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COMPREHENSIVE EVALUATION OF THE PHYTOREMEDIATION ABILITY OF A NUMBER OF AGRICULTURAL CROPS FOR THE RESTORATION OF POLLUTED WITH HEAVY METALS SOILS

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During the work, the method of X-ray fluorescence analysis (XRF) has been used, which allows analyzing the content of various chemical elements from sulfur to uranium in samples of various nature (soil, plants). This method allows analyzing samples without complicated and lengthy sample preparation. In the course of work, model systems for heavy metals («soil–lead», «soil–cadmium», «soil–cadmium–lead») within the limits of 1–3 maximum permissible concentrations (MPC) for growing colza & flax have been compiled, bioelement gross concentrations have been determined. As a result of the work, one of the possible ways of removing heavy metals (HM) from contaminated soils, monitoring the environmental safety of a soil cover, as well as the opportunity to assess the phytoremediation ability of agricultural crops are proposed. A quantitative assessment of the content of trace and macroelements in the sod-podzolic soil as well as in model compositions based on it is presented. Bench studies on the cultivation of technical colza and flax on the proposed mixtures have been carried out. It has been shown that different concentrations of cadmium and lead have different effects on seed germination and plant growth dynamics. An increase in the concentration of heavy metals in the soil reduces the percentage of seed germination. Seed germination on the clean sod-podzolic soil is the highest – 80 %. The degree of the environmental safety of soils before and after the cultivation of technical colza and flax has been assessed. In some cases, the cultivation of technical colza and flax made it possible to reduce the total soil pollution by 3.1 times. It is shown that during the cultivation of industrial crops, metals are intensively absorbed and largely accumulate in the aboveground organs of plants. After harvesting the phytomass, the level of soil pollution risk is significantly reduced. Thus, the removal of chemical elements from the soil with industrial crops contributes to its self-cleaning. The results of the study will make it possible to expand the range of optimal crops for the phytoremediation of the soils contaminated with lead and cadmium.

Key words: X-ray fluorescence analysis; soil; heavy metals; agricultural crops, restoration of polluted soils.

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КОМПЛЕКСНАЯ ОЦЕНКА ФИТОРЕМЕДИАЦИОННОЙ СПОСОБНОСТИ РЯДА СЕЛЬСКОХОЗЯЙСТВЕННЫХ КУЛЬТУР ДЛЯ ВОССТАНОВЛЕНИЯ ЗАГРЯЗНЕННЫХ ТЯЖЕЛЫМИ МЕТАЛЛАМИ ПОЧВ

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Использован метод рентгенофлуоресцентного анализа (РФА), позволяющий анализировать содержание химических элементов (от серы до урана) в образцах различной природы (почвы, растений), без сложной и длительной пробоподготовки. Составлены модельные системы по тяжелым металлам (ТМ) «почва – свинец», «почва – кадмий», «почва – кадмий – свинец» в пределах 1 ПДК и 2–3 ПДК для выращивания рапса и льна, определены валовые концентрации биоэлементов. Предлагается один из вариантов удаления тяжелых металлов из загрязненных почв, контроля экологической безопасности почвенного покрова и определения фиторемедиационной способности сельскохозяйственных культур. Дана количественная оценка содержания микро- и макроэлементов в дерново-подзолистой почве и в модельных композициях. Проведены стендовые исследования по выращиванию ярового рапса и льна на предложенных смесях. Показано, что определенные концентрации кадмия и свинца оказывают различное влияние на всхожесть семян и динамику роста растений. Следует отметить, что увеличение концентрации тяжелых металлов в почве снижает процент всхожести семян, при этом на чистой дерново-подзолистой почве она самая высокая – 80%. Проведена оценка степени экологической безопасности почв до и после выращивания технического рапса и льна, что в некоторых случаях позволило снизить суммарный показатель загрязнения почвы в 3,1 раза. При выращивании технических культур металлы интенсивно поглощаются и в значительной степени накапливаются в надземных органах растений, поэтому после уборки фитомассы значительно снижается уровень опасности загрязнения почвы. Таким образом, вынос химических элементов из почвы с техническими культурами способствует ее самоочищению. Полученные результаты исследования дадут возможность расширить спектр оптимальных культур для фиторемедиации почв, загрязненных свинцом и кадмием.

