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Preliminary analysis of the leachability of organic compounds into surface waters from waste reclaimed asphalt

*Wstępna analiza wymywalności związków organicznych
do wód powierzchniowych z odpadowego destruktu asfaltowego*

DOI: 10.15199/33.2024.06.08

Abstract. The Regulation of the Minister of the Environment of 2021 allows for the removal of the waste status from reclaimed asphalt. However, such a material must meet guidelines regarding, among others, the content of organic pollutants, in particular polycyclic aromatic hydrocarbons (PAHs). Unfortunately, the use of material without analysis may involve a number of threats to existing ecosystems, including aquatic ecosystems. In the article the authors present the results of the analysis of the leachability of organic compounds into the aquatic ecosystem. The results of testing samples of reclaimed asphalt for 10 streets in the city of Płock are presented. The research were carried out to analyze chemical properties that could pose a potential threat to the environment.

Keywords: recycled asphalt; natural environment; waste status; pollution.

Streszczenie. Rozporządzenie Ministra Środowiska z 2021 r. pozwala na zdjęcie statusu odpadu z destruktu asfaltowego. Materiał taki musi jednak spełniać wytyczne dotyczące m.in. zawartości zanieczyszczeń organicznych, w szczególności wielkocząsteczkowych węglowodorów aromatycznych (WWA). Zastosowanie materiału bez odpowiedniej analizy może wiązać się z wieloma zagrożeniami w stosunku do istniejących ekosystemów, w tym wodnych. W artykule zaprezentowano wyniki analizy wymywalności związków organicznych do ekosystemu wodnego. Przedstawiono wyniki badań próbek destruktu asfaltowego pobranych z 10 ulic w obrębie miasta Płocka. Badania realizowano pod kątem analizy właściwości chemicznych mogących stanowić potencjalne zagrożenie dla środowiska.

Słowa kluczowe: destruktu asfaltowy; środowisko naturalne; status odpadu; zanieczyszczenia.

According to the norm [1], asphalt destructure is defined as material from the designed asphalt layers or plates broken from the asphalt surface. The destructure is also called mineral-asphalt mixtures (MMA) rejected from production or, which is a production surplus, which can be a component of the newly created MMA after meeting the criteria indicated by the norm [2].

Until 2021, asphalt destructure in Poland was treated as waste. This waste could not be supplemented with cavities in road surfaces or cure local roads. The abolition of waste status was aimed at

facilitating the use of material in road investments from both the administrative and implementation. Regulation [3] determining the detailed conditions for the loss of waste status for asphalt destructure waste defines a number of guidelines, after which asphalt destructure, i.e., recycled material of asphalt surfaces, will cease to be treated as waste [4].

The use of asphalt destructure in road projects is associated with economic and ecological benefits. First, the secondary use of the designed surfaces allows you to reduce greenhouse gas emissions and energy consumption, especially in the aspect of limiting the operation of natural aggregate deposits [5 – 7]. In addition, research conducted by Zhang et al. [8], Giani et

al. [9] and Hasan et al. [10] confirm that the introduction of destructure to the newly created mineral-asphalt mix has a positive effect on the strength parameters of the shaped surface. This allows the implementation of materials with comparable or even better mechanical properties relative to the mixture without destructure [8 – 10]. Among the leading economic benefits related to the re-building of the destructure, there is primarily a reduction in the total costs of making the surface relative to mixtures created exclusively based on ingredients of primary origin. In addition, the storage of destructure generates costs, and efficient re-introduction of material in circulation also allows savings in this respect [11, 12].

