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Study of the Raveling Resistance of Porous Asphalt Pavements Used in Sustainable Drainage Systems Affected by Hydrocarbon Spills

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Abstract: Permeable pavements are one of the most commonly-used sustainable drainage systems (SuDS) in urban areas for managing stormwater runoff problems. Porous asphalt is widely used in surface layers of permeable pavement systems, where it can suffer from accidental oil spills from vehicles. Oil spills affect bituminous mixes through the solvent action of the hydrocarbons on the bitumen, reducing the raveling resistance of asphalt pavements. In order to assess the raveling resistance in porous asphalt pavements, the Cantabro abrasion test was performed on 200 test samples after applying controlled oil spills. Three different types of binders were used: conventional bitumen, polymer-modified bitumen and special fuel-resistant bitumen. After analyzing the results, it was concluded that the most suitable bitumen to protect against oil leakages is the polymer-modified one, which is far better than the other two types of bitumen tested.

Keywords: permeable pavements; sustainable drainage systems; SuDS; open graded friction course; raveling resistance

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1. Introduction

Sustainable stormwater management has become one of the most important factors in the sustainable development of urban areas. The massive waterproofing of natural land due to continuous urban sprawl has disturbed the natural water cycle [1], generating flooding problems in lowland areas, the loss of serviceability of urban infrastructures and non-point pollution effects. Moreover, the increasing runoff volumes coupled with the predominance of combined sewerage systems can increase the frequency of combined sewer overflows (CSOs) polluting the natural water bodies [2].

Permeable pavements are one of the most widely-used sustainable drainage systems (SuDS) in urban environments, helping with managing stormwater runoff and reducing the non-point pollution effects through the filtration provided by the different permeable layers that comprise their structure [2]. These sustainable systems have been widely used in low-traffic roads and in open air parking areas. One of the most common types of permeable surfaces in permeable pavements is porous asphalt, which is a special type of asphalt mix, with high void ratios, normally greater than 15%–20%, and high permeability values allowing water infiltration.

Different studies pointed out that hydrocarbon spills are one of the main pollutant sources in road infrastructures [3], producing toxic effects in natural water bodies [4]. Moreover, one of asphalt's drawbacks is its resistance loss when affected by hydrocarbon spills, limiting its use in areas with high hydrocarbon spill risk [5,6]. Motor oils and petroleum fuels are basically composed of hydrocarbons, similar to the asphalt bitumen used in porous asphalt surfaces. Therefore, by an affinity mechanism, hydrocarbon spills partially dissolve the asphalt binder [7], affecting the mechanical characteristics of bituminous mixtures, reducing their operational life and, consequently, their sustainability.

Different solutions have been developed during the last few decades to increase the resistance of bituminous binders to hydrocarbon spills. McBee and Sullivan [8] assessed the resistance of bituminous mixtures with sulfur additions, finding that the higher the sulfur content, the better the resistance to hydrocarbon damage. Moreover, until the end of the last decade, it was normal to use coal tar for treating asphalt pavements used in airports and industrial areas [9], because its chemical structure provided more resistance to the solvent action of hydrocarbons. Nevertheless, in the 1990s, tar was classified as a carcinogenic agent, and its effectiveness over time was questioned [7,10].

The rapid growth of polymer-modified bituminous binders has provided another option for asphalt pavements at risk of hydrocarbon spills. Corun *et al.* [10] pointed out the good performance of polymer-modified asphalt in the presence of jet fuel leakage, showing good mechanical response after exposure to hydrocarbons. The recent development of crumb rubber-modified asphalt (CRM) provided another possible solution to fuel spills due to the improvement in the rheological properties of the binder and the strength of the mixtures. Merussi *et al.* [11] studied the effect of kerosene on the properties of crumb rubber-modified binders, concluding that the addition of crumb rubber reduces the solubility of the binder in kerosene. Nonetheless, this reduction in solubility is not accompanied by a similar improvement in the mechanical properties, CRM mixtures providing similar performance to conventional binders.

On the other hand, the damage produced in asphalt pavements is related to the porosity and permeability of the mixtures due to the different depth of influence of the hydrocarbon spills. Prowell *et al.* [12] tested the performance of stone mastic asphalt (SMA) and dense graded mixtures, with void ratios of less than 7%, after undergoing a fuel spill. They concluded that the samples were only affected in their outer portion and retained about 80% of their original strength. This finding seems to indicate that virtually impervious asphalt pavements sustained only superficial damage due to the hydrocarbon leakage, so its main effect appears to be the loss of raveling resistance.

