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THE INFLUENCE OF WASTE WATER SEDIMENT AS A COMPONENT OF ASPHALT CONTAININGHEAVY METALS AND DETECTING ITS EFFECT ON SOILS UNDER MODEL EXPERIMENTAL CONDITIONS

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Abstract. *Waste water sediment (WWS) is an ecological hazard to the environment and the search for its effective disposal is an urgent task. The main limitation of the WWS use is associated with the presence of the different ingredients - heavy metals, which can have a negative impact on the environment. There is asphalt production technolologies using WWS. This work considers the possibility of using asphalts with the WWS additives from car washes. This technology will reduce the negative influence on the environment due to the WWS recycling,as well as due to the possibility of using WWS instead of pure sand by replacing it in the standard asphalt production technology. In this work, a heavy metal content was determined by the ICP-OES method using an Agilent 5110 spectrometer; in the course of a model experiment, the effect on the soil of three model objects was studied: WWS from car washes in Moscow; the standard technology for asphalt production; asphalt fused with car wash WWS. We studied the change in the content of various forms of heavy metals penetrating the soil from wash-offs of these objects, and by comparing the results and assessing their absolute content, we assessed thepossibility of using fused asphalt. The studies have shown that during the model experiment, the total content of heavy metals increased up to 7-10 times for Zn, Ni and Cr. In all variants of the experiment, the content of mobile forms of all Cr, Cu, Mn, Ni, Pb, V and Zn significantly decreased by the third stage of the experiment to 50--95%, which indicates the transition of mobile forms of heavy metals to the total content due to the processes of sorption and precipitation.*

Keywords*: asphalt, ecological technology, waste water sediment, the effect of asphalt, heavy metals, soil pollution*

INTRODUCTION. Today one of the most important environmental problems is the problem of accumulation and disposal of production and consumption waste. In the Russian Federation, more than 80 billion tons of solid waste have been accumulated in landfills and landfills, about 7 billion tons more are generated and disposed of annually, while the country has introduced an insufficient number of waste disposal and detoxification technologies [1].

Heavy metals (HM) are hazardous environmental pollutants, since they have a significant toxic effect. Heavy metal contamination disrupts the normal functioning of soils, inhibits crops and hinders their development. The accumulation of heavy metals in plants leads to their transfer along the food chain, which can affect human health [2].

Waste water sediment (WWS) is a group of wastes generated at mechanical, biological and physicochemical treatment facilities for surface and underground waters, waste waters of settlements and industrial waste waters close to them in composition. So, the sediment of

domestic wastewater has a relatively homogeneous composition, they contain a significant amount of organic matter 75-80%, nitrogen, phosphorus and other biophilic elements. Thecomposition of WWS from industrial enterprises is different and directly depends on the type of production, the composition of the feedstock and the technological process. There is no strict classification of WWS in the literature. It is customary to divide WWS by the type of their constituent impurities - coarse precipitation, which are retained by gratings and sieves, their composition is variable, represented by various large floating impurities - paper, wood fragments, plastic waste, etc.; heavy precipitation - is retained by sand traps, consists mainly of mineral fragments - sand, glass fragments, bricks, etc., are not very susceptible to decay; floating
- are retained by grease traps, represented by fats and oils, respectively, as well as primary and secondary sediments, which are separated from the water by various sedimentation tanks [3].

In this work, one of the objects of research is the waste water sediment of car washes. Car washes consume significant amounts of clean water, and it has been estimated that an average car wash station generates between 150 and 600 liters of wastewater. In this regard, there is a need for effective water purification and reuse. However, the composition of wastewater from car washes is complex and varied; the components contained in wastewater from car washes can include a variety of impurities and pollutants that complicate treatment technologies. So, waste water from car washes can contain various mechanical impurities, such as sand and dust, oil products, lubricants and cooling liquids, motor oils, surfactants and heavy metal salts [4-6].