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The secondary use of the designed asphalt material is associated with the possibility of adverse impact of material on the environment, especially the aquatic environment. Traffic taking place on the road is more burdensome for the environment than the construction and maintenance of the road through the period of use [11, 13]. However, because of sunlight, wind and rainwater, the components of the mixture, including toxic, harmful compounds, are released into the groundwater [14]. According to Yang et al. [14] and Chen et al. [15] the weathering or aging of asphalt showed a clear effect of reducing the concentration of inventive metal concentration in experiments on the flow column, such as AS, CR, CU, Ca, K, Li, Mg, Mn, V. In many literature positions, threats related to leaching have been shown in many literature positions by water, trace amounts of heavy and multi-line aromatic hydrocarbons (WWA). In addition, the older the destructors, the higher the leaching of the ingredients [16, 17]. It is influenced by the dynamic development of the surface technology. Unwanted substances can also arise during road use, which is influenced by, among others, vehicle consumption and the condition and type of tires [18].

Building asphalt destructors in previously undeveloped areas reduces the infiltration of precipitation to groundwater. As a result, surface rafting is mixed with chemical compounds contained in the building material of the newly created surface [19]. The harmfulness of accumulating pollution results, among others, from their type, as well as from the technical condition of vehicles moving on the surface and fuels used by them. Additional factors affecting the quality of asphalt destruct are local speed limits and permissible tonnage of vehicles, road maintenance methods, as well as the geology of the building area [20].

Based on research [17, 21, 22] regarding the leaching of asphalt pollutants, it is known that in addition to the type and method of processing asphalt materials to the pace and level of leaching of chemical compounds, the temperature of the liquid phase (surface water) and the fixed phase

(asphalt), as well as pH of solutions formed from rainwater. The study [22] showed that the highest PAW concentrations were recorded for pH = 4 [22]. Funds based on sodium chloride, i.e., salts, having neutral reaction used to maintain roads in winter, can affect the properties of asphalt and contribute to the penetration of unwanted compounds into surface water. According to the research [23], salt dissolving reduces asphalt durability with long-term surface use. Salt dissolution adversely affects the properties of the surface because they exert the greatest impact on the water resistance of mineral-asphalt mixtures, followed by elevated temperatures, and in a lesser extent to less resistance to cracking at low temperatures [23].

Despite the growing outlays allocated to the renovation and construction of new roads in Poland, the percentage of dirt roads, especially municipal roads, is still high. The lack of sufficient funds for the construction of roads, forces managers to take actions aimed at curing dirt roads with various materials available on the market. In these circumstances, broken aggregate found widely [24]. The biggest drawback of this type of surface, apart from dusting, is, however, the need for regular procedures ensuring equality of surfaces. The search for more durable material, more resistant to the formation of unevenness, meant that the managers to harden dirt paths began to use asphalt destruct. Material, which in connection with numerous repairs of Polish roads appeared a lot on the market and was easily available. Classification of destruct as waste did not prevent the managers in its use before 2021, i.e., before the period of departure [3] specifying the detailed conditions for the loss of waste status for asphalt waste. The asphalt destructures used were not subject to basic tests and it is difficult to determine today whether they are safe for the environment after the embodiment.

The aim of the work was to analyze the leaching of organic compounds that are a component of binder in asphalt destruct. Research was focused on the quantitative designation of compounds

dissolving in various environments, which allowed to assess the impact of weather conditions, rain on the asphalt destructures used. Test material was downloaded from ten sections of roads hardened by destructure in the city of Płock. As part of the presented work, an assessment of potential threats to the environment related to the possibility of leaking by rain by organic compounds to surface waters and resulting from the construction of secondary materials.

Methodology

In Poland, guidelines regarding the construction of asphalt destructure into the structure of the surface were defined in the regulation of the Minister of the Climate and the Environment [3]. There are a number of conditions necessary to meet that the recovered asphalt material is not qualified for waste. The provisions contained in the regulation include requirements for the content of pollution other than those formed during the production, use and use of mineral and-asphalt mixtures, and the occurrence of multi-line aromatic hydrocarbons. In addition, the content indicates standards specifying the methods of taking sampling for testing and requirements set to asphalt granules.

As part of the research program, the samples were examined for the presence of multi-line aromatic hydrocarbons in accordance with the guidelines contained in Annex 2 of the Regulation [3] and the designation of the concentration of organic compounds using solutions with acid, neutral and alkaline reaction.

