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Analysis of the existing methodological approaches to the problem of establishing the boundaries of soil pollution with the main pollutants and metal-containing nanoparticles in the areas of location of unauthorized dumps

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ABSTRACT: Introduction. Unauthorized dumps are territories that are not permitted and not equipped in accordance with the current legislation of the territory, where waste products are disposed of. Most of the unauthorized dumps, both industrial and municipal solid waste (MSW), are objects of accumulated harm. In connection with the increase in the use of nanoparticles of various genesis in engineering materials, there is a need to assess their impact on environmental components and human health. One of the main marker substances that determine the ecotoxicological effect of dumps are heavy metals, including those entering the environment in the form of metal-containing nanoparticles. Their identification, determination of the processes of horizontal and vertical migration of pollutants (P), identification of the intensity of negative processes are necessary information for making decisions on the choice of methods and technologies for recultivation of disturbed areas. Currently, there are no legislative acts (LA) that take into account the specifics of such objects in the study of territories within the framework of environmental surveys (ES), which necessarily precede the implementation of project documentation for recultivation. A vital task is to develop a methodological approach to diagnosing the state of territories disturbed as a result of waste disposal, and to reliably determine the boundaries of soil pollution, taking into account modern trends in the development of nanotechnology. Methods and materials. The analytical study was based on the results of environmental surveys at four unauthorized dumps of the Republic of Bashkortostan (three dumps for municipal solid waste (MSW) and one dump for industrial waste), carried out in 2019–2020. Results and discussion. A study of the reliability and completeness of determining the level of soil pollution in the territory of the dumps was made. Deviations from the requirements of CS 11-102-97, GOST 17.4.3.01-83 in terms of the number of sampling points and their spatial location were revealed. It was found that the requirements of Article 1 and Article 80.1 of the Federal Law of 10.01.2001 No.7-FL "On Protection of Environment" on identifying negative environmental changes and establishing the ability of pollutants to migrate to other components of the environment were not taken into account. Conclusions. The existing methodological approaches to establishing the boundaries of soil pollution in the areas where unauthorized dumps are located require revision and specification.

KEY WORDS: waste, unauthorized dumps, soils, pollutants, metal nanoparticles, environmental surveys (ES), environment.

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INTRODUCTION

U nauthorized dumps are one of the most common objects of accumulated harm (negative impact resulting from past economic and other activities, the obligations

to eliminate which were not fulfilled or were not fulfilled in full) and sources of a whole spectrum of pollutants entering the environment [1-4].

According to the Federal Target Program "Elimination of Accumulated Environmental Damage" for

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2014–2025 [5], as a result of economic activity, 31.6 billion tons of waste have been accumulated in the Russian Federation by now, of which 2–2.3 billion tons are toxic. Of the 121 particularly hazardous objects of accumulated environmental harm on the territory of the Russian Federation included in the register, 69 are dumps and waste landfills [6].

The specificity of unauthorized dumps as objects of pollution, first of all, is the lack of information about the waste disposed on them, and, as a consequence, the unpredictability of the nature and levels of impact. On the territories of unauthorized dumps, the status and composition of MSW is not monitored, and the established period for the placement of MSW is not observed [7].

In this case, the main component of the environment, experiencing a negative effect, are soils in which the morphological and physical properties deteriorate, the water, air, thermal and redox regimes are disturbed, the conditions necessary for the existence of soil animals and microorganisms, the growth and development of plants change.

As studies show [8], under soil pollution, the soil matrix system and the associated work of all nanoreactors are significantly affected. At the same time, nanotechnological processes in soils (the formation of aggregates, humus, litter, etc.), which are directly related to soil fertility, are not taken into account in the system of environmental regulation, although they could provide decisive information about the degree of degradation of soil ecosystems.

The soil is the main accumulator of technogenic nanoparticles entering the environment as a result of economic and other activities. Technogenic metal nanoparticles in soil are mainly represented by the following compounds TiO₂, ZnO, AlO, CuO, Ag, MgO, FeO_x, NiO, MnO_x [9–13]. The sources of their entry from dumps into soils are waste of packaging materials, household chemicals, agrochemicals, spent industrial catalysts, ash from thermal waste disposal, etc.

In the structure of normative legal acts [14–18], regulating the scope of research to determine the admissibility of negative impact on soils, we are faced with the traditionally accepted definition of the degree of degradation by the level of chemical pollution.

This approach does not take into account the possibility of joint action of harmful factors, the dependence of the impact on the symbaticity of factors, and the determination of environmental risks, and, moreover, the determination of the form of chemical compounds (mobile, immobile, macrodispersed, nanoparticles, etc.).

