Environmental aspects of the use of sewage sludge as fertilizer materials

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Abstract. The possibilities of using sewage sludge as fertilizer materials are determined by their chemical composition, sanitary-toxicological and physical condition. In each case, the use of these wastes must be preceded by studies proving not only their nutritional value, but also safety, and be accompanied by the development of an environmentally friendly technology for their disposal. The paper provides data on the analysis of industrial soil based on sewage sludge with the aim of using it for the restoration of disturbed and contaminated lands. In an experiment with the cultivation of cereal grasses, the limiting doses of introducing waste were determined. An increase in the amount of sewage sludge led to an increase in the gross content of heavy metals in the industrial soil, and the cadmium content exceeded the permissible standards for close to neutral and neutral soils. A clear correlation between the content of toxicants in the soil and their accumulation in plants was not observed.

1. Introduction

The problem of the placement of sewage sludge in Russia is caused by the discrepancy between the volumes of incoming waste and the technical capabilities of their disposal. 70-80 million tons of precipitation with a moisture content of 96-97%, which is about 2.5 - 3 million tons of dry matter, is formed annually at treatment facilities of the Russian Federation [1].

This problem is noted in many Russian cities, the treatment facilities of which are overflowed with unused waste. For example, in Moscow Region, more than 120 million tons of sewage sludge have been accumulated, the amount of which is increasing by 14–20 million tons every year [2].

Sewage sludge is formed at treatment plants as a result of wastewater treatment at urban aeration stations. According to GOST R 54534-2011, wastewater sludge (WWS) includes wastes generated at the facilities of mechanical, biological and physico-chemical treatment of surface and underground waters, wastewater from settlements and similar wastewater composition. The amount and composition of precipitates formed is determined by the methods of their purification and the content of pollutants.

WWS contain in its composition nutrients - humus, nitrogen, phosphorus, potassium, trace elements, as well as many groups of microorganisms. Their humidity is 85 - 99%. The composition of the sediments includes organic matter - up to 50% or more, total nitrogen - from 1 to 2%, phosphorus 3-4%, potassium 0.3 - 0.6%, trace elements. The amount of annually accumulating nutrient compounds of WWS is 524 thousand tons, including 448 thousand tons of nitrogen, 64 thousand tons of phosphorus and 12 thousand tons of potassium [3].
WWS of the second and third classes related to highly hazardous and hazardous waste are accumulated on silt maps and dumps. As a result of the limited territory of silt fields, in many cases the problem of the placement of new precipitation arises.

Storage of WWS is associated with adverse environmental effects. During storage of WWS, gases are released that can exceed the MPC by several times. If stored improperly, there is a likelihood of contaminants entering natural water bodies and groundwater, soil, plant communities.

Currently, the most common methods for the disposal of WWS are thermophilic digestion in digesters to produce biogas; production of activated carbon (sorbents); use as an additive in the production of expanded clay and other building materials; composting; vermicomposting; use as fertilizer, recultivant of disturbed lands and landfills; burning; pyrolysis; burial and others [4].

The main methods for the disposal of WWS in different countries are shown in table 1.

**Table 1. The main methods of disposal of sewage sludge (%) in modern conditions [5].**

| Country    | Using in agriculture | Waste disposal in landfills | Burning | Discharge into the sea, ocean and other technologies |
|------------|----------------------|-----------------------------|---------|------------------------------------------------------|
| England    | 53                   | 16                          | 7       | 24                                                   |
| Austria    | 20                   | 49                          | 31      | –                                                    |
| Germany    | 25                   | 55                          | 15      | 5                                                   |
| Danmark    | 45                   | 28                          | 18      | 9                                                   |
| USA        | 25                   | 25                          | 35      | 15                                                  |
| Italy      | 20                   | 60                          | –       | 20                                                  |
| Finland    | 40                   | 41                          | –       | 19                                                  |
| Switzerland| 50                   | 30                          | 20      | –                                                   |
| Sweden     | 60                   | 30                          | –       | 10                                                  |
| France     | 23                   | 46                          | 31      | –                                                   |

When WWS is disposed of by burning, phenol, benzapyrene, dioxins, and other polluting substances can be released. For the burial of large volumes of precipitation, the alienation of large areas under landfills is required. One of the common methods is the use of WWS as fillers in the manufacture of concrete, building ceramics, glass, pigments, paints, etc.

There is data on the restoration of natural landscapes of WWS, together with excess soil wastes generated during the construction of buildings and structures. Remaining quarries after mining are filled with a recultivant. As a recultivant, WWS and its mixtures with various soils are used. One of the problems with the use of WWS in this quality is its low density and bearing capacity. It was shown that the density and bearing capacity of the mixture increases when sandy and clay soils are added to WWS. WWS is mixed with soils of hazard classes 4 and 5 remaining after construction, thereby reducing the hazard class of the mixture [2].