The collection of material for the tests was carried out in accordance with the PN-EN 12697-27 standard by drilling from a concentrated layer. With the help of a drill, spiral samples with a diameter of 100 mm and height covering the full thickness of the analyzed surface of the surface were drew [25].

Sampling of samples for testing was carried out on the streets, located within the city of Płock, where asphalt destructing was used to harden the ground surface. As part of research in consultation with the Municipal Road Administration in Płock – an institution

managing public roads within the administrative borders of Plock, all roads, which in 2018 – 2023 were hardened with asphalt destruct in 2018 – 2023. Due to the identification of ten such streets (Zajęcza, Jeziorna, Lowen, Boryszewska, Browarna, Kraków, Parcele, Powiśle, Janówek and Rozajna), in order to make the most reliable analysis of chemical properties that could pose a potential threat to the environment, it was decided to take samples on the samples each of them. The location of individual streets is shown in Figure 1.



Fig. 1. Map of Plock with marked streets where samples were taken for testing; streets: 1 – Zajęcza; 2 – Jeziorna; 3 – Nizinna; 4 – Boryszewska; 5 – Browarna; 6 – Krakówka; 7 – Parcele; 8 – Powiśle; 9 – Janówek; 10 – Urodzajna

Rys. 1. Mapa Płocka z zaznaczonymi ulicami, z których pobierano próbki do badań; ulica: 1 – Zajęcza; 2 – Jeziorna; 3 – Nizinna; 4 – Boryszewska; 5 – Browarna; 6 – Krakówka; 7 – Parcele; 8 – Powiśle; 9 – Janówek; 10 – Urodzajna

Properly prepared samples of secondary asphalt material with a mass of 10 g were flooded with one of three solutions – alkaline NaOH, acid HNO₃ and indifferent H₂O, which is to correspond to the varied conditions of surface operation. The degree of invention of organic compounds was analyzed after one, three and five months after the preparation of the samples. Then, after a specified time, the water phase was examined, in which samples were placed to determine the level of

leaking organic compounds. The analysis was performed in the Shimadzu TOC-L analyzer. Total carbon analyzer enables the testing of total carbon content (TC) (in the range of 50 ppb – 300,000 ppm). The TOC-L analyzer had NDIR infrared detector. Samples of water lifts were placed in the automatic sample feeder and analyzed TC (total coal). As a result of the combustion of TC, carbon dioxide formed in the pipe, which after cooling and dehydration moved through the halogens catching captable leading to its detection in the NDIR detector (infrared non-terrifying).

Analog NDIR detection signal created a peak whose field is proportional to the TC concentration in the sample. Based on calibration curves, the percentage content of organic compounds in the examined asphalt destructors was determined.

The test for the presence of multi-line aromatic hydrocarbons was performed using a simplified method, in accordance with the guidelines contained in the regulation [3]. Before the examination, the sample was dried and cleaned of particles applied during consumption and transport. Then the surface of the sample was covered with Pak Detector Spray (Soppec Construcion). The preparation occurs

in the form of spray paint, which facilitates the identification of the presence of tar at road surfaces by detecting multi-linear compounds of aromatic hydrocarbons (WWA). As a result of PAW in the concentrations below 0.5 to 25 mg/kg, the preparation changes color from white to yellow. The study was carried out in two lighting conditions, in daylight and in a dark room with UV rays. The need to conduct a study in UV light results in the guidelines contained in Annex 2 of the Regulation [3], where in the

absence of a change in the color of the preparation in daylight, the samples should be illuminated with ultraviolet radiation.

