES5
Naphthalene	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
Acenaphthylene	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
Acenaphthene	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
Fluorene	0.007	0.005	nd	nd	nd	nd	0.003	0.003	nd	nd	nd	0.008	0.004	nd	nd
Phenanthrene	0.021	0.04	nd	nd	nd	0.038	nd	nd	nd	nd	nd	nd	nd	nd	nd
Anthracene	nd	0.014	0.008	0.003	0.008	nd	0.019	0.009	nd	0.006	nd	0.026	0.007	nd	0.01
Fluoranthene	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
Pyrene	nd	0.013	nd	nd	0.017	nd	nd	0.014	nd	0.008	nd	nd	0.015	nd	0.017
Benz[a] anthracene	0.036	0.027	0.031	0.004	0.021	0.084	0.026	0.014	0.017	0.014	nd	0.027	0.022	0.013	0.008
Chrysene	0.045	nd	nd	0.001	0.068	0.101	nd	nd	0.062	0.043	nd	nd	0.018	0.043	0.034
Benzo[b] fluoranthene	nd	0.011	nd	nd	nd	nd	0.017	nd	nd	nd	nd	0.002	nd	nd	nd
Benzo[k] fluoranthene	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
Benzo[a] pyrene	0.047**	0.073*,**	0.051***	0.023**	0.016**	0.054**	0.012**	0.051**	0.149*,**	0.009**	nd	0.003**	0.007**	nd	nd
Indeno[1,2,3-cd] pyrene	nd	0,001	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
Dibenz[a,h] anthracene	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
Benzo[g,h,i] perylene	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd

CLS: column leaching sample; SLS: shaker leaching sample; SES: solubilized extract sample; nd: not detected; *above maximum concentration allowed by Annex G of ABNT NBR 10004:2004 (ABNT, 2004); **above maximum concentration allowed by Annex F of ABNT NBR 10004:2004 (ABNT, 2004).
Source: elaborated by the authors.

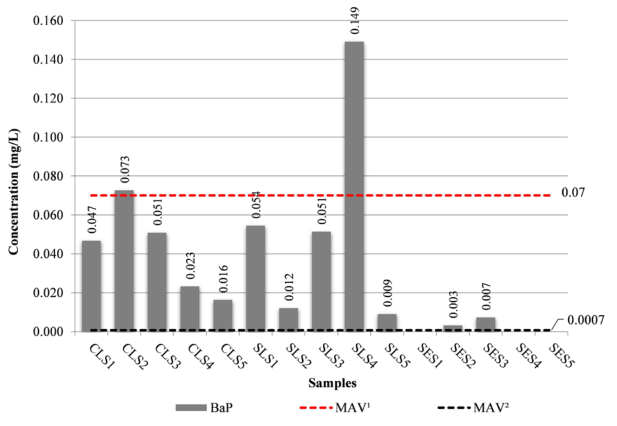
BaA, BkF, Chr, and DahA. Mehta *et al.* (2017), in a study with RAP from New Jersey (USA), found only BaA and Chr of the seven PHA with the greatest carcinogenic potential. In batch tests, the authors noted that benzo(a)anthracene was the only PAH detected at worrying levels.

In this study, BaP was present in all leachate samples, also showing the highest concentrations among the PAH detected. BaP concentrations ranged between 0.003 and 0.149 mg·L^{-1}. In samples CLS2 (0.073 mg·L^{-1}) and SLS4 (0.149 mg·L^{-1}) the concentrations of BaP were above the maximum value allowed by Annex F (Concentration — Maximum limit in the extract obtained by the leaching test) of ABNT NBR 10004:2004 (Solid waste — Classification), which is 0.07 mg·L^{-1}. In all samples of leachate by column and shaker, the detected concentration of BaP extrapolated the maximum value allowed by the Brazilian Drinking Water Legislation (0.007 mg·L^{-1}) (BRASIL, 2021). For the solubilized extract, only in samples SES2 and SES4 were detected concentrations of BaP, and their values were above the water potability standard (Figure 3).



Source: elaborated by the authors.

Figure 2 – Concentrations of the PAH found in leaching and solubilized extracts samples.



MAV: maximum allowed value: 1) for waste classification purpose in ABNT NBR 10004:2004. 2) Brazilian water potability standard (Brasil, 2021).
Convention: CLS: Column Leaching Sample; SLS: Shaker leaching Sample; SES: Solubilized Extract Sample.
Source: elaborated by the authors.

Figure 3 – Concentration of BaP detected in leaching and solubilized extract samples.

In a study with RAP in Florida (USA), Mehta *et al.* (2017) detected average PAH concentrations between 0.004 and 0.035 mg·L^{-1}. In the “fresh” sample, the lowest concentrations of PAH were detected when compared to RAP. These data demonstrate that traffic actions contribute significantly to the concentrations of PAH on pavements and, therefore, in RAP.

It is a high molecular weight PAH characteristic, such as BaP, to be emitted in the particulate phase in traffic emissions, are deposited on the runways, and consequently in the RAP, and subsequently carried out to soils and water by rainwater (RAVINDRA; SOKHI; VAN GRIEKEN, 2008; GATEAUILLE *et al.*, 2014).

Based on the information presented in this research, compared to the studies by Brown and Peak (2006), Butera, Christensen and Astrup (2014) and Zhang *et al.* (2015), it was possible to observe that rain is a potential agent for conducting organic pollutants to soils and water bodies. The disposal of this type of waste directly on the ground can impact environmental compartments, which can affect environmental health.