Although the effect of aging on porous asphalt mixtures has been studied before [5,13,14], the resistance of porous bituminous mixtures to hydrocarbon spills has not been deeply studied yet. Great efforts have been made by some authors [15,16] studying fuel damage to asphalt mixtures, including porous asphalt mixtures, through the European standard EN 12697-43 [17]. These research projects provide interesting mathematical models for predicting the abrasion losses on fuel spilled samples depending on mixture, binder and fuel characteristics, but lacked an analysis of the influence of the binder characteristics, proposing its study for future work.

The main aim of this research was to assess the influence of the most common types of hydrocarbon spills on the resistance to raveling of different porous asphalt mixtures in order to increase their sustainability over time. To achieve this aim, the Cantabro abrasion test [18] was performed on three different porous asphalt mixtures made with different bituminous binders. Different types of hydrocarbon were assessed, and various spilling loads were tested. Moreover, the void ratios and permeability of all of the porous asphalt mixtures were measured in order to check the influence of these parameters on the abrasion losses of the porous asphalt mixtures tested. Finally, the results obtained were statistically analyzed when necessary to assess the statistical significance of the differences.

2. Materials and Methods

Three different porous asphalt mixtures (PAM) were produced using three different bituminous binders: conventional bituminous binder (CBB), polymer-modified bituminous binder (PMBB) and fuel-resistant bituminous binder (FRBB). All three types of bitumen fulfilled the requirements of the Spanish technical standard [19] for its use in PAM, and their characteristics are shown in Table 1.

Table 1. Properties of bituminous binders: conventional bituminous binder (CBB), polymer-modified bituminous binder (PMBB) and fuel-resistant bituminous binder (FRBB).

Properties	**Unit**	**Standard**	**Binder**		
			CBB	**PMBB**	**FRBB**
Original Penetration (o.p) 25 °C; 100 g; 5 s	0.1 mm	EN 1426	60–70	45–80	35–50
Retained Penetration	% of o.p	EN 1426	>50	>60	>65
Softening Point (Ring and Ball)	°C	EN 1427	46–54	>60	>85
Softening Point Variation	°C	EN 1427	<9	<10	<5
Difference in Penetration	0.1 mm	EN 13399	-	<9	<5
Difference in Softening Temperature	°C	EN 13399	-	<5	<5
Fraas Breaking Point	°C	EN 12593	<–8	<–12	<–13
Solubility in Toluene	%	EN 12592	>99		>95
Elastic Recovery at 25 °C	%	EN 13398	-	>50	>15
Relative Density (25 °C/25 °C)	-	EN 3848	-		>1.025
Mass Variation	%	EN 12607-1	<0.5	<1	<0.3

Although the used binders showed some differences in their penetration range, they are reasonably similar, limiting the possible influence of this factor on the abrasion losses of PAM in relation to other binder characteristics associated with the binder type, such as the binder adhesiveness and viscosity. The binder-to-aggregate ratio was fixed at 4.5%, and the filler-to-aggregate ratio was established at 4%, both by the weight of the mixture. An ophitic material with a specific weight of 2746 kg/m^3 was used as a coarse aggregate in all mixtures, and Portland cement was used as a filler material. The coarse aggregate gradations used in all mixtures are shown in Table 2.

Table 2. Aggregate gradation used for the Marshall samples of porous asphalt.

Sieve	**20 mm**	**12.5 mm**	**8 mm**	**4 mm**	**2 mm**	**0.5 mm**	**0.063 mm**
% passing	100	85	50	14	9	5	3

In order to obtain the same viscosity values during the mixing process, the different PAM produced were mixed at different temperatures according to the manufacturer's indications, which ensures a thorough aggregate coating by the binder film. The mixing temperatures were 160 °C for CBB, 180 °C for PMBB and 155 °C for FRBB.

Cylindrical specimens of the tested PAM with a 102-mm diameter, also called Marshall samples, were compacted by 50 blows per side of a Marshall hammer, resulting in a specimens height in the range of 60–65 mm. For all of the tested samples, the total void ratio was measured by the geometrical-gravimetrical method according to the Spanish Transportation Laboratory Standard NLT-168/90 [20], and the permeability was tested by using a falling head permeameter (Figure 1).