Ключевые слова: рентгенофлуоресцентный анализ; почва; тяжелые металлы; сельскохозяйственные культуры; восстановление загрязненных почв.

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Introduction

The intensification of industrial and agricultural production, the development of transport, and the intensification of mining operations inevitably lead to the pollution of natural ecosystems with heavy metals. In this case, one of the main objects of pollution is soil. As a result of pollution, the quality of soils and the value of agricultural land are being reduced. One of the most serious aspects of this problem is that heavy metals and the products of their transformation entering the soil are absorbed by plants and accumulate in them in concentrations that are harmful to human and animal health. Currently, industrialized countries are actively developing economical and soft remediation technologies for the soils contaminated with heavy metals, which are based on the ability of specially selected species of higher plants and the associated microbiota to absorb and accumulate heavy metals in their biomass in quantities significantly exceeding their content in the growth environment. Subsequently, contaminated biomass is removed and utilized. Currently, about 400 species of hyperaccumulators of various metals from 22 families have been identified in the world, the use of which as phytoremediants is of great interest to researchers. At the same time, the study of phytoremediation processes raises a wide range of issues regarding the behavior of heavy metals in a soil-plant system, which enhances the relevance of research on this topic [1–10].

In addition, there remains an urgent issue of the suitability of the chosen territory for the safe functioning of the population, because, for example, in Ukraine, the lands subject to a strong anthropogenic impact with anthropogenic pressure of 1,5–3,0 MPC, which corresponds to weak and medium levels of pollution according to V. B. Ilyin (1995), are allocated for the development of agro-social complexes. Therefore, there is a need for the selection of crops resistant to heavy metal pollution. It should be noted that in conditions of low and medium pollution levels, phytostabilization is of great importance – the cultivation of crops that do not remove toxicants from the soil and, accordingly, do not accumulate them in their biomass. At a high level of pollution, phytoextraction is used – cleaning the soil due to the absorption of heavy metal cations by a root system, followed by their accumulation in the aerial parts of plants. Plants with high values of biological absorption and translocation coefficients

as well as with the basipetal distribution of chemical elements across organs will be the most promising phytoextractors, While the main requirement of phytostabilization is the acropetal distribution of toxicants, which, in the presence of a high geochemical barrier and protective mechanisms of the plant itself, will provide high-quality crop products that comply with sanitary standards with heavy metals in the commodity part within the MPC. However, it is known that the degree of tolerance to heavy metals varies greatly not only among different crops, but even by variety within a given crop.

The excessive content of heavy metals in soils negatively affects the growth and development of agricultural plants, deteriorates the quality of products. The latter occurs mainly not due to changes in chemical composition, but as a result of excessive accumulation of heavy metals. There is a direct, but far from adequate connection between the content of heavy metals in the soil and the culture grown on it – on highly contaminated, but highly protected soil, it is possible to obtain a hygienically acceptable crop. With the excessive intake of heavy metals through roots, there work plant protective mechanisms of nonspecific nature, which restrict the penetration of heavy metals into the aboveground organs and metabolic centers of cells. In relation to various heavy metals, the protective capabilities of the plant are not the same: lead is mainly retained already in roots, cesium penetrates relatively easily into the aboveground organs of plants [11–13].