Comparing extraction methods, leaching extraction was more efficient in PAH extraction than solubility extraction methods, which can be explained by the physical and chemical characteristics of PAH, since they are directly linked to the transport and availability of these substances in environmental compartments (PEREIRA NETTO *et al.*, 2000). The low solubility of PAH can difficult extraction in water, as in the case of solubilization.

Acute toxicity with Daphnia magna

The calculated Daphnia Toxicity Factor (DTF) was between 16 and 64 for column and shaker leachate extract methods. For the solubilized extract, samples S1 and S3 showed DTF above 2, presenting toxicity to the *Daphnia magna* organism. The results of the toxicity analyses were validated through control evaluation, which presented DTF equal to 1. All results of the toxicity tests are summarized in Table 4.

All samples of extract leached by column, without pH correction, showed DTF of 64, that is, a dilution of 64 times would be necessary so that they did not present toxicity. For sample CLS3, which required pH correction due to its pH below 5, DTF was 16. These results expressed the acute toxicity of the samples. For the shaker’s leached extract, with the original pH of the samples, DTF was 64, and when pH was corrected, DTF was 16 for all samples.

As the extraction fluid of the leaching tests is composed of water, glacial acetic acid, and sodium hydroxide, uncertainty was generated as to whether the toxicity presented was related to the extraction fluid or the organic (PAH) and inorganic (metals) contaminants found in all samples. In this sense, the need was felt to test the toxicity of the extraction fluid, without contact with the RAP, and due to the acidity of the fluid, pH correction was necessary. DTF for the extractor fluid without pH correction was 64 and with correction was 16, that is, the toxicity presented for the leachate samples was due to the composition of the extractor fluid and not necessarily to the contaminants present in it. In a toxicity study with RAP, Mehta *et al.* (2017) also suggested that the extracted fluid was the cause of the toxicity.

For the solubilized extract, with water as the fluid extraction, samples SES1 and SES3 presented DTF equal to 2, that is, they presented acute toxicity to *D. magna*. It is worth mentioning that sample S1 was the one with the highest concentration of total PAH (Σ16PAH), which may be the cause of toxicity in *Daphnias*.

Table 4 – Results of acute toxicity of RAP in leached and solubilized extracts/

Test	Sample	pH	DTF
1	CLS1	5.07	64
2	CLS2	5.06	64
3	CLS3	4.88	64
4	CLS4	5.06	64
5	CLS5	5.07	64
6	CLS3*	6.39	16
7	SLS1	4.95	64
8	SLS2	4.98	64
9	SLS3	4.88	64
10	SLS4	4.98	64
11	SLS5	5.02	64
12	SLS1*	6.98	16
13	SLS2*	6.85	16
14	SLS3*	6.72	16
15	SLS4*	7.01	16
16	SES1	6.91	2[#]
17	SES2	7.12	1
18	SES3	7.24	2[#]
19	SES4	7.13	1
20	SES5	7.52	1
21	Extraction fluid	4.96	< 64
22	Extraction fluid*	6.79	16
23	Control Sample	-	1

[#] Samples that showed acute toxicity. CL: column leaching; SL: Shaker leaching; SE: Solubilized extract; *pH of the samples was corrected, in compliance with the standard method.
Source: elaborated by the authors.

Based on this information, it was possible to verify that samples SES1 and SES3 presented acute toxicity for the indicator organism *D. magna*. It is worth mentioning that the solubilized sample SES1 also presented concentrations of Mn above the value allowed by annex G (standards for the solubilization test) of ABNT NBR 10004:2004 (Solid waste — Classification). SES3, on the other hand, extrapolated the maximum allowed value of Cd, for annex G of ABNT NBR 10004:2004, and also for BaP for the Brazilian water potability standard. Both Cd and BaP are listed in Annex C of ABNT NBR 10004:2004 and are considered substances that render toxicity to waste.

CONCLUSION

The study demonstrated the influence of the method used for the preparation of leachate and solubilized extract from asphalt pavement residues. Although with similar granulometry and bitumen contents, there were significant differences in the presence of metals and the acute toxicity factor for daphnia. Regarding the concentrations of metals, although the leaching methods (column and shaker) have presented varying concentrations of metals for the same sample, the difference is evident compared to the solubilized extract samples. This result demonstrates that dynamic extraction is more effective and, considering the natural process of leaching of waste from asphalt pavements disposed on unprotected soils, the values obtained in the leaching methods are close to what can occur with the natural leaching process.

In relation to the analysis of PAH, the values found between the leachates and the solubilized extract were convergent and demonstrate that, for this class of environmental contaminants, the preparation method does not interfere in the analysis. This result is justified due to the low water solubility of these compounds, which tend to remain adsorbed to soil organic matter, sediments and, sometimes, to the RAP aggregate itself. Acute toxicity for daphnia magna showed significant variation between the different preparation methods and indicates the presence of toxic contaminants solubilized in the leaching methods more effectively than in the solubilized extracts.

AUTHORS' CONTRIBUTIONS

MORAES, D.: Investigation, Writing — Original Draft. SEVERINO, M.A.: Formal analysis. FREITAS, A.M.: Methodology. NAGALLI, A.: Conceptualization, Writing — Original draft, Writing — Review & Editing, Project administration, Funding acquisition. MARTINS, L.R.R.: Conceptualization, Methodology, Validation, Supervision, Resources. PAGIORO, T.A.: Methodology, Formal analysis, Writing — Review & Editing, Resources.

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