And while chemical pollution of soils and grounds within the framework of ES is assessed by the total indicator (Zc), which takes into account the additivity of pollutants, it is an indicator of the adverse impact only on the health of the population. This methodological approach is complicated by the lack of requirements to the scope of research during surveys (the number of samples) for such complex objects as unauthorized dumps, accordingly, it becomes difficult to determine the zones of influence of objects and the degree of soil degradation.

According to RP 1.2.2639-10 "Use of methods for quantitative determination of nanomaterials at nanoindustry enterprises" [19], it is recommended to determine the presence of nanoparticles in environmental components (atmospheric air, water bodies, soils) in a number of controlled objects. The presence of this document reflects the importance of an integrated approach to identifying the degree and nature of contamination.

Within the framework of the state environmental expertise, all the above points are of particular importance, since materials on environmental impact assessment should be scientifically substantiated, reliable and reflect the results of research carried out taking into account the relationship of various environmental factors [20].

METHODS AND MATERIALS

This work was based on the results of the ES analysis for 2019–2020 in terms of the reliability and completeness of determining the level of soil pollution in the territory of the location of four unauthorized dumps of the Republic of Bashkortostan. The study of the compliance of the ES materials with the requirements of the current regulatory documentation for geoecological soil testing was carried out.

Dump $N \ge 1$ is an unauthorized MSW dump. The year of the beginning of the exploitation of the dump is 1993. The landfill body is a mixture of MSW covered with layers of insulating soil. The proposed waste storage system is based on the maps using the thrust method.

Dump N_{2} is an unauthorized MSW dump. The dump body is an embankment with steep slopes, partly overgrown with weeds. There are no intermediate layers of insulating primer in the body of the dump.

Dump N_{0} 3 – Industrial waste dump. The beginning of waste storage – 1970s. The dump is an open pit where solid and resinous wastes (various spent catalysts containing heavy metals) were stored and liquid heavy hydrocarbons were placed in technological maps.

Dump N_{2} 4 is an unauthorized MSW dump. The dump body is an embankment of garbage, mainly covered with soil and overgrown with weeds of the grass layer.

RESULTS AND DISCUSSIONS

The first question that we face when assessing the sufficiency of the ES materials is the number and spatial location of the samples taken (determining the completeness and representativeness of research results).





In clause 4.19 of the Code specification CS 11-102-97 [14], a reference is provided to GOST 17.4.3.01-83 "Environmental protection. Soils. General requirements for sampling" [15], which contains the following recommendation on the number of samples: at least one combined sample per 0.5-1.0 hectares with a heterogeneous soil cover and at least one combined sample per 1.0-5, 0 ha – with uniform. Scope [15] – for general and local pollution.

Further, according to the text of clause 4.19 [14], it is indicated that the number and location of samples, as well as the distance between samples, are set in the *survey program* depending on the type and purpose of the projected object, the natural and technogenic conditions of the research area and the stage of design and survey work.

Table 1 shows the features of the dumps under consideration that affect the spread of pollutants in the soil profile and the possibility of the occurrence of foci of secondary soil pollution.

Clause 4.16 [14], which establishes the requirements for assessing pollution in the zone of influence of economic objects, also gives a reference to the survey program: "The location of sampling points is established in the survey program depending on the expected structure of the pollution field, the geological structure of the territory."

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Thus, the quality of the work is directly related to the responsibility of the Customer, who draws up the Terms of Reference for the ES. In ES, for all objects in the survey programs, the scope of work is limited and/or tied to the "territory of the object location" in accordance with the data of the development plan for a land plot [21].

In this context, many legal restraints arise, and sampling in adjacent areas is, at best, carried out only within the sanitary protection zones (SPZ) of the facilities. Although soil pollution at landfill sites is often secondary, some of the pollutants are washed out with precipitation and groundwater, ensuring the accumulation of pollutants at a certain distance from the objects [22–24].

The same clause 4.16 [14] regulates that "the adopted sampling system should ensure the study of the pollutional zone in plan and in vertical section by the main components of the environment, identification of pollution sources, migration routes, areas and fluxes of dispersion and accumulation of pollutants".