They are many ways to utilize WWS, but this mainly concerns the utilization of WWS containing a significant amount of organic matter - i.e. domestic WWS. WWS often contain a large amount of HMs, which limit the possibilities of their utilization [7-9]. Subject to compliance with the standards, such precipitation is used as fertilizer and for reclamation of disturbed lands. In addition, WWS is often disposed of by pyrolysis. Pyrolysis products - slags are later used as additives in cement, concrete and other building materials [9-12].

WWS are often stored at landfills or pumped into the voids of the landfill, which can have a negative impact on the environment [13].

WWS from car wash sewage differs from domestic WWS, primarily due to the low amount of organic matter and high content of mineral components, approaching in its composition to the components deposited on sand traps. There are 6,400 washing stations and more than 2,000 washing enterprises in Moscow, in Tashkent there are about 200 washing posts, as a result of which a large amount of WWS is formed, which are unsafe for environment. When looking for approaches to recycling the use of WWS from car washes, it is important to have in mind the possible consequences for the environment, which determines the relevance of this work.

There are technologies for the production of asphalt using WWS. Thus, in a study by N de Souza Campelo (2020), it was shown that asphalt mixtures with the addition of slag from WWS meet Brazilian standards for the quality of asphalt concrete pavement [14]. In the studies of other authors [15,16], it was proved that the addition of WWS to asphalt concrete mixtures improves their rheological characteristics.

From an environmental point of view, the impact of asphalt pavements on the environment and, in particular, on soils is ambiguous. There are many studies of soils under asphalt, so-called "sealed" soils. They are largely protected both from aerosol deposition of pollutants and from the influx with atmospheric precipitation and melt water. On the other hand, studies have shown that evapotranspiration is reduced in sealed soils, they have a significantly higher density than uncovered soils, which is associated with high vibration loads. Asphalt concrete pavements significantly change the thermal regime of soils - the temperature of the sealed soils sometimes turns out to be several degrees higher than the background ones. The chemical properties of the sealed soils are also noticeably different from the background, they have a more alkaline reaction of the environment, and the content of carbon and nitrogen in themdecreases. Soil shielding has the most significant effect on the microbiological community. Thus, in sealed soils, nitrification processes are suppressed, and the composition of the microbial community changes. All these factors negatively affect the state of urban soils [17-20].

The side effect of asphalt concrete pavements on the soils of cities is also significant - asphalt roads are directly related to vehicles, the negative impact of which on roadside soils is widely discussed. It was shown that roadside soils are significantly affected by salinization, the source of which is anti-icing reagents [21]. Road transport is a source of soil pollution with oil products and polyaromatic hydrocarbons (PAHs). Most PAHs are potent carcinogens, and there are many different sources of PAH contamination in today's urban environment, including those generated by the combustion of fossil fuels. As a result of combustion, PAHs are emitted into the atmosphere in the form of particles and gases, after which they settle and accumulate on the soil surface [22]. It is well known that road transport is one of the main sources of soil pollution with heavy metals; heavy metals enter the soil with engine oil particles, fuel combustion products and dust from tire abrasion [23, 24].

Thus, both asphalt and its components can have a negative effect on soils, the purpose of this study is to study the effect of WWS as a component of asphalt on the content of heavy metals in soils.

EXPERIMENTAL PART. The object of the study was the soil of the Zaryadye park in the city of Moscow from the recreated natural community ―Berezovaya Roscha‖. Soil samples were taken in four replicates, dried and sieved through a sieve with a mesh diameter of 1 mm.

As model objects were taken 1) WWS from car washes in the city of Moscow, selected by JSC "Combine of ecological services" for disposal, dried; 2) asphalt made according to standard technology, crushed to a size of 1-2 cm; 3) asphalt fused with car wash WWS.

The experiment was carried out in four replicates with three model objects that can have a negative impact on the soil as a component of the environment.