Results

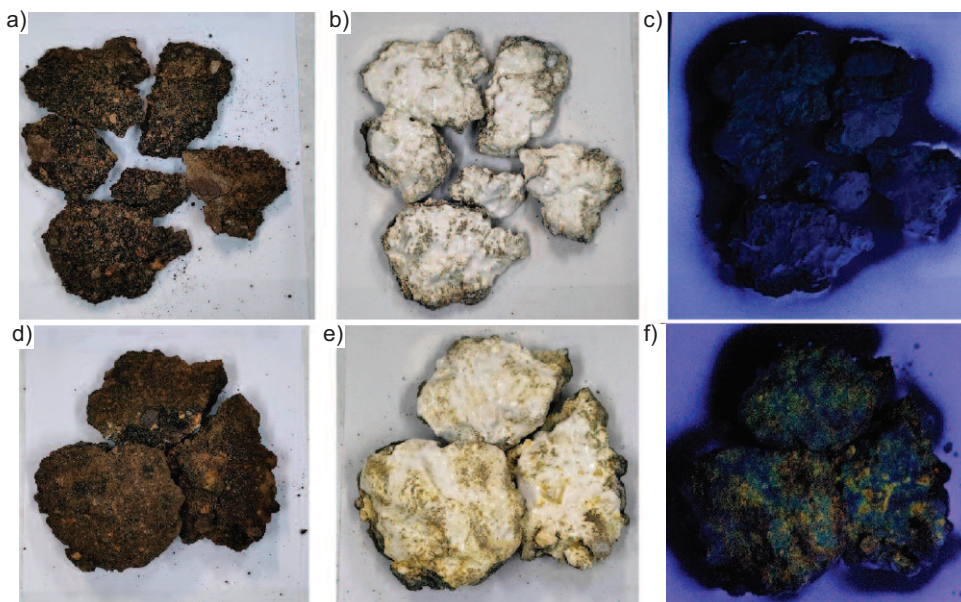
Results of research for the presence of multi-line aromatic hydrocarbons (Table) indicate that on two out of ten analyzed streets, on Nizinna and Browarna Streets, changes in the color of the preparation covering the surface of white to yellow samples, both in daylight and in UV (Photo). This means that the structure of the material contains undesirable compounds of WWA.

The simplified test method only allows you to demonstrate the presence of multi-line aromatic hydrocarbons in asphalt destruct. Pursuant to the Regulation [3], for the destruct to be deprived of the status of waste, the content of benzo(a)pyrene (BaP) and the total PAW content may not exceed the maximum permissible concentrations indicated in the document. Because the preparation did not indicate a pure color change over the entire surface of the sample, but only on its fragment did not take the quantitative analysis of the content of individual WWA.

As a result of leaching carbon compounds from destructive samples in sodium hydroxide solution, the concen-

Test results (simplified method) for the presence of polycyclic aromatic hydrocarbons
Wyniki badań (metodą uproszczoną) obecności wielopierścieniowych węglowodorów aromatycznych

Street	Color light change	Color change in UV light
Zajęcza	no changes	no changes
Jeziorna	no changes	no changes
Nizinna	change to yellow	change to yellow
Boryszewska	no changes	no changes
Browarna	change to yellow	change to yellow
Krakówka	no changes	no changes
Parcele	no changes	no changes
Powiśle	no changes	no changes
Janówek	no changes	no changes
Urodzajna	no changes	no changes



Samples for testing for polycyclic aromatic hydrocarbons (PAHs): a and d – material for testing before covering with white preparation; b and e – examination in daylight; c and f – examination under UV light; a ÷ c – reclaimed sample taken at Boryszewska – no color changes; d – e – samples taken at Nizinna – clear color changed to yellow

Próbki do badań na obecność wielopierścieniowych węglowodorów aromatycznych (WWA): a i d – materiał do badań przed pokryciem białym preparatem; b i e – badanie w świetle dziennym; c i f – badanie w świetle UV; a ÷ c – destrukcja pobrana na ul. Boryszewskiej – brak zmian koloru; d – e – próbki pobrane na ul. Nizinnej – wyraźna zmiana koloru na żółty