As you can see from the table 2, the number of sampling points (sites) does not meet the requirements of [14, 15] in terms of completeness of research: the sampling

Indicator name	Dump № 1	Dump № 2	Dump № 3	Dump № 4
Dump area, ha	18,12	12,05	4,13	10,07
Volume (mass) of accumulated waste	1 434 369 m ³	442 472 m ³	30 000 t	850 000 m ³
Features of hydro- geological conditions	Wetland area	Wetland area	_	Wetland area, confined groundwater
Existence of exceed- ing the MAC for pollutants in ground (underground wa- ters)	Not available	Sulfate ion, calcium, magnesium, lithium	Benz (a) pyrene, phenol, nickel	BOD, dry residue, chloride ion, chromium, lead, iron, cadmium, magnesium, mercury, barium, lithium, petroleum products
Existence of exceed- ing the MAC for pollutants in surface water bodies (includ- ing bottom sedi- ments)	BOD, suspended solids	Copper, arsenic, chromium, mercury, lead, petroleum products	Benz (a) pyrene, phenols, nitrate ions	Copper, arsenic, chromi- um, mercury, lead, petro- leum products
The presence of leachate in the body of the dump	Yes	No	No	Yes
Soil type of the sur- rounding area	Urbanozems, carbonate cher- nozems	Urbanozems, cher- nozems, leached medium humus me- dium thick	Urbanozems, leached cherno- zems	Urbanozems, leached cher- nozems

Table 1General data on research objects



Table 2

The scope of research within the framework of the ES to establish soil pollution in the areas of dumps

Indicator name	Dump № 1	Dump № 2	Dump № 3	Dump № 4
Study area boundaries	Within the SPZ (500 m from the bor- der of the dump)	Within the SPZ (115 m from the bor- der of the dump)	Within the SPZ (1000 m from the border of the dump)	Within the SPZ (320 m from the bor- der of the dump)
Number of sampling points and/or sam- ple ares (SA)	5 wells along the pe- rimeter of the dump body, 4 SA within a radius of 50 m from the dump body	3 SA within a radius of 50 m from the dump body	3 wells within a ra- dius of 100 m from the dump body	5 wells along the pe- rimeter of the dump body, 3 SA within a radius of 50 m from the dump body
Number of samples	20 well samples layer by layer 4 pooled samples (envelope method with SA)	3 pooled samples (envelope method) with SA	18 well samples layer by layer	20 well samples layer by layer 4 pooled samples (envelope method with SA)
Vertical depth survey	0.0–3.0 m layer by layer in wells 0.0–0.2 m on SA	0.0–0.2 m on SA	0.3–15.0 m layer by layer in wells	0,0-3,0 m layer by layer in wells 0,0-0,2 m on SA
Presence of a base- line sample taken outside the sphere of local anthropogenic impact	No	No	No	Yes
Assessment of soil fertility	No	No	No	Yes
Analysis of pollution by toxic-chemical indicators	Nickel, copper, zinc, lead, cadmium, arse- nic, mercury, cobalt, manganese, chro- mium, petroleum products, benz (a) pyrene	Nickel, copper, zinc, lead, cadmium, arse- nic, mercury, cobalt, petroleum products, benzo (a) pyrene	Nickel copper, zinc, lead, cadmium, arse- nic, mercury, petro- leum products, benz (a) pyrene	Nickel, copper, zinc, lead, cadmium, arse- nic, mercury, man- ganese, chromium, cobalt, petroleum products, benzo (a) pyrene
Determination of mobile forms of metals	No	No	No	No
Determination of nanoparticles, including metal- containing ones	No	No	No	No
Dump base soil sur- veys	Absent	Absent	4 points	2 points

depth was chosen incorrectly. In case of chronic anthropogenic impact (from 30 to 50 years for research objects), which develops at unauthorized dumps, it is unacceptable to analyze only the surface layer, soil sampling should be carried out in each engineering-geologic element.

According to the table 1, pollution of surface and ground waters is present in all objects, which, when analyzed together with soil research data (table 2), can only

indicate that the soil control points were chosen incorrectly, and migration routes and reliable depth of pollutant accumulation have not been established.

An important point showing the quality of research work is the definition, along with the bulk forms of metals, their mobile forms. The amount of mobile metal compounds is used to assess the amount of trace elements available to plants, as well as the ecological state



of contaminated soils. The main mechanisms of the action of mobile forms on the soil are complexation and ion exchange. It has been established that the form of input of heavy metals into the soil significantly affects the transformation of technogenic compounds and their distribution [25], namely, heavy metals are the main pollutants entering the environment components from waste disposal facilities [26].

Currently, there are not many studies to determine the migration paths of nanoparticles in soils and to determine their role in the degradation of soil ecosystems. However, it was established [9] that natural soil colloids are carriers of metal nanoparticles through soil profiles. The main trends in the distribution of metal-containing nanoparticles have a similar picture with the distribution of other pollutants in soils, while the nanoparticles have a higher migration ability and bioavailability.

