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Innovative Bioremediation Technology of Lands Polluted with Chlororganic Pesticides

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Abstract. A complex of *ex situ* bioremediation technology proposed for lands contaminated with organochlorine pesticides, which differs from the existing ones by significantly lower material and time costs and accelerated return of polluted territories to the economy. To establish the contour of the pollution area it is proposed to use mathematical modeling of the pollution spread. The calculated dependencies for point and linear surface sources of pollution are established. The technology based on the use of humus as an affordable and inexpensive carrier of microflora, carrying out the metabolic decomposition of pesticides by microorganisms and accelerated low-cost determination of the volumetric configuration of land contaminated with organochlorine pesticides more of their standard concentration. It is established that the best bio-stimulation of autochthonous natural microflora in the studied process is 45% humus in a mixture contaminated with organochlorine pesticides.

Key words: ex situ bioremediation, organochlorine pesticides, low time and material costs, microflora carriers, pollution.

Introduction

The increasing global demand for food products has required intensification of their production (Royal Society, 2009). One of its directions was the development and use of pesticides in agriculture (Aktar et al., 2009). Their use increased the number of food products but caused an acute, ever-increasing and difficult to solve the problem of chemical pollution of land (Zhang et al., 2011), which, in turn, led to a deterioration in the quality of agricultural products and a decrease in food safety (Carvalho, 2017). The solution to this problem has long been recognized as an urgent task, but, nevertheless, the area of pesticide-contaminated land around the world is constantly growing. It is due to the lack of fairly simple, low-cost technologies that can quickly stop the migration of pesticides in the environment and quickly restore the contaminated area (FAO, 2011). This work is devoted to the development of just such a technology.

Organochlorine pesticides (OCPs) are among the most dangerous pollutants of agricultural land because they have high toxicity, stability, long migration in ecosystems (Cernansky, 2015), and high cumulative effect (Aichner et al., 2013; Solovyanov et al., 2017).

For any content of OCPs in agricultural lands, it is necessary to remediate them, but urgent attention should be paid to plots of land contaminated with OCPs above their normative level since over time the area of dangerous pollution and all kinds of damage from it increase significantly (Yankevich et al., 2015).

We will conduct a brief critical analysis of the most common methods for soils, groundwater, and surface water treatment that are worldwide used:

- adsorption methods (Sakalova et al., 2019; Zelenko et al., 2019). These methods enable to achieve a high degree of purification but require a large number of adsorbents (the most perspective is the use of natural adsorbents: zeolites, bentonites, glauconites). Further utilization of spent sorbents is also a problem;
- reagent methods (Tulaydan et al., 2017). These methods enable to achieve almost complete purification but often require special equipment and conditions for the implementation of chemical reactions and require the use of special, often expensive reagents. Quite often there is a danger of secondary contamination by reaction products;
- biological methods (Malovanyy et al., 2016; Malovanyy et al., 2019) allow achieving the required degree of purification at a minimum cost. In the case of anaerobic methods, the decomposition of biomass is accompanied by the synthesis of biogas which is a valuable energy source (Nykyforov V. et al., 2016). The disadvantage is the significant duration of biological processes.

Numerous ex situ, in situ and on situ remediation technologies for contaminated land based on physicochemical, chemical, thermal, extraction and separation processes have been proposed, but their implementation limited by many factors, that reduces their effectiveness and use. Among these key factors are the technological complexity of the processes, significant material costs of implementation, and the likelihood of negative impacts on local natural processes (Yurchenko et al., 2014). Therefore, these technologies are not widely used in the practice of bioremediation of lands contaminated with OCPs and were used mainly on experimental sites.

During bioremediation of lands contaminated with OCPs, technologies based on the use of pesticide decomposition processes by various microorganisms in their metabolism are most prevalent. The main advantage of these technologies in ex situ, in situ and on situ implementations is the natural course of the processes and their low cost, and the common disadvantage is a long time of the destruction of OCPs, during which large areas of the contaminated area do not return to economic circulation. Also, with in situ and on situ technologies, prolonged migration and accumulation of OCPs in the components of the environment and the various negative consequences associated with this (Yurchenko et al., 2014) remain. In this regard, ex situ technologies are preferable.