For assessing the hydrocarbons' influence on PAM, two tests were designed and used: the semi-immersion test and the dripping and runoff test. The semi-immersion test was developed in order to assess the influence of the different types of hydrocarbons on the abrasion losses of PAM, which was based on the EN 12697-43 used for testing the resistance of asphalt mixtures to fuel action. On the other hand, the dripping and runoff test was developed in order to determine the influence of drop-by-drop hydrocarbon spills, the most common spills on roads, onto PAM surfaces when used in permeable pavements. To decide the amount of hydrocarbon to use, previous investigations on hydrocarbon retention and biodegradation in permeable pavements were taken as references [21–23].

After treating the samples with the different treatments applied, the resistance to raveling of the specimens was tested through the Cantabro abrasion test performed according to European Standard EN 12697-17 [24]. The Cantabro test have been widely used for assessing the resistance to raveling of asphalt pavements. In this test, the Marshall samples were conditioned at 15–25 °C, initially weighed (W_1) and placed in a Los Angeles steel drum for 300 gyrations at 30 rpm. Afterwards, the samples were weighed again (W_2), and the abrasion losses (AL) were calculated according to Equation (1).

$$AL(\%) = 100 \times \frac{(W_1 - W_2)}{W_1} \quad (1)$$

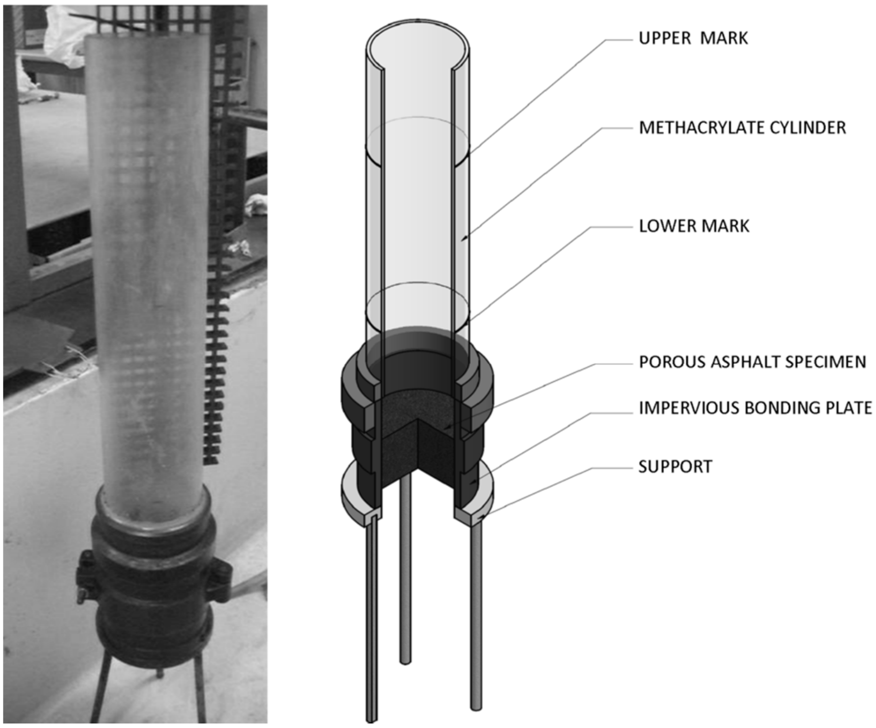


Figure 1. Falling head permeameter.

Lower abrasion losses of the specimens mean better cohesion of the mixture and better resistance to raveling, leading to higher durability of the mixture. The different road agencies that use this durability test established the limit value of the abrasion losses for the tested samples, normally being in the range of 20%–30%, depending on the traffic level and the country [25]. In addition, some road agencies also performed this test on aged and conditioned samples, increasing the limit of abrasion losses to the range of 30%–40% [25].

2.1. The Semi-Immersion Test

The semi-immersion test was developed in order to assess the aging effect of different hydrocarbons on PA samples. In this test, groups of 4 Marshall samples of each bituminous binder were placed in separate plastic buckets. Plastic buckets and samples were placed in a conditioned chamber at 20 °C. Three different treatments were assessed, and each treatment was applied to a group of samples of each bituminous binder. The different treatments correspond to the different tested hydrocarbons: gasoline, diesel fuel and used motor oil. A fixed load of 339 mg of the tested hydrocarbon was directly applied onto the PAM specimens, and 200 mL of distilled water were also applied to simulate the rainfall runoff effects. This process was repeated 4 times in two days, once each 12 h, resulting in a total hydrocarbon load of 1356 mg and 800 mL of distilled water applied onto each Marshall sample. Afterwards, the specimens were left in the buckets for three days, partially submerged in the water, and the hydrocarbons drained from the samples, reaching depths in the range of 3.5 cm. Finally, the samples were cleaned, air dried for 24 h at 20 °C and tested according to the Cantabro test, fixing the test temperature at 20 °C. In addition, two control treatments were also used for comparing the results obtained. One control treatment consists of applying the above-mentioned procedure, but only using distilled water. On the other hand, the other control treatment consists of maintaining the Marshall samples in the conditioned chamber at 20 °C without applying any other treatment during the full five days of the test.