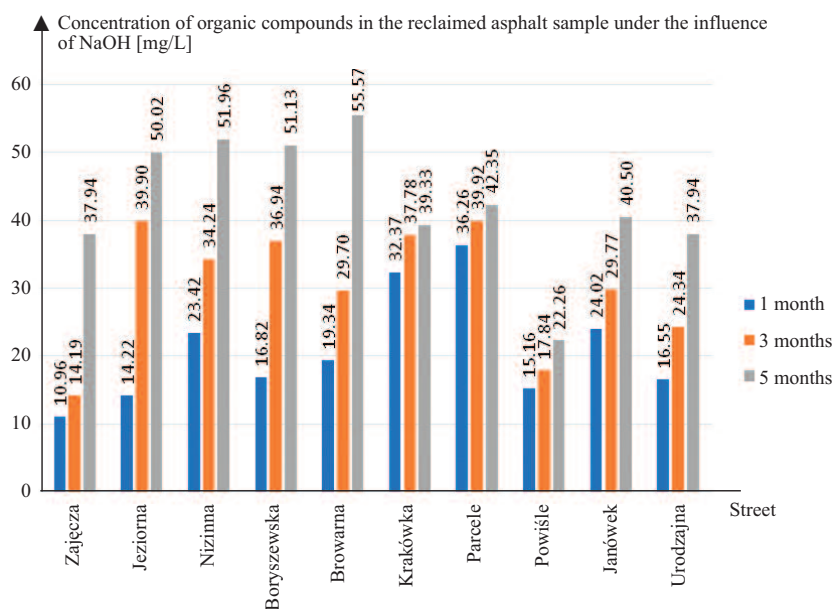


Fig. 2. Results of testing the concentration of organic compounds in samples of reclaimed material stored in NaOH solution at pH = 10 for one, three and five months

Rys. 2. Wyniki badań stężenia związków organicznych w próbkach destrukcji przechowywanego w roztworze NaOH o pH = 10 przez jeden, trzy i pięć miesięcy

tration of organic compounds varies depending on the place of material intake and sample storage time (Figure 2). The designation carried out after a month showed results in the range from 10 mg/l to about 37 mg/l, after three months the results increased

from 10% (samples from Parcele Street), to 180% (samples from Jeziorna Street) in relation to the state after a month. After five months, another increase was recorded in the range from 4 (Krakówka Street) to 167% (Zajęcza Street) relative to the preceding study.

The research on the leaching of organic compounds from destructors after storing samples in the alkaline solution shows that the lowest susceptibility to leaching in the analyzed periods is characterized by samples from Powiśle Street. In the case of Krakówka and Parcele streets, between the degree of leaching after a month, and after five months of impact of the solution, minor changes were observed, up to 7%. With the passage of time, the ranking of sample from samples on the Zajęcza Str., Jeziorna Str. and Boryszewska Str. (more than a three-time increase in concentration after five months of leaching in NaOH in relation to the results after a month).

The results of testing the sample that were in the solution of nitric acid (V) are characterized by less diversity relative to sodium hydroxide samples (Figure 3).

The designation carried out after a month showed results in the range from 7 mg/l to about 26 mg/l, after three months the results increased from 3% (Powiśle Street), to 270% (Nizinna Street) relative to the state from a month ago. After five months, another increase in the range of 10% (Janówek Street) to 50% (Urodzajna Street) compared to the study after three months. In addition, in the case of samples of destruct from Zajęcza Street after five months of leaching, there was a decrease in the concentration of leaked compounds relative to the study conducted after three months. This means that organic compounds found in the sample were washed out.

The designation of the concentration of organic compounds in destructors after storing samples in the acid solution shows that the greatest susceptibility to leaching is characterized by a sample taken from Krakówka Street. In the sample with Nizinna Street there was almost five times an increase in the concentration of organic compounds after five months of leaching in HNO₃. With the passage of time, the degree of leaching from samples on the Powiśle and Zajęcza Streets increased.