The process of destruction of OCPs during bioremediation can be enhanced by using various biological products containing artificially obtained microorganisms destructors (Yurchenko et al., 2014). In this work, this direction, as a possible modern technology, was not considered, since it is in the stage of experimental research. The most promising at the present is a simple and inexpensive technology in which bio-stimulation of autochthonous microflora destructors carried out. It should be noted that this process in each case requires the development of its technological parameters since they directly depend on the type of land, microflora contained in it, types of OCPs and their concentration.

The most promising direction in the bioremediation of lands contaminated with OCPs is a simple and inexpensive composting technology in which an environmentally friendly process of decomposition of environmentally hazardous products into non-toxic products occurs. A compost mixture prepared by mixing contaminated soil with biological mass. Livestock manure, bird droppings, activated sludge from municipal sewage treatment plants, alfalfa, hay, straw, peat, grass, etc. can be used as a biological mass (Yurchenko et al., 2014).

It should be noted that this process in each case requires the development of the composition of the compost mixture since it directly depends on the type of land, its level of pollution and the applied biological mass.

One of the key problems in the remediation of contaminated lands is the determination of their spatial configuration and volume. Existing approaches are based on the collection of a large number (up to several hundred) of samples by area and depth for contouring the pollution zone. At the same time, sampling from depths of more than 2 m is technologically complicated and highly expensive, although according to available data, soil pollution above standard values can be observed at depths of up to 6 m (Ukrainian Research Institute for Environmental Issues, 2013). Therefore, field studies, as a rule, make it possible to determine with sufficient accuracy only the area, but not the spatial configuration and the amount of pollution.

Therefore, the development of ex situ bioremediation technology for contaminated lands, devoid of these shortcomings, is an urgent and practically important task.

The study aimed to create a new ex situ bioremediation technology for lands contaminated with OCP, which differs from the existing ones by significantly lower material and time costs.

To achieve this goal, it was necessary to solve the following tasks:

– to create a way of the accelerated, low-cost determination of the spatial configuration of land contaminated with OCPs above the normative value;

– to develop a methodology for determining the composition of the mixture for composting;

– to develop a low-cost technological scheme of isolated bioremediation of lands with the accelerated return of contaminated territories to economic circulation.

Materials and Methods

Determination of the spatial configuration of the land contaminated with OCPs above the standard value. The need for the extraction and relocation of significant volumes of land, which is characteristic of ex situ technology, makes it important to reduce the time and money costs associated with determining the spatial configuration of the pollution area. Existing approaches are based on the collection of a large number (up to several hundred) of samples by area and depth for contouring the pollution zone. At the same time, due to technological and financial limitations, full-scale studies at depths exceeding 2 meters are rather rare.

In this paper, we propose a method based on the use of a mathematical model. In this case, full-scale studies are necessary to identify the parameters of the model, to verify it, and to carry out refinement analyzes on the calculated contour of the pollution area, which can significantly reduce the volume of full-scale studies and, therefore, the associated costs.

The standard concentration of OCP adopted as a criterion for determining the boundary of the pollution zone.

The proposed methodology developed for two types of surface sources of pollution - point and linear.

As a calculation basis in the case of a point source based on preliminary studies (Ukrainian Research Institute for Environmental Issues, 2013), an exponential regression model of the form was chosen:

$$C = C_{\text{мах}} e^{\beta h + \alpha r}, \qquad (1)$$

where C – is the concentration of OCP at the calculated point, mcg/kg; $C_{мах}$ – the concentration of OCP in a point source, mcg/kg; r – the distance from a point source of pollution, m; h – depth, m, a and b – model parameters.