2.2. The Dripping and Runoff Test

The dripping and runoff test was developed based on previous studies [9] in order to assess the aging effect of hydrocarbon spills onto PAM surfaces in the real conditions that occur in permeable pavements. In this test, used motor oil was utilized as the hydrocarbon (Table 3), and different spill loads were tested: 271, 814, 1356, 1898 and 2441 mg. Each sample was placed in stands that let them drain from the bottom, and a quarter of the total oil spill loads tested was applied by a drop-by-drop flow. Afterwards, 200 mL of distilled water were poured by long-term dripping for 7 days, during which both the hydrocarbon and water have time to penetrate into the mixture. This process was repeated four times, and the examination took place on the 28th day, taking the samples out of the stands and keeping them at 20 °C for 24 h before testing their raveling resistance through the Cantabro test, fixing the test temperature at 20 °C. In this test, the samples and stands are also placed in a conditioned chamber with a fixed temperature of 20 °C during the whole test.

Table 3. Chemical composition of used motor oil.

HYDROCARBON BANDS					**COMPOSITION**		
Band	**Area**	**Area (%)**	**Aliphatic (% in Weight)**	**Aromatic (% in Weight)**	**Compound**	**Area**	**Area (%)**
C5-C6	237,608	0.71	0.71	0.00	Benzene	1154	0.00
C6-C8	273,796	0.82	0.62	0.19	Toluene	64,953	0.19
C8-C10	590,169	1.76	0.03	1.73	Ethyl-Benzene	34,932	0.10
C10-C12	166,720	0.50	0.03	0.47	Xylene	19,984	0.60
C12-C16	201,756	0.60	0.26	0.35	Octane	6777	0.02
C16-C21	1,191,096	3.55	2.16	1.40	Nonane	3801	0.01
C21-C35	5,783,999	17.26	15.98	1.27	Decane	6446	0.02
>C35	25,069,548	74.80	-	-	Undecane	3517	0.01

The experimental design adopted for the semi-immersion test and the dripping and runoff test is summarized in Table 4. The data obtained from the laboratory experiments were statistically analyzed by using IBM SPSS 22® software with the aim of establishing the representativeness of the results obtained and finding the relationship among the void ratio, permeability and abrasion loss. In order to establish the significance of the differences observed among the results obtained for each binder and treatment, different statistical tests were done: a non-parametric test for non-normally-distributed parameters and a parametric test for homoscedastic parameters which follows a normal distribution [26]. All of the statistical analyses were performed at a confidence level of 95%, accepted as a standard in the statistical analysis [27]. Furthermore, the tested hypothesis in the performed statical analyses was accepted as true when the statistical significance (sig) is lower than 0.05.

Table 4. Experimental design for the semi-immersion test and the dripping and runoff test.

Hydrocarbon Spill Test	**Binder Type**	**Treatment**	**Number of Samples (Ni)**
Semi-Immersion test	CBB, PMBB, FRBB	Gasoline: 1356 mg + Distilled water: 800 mL	4 *
		Diesel: 1356 mg + Distilled water: 800 mL	4 *
		Used motor oil: 1356 mg+ Distilled water: 800 mL	4 *
		Distilled water: 800 mL	4 *
		No treatment	4 *

Table 4. *Cont.*

Hydrocarbon Spill Test	Binder Type	Treatment	Number of Samples (Ni)
Dripping and Runoff test	CBB	Used motor oil: 271 mg, 814 mg, 1356 mg + Distilled water: 800 mL	12 **
		Distilled water: 800 mL	12
		No treatment	12
	PMBB	Used motor oil: 814 mg, 1356 mg, 1898 mg + Distilled water: 800 mL	12 **
		Distilled water: 800 mL	12
		No treatment	12
	FRBB	Used motor oil: 1356 mg, 1898 mg, 2441 mg + Distilled water: 800 mL	12 **
		Distilled water: 800 mL	12
		No treatment	12

* Per each one of the three binder types (12 in total); ** Per each one of the three quantities of used motor oil in the treatment (36 in total).