In the framework of the model experiment, soil samples were contaminated with water flowing down from the model objects as follows. Samples of uncontaminated soils were placed in containers of the same volume (350 ml), above them were containers of the same volume 1) with WWS, 2) with asphalt made using standard technology (Ast) 3) with asphalt fused with sediment waste water (Awws). There was no drainage, the activity of microorganisms was not suppressed; humidity was not maintained at a constant level, while the soil was filled with distilled water based on an annual precipitation rate of 700 mm / year, which is typical for Moscow. The samples were mixed after each watering. Sampling for analysis was carried out after each spill of the monthly precipitation rate. In the samples, the content of water-soluble, mobile (ammonium acetate extract with pH 4.8) forms of heavy metals was determined, as well as the gross content of heavy metals (nitric acid extract) in the soil according to environmental regulatory document of the federal level (PND F) 16.2.2: 2.3.71-2011. The analysis of the content of heavy metals was carried out by the ICP-OES method on an Agilent 5110 spectrometer. Statistical processing was carried out using the OriginPro2019 and STATISTICA 10.0 software.

RESULTS AND DISCUSSION. Before starting the experiment, the initial contents of Cd, Cr, Cu, Mn, Ni, Pb, V, Zn were determined in the model objects. The highest contents of most HMs were shown for WWS. Figure 1 shows a significant excess of copper content in WWS compared to the rest of the objects - 420.3 mg / kg, versus 14.9 and 7.6 in Ast and Aosv, respectively. The maximum manganese content in the soil is 438 mg / kg. The highest zinc content is shown for Ast - 606.1 mg / kg. At the time of the experiment, the content of HM in thesoil of the Zaryadye Park did not exceed the MPC (OEC) for the hygienic standard (GN) 2.1.7.2041-06 and the sanitary norms and rules (SanPiN) 2.1.7.1287-03.

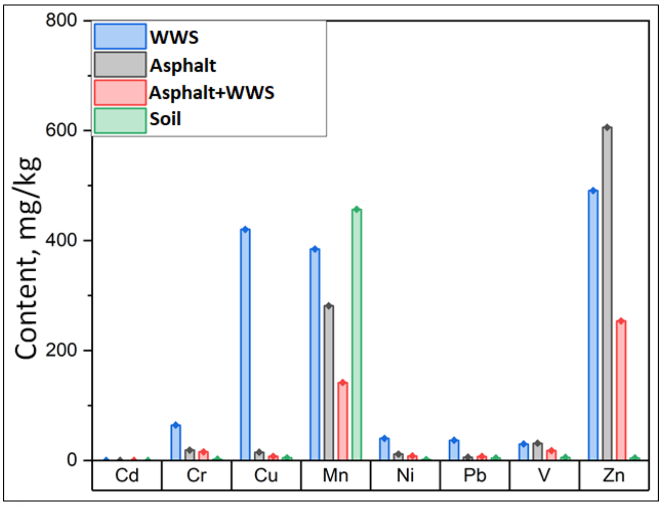


Figure 1. The content of HM in the soil and model objects at the start of the experiment.

As a result of the experiment, the HM contents in soil samples were obtained after adding first, second and third precipitation rates - Point 1, Point 2 and Point 3, respectively. At the same time, in fact, samples for Point 1 were taken 3 months after the start of the experiment, for Point 2 - after 6 months, and for Point 3 - after 12 months, respectively, the total time of polluting ingredients from the model objects in the soil at the time of the end of the experiment was 1 year.

After spilling the first monthly norm of precipitation and analyzing the results, a significant difference was shown in the content of water-soluble forms of manganese and nickel: their content in WWS was significantly higher than in other variants of the experiment (medians: WWS - 2.16 mg / kg Mn and 0 , 19 Ni; Ast 0.07 and 0.05; Awws - 0.06 mg / kg Mn, and the nickel content in this variant was below the detection limit of the device).