Taking into account that the boundary of the pollution zone is the achievement of the standard concentration of OCP Cs, based on equation (1), it is possible to determine the boundaries of the pollution zone. For this, we take the logarithm (1) and obtain:

$$\ln C_s = \ln C_{max} + \alpha r_s + \beta h_s, \quad (2)$$

where C_s – is the standard concentration, g/m^3; r_s – the radius of the area of pollution on the surface of the earth, m; h_s –the depth of the pollution zone, m.

Equation (2) shows that the pollution zone has the configuration of an inverted cone with an axis perpendicular to the surface of the earth and passing through the point of the pollution source (Fig. 1).

The characteristics of the pollution cone – the radius $r_{s\ max}$ and the height $h_{s\ max}$, , which determine the maximum depth and diameter of the pollution zone, can be determined from equation (2) with $h_s = 0$ and $r_s = 0$, respectively, and $C = C_s$:

$$r_{smax} = \frac{1}{\alpha}\ln\frac{C_s}{C_{max}}; \; h_{smax} = \frac{1}{\beta}\ln\frac{C_s}{C_{max}}. \quad (3)$$

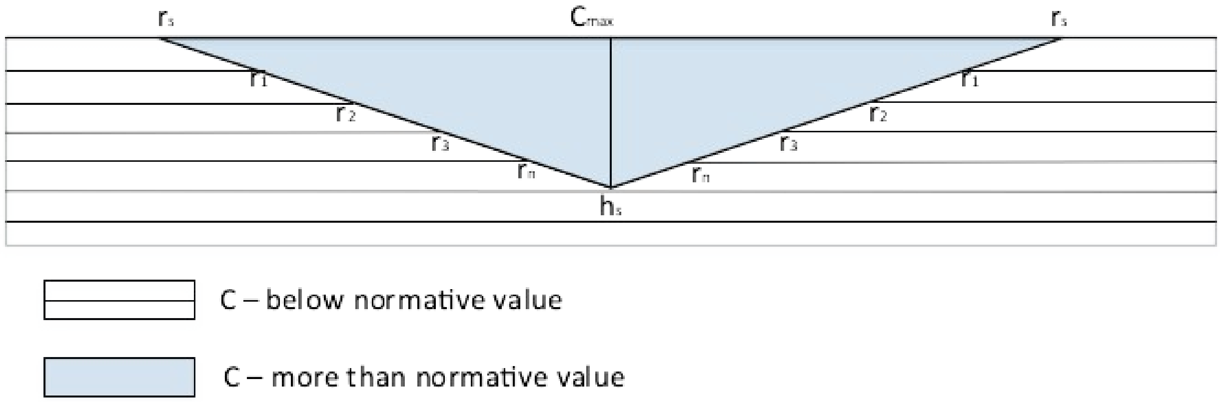

Fig. 1. Configuration of the pollution zone.

Determining the configuration of the pollution zone using the proposed model involves the following sequence of actions:

- based on a limited arbitrary set of field data of the measurements OCP concentrations are determined using the least square method and the parameters of the linear regression equation (2). Theoretically, four measurements are enough for this, but in practice, there should be more measurements (about 10-15). It is mandatory to have samples from various depths, although the depths themselves are not fundamental and may be limited to relatively small values (up to 1 m);
- an assessment made of the adequacy of the model based on one of the statistical criteria. As the latter, in particular, the coefficient of determination or the Fisher criterion can be used. In the absence of adequacy, an additional set of field data and the repetition of this stage are necessary;
- based on independent field data, the reliability of the model evaluated;
- based on the model, the boundaries of the pollution zone calculated.

The calculated values used to determine the control sampling points confirming the correctness or inaccuracy of the calculated data. In the latter case, the obtained data used to refine the model parameters and conduct repeated calculations with their subsequent control.

Thus, it is possible to use a small number of sampling points of soil components at an arbitrary distance, for example, from 1 m to 100 m from the center of pollution and, at a shallow depth, for example, from 0.1 m to 1 m, which significantly reduces the time of sampling and analysis samples and reduces the cost of this work.