3. Results and Discussion

3.1. Semi-Immersion Test

The results obtained in the semi-immersion test were used to check the influence of the different types of hydrocarbon normally spilled on roads. Figure 2 shows the air voids, permeability and abrasion losses of the tested PAMs, for the different treatments applied onto the porous asphalt samples. It can be observed that the abrasion losses for the hydrocarbon-treated samples were very similar to those obtained for water-treated samples and higher than those obtained for air conditioned samples, indicating the important effect of water in the abrasion losses of PAM. However, it can also be observed that diesel fuels and used motor oil seem to have a higher degradation capacity than gasoline fuels or water.

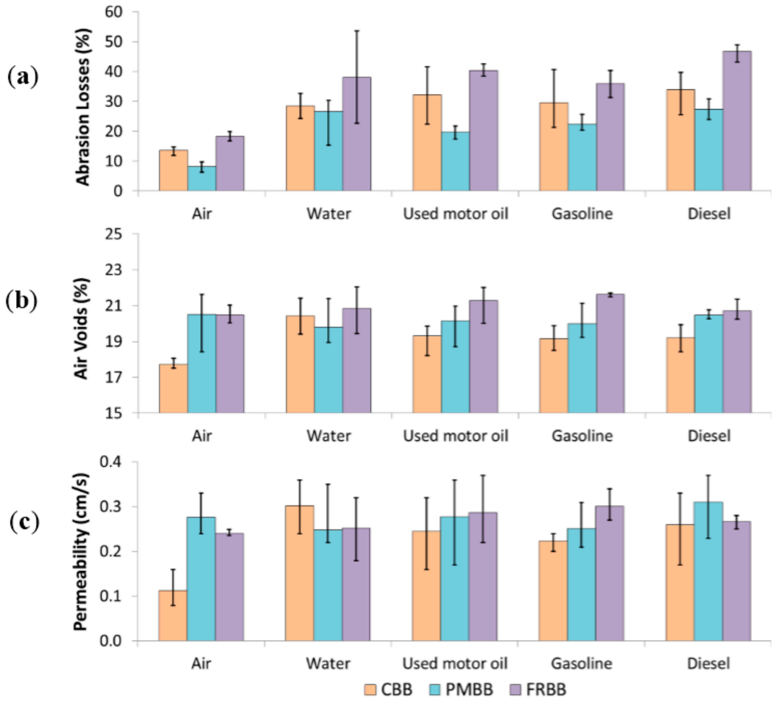


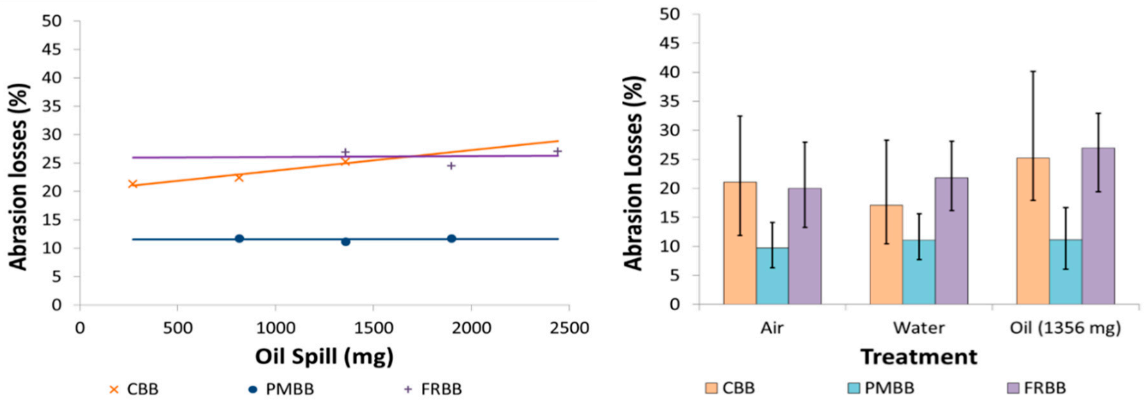
Figure 2. Results of the semi-immersion test: (**a**) abrasion losses; (**b**) air void content; and (**c**) permeability.