Up to 25 % of the territory of the Republic of Belarus is contaminated with heavy metals, radionuclides and, as a result, is excluded from agricultural circulation. It is shown that technical colza (*Brassica napus*) as the most promising crop for cultivation in the Republic is not afraid of soils contaminated with radionuclides and heavy metals. According to preliminary data, the land previously withdrawn from agricultural circulation can potentially be used as land for growing raw materials used in the production of biofuels, in particular, based on technical colza. In addition, the Republic will be able to obtain significant economic benefits, since the production capacities of Belarus will be sufficient not only for the «closur» of domestic consumption, but also for the potential export of surplus biofuels to the EU countries [14–15].

Another crop resistant to pollution is ordinary flax or sowing (*Linum usitatissimum*). In the whole country, the area of suitable soils for flax makes up 28,1 % of the total area of arable land. It ranges from 37,9–38,9 % in the Grodno and Mogilev regions to 8,0–8,7 % in the Brest and Gomel regions; in districts – from 40–50 % in the northern districts of the Grodno, Mogilev, Vitebsk and Minsk regions, to almost complete absence in the southern districts of the Brest and Gomel regions. Flax cultivation in Belarus is planned to be given special attention, and in the near future, a program will be developed for the integrated development of the flax growing industry in Belarus and Russia [16].

For a more objective assessment of the influence of heavy metals under conditions of agrocenosis, several additional indicators are introduced:

- environmental safety coefficient (ESC);
- mobility coefficient of heavy metals in soils;
- «The zincous equivalent».

The environmental safety coefficient (ESC) is the ratio of the MPC for the soil to the gross metal content in this soil. The ESC is always higher on the light-sized soils and soils with low buffering ability. The authors of [17] believe that the environmental safety coefficient should be $MPC / C_{HM} > 2.0$ for the sod-podzolic, gray forest soils. If this condition is not satisfied, it can't guarantee the production of clean products. The determination of the mobility coefficient of HM in various types of soils gives a real idea of its migration ability within agricultural lands. It is proposed on the basis of comprehensive monitoring definitions. For example, the maximum permissible concentration of lead for soil is 30,0 mg/kg, and its translocation index is 35,0. For cadmium APC (approximately permissible concentration) – 0,5–2,0 mg / kg. For lead, OEC is 32–130 mg / kg. In both cases, the hazard class is the first [17–23].

Thus, the proposed topic is relevant and fits into international environmental trends.

The aim of the work is to study the possibility of using crops as phytoextractors or phytostabilizers in conditions of soil pollution with heavy metals.

Materials and research methods

Characteristics of the research object, sampling and sample preparation. Soil compositions were created on the basis of the light loamy sod-podzolic soil with the addition of heavy metal salts: cadmium sulfate ($CdSO_4$) and lead nitrate ($Pb(NO_3)_2$) in various ratios of 1–3 MPC. A total of 12 samples were created for further posterior observations. Further three containers were prepared with each of the soil compositions proposed above – 36 in total. Each container had a volume of 1,14 dm³, and the area for sowing was 0,143 m², respectively.

To account for the total heavy metal accumulation in plants in the «soil-plant» system, technical colza and flax were selected. Seeding rates for these crops were reduced to 80 seeds per 1 m², so that no more than 9 seeds were sown in each prepared container, since a denser sowing could lead to stretching and weakening of the aerial part of the plants.

- 1) Soil 100 % (3 samples);
- 2) Soil 90 % + 2–3 MPC of cadmium (3 samples);
- 3) Soil 90 % + 2–3 MPC of lead (3 samples);
- 4) Soil 90 % + 2–3 MPC of cadmium and lead (3 samples).

The samples of soil and plants were taken from each composition for further sample preparation and research by the XRF method according to the methodologies of MP.MN 4092-2011, MP.MN 3272-2009.

The stages of sample preparation for X-ray fluorescence analysis are presented in fig. 1.