The use of WWS in the reclamation of disturbed lands is regulated by GOST R 54534-2011 “Resource saving. Sewage sludge. Requirements for use for restoration of disturbed lands”. The permissible content of pollutants in WWS and their processed products when used for the restoration of disturbed lands is shown in table 2.
Table 2. Permissible content of pollutants in WWS and products of their processing when used for the restoration of disturbed lands (according to GOST R 54534-2011).

| Name of indicator, mg/kg dry matter, no more | Norm                  |
|---------------------------------------------|-----------------------|
|                                             | For technical remediation | For biological remediation |
| Mercury                                     | 30                    | 15                     |
| Chromium                                    | 2000                  | 1000                   |
| Lead                                        | 1000                  | 500                    |
| Cadmium                                     | 60                    | 30                     |
| Nickel                                      | 800                   | 400                    |
| Copper                                      | 1500                  | 750                    |
| Zink                                        | 7000                  | 3500                   |
| Arsenic                                     | 40                    | 20                     |

The level of use of WWS in Russian agriculture is, according to various estimates, from 4 to 7%. Agriculture in different countries uses from 10 to 90% of WWS, for example, 30–40% in Western Europe and about 60% in the USA [5–7].

The use of WWS as a fertilizer is regulated by the following legal documents: SanPiN 2.1.7.573-96 “Hygienic requirements for the use of wastewater and their rainfall for irrigation and fertilizer”, GOST R 17.4.3.07-2001 “Nature protection. The soil. Requirements for the properties of WWS when using them as fertilizers”, GOST R 54651-2011 “Organic fertilizers based on sewage sludge. Technical conditions”.

According to several authors, sewage sludge is a promising remediation agent [1, 8–9]. It is proposed to use WWS as organo-mineral fertilizers that have a positive effect on soil fertility, soil structure and components. At the same time, there is an increase in the content of organic matter, conservation of moisture and aggregation, and prevention of soil erosion.

There is evidence of a decrease in soil density, an increase in its total porosity, moisture capacity and content of water-resistant aggregates. It was shown that the introduction of precipitation into the soil has a prolonged effect, i.e. the impact persists for the next few years [10].

It was noted that the processes of humus formation during the introduction of WWS are enhanced. It has been established that in terms of dry matter content and basic nutrients, 10 million tons of WWS are equivalent to using about 50 million tons of organic fertilizers in the form of manure [1].

There are many studies in the literature proving an increase in crop productivity when rainfall is applied to the soil. The positive effect of WWS as a fertilizer on the bioproductivity and biological activity of the soil was noted. There is experience in the application of WWS in the cultivation of ornamental crops. Their positive effect on the growth and development of plants is noted [8].

However, many works do not provide data on the amount of environmentally harmful substances entering the soil and plants from WWS. In this case, the conclusions encountered in a number of works on improving the sanitary and environmental situation as a result of the introduction of WWS into the soil can be considered unreasonable.

In addition to impurities of organic and mineral substances, WWS incorporate general toxic, toxicogenetic, embryotoxic and carcinogenic compounds. According to several researchers, WWS contain heavy metals: chromium, cadmium, mercury, copper, lead, cobalt, zinc, molybdenum. They also contain toxic substances, pesticides, polychlorinated biphenyls, aliphatic compounds, esters, mono- and polycyclic aromatic substances, phenols, nitrosamines, nitrates. Of the biological objects - bacteria, protozoa, helminths, viruses [2].

The content of HM in WWS varies by city, which should be taken into account when using rainfall for remediation. The increased cadmium content was noted in Kazan and Novosibirsk, nickel in Nizhny
Novgorod, zinc in Novosibirsk and Nizhny Novgorod, and chromium in Kazan, Novosibirsk and Nizhny Novgorod [1].

To date, some methods have been developed to prevent the release of HM into the natural environment. One of the methods for reducing the concentration of HM is the creation of mixtures of WWS with neutral components in this respect - straw, sawdust, manure, peat, tree bark, etc. Composting WWS with various materials is one of the ways to prepare WWS for use, leading to the established standards the amount of toxic substances contained in them.

Experience with the use of composts based on WWS in agroecosystems showed an increase in yield, biological activity of the soil and did not cause the accumulation of HM in soil and plant products [5]. On the basis of experimental data, environmentally friendly doses have been established for the use of sewage sludge, increasing productivity, improving agrochemical parameters of the soil. At the same time, the concentration of HM in the soil and crop production remained within acceptable limits. An increase in crop productivity by 10-30% without a decrease in crop quality when using WWS is shown. It has been shown that chemical cleaning and vermicomposting techniques increase the safety and agronomic value of WWS [11].

Other studies have recommended the use of WWS only for fertilizing non-food crops such as lawns, flower beds, and nurseries. When making WWS under crops, the accumulation of HM in soil and products [12]. On the example of long-term experiments (5 rotations of a 7-field crop rotation), it is shown that the systematic application of WWS increases the content in the sod-podzolic soil of the content of total forms of HM 1.1–2.7 times, acid-soluble and mobile forms from 2.1 to 6 times. An increase in the concentration of HM in the soil upon the introduction of WWS leads to their accumulation in plants [13, 14].