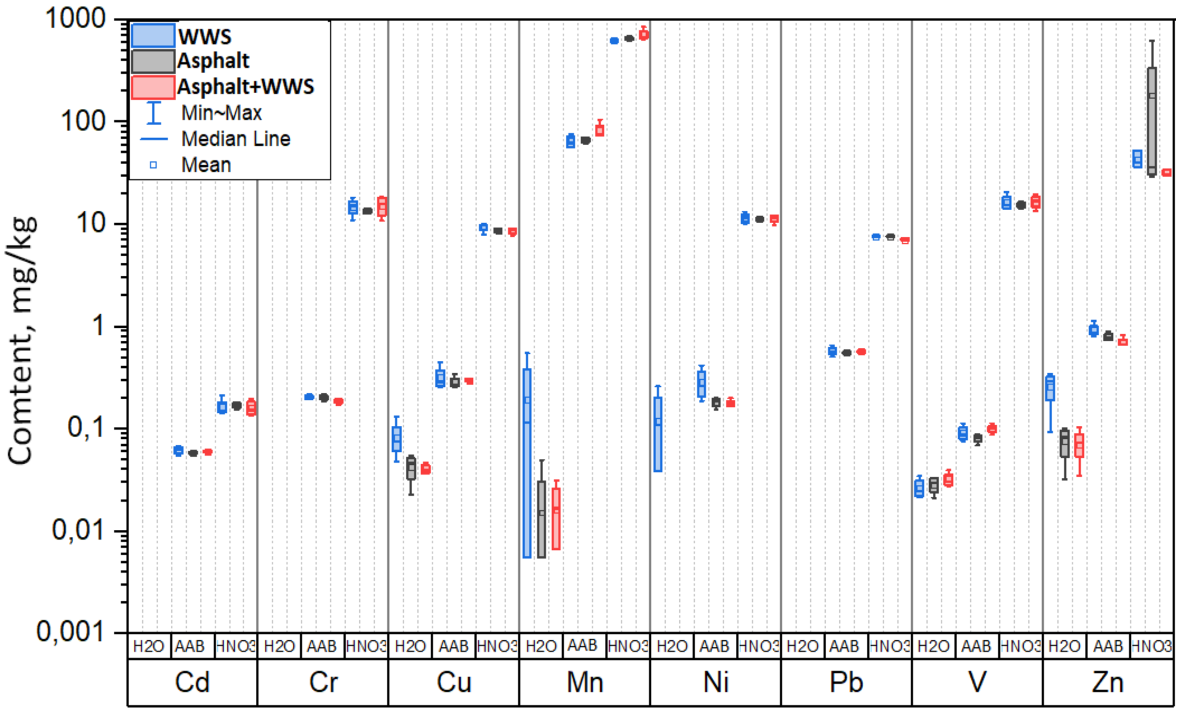


Figure 2. Diagram of the median HM contents and limits after the second monthly norm of precipitation (Point 2).

The content of water-soluble zinc was significantly higher in the variant with WWS than in the third variant of the experiment (Awws): 0.59 mg / kg and 0.12 mg / kg, respectively. The content of water-soluble cadmium and lead in all variants of the experiment at this stage was below the detection limit of the device.

The content of mobile forms of HM did not differ significantly in all variants of the experiment. The gross chromium content in the variant with standard asphalt was significantly lower than in other variants (medians: Ast - 2.12 mg / kg; WWS - 2.21; Awws - 3.27), as well as a tendency to a lower content in this variant is also noticeable for other metals.

After the spilling of the second norm of precipitation, the contents of water-soluble forms of cadmium, chromium, nickel and lead were below the detection limit of the device. In general, no significant differences were shown between the variants in the contents of water-soluble forms of metals in the three experimental variants. In the case of mobile forms and the total HM content, there are also no significant differences between the variants (Figure 3). However, there is a tendency to a lower content of mobile forms of chromium, copper, nickel and zinc in the Awws variant compared to other variants, at the same time, the median content of mobile manganese in this variant is noticeably higher than in other variants (OSV - 64.04; Ast - 67.38; Awws - 77.08), it is possible that these metals are sorbed on manganese compounds and become less mobile.