The statistical analysis showed that the abrasion losses of the samples sorted by treatment and binder were normally distributed and homoscedastic. Furthermore, parametric statistics can be used to analyze the influence of these parameters on the abrasion losses. A factorial ANOVA test was performed in order to assess the influence of the bitumen type and the treatment applied on the abrasion losses of the samples. The results of this test showed that there are significant differences among the different treatments and the binders used. Tukey HSD post-hoc analysis was performed to define homogenous groups of samples depending on the abrasion losses, in relation to the treatments applied and the bituminous binder used. The results of these tests showed that each binder displayed significantly different abrasion resistance to each other (sig < 0.017) with higher resistance to raveling for PMBB. Moreover, air-treated samples showed significantly lower abrasion losses than the other samples (sig < 0.001), while there are no significant differences among the abrasion losses for the other treatments independent of the binder used (sig > 0.265), indicating similar degrading effects of all of the hydrocarbons tested, similar to those obtained with only distilled water. The effects of the air void content and permeability were assessed by performing a factorial ANCOVA test, which showed that there is no significant influence of these parameters on the abrasion losses of the samples for the different treatments and binders (sig > 0.138). In fact, the R squared provided by the factorial ANOVA performed to check the influence of binder type and treatment on the abrasion losses ($R^2_{\text{ANOVA}} = 0.794$) was very similar to the one obtained by the factorial ANCOVA with the same parameters while controlling the effects of air voids and permeability ($R^2_{\text{ANCOVA}} = 0.796$). Furthermore, according to this test, the different hydrocarbons tested have similar degrading effects on PAM specimens independent of the air voids of the mixtures and similar to those obtained for water-treated specimens. It should be noted that the results obtained are only from four samples of each PAM affected by each treatment, considered enough according to the standards selected as the reference, EN 12697-43 and EN-12697-17, in which the number of required tested samples is three.

3.2. Dripping and Runoff Test

The dripping and runoff test was performed to assess the influence of drop-by-drop spills, the most common type of spills from vehicles, on the abrasion losses of PAMs in permeable pavement systems. Used motor oil was selected as the most common hydrocarbon source, taking into account that the results obtained in the semi-immersion test showed similar degrading effects obtained for all of the hydrocarbons tested.

The results obtained for this test are shown in Figure 3. Analyzing the average abrasion losses for the 12 samples tested for each level of used motor oil load (Figure 3a), it can be stated that the variation in the hydrocarbon loads only affected CBB mixtures, increasing the abrasion losses, while PMBB and FRBB mixtures showed similar abrasion resistance independent of the oil spill load.



(**a**) (**b**)

Figure 3. Results of the dripping and runoff test: (**a**) average abrasion losses for the tested levels of used motor oil (Ni = 12); (**b**) abrasion losses for the common treatments applied to the different porous asphalt mixtures (PAM) (Ni = 12)

Analyzing the abrasion losses for an oil spill load of 1356 mg and 800 mL of distilled water and comparing them to the control treatments, it can be observed that the oil spill increased the abrasion losses of CB mixtures and FRB mixtures in relation to control treatments. PMBB mixtures showed the best performance for the analyzed treatments in the dripping and runoff test, showing average abrasion losses of 10.0%, 11.1% and 11.2% for air-treated samples, water-treated samples and oil spilled samples, respectively, as can be observed in Figure 3b. CBB and FRBB mixtures have the worst performance and suffer average abrasion losses greater than 20% for all of the analyzed treatments. The statistical analysis showed that the abrasion losses sorted by binder and treatment were non-normally distributed or non-homoscedastic. Furthermore, non-parametric statistics were used for further analysis. A non-parametric Kruskal–Wallis H-test was performed to determine whether there are significant differences among the abrasion losses depending on the treatment applied and the binder used. The results of these tests showed that for the same treatment, there are significant differences in the abrasion losses depending on the binder used (sig < 0.002). In addition, for the same bituminous binder, there are significant differences among the different treatments for CBB and FRBB mixtures (sig < 0.002), while PMBB mixtures showed similar abrasion losses independent of the treatment (sig < 0.392). Multiple pairwise Mann–Whitney U-tests were performed to determine among which groups there are significant differences. The results obtained showed similar abrasion losses of CBB and FRBB mixtures for air-treated samples and water-treated samples (sig > 0.291). However, these PAM showed significantly higher abrasion losses for oil-treated samples in relation to the other treatments analyzed (sig < 0.002), there being no significant differences between these PAM with oil spilled samples (sig > 0.068).

The influence of the air voids and permeability on the abrasion losses for the different mixtures was analyzed in three scenarios, as is shown in Figure 4:

-Air conditioned samples at 20 °C;
-Air conditioned samples at 20 °C with 800 mL of distilled water;
-Air conditioned samples at 20 °C with 1356 mg of used motor oil and 800 mL of distilled water.