Promising methods of removing harmful substances from the environment include phytoremediation - growing plants that accumulate an increased amount of HM in biomass. Studies have established the types of agricultural and wild plants that are most suitable for this method. However, the use of this method is limited by a number of conditions: the concentration of HM in the soil, under which plants can develop; the possibility of placing biomass; long recovery period.

There are other methods for the neutralization of solid waste from urban wastewater. One of these methods consists in the interaction of HM with actinomycete Str. chromogeness.g 0832, showing high specificity for protein contaminants. The degree of wastewater treatment ranged from 91.2 to 98.8% [8].

Another effective reagent used to neutralize HM are humic substances [1]. The ions of the reaction groups of humic substances react with HM ions, which leads to their retention in the composition of humus. However, humic substances in nature are decomposed by microorganisms, which leads to the release of HM and their inclusion in the biological cycle [15].

Thus, one of the directions for the placement of WWS in the medium can be their introduction into the soil – plant system. However, none of the methods guarantees reliable neutralization of toxic substances. Therefore, this problem requires further research and development of affordable and environmentally friendly ways of using WWS as fertilizers.

2. Object and methods of research

In order to determine the possibility of using sewage sludge for the preparation of remediation soils, a toxicological study of excess activated sludge after treatment of municipal wastewater was carried out. The studied sewage sludge contained nutrients: N - 0.45%, P₂O₅ - 0.88%, K₂O - 0.65% and had an alkaline reaction (pH – 10.7). The amount of heavy metals did not exceed the permissible gross content in accordance with GOST R 17.4.3.07-2001.

To determine the maximum dose of waste, laboratory experiments were carried out with the cultivation of cereal grasses. The experimental design consisted of five options: 1) control (acid peat); 2) background (calcareaeous peat); 3) peat + WWS 1:0.25; 4) peat + WWS 1:0.5; 5) peat + WWS 1:0.75; 5) peat + WWS 1:1.

3. Results and discussion
The results of the toxicological analysis of the soil in the experimental variants are presented in Table 3. An increase in the proportion of waste in the soil increased the gross content of all heavy metals. The maximum number of toxicants of the first hazard class was noted in the variants with the ratio of peat and WWS 1:0.75 and 1:1. Moreover, the cadmium content more than doubled the permissible standards for close to neutral and neutral soils.

**Table 3.** The content of heavy metals in the soil.

| Variants | Pb (mg/kg) | Ni (mg/kg) | Cu (mg/kg) | Cd (mg/kg) |
|----------|------------|------------|------------|------------|
| 1        | 3.43       | 6.61       | 11.07      | 0.48       |
| 2        | 7.70       | 5.97       | 17.61      | 0.60       |
| 3        | 18.35      | 5.26       | 26.34      | 1.60       |
| 4        | 24.99      | 24.99      | 41.86      | 2.99       |
| 5        | 45.94      | 14.95      | 64.16      | 5.26       |
| 6        | 39.24      | 15.67      | 79.74      | 5.40       |
| ODK      | 130.0      | 80.0       | 132.0      | 2.0        |
| HCP95    | 22.22      | 7.35       | 34.92      | 2.77       |

The copper content increased in proportion to the dose of waste, reaching a maximum in option 6 – 79.7 mg/kg. In general, the content of heavy metals of the second hazard class in all the studied variants does not exceed the permissible values.

An increase in the content of heavy metals in the studied soils had an ambiguous effect on their accumulation in plants. Figures 1–4 show the data of plant analysis in experimental variants. It is believed that elements such as lead, cadmium, nickel and copper have a great affinity for physiologically important organic compounds and are actively accumulated by plants. Entering plants, heavy metals are distributed very unevenly in their organs. The mechanisms of absorption and distribution of each element are closely related to the species characteristics of cultivated plants and the influence of environmental factors. Apparently, this caused differences in the accumulation of heavy metals in the aerial part of cereal grasses. In our studies, a clear relationship between the content of heavy metals in the soil and their entry into plants was noted only in the case of lead accumulation. The amount of nickel and copper in plants grown on soils with a maximum dose of waste increased by two to three times. The increased contamination of the recultivant with cadmium did not affect its entry into plants.
Figure 1. Lead content in plants, mg/kg.

Figure 2. Nickel content in plants, mg/kg.

Figure 3. Cadmium content in plants, mg/kg.

Figure 4. Copper content in plants, mg/kg.

4. Conclusion
Based on the studies, the following conclusions can be drawn: the ratio of peat and a mixture of WWS and ash can be considered environmentally safe – 1:0.25. The critical factor determining the limits for the use of municipal waste in the preparation of remediation soils is the cadmium content. The accumulation of heavy metals in plants is affected by soil pollution. Significant dependence was noted in the accumulation of lead by cereal herbs.

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