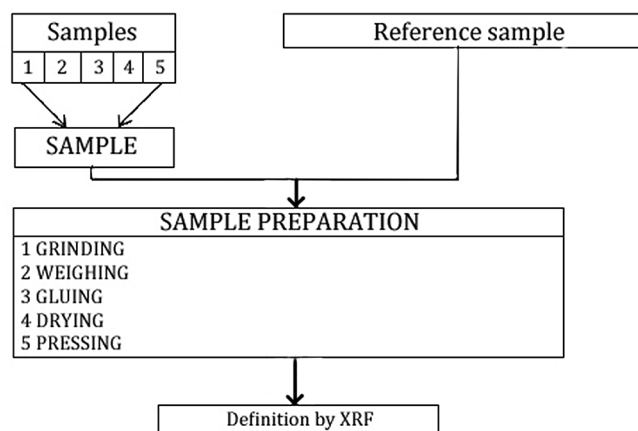


Fig. 1. Stages of sample preparation for XRF

X-ray fluorescence analysis is one of many modern physicochemical methods of measurement and widely used for a qualitative, semi-quantitative and quantitative determination of the elemental composition of substances.

Results and discussion

The gross content of chemical elements, which characterizes the degree of danger of soil pollution and allows controlling it, is an important indicator of soil pollution. When conducting a quantitative analysis of the studied soil compositions as fertilizers, it has been revealed that all samples contain 13 chemical elements that pose the greatest environmental hazard: As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sn, Ti, Zn, Zr (according to Directive 86/278/EEC of June 12, 1986).

Determination of biological absorption coefficients for crops: technical colza, flax at various levels of soil pollution.

The results of calculating the biological absorption coefficients of heavy metals for the phytomass of technical colza and flax grown on the soils with various degrees of contamination with lead and cadmium are presented in tabl. 1–2.

As can be seen from the data presented, severe accumulative metals have not been detected. Lead and zinc are strongly accumulative. Arsenic, copper, lead and strontium are weak accumulative or secondary gripping elements. Other metals belong to a weak capture group.

Translocation coefficient (TC) – is the ratio of the content of an element in the aerial part to the content in roots. The translocation coefficient is calculated by the formula 1:

$$TC = C_a/C_r, \quad (1)$$

where C_a – concentration of an element in the aerial part of a plant;

C_r – concentration of an element in the root of a plant.

The translocation coefficients of heavy metals for technical colza and flax grown in the soils with different degrees of contamination have been calculated under obtained data. The results are presented in tabl. 3–4.

As can be seen from the data presented in tabl. 3–4, cadmium, manganese, tin, and strontium are most intensively accumulated in colza phytomass. Moreover, cadmium most intensively passes into the aboveground organs of the plant with a dominant soil contamination.

In the flax phytomass, cadmium, tin, and zinc accumulate most intensively. The accumulation of cadmium in the leaves intensifies with an increase in its concentration in the soil. Tin to a greater extent passes from the roots to the aerial part of the plant at moderate lead contamination. With increasing the concentrations of lead and cadmium in the soil, the transition of zinc to the phytomass becomes more difficult.

Table 1

Biological absorption coefficients of heavy metals for colza grown on soils with various degrees of contamination

Plants	Biological Absorption Coefficients										
	As	Cd	Co	Cu	Fe	Mn	Pb	Sn	Sr	Zn	Zr
Colza, grown on pure soil	–	0,0301	–	0,3068	0,0288	0,0437	0,3144	0,9633	0,2990	1,6642	0,0223
Colza, grown on soil with 1 MPC of Cd	0,1685	0,012	–	0,1588	0,0264	0,0544	0,1441	0,5699	0,4444	0,9857	0,0482
Colza, grown on soil with 3 MPC of Cd	0,4118	0,0056	–	0,2563	0,0307	0,0608	0,1262	0,8882	0,4085	1,15	0,0442
Colza, grown on soil with 1 MPC of Pb	–	0,0640	–	0,3889	0,0186	0,0414	0,0624	1,0006	0,3380	1,1392	0,0273
Colza, grown on soil with 3 MPC of Pb	0,191	0,0541	–	0,1842	0,0258	0,0612	0,0624	0,8857	0,4357	0,8912	0,0498
Colza, grown on soil with 3 MPC of Cd & Pb	0,3015	0,0085	–	0,2527	0,0144	0,041	0,0912	0,7111	0,03401	1,2091	0,0764