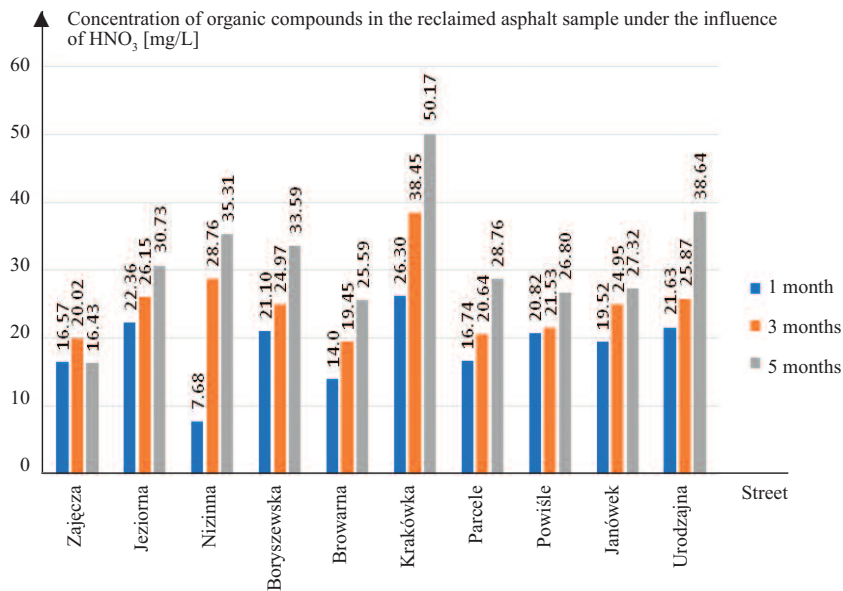


Fig. 3. Test results for the concentration of organic compounds in samples of reclaimed material stored in a solution of HNO₃ at pH = 4 for one, three and five months
 Rys. 3. Wyniki badań stężenia związków organicznych w próbkach destruktu przechowywanego w roztworze HNO₃ o pH = 4 przez jeden, trzy i pięć miesięcy

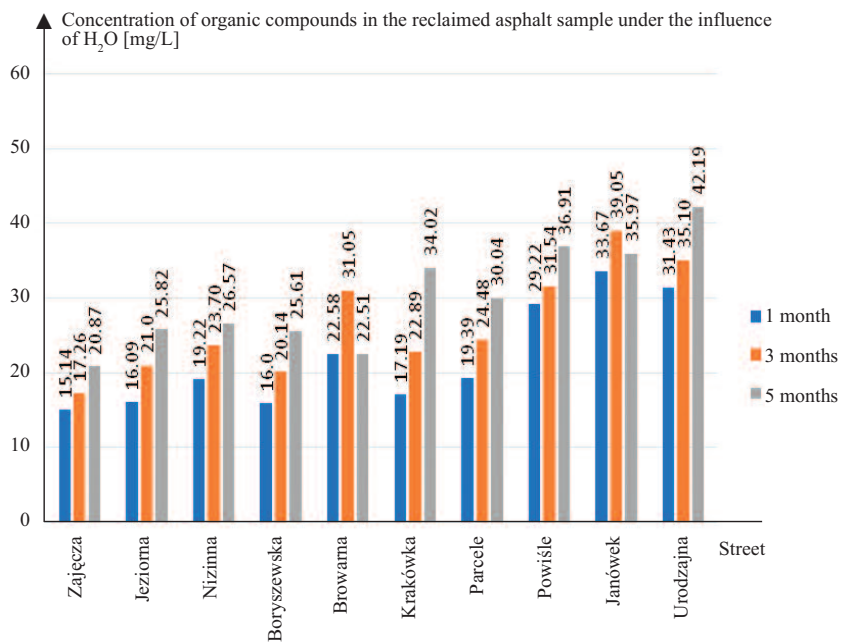


Fig. 4. Test results for the concentration of organic compounds in samples of reclaimed material stored in distilled water (H₂O) at pH = 7 for one, three and five months
 Rys. 4. Wyniki badań stężenia związków organicznych w próbkach destruktu przechowywanego w wodzie destylowanej (H₂O) o pH = 7 przez jeden, trzy i pięć miesięcy