Observing the relationships between void ratios and abrasion losses, it can be stated that total void content and permeability were generally correlated with abrasion losses in CBB mixtures for the treatments analyzed. Analyzing the performance of PAMs with modified bituminous binders, it can be observed that abrasion losses in PMBB mixtures do not seem to be affected by void content for control treatments, while a significant correlation was observed between void content and abrasion losses for oil-treated samples, indicating some influence of contact points between binder and oil on the abrasion losses of PMBB mixtures. On the other hand, abrasion loss of PAM with FRBB shows significant correlation levels between abrasion loss and void content for control treatments, indicating some influence of the compactness of PAM and water contact points on abrasion loss. Interestingly, FRBB showed similar abrasion losses independent of the air void content and permeability when affected by oil spills, but with very high abrasion loss values, higher than those obtained for similar air voids with only distilled water.

The higher the air void ratio in the mixtures, the higher the abrasion losses. In fact, higher values of void content imply lower compactness of the mixture, theoretically reducing the resistance to raveling. Moreover, the higher the air voids of PAM, the deeper the influence of water and hydrocarbons into the mixture, increasing the contact points of hydrocarbons with the binder, thus increasing the effect on the bitumen. According to the results obtained, the use of void ratios not greater than 20% could be recommended in order to ensure abrasion losses under the range of 20%–30% for the conditions analyzed in Figure 4.

Finally, comparing the results obtained for the semi-immersion test and those measured in the dripping and runoff tests for the common treatments, oil spill load of 1356 mg and 800 mL of distilled water and the control treatment with distilled water (Figure 5), it can be observed that the degrading effect of water and hydrocarbon spills was higher for the semi-immersion test.