Soil quality control by chemical parameters and determination of the degree of hazard was carried out in accordance with SanPiN 2.1.7.1287-03 "Sanitary- epidemiological requirements for soil quality" [27], which set requirements for the quality of soils in populated areas and agricultural land (table 3). An additive assessment of the degree of chemical pollution of soil and ground at the survey sites was carried out on the basis of the total indicator of chemical pollution (Z_c), which is an indicator of adverse impact on the population. In the context of unauthorized dumps, it is important not only to comply with hygienic standards, but also to take into account other criteria for the functioning of ecosystems: the enzymatic activity of soil microbiota, potential fertility, potential phyto- and zootoxicity of soils, and the intensity of nanotechnological soil processes. It is they which can provide reliable information on the magnitude of environmental risk and a long-term forecast for the restoration of territories, that can directly affect technological solutions for eliminating the negative impact on the environment and reclamation of disturbed lands.

Another approach to comparing and assessing the levels of chemical pollution is to compare the concentrations of pollutants with "background" values – the content of analyzed substances in anthropogenically undisturbed natural objects-analogues. However, this approach was not reflected in the ES for three objects out of four analyzed.

Another requirement of CS 11-102-97 [14] in terms of the analyzed pollutants was not taken into account in research at unauthorized dumps: Clause 4.29 regulates soil testing for the content of volatile toxicants (benzene, toluene, xylene, ethylbenzene, chlorinated hydrocarbons, oil and oil products) and other pollutants that penetrate into subsoil horizons to a depth of 3.0–3.5 m.

In dumps \mathbb{N}_{2} 1 and 4, studies on petroleum products were carried out to a depth of up to 3.0 m in wells and up

Table 3

Research area	Pollution category according to SanPiN 2.1.7.1287-03 "Sanitary-epidemiological requirements for soil quality"					
	Dump № 1	Dump № 2	Dump № 3	Dump № 4		
Dump base soils	-	_	Permissible	Permissible		
Adjacent area	Wells: 0.00–0.20 m (moderately dangerous); 0.2–3.0 m (permissible) SA: moderately dangerous permissible	SA: moderately dangerous permissible	Wells: 0,3-1,0 m; 1,0-2,0 m; 2,0-3,0 m; 3,0-5,0 m (permissible); 5,0-10,0 m; 10,0-15,0 m (clean) 1,0-2,0 m; 5,0-10,0 m (permissible); 0,3-1,0 m; 2,0-3,0 m; 3,0-5,0 m; 10,0-15,0 m (clean) 1,0-2,0 m; 3,0-5,0 m (permissible); 0,3-1,0 m; 2,0-3,0 m; 5,0-10,0 m; 10,0-15,0 m (clean)	SA: permissible		

Category of soil pollution according to SanPiN 2.1.7.1287-03 [27]



to 0.2 m at the SA, and at dump $\mathbb{N} = 2$ — only at the SA up to 0.2 m. At the industrial waste dump, where organic pollutants could have been disposed of, all petroleum hydrocarbons were accounted for as "petroleum products", although in the context of toxicity and impact on soil processes, aromatic hydrocarbons (benzene), unlike petroleum products, have established MAC values and a higher hazard level [28].

CONCLUSIONS

Currently, a large scientific and practical experience has been accumulated in the diagnosis of soil ecosystems exposed to unauthorized disposal and disposal of waste. However, the requirements of environmental regulatory documents that determine the composition and structure of research within the framework of ES are more focused on construction objects (rather than elimination of accumulated harm) and compliance with the requirements of hygienic standards in the light of the impact of pollutants on human health. At the same time, the term "negative impact on the environment" in the legislation is defined much broader – as "the impact of economic and other activities, the consequences of which lead to negative changes in the quality of the environment" [29].

With the development of scientific research, new technologies and industries, the list of substances required for diagnosis and control is also expanding. Due to the presence of dissimilar engineering materials in dumps and the formation of special pollution due to the migration of heavy metals, it is recommended to expand approaches to assessing the impact of these objects on the environment, as well as taking into account the ecotoxicological effect of metal-containing nanoparticles.

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Currently, the completeness and reliability of research directly depend on the qualifications of the developers of the survey and the requirements of the Customer of the design documentation for the elimination (reclamation) of unauthorized dumps.

The existing methodological approaches to establishing the boundaries of soil pollution in areas where unauthorized dumps are located require revision and specification in terms of:

- requirements for the number and spatial distribution of sampling points, taking into account the specifics of waste disposal facilities;
- requirements for the list of analyzed substances with the establishment of marker substances for each object;
- taking into account the structural, genetic, biochemical characteristics of soils, for which the impact on the environment is assessed;
- taking into account the assessment of the ecological potential of soils, their resistance to impact and the ability to restore.

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