Determination of the concentration of organic compounds in destructors stored in distilled water, i.e., indifferent environment, characterized by the slightest spread of results from among three analyzed environments (Figure 4). The results of the research conducted after a month are in the range of 15 mg/l to 34 mg/l. After three months, the

concentration of organic compounds increased compared to a month after 8% (Powisłe Street) to 38% (Browarna Street). After five months for two samples (Browarna Str. and Janówek Str.), the content of organic compounds in relation to the results of markings carried out after three months of storage of samples in water was obtained (after

three months of storage of destruct in water in water, the cleaning of organic compounds stepped down). The least susceptible to leaking destruct was from Zajęcza Street. Water over the 5-month analyzed was the most dynamically leached organic compounds in samples taken on Krakówka Street.

Conclusion

Asphalt destructors built in Płock before 2021 were not deprived of the status of waste. This material has been used over several years and it is currently not possible to determine the exact date of building the tested destructors. It is also problematic to indicate the construction site, i.e., the place from which the destruct the first age of the originally produced material was collected.

Different composition of mineral – asphalt mixtures, their construction time and location – both the primary and re-building of asphalt destructors have a direct impact on the results of tests of the concentration of organic compounds in the analyzed solutions. Depending on the traffic intensity, the type of vehicle supported by the road changes the surface structure and its susceptibility to leaching. In addition, atmospheric precipitation or de-icing agents used in winter periods change the content of organic compounds, which was indicated using solutions with highly alkaline, acid, and neutral solutions (distilled water). There was no sample of the destruct in the study, which would generate similar concentrations in each of these three solutions. It follows that, depending on all the previously mentioned factors and operating conditions, specific destructures are susceptible to the washing of organic compounds. According to the tests carried out, the highest level of carbon reinforcement was observed in the alkaline environment. This indicates the lack of leaching of carbonates, which in this environment occur in the form of salt. A significant increase in the total carbon content in leachate after leaching testifies to the leaching of compounds formed during oxidation, i.e., most likely organic acids with complex and complicated structure [26]. Leaching in an acid and neutral environment indicates a small amount of compounds release from the materials.

In order to assess potential environmental threats, the chemical analysis of substances leached from destructive samples and verification of the concentration of WWA compounds in two streets should be performed, where the simplified method enabled them to be detected, but only at a level below 25 mg/kg, which indicates that according to the regulation [3] The destruct should not be qualified as waste. WWA as aromatic hydrocarbon do not easily oxidize in ambient conditions, even at elevated temperatures in the summer [27, 28]. Therefore, the compounds from asphalt binder used during the formation of roads from which destructives have been collected.

Based on the research, it can be concluded that the highest level of organic compounds removal from asphalt destructors occurred using a highly alkaline solution. The designation conducted after a month showed results in the range from 10 mg/l to about 37 mg/l, after three months the results increased to even 180%, and after five months another increases in the range of 10 to 167% was recorded. The designation conducted on samples undergoing acid with $\text{pH} = 4$ after a month showed results from 7 mg/l to about 26 mg/l, after three months the results increased from 3% to 270%, and after five months there was another increase in the scope from 10% to 50%. The slightest disposal of the results was observed for destructive destruction destructors. The results of the research conducted after a month are in the range of 15 mg/l to 34 mg/l. After three months, the concentration of organic compounds increased in relation to the state after a month to a maximum of 38%, and after five months to 49%. The reaction of rainwater oscillates in the range from $\text{pH} = 4$ to $\text{pH} = 7$, therefore the washing of organic compounds from asphalt destructors to surface water due to atmospheric precipitation does not pose such a high environmental threat as in the case of strong alkaline solutions. Solutions with $\text{pH} = 4$ and $\text{pH} = 7$ generate the increase in organic compounds extended from destruct over time much less dynamically than in the case of a solution with $\text{pH} = 12$.

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Accepted for publication: 22.05.2024 r.