Table 2

Biological absorption coefficients of heavy metals for flax grown on soils with various degrees of contamination

Plants	Biological Absorption Coefficients										
	As	Cd	Co	Cu	Fe	Mn	Pb	Sn	Sr	Zn	Zr
Flax, grown on pure soil	–	0,0452	0,0192	0,3378	0,0139	0,0352	0,1394	0,5691	0,1618	1,4938	0,0105
Flax, grown on soil with 1 MPC of Cd	0,1797	0,026	0,0489	0,3281	0,0365	0,0820	0,1412	0,5342	0,2611	1,4500	0,042
Flax, grown on soil with 3 MPC of Cd	0,4967	0,0074	0,0276	0,1750	0,0319	0,0610	0,0615	0,4929	0,2554	0,916	0,0609
Flax, grown on soil with 1 MPC of Pb	–	0,0327	–	0,5278	0,0066	0,0725	0,0402	0,4343	0,1901	1,9303	0,0199
Flax, grown on soil with 3 MPC of Pb	0,1348	0,0348	0,0772	0,1807	0,0274	0,0633	0,039	0,4780	0,2028	0,601	0,488
Flax, grown on soil with 3 MPC of Cd & Pb	–	0,0058	0,1552	0,1771	0,0116	0,0273	0,0162	0,4053	0,2139	1,0242	0,0333

Table 3

Translocation coefficients of heavy metals for colza grown on soils with various degrees of contamination

Plants	Translocation coefficients										
	As	Cd	Co	Cu	Fe	Mn	Pb	Sn	Sr	Zn	Zr
Colza, grown on pure soil	–	2,545	–	0,071	0,508	1,903	0,330	2,589	1,089	0,895	0,336
Colza, grown on soil with 1 MPC of Cd	0,100	8,182	–	0,057	0,603	2,399	0,229	1,467	1,199	0,728	0,693
Colza, grown on soil with 3 MPC of Cd	0,201	7,962	–	0,093	0,699	2,673	0,192	2,284	1,102	0,846	0,550
Colza, grown on soil with 1 MPC of Pb	–	4,363	–	0,158	0,469	1,568	0,420	3,095	0,892	0,888	0,268
Colza, grown on soil with 3 MPC of Pb	0,122	3,318	–	0,069	0,610	2,399	0,288	2,841	1,143	0,842	0,633
Colza, grown on soil with 3 MPC of Cd & Pb	0,295	3,944	–	0,107	0,375	1,900	0,159	2,818	0,983	0,955	0,667

Translocation coefficients of heavy metals for colza grown on soils with various degrees of contamination

Plants	Translocation coefficients										
	As	Cd	Co	Cu	Fe	Mn	Pb	Sn	Sr	Zn	Zr
Flax, grown on pure soil	–	0,910	0,006	0,122	0,071	0,373	0,099	4,830	0,792	2,745	0,073
Flax, grown on soil with 1 MPC of Cd	0,08	3,65	0,246	0,235	0,227	0,475	0,086	9,911	0,605	2,152	0,303
Flax, grown on soil with 3 MPC of Cd	0,170	5,346	0,138	0,125	0,195	0,353	0,035	8,671	0,528	1,360	0,399
Flax, grown on soil with 1 MPC of Pb	–	0,446	–	0,436	0,046	0,368	0,103	9,646	0,433	3,034	0,087
Flax, grown on soil with 3 MPC of Pb	0,060	0,429	0,404	0,135	0,177	0,327	0,520	11,000	0,454	1,149	0,311
Flax, grown on soil with 3 MPC of Cd & Pb	–	1,355	0,877	0,151	0,082	0,169	0,044	11,544	0,529	1,633	0,146

The assessment of the chemical pollution of agricultural field soils (control soil) by gross forms of chemical elements using the total pollution indicator has shown that the initial soil is characterized by a moderately hazardous level of pollution initially (before growing) and after collecting flax and colza ($Z_c = 23,9$, $Z_c = 18,5$ and $Z_c = 20,6$ respectively).