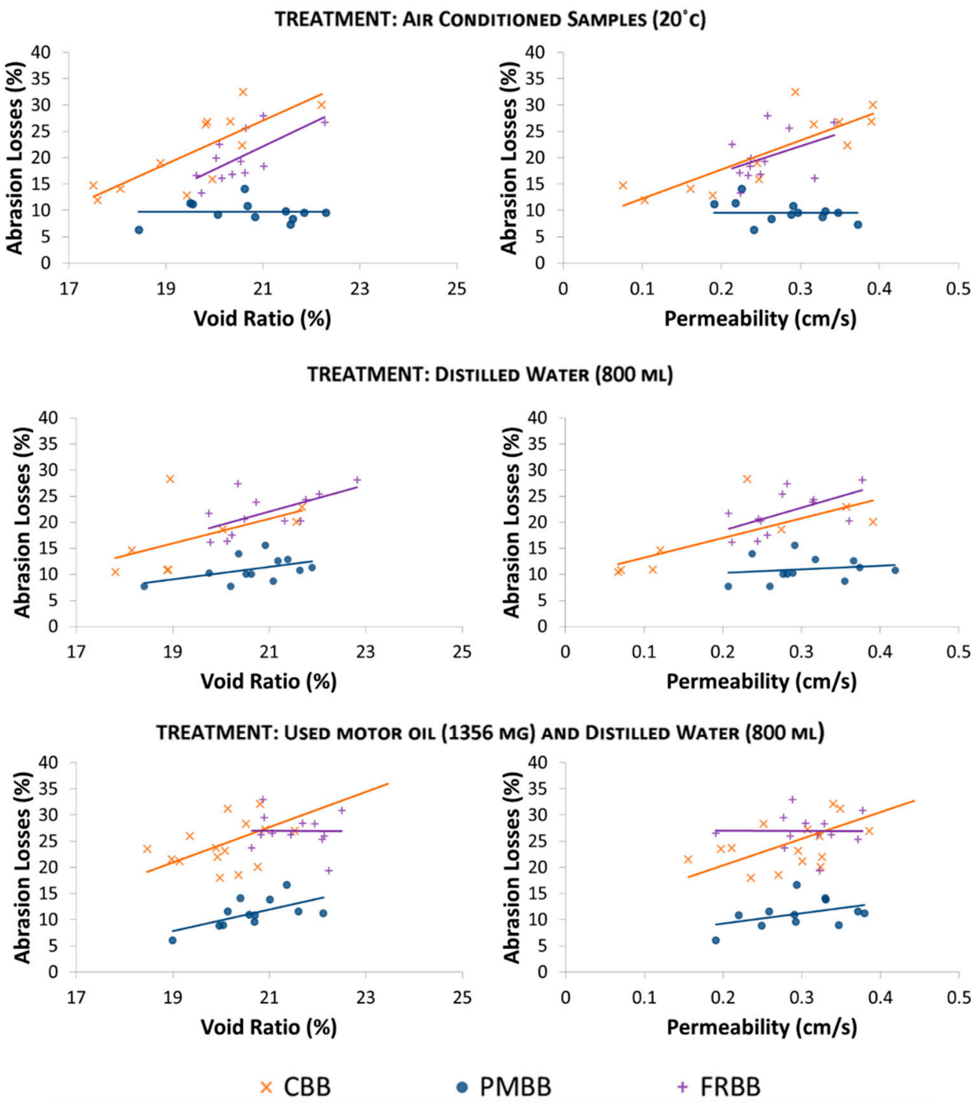


Figure 4. Relationships of the air void content and permeability to the abrasion losses of the samples for the three common treatments applied to the different bituminous binders tested.

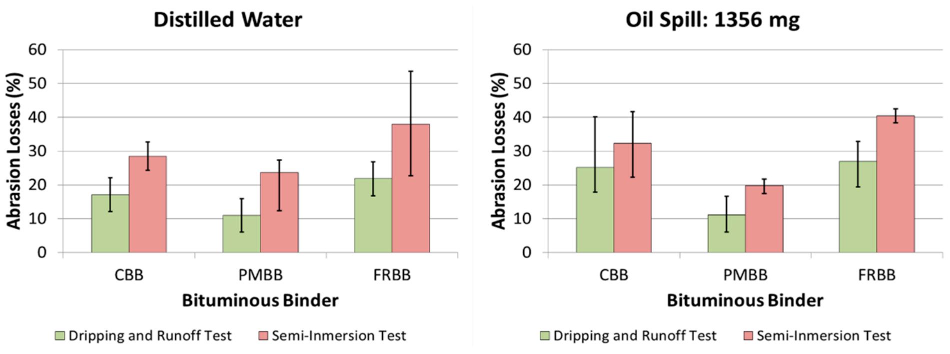


Figure 5. Comparison of the abrasion losses obtained for the semi-immersion test and dripping and runoff test.

The special conditions of permeable pavements were reflected in the dripping and runoff test, in which the water and the spilled hydrocarbons can flow through the surface to the lower layers, reducing the contact time of the hydrocarbons with the bituminous binder, resulting in lower abrasion losses. In this situation, the conditions imposed by the semi-immersion test were unrealistic due to the permeability of the whole cross-section of the permeable pavements. Furthermore, the semi-immersion test tends to overestimate the abrasion losses.

4. Conclusions

The semi-immersion test results showed similar abrasion losses for the PAMs studied independent of the air void content and permeability of the mixtures, for all of the hydrocarbons tested, which were similar to those obtained for water-treated samples.

The dripping and runoff test results showed the influence of the air void content on the abrasion losses, also enabling the identification of significant differences in the abrasion losses for hydrocarbon-treated samples in comparison with both water-treated and air conditioned samples. Furthermore, the dripping and runoff test provides a more accurate method than the semi-immersion test for assessing the influence of hydrocarbon spills on PAM used in permeable pavement systems.

PMBB mixtures have shown the lowest abrasion losses for all of the treatments applied in both the semi-immersion test and the dripping and runoff test, not being affected by the different types or loads of hydrocarbons spilled on the mixtures.

FRBB mixtures showed the highest abrasion losses, for all of the hydrocarbons tested and for the different oil spill loads. However, the different loads of hydrocarbons resulted in similar abrasion losses, indicating that this binder, although affected by hydrocarbons, was not influenced by the quantity range applied.

The void content and permeability of the PAMs have been demonstrated to influence the effect of the hydrocarbons on the abrasion losses of the CBB and PMBB mixtures tested by the dripping and runoff tests: the higher the air void content in mixtures and the greater their permeability, the higher the abrasion losses.

Considering all of the results obtained, PMBB has shown the best resistance to abrasion loss, being the best option to be used in PAMs to ensure their sustainability over time, even in areas with high risks of oil spills. On the other hand, CBB and FRBB mixtures affected by hydrocarbon spills underwent abrasion losses that preclude their use where hydrocarbon spills are expected.

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Author Contributions: Miguel A. Calzada-Pérez, Ángel Vega-Zamanillo and Daniel Castro-Fresno designed the research and discussed the results. Jorge Rodriguez-Hernandez performed the research and wrote the paper. Valerio C. Andrés-Valeri analyzed the data, discussed the results and wrote the paper. All authors read and approved the final manuscript.

Conflicts of Interest: The authors declare no conflict of interest.

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