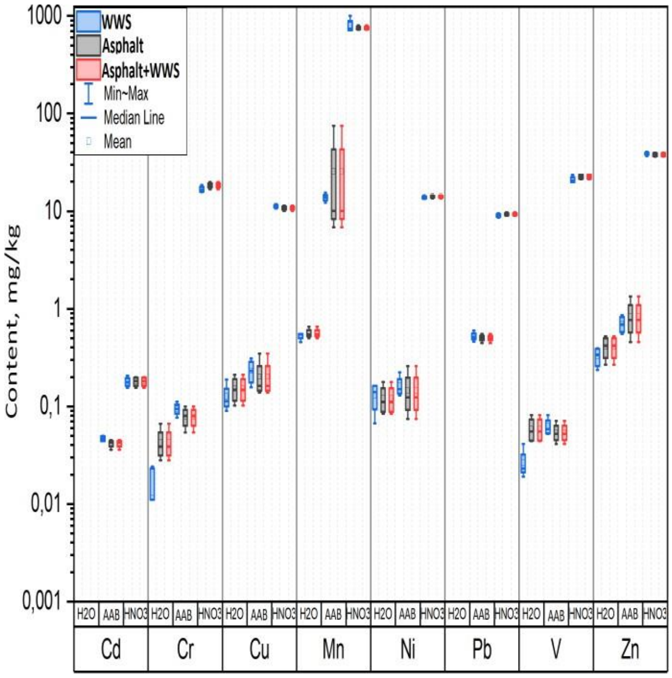
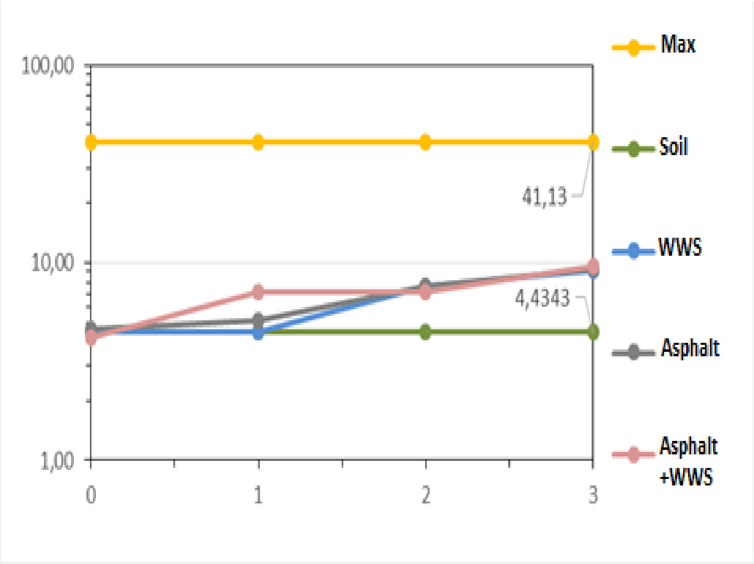


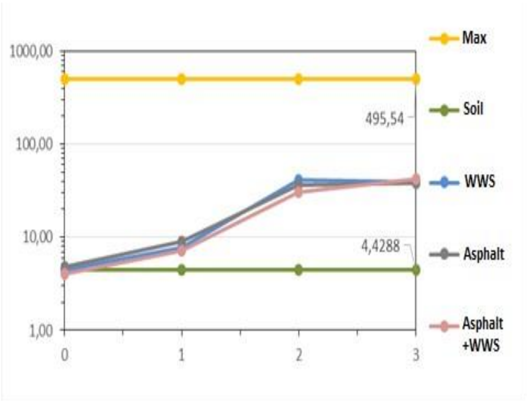
Figure 3. Diagram of median contents and limits after the third monthly precipitation norm (Point 3).

After the spilling of the third monthly precipitation norm, the contents of water-soluble forms of cadmium and lead were not determined as in the previous points. No significant differences were shown in the content of water-soluble forms of HM, with the exception of the content of water-soluble chromium, the median of its content in the variant with WWS was two times lower than in the variant Awws and almost 4 times lower than in the variant Ast (0.01 mg /kg - in WWS, 0.04 - Ast and 0.039 in the variant with Awws). Figure 4 clearly shows that the values of the contents of mobile forms of all metals to one degree or another approached the contents of water-soluble forms, the only exception to this trend is mobile manganese. The gross HM contents showed no differences between the experimental variants.