According to the indicative scale of pollution hazard, the soil with 1 MPC of cadmium is assessed as hazardous ($Z_c = 41,7$), and after collecting flax and technical colza, it is already assessed as moderately hazardous ($Z_c = 27,5$, $Z_c = 28,9$ respectively). The following pattern is traced by the accumulation of chemical elements: $Fe > Cd > Sn > Mn > Cu > Co$.

The soil with 3 MPC of cadmium is characterized by a dangerous level of contamination initially and after harvesting flax and technical colza ($Z_c = 98,1$, $Z_c = 77,2$, $Z_c = 89,9$ and $Z_c = 38,7$ respectively). Elements enter the soil to varying degrees as follows: $Cd > Fe > Mn > Cu > Co$.

The soil with 1 MPC of lead is characterized by a moderately hazardous level of contamination initially and after harvesting flax and technical colza ($Z_c = 28,7$, $Z_c = 21,8$ and $Z_c = 25,5$ respectively). Elements enter the soil to varying degrees as follows: $Fe > Sn > Pb > Cd > Mn > Cu > Co$.

According to the averaged total indicators of contamination by the gross forms of the studied elements, the soil with 3 MPC of lead is assessed as dangerous ($Z_c = 37,0$). After collecting flax and technical colza, this soil is already assessed as moderately hazardous ($Z_c = 27,3$ and $Z_c = 28,4$ respectively). Elements enter the soil to varying degrees as follows: $Fe > Sn > Pb > Mn > Cu > Co > Cd$.

The soil with 3 MPC of cadmium and lead is characterized by a dangerous level of contamination initially and after flax collection ($Z_c = 60,7$ and $Z_c = 37,8$ respectively). After collecting technical colza, this soil is moderately hazardous ($Z_c = 19,3$). Elements enter the soil to varying degrees as follows: $Fe > Cd > Sn > Pb$.

The dynamics of changes in the degree of soil hazard after growing technical colza and flax are shown in fig. 2–7.

From the data shown in Figures 2–7, it is seen that the degree of danger of the original soil after growing flax is reduced by 1,3 times, technical colza – by 1,2 times. The total pollution index for the soil with 1 MPC of Cd after growing flax decreases by 1,5 times, colza – by 1,4 times. The risk of the soil with 3 MPCs of Pb after growing flax is reduced by 1,4 times, technical colza – by 1,3 times. The degree of soil hazard from 1 MPC of Pb after growing flax is reduced by 1,3 times, technical colza – by 1,1 times. The total pollution index for the soil with 3 MPCs of Cd after growing flax is reduced by 1,3 times, technical colza – by 1,1 times. The risk of the soil with 3 MPCs for Cd and Pb after growing flax is reduced by 1,6 times, technical colza – by 3,1 times. With simultaneous contamination of the soil with several heavy metals, the phytostabilizing ability of colza is also activated.

It is shown that the removal of chemical elements from the soil with industrial crops (colza and flax) contributes to its self-cleaning.