The process of determining the spatial configuration of the area of contamination of soil components by a linear source of pollution is similar to the procedure for a point source of pollution. The difference lies in the use of a regression model, in which the coordinates of the calculated point will be characterized by three parameters measured relative to the center of the pollution line: depth and distance along the surface of the earth in directions parallel and perpendicular to the direction of placement of the linear source on the ground.

In this case, the regression equation represented as:

$$C = C_{\text{мах}} e^{\beta h + \alpha f(x,y)}, \quad (4)$$

where x and y – are the distances along the surface of the earth from the center of the linear source in the directions parallel and perpendicular to the direction of its placement, respectively, m.

$$f(x,y) = \begin{cases} y & x \le d \\ \sqrt{(x-d)^2 + y^2} & x > d \end{cases}, \quad (5)$$

where d – is the distance from the center of the linear source to its boundary, m.

Then, after linearization, the equation for calculating the boundaries of the pollution zone will take the form:

$$h_s = -\frac{\alpha}{\beta} f(x_s, y_s) + \frac{1}{\beta} \ln \frac{C_s}{C_{max}}. \quad (6)$$

where x_s and y_s – are the coordinates of the boundary of the pollution zone on the surface of the earth, m.

The maximum depth of the contamination zone $h_{s\,max}$ was reached on the axis of the source $y_s = 0$ within its length $x_s = 0$ (for $x_s < d$):

$$h_{s\,max} = \frac{1}{\beta} \ln \frac{C_s}{C_{max}}; \quad (7)$$

The depth of the contamination zone within the length of the source decreases linearly in the direction perpendicular to the source. On the axis of the source outside, the depth of the contamination zone decreases linearly with distance from the source.

The boundary of the pollution zone on the earth's surface is determined from (6) at $h_s = 0$:

$$f(x_{s\,max}, y_{s\,max}) = \frac{1}{\alpha} \ln\left(\frac{C_s}{C_{max}}\right). \quad (8)$$

The contamination area in the plan limited by straight lines parallel to the source within its length and semicircles with centers at the ends of the source - outside its length.

Method for determining the composition of the mixture for composting. OCP biodegradation is the result of the inclusion of many organisms in the metabolic decomposition processes, but bacteria play a key role. The intensity of bioremediation processes depends on several factors. First of all, it is the presence of bacteria adapted to this type of OCP, and environmental factors - soil structure, temperature, pH, oxygen regime. Bioremediation is a technology aimed at creating favorable conditions for the growth of microorganisms that transform and decompose OCPs. In general, this technology may include the addition of microorganisms (bio-augmentation), the use of nutrients to stimulate natural microorganisms (bio-stimulation), the ventilation of the soil to saturate it with oxygen, and the removal of gases generated during the decomposition of OCP (bioventilation).

Preliminary studies of the authors showed that contaminated sites contain microorganisms adapted to OCP. However, usually, the use of OCPs in the metabolism of microorganisms cannot serve as a complete source of energy for them. Therefore, it is critically important to create conditions for bacterial cometabolism by introducing a substrate suitable for microorganisms with the formation of a compost mixture. Given the large volumes of land being restored, such a substrate should have accessibility and low cost. Therefore, for these purposes, affordable and cheap materials of natural origin were selected - peat, humus, and straw.

The method of determining the composition of the mixture for composting was developed by us (Yurchenko et al., 2014) and adapted to the conditions of the study. When developing a methodology for determining the composition of a mixture for composting, a compost mixture was prepared by mixing contaminated DDT soil with a biological mass in a quantitative ratio of 10 - 95% of contaminated soil to 30 - 70% of the biological mass. As a biological mass, cattle manure, straw, and peat were used.

Composting was carried out for seven weeks. The soil contaminated with DDT (180 mg/kg) was mixed with the biomass of the above species and placed in laboratory composters, 65 g of the mixture in each composter.