Changes in the total HM contents by the stages of the experiment. To assess changes in the total content of heavy metals in the studied soil, graphs were drawn, the yellow line on the graphs is the theoretical maximum metal contentat the time of the end of the experiment (the sum of the metal content in the uncontaminated soil

Figure 4. Medians of gross lead contents by the stages of the experiment



and in the sewage sludge), and the green line is the initial metal content in uncontaminated soil. From Figures 5-7, it can be seen a smoother change in content in the Awws variant for all metals, except for zinc.

The gross contents of all heavy metals during the experiment increased - Zn and Pb - 10 times, Cr - 7 times, Cd - 3 times, Mn - 1.5 times, Cu - 2 times, V - 4 times, and Ni - 9 times.

Figure 5. Medians of gross zinc contents by stages of the experiment

At the same time, in almost all variants at the third stage of the experiment, the differences in the gross contents of heavy metals were not so significant.

Assessment of the degree of soil contamination. To assess the degree of pollution at each stage of pollution, concentration factors were calculated for all studied heavy metals (Table 1.)

The table shows that at each stage of the experiment, the highest Kc is characterized by the zinc content in all variants of the experiment.

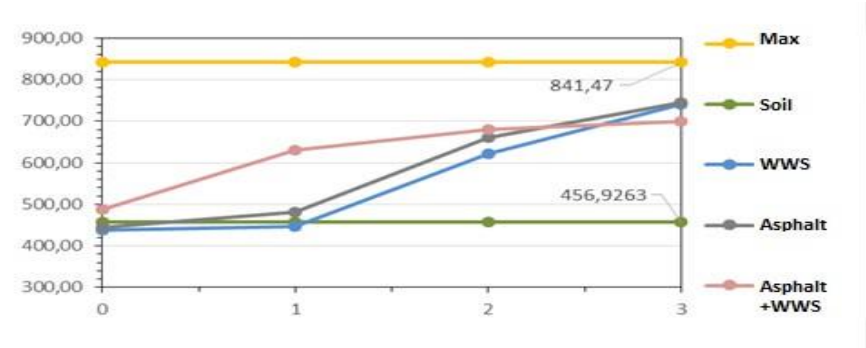


Figure 6. Medians of gross chromium contents by stages of the experiment

Based on the data obtained, the hazard of soil contamination was assessed according to the total pollution coefficient and the corresponding category of soil contamination was assigned. After the first stage of the experiment, the category of soil contamination for all variants of the experiment was determined as permissible, after the second - as moderately hazardous, and at the last stage, the variants with WWS and standard asphalt were at the upper boundary (in terms of Zc)of the moderately hazardous soil category, and the soil in the version of asphalt with WWS was already classified as a hazardous pollution category.

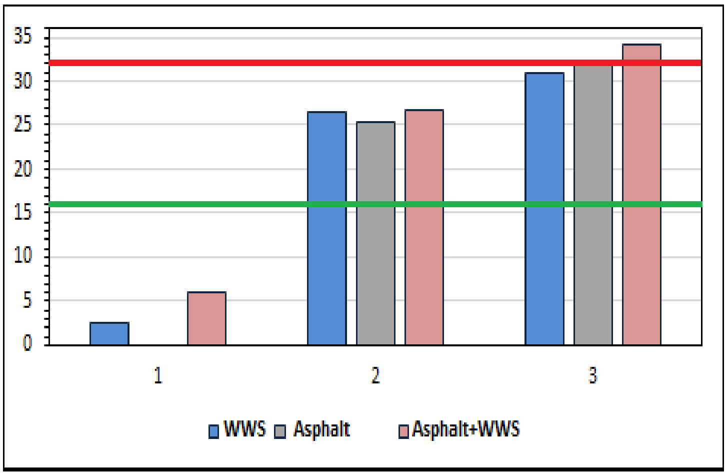


Figure 7. Diagrams of changes in the total coefficient of soil pollution (Zc) by the stages of the experiment

Table 1.