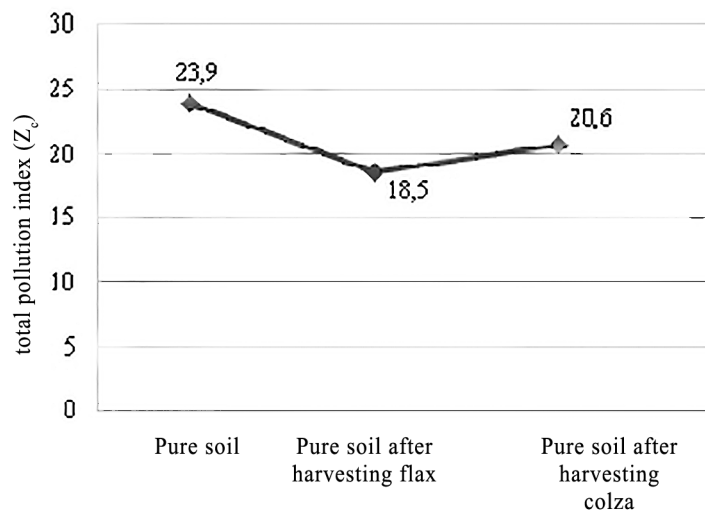


Fig. 2. Change in the degree of danger of the initial soil during the cultivation of flax and technical colza

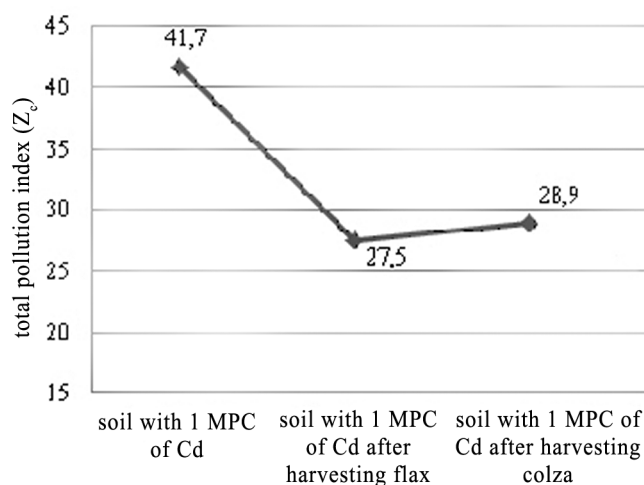


Fig. 3. Change in the degree of soil hazard from 1 MPC of Cd when growing flax and technical colza

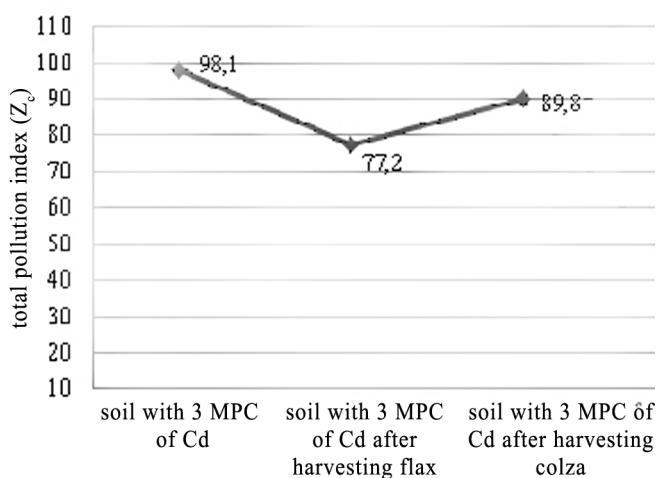


Fig. 4. Change in the degree of soil hazard from 3 MPC of Cd when growing flax and technical colza

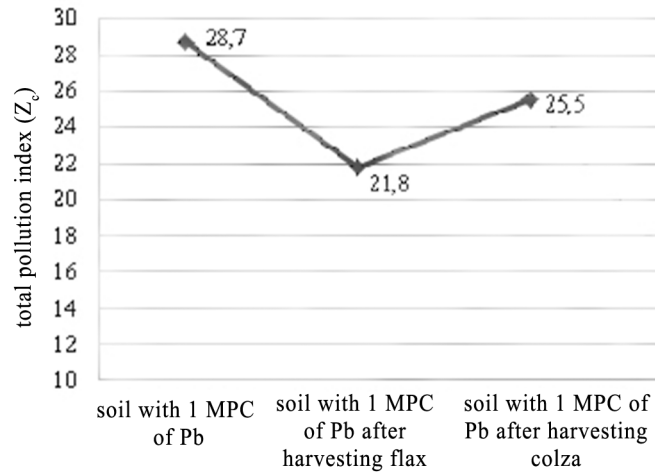


Fig. 5. Change in the degree of soil hazard from 1 MPC of Pb when growing flax and technical colza