Values of the concentration factor for the studied metals by stages of the experiment

	Concentration factor (Kc)								
	Point 1			Point 2			Point 3		
Metall / object	WWS	Asphalt	Asphalt+ WWS	WWS	Asphalt	Asphalt + WWS	WWS	Asphalt	Asphalt+ WWS
Cd	1,72	1,68	2,34	2,50	2,92	2,65	2,96	3,29	2,94
Cr	0,99	0,98	1,59	6,64	6,02	7,57	7,34	8,26	8,47
Cu	0,99	0,90	1,46	2,05	1,86	1,97	2,36	2,24	2,30
Mn	1,02	0,88	1,29	1,42	1,51	1,39	1,69	1,68	1,43
Ni	0,98	0,90	1,34	7,34	7,28	7,62	8,91	9,47	9,17
Pb	0,99	1,11	1,70	1,69	1,69	1,70	2,03	2,01	2,28
V	1,00	1,03	1,62	2,81	2,83	3,21	3,72	3,97	4,23
Zn	1,73	1,91	1,79	9,14	8,18	7,57	8,81	7,96	10,41

CONCLUSION

The gross HM contents increased during the year of the experiment - the maximum increase was observed for Zn in the Awws variant - 10 times. The least change in the Mn content was 1.4 times in the same variant of the experiment. A noticeable increase was shown for Ni and Cr - in all variants of the experiment, their content increased by at least 7 times.

The content of mobile forms of all HMs markedly decreased by the third stage of the experiment by 47% or more. The maximum decrease is shown for Mn in all variants of the experiment - by 95 - 98%. In all variants of the experiment, there is a tendency to a decrease in the content of mobile forms of Cr, Cu, Mn, Ni, Pb, V, and Zn - over time with an overall increase in the total content of these HMs, which may indicate the fixation of HMs in the soil.

An assessment was made of the degree of soil pollution with metals according to the total pollution index (Zc). The soil in all variants of the experiment was assigned to the permissible category of pollution at the first stage. Whereas at the third stage, the pollution category for the WWS variants and with standard asphalt was determined as moderately hazardous, and in the WWS variant, the asphalt with WWS was determined as hazardous.

Based on the results, it can be concluded that no significant differences between the experimental variants were observed, and the use of WWS as a component of asphalt is permissible.

REVIEW. Every year, as a result of the activity of car wash stations, a large amount of WWS is formed, which is environmentally unsafe. When looking for approaches to recycling the use of WWS from car washes, it is important to understand the possible consequences for the environment, which determines the relevance of this work. The main limitation of the use of sewage sludge is associated with the presence of accompanying ingredients - heavy metals, which can have a negative impact on the environment. There are technologies for the production of asphalt using WWS. The paper considers the possibility of using asphalts with the addition of WWS from car washes, a model experiment was carried out in four-fold repetition of soil contamination of the Zaryadye park in Moscow from the recreated natural community "Berezovaya Roscha" by gravitational water effluents from three model objects 1) sediment waste water from car washes in Moscow; 2) asphalt produced using standard technology; 3) asphalt fused with car wash WWS. Sampling for analysis was carried out after each spill of the monthly norm of precipitation; in total, sampling was carried out three times. The samples determined the content of water-soluble, mobile forms of heavy metals, as well as the total content (gross) of heavy metals in the soil. The analysis of the content of heavy metals was carried out by the ICP-OES method on an Agilent 5110 spectrometer. Statistical data processing was carried out using the OriginPro2019 and STATISTICA 10.0 software. We studied the change in the content of various forms of heavy metals entering the soil from washings from different variants. Comparison of the results obtained made it possible to assess the possibility ofusing asphalt prepared using the new technology. The studies have shown that during the model experiment, the total content of heavy metals increased up to 7-10 times for Zn, Ni and Cr. In all variants of the experiment, the content of mobile forms of all Cr, Cu, Mn, Ni, Pb, V and Zn significantly decreased by the third stage of the experiment to 50--95%, which indicates the transition of mobile forms of heavy metals to the total content due to the processes of sorption and precipitation. Based on the results of the work, it can be concluded that no significant differences between the experimental variants were observed, and the use of WWS as a component of asphalt is permissible. This technology will reduce the environmental burden on the environment due to the recycling of WWS, as well as due to the possibility of using WWS instead of pure sand by replacing it in the technology of preparing standard asphalt.

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