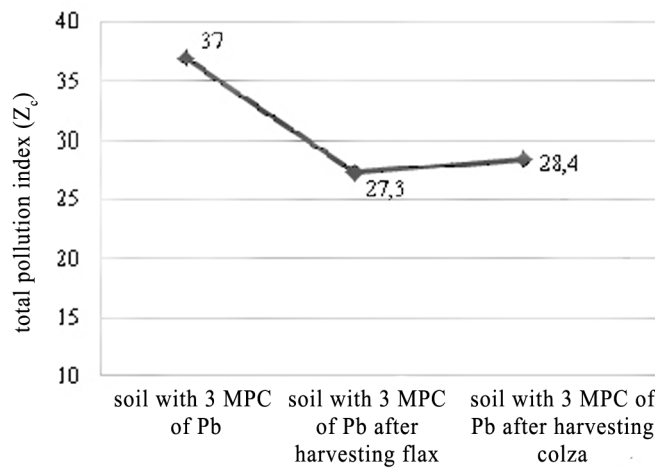


Fig. 6. Change in the degree of soil hazard from 3 MPC of Pb when growing flax and technical colza

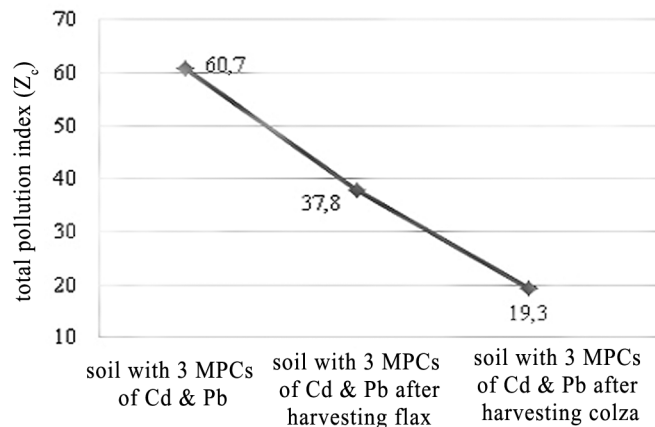


Fig. 7. Change in the degree of soil hazard from 3 MPCs of Cd & Pb when growing flax and technical colza

In the areas of industrial emissions and possible soil pollution by heavy metals, it is necessary to select industrial crops as phytoremediants with barrier-free functions with respect to the influx of heavy metals into them, which will allow for the ecologically safe restoration of the soils previously removed from crop rotation.

Thus, the possibility of using technical colza as a promising phytoremediant for the Belarusian soils contaminated with other chemical elements, except lead and cadmium, needs to be studied further.

Conclusion

1. The possibility of using the method of X-ray fluorescence analysis to assess the content and migration of heavy metals of various degrees of danger in a soil-plant system has been shown.
2. It has been established that cadmium, manganese, tin and strontium accumulate most intensively in the phytomass of industrial colza; cadmium, tin, zinc accumulate in the flax phytomass.
3. Based on the data obtained, it can be assumed that the degree of danger of the soil, depending on the nature of its pollution with heavy metals, is reduced by 1,3–1,6 times after growing flax, by 1,2–3,1 times after growing colza. Moreover, the phytostabilizing properties of technical colza are manifested to a greater extent with the initially high values of the total soil pollution index.
4. It has been shown that when growing industrial crops, metals are intensively absorbed and accumulate in the aboveground and underground organs of plants, which significantly reduces the level of risk of soil pollution. The removal of chemical elements from the soil with industrial crops (colza, flax) contributes to its self-cleaning.

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