During composting, the mixture in test composters was saturated with oxygen (within 5 days) and nitrogen (within 2 days). To maintain the temperature in the range of 25-50 °C, the mixture was placed in an incubator. The humidity of the mixture was maintained at a level of 60 - 80% by adding water twice a week, and a pH of 5.25 - 9.0. After seven weeks, the samples were treated with methylene chloride and the resulting extract was analyzed on a chromatograph with an ECD detector to determine the residual content of DDT.

Development of a method for isolated bioremediation of contaminated OCP land above standard values. The authors (Yurchenko et al., 2014) developed and adapted to the research conditions a method of determining the composition of the composting mixture. The destruction of OCPs contained in the earth is proposed to be carried out in isolation from uncontaminated land, which will prevent the migration of pollutants during the process. Initially, it extracts the soil contaminated with OCPs above their standard value and mixes it with humus in the ratio of 55% and 45%, respectively. At the same time, on a non-economic territory, a corresponding depression is created with a waterproof bottom and walls and a mixture of contaminated land with humus placed in it. In the resulting isolated volume of the mixture, the destruction of OCP by microflora will occur. Isolation of the remediation volume of the earth prevents the penetration of OCPs with various natural aqueous solutions into unpolluted lands and interrupts their movement along trophic chains. Uncontaminated soil components that removed during the creation of the recess filled with a foundation pit formed after removal of soil components contaminated with OCP. The stages of the process of this remediation:

1. Determine the volumetric configuration of the land contaminated with OCP above the standard value.

2. Dig up the soil contaminated with OCP above the standard value, forming a depression. The excavated land moved to the non-economic territory.

3. Mix the displaced soil contaminated with OCP above the standard value with humus

4. On a non-economic territory, a recess is created for bioremediation of land contaminated with OCPs above the normative value, and its bottom and walls are insulated to stop the migration of OCPs.

5. Fill the isolated recess with a mixture of soil contaminated with OCP above the standard value, and humus, and fill the recess obtained during the excavation of the

soil contaminated with OCP above the standard value, not contaminated land removed when creating the recess with insulation.

Results and Discussion

Determination of the volumetric configuration of the OCP contaminated soil. During development of a method for fast, low-cost determination of the volumetric configuration of the OCP contaminated soil by a point source the plot studies data were obtained on agricultural territories of former pesticide storage warehouses in the Kharkiv Region, characterized by a small area which allows us to consider them as the main pollution sources. At present warehouses have been liquidated but the residual pollution of OCPs according to the monitoring data is many times higher than the standard values.

The use of the methodology is illustrated by the example of field data on the content of OCP in the soil in the vicinity of the liquidated warehouse in s. Dobropolie Valkovsky District of Kharkiv Region (Table 1).

Sample 1 - 4 and 6 were used to identify model parameters and evaluate its adequacy. Sample 5 was used to verify the model.

For all three indicators, dependences of the form (1) were obtained. An assessment of the adequacy by Fisher's criteria and determination coefficient confirmed the adequacy of the model. The values of the determination coefficient for a-HCH, b-HCH and g-HCH were 0,99, 0,96 and 0,9, respectively, and the values of the Fisher criterion are 88,9, 28,3 and 9,8, which exceeds the critical value of 9,3. The verification of the model showed that the predicted value calculated for control point 5 does not go beyond the boundaries of the confidence corridor. This made possible to use the model to determine the configuration of the pollution cone, in particular, to determine the diameter and depth of the pollution zone from relation (4) for each OCP. It allows us to determine the maximum boundaries of the contaminated area and the amount of land requiring bioremediation (Table 3).

The volume of land requiring bioremediation determined by the ratio:

$$V = \frac{\pi r_{s\max}^{2} h_{s\max}}{3}, \qquad (12)$$

where V – is the volume of contaminated land, m^3.

Similarly, the configuration and volume of pollution zones were determined by DDT and its metabolites and HCH isomers for some of the sites in the Kharkiv Region. The results are shown in Table 2.

Field studies using the method of accelerated, low-cost determination of the volumetric configuration of the OCP contaminated soil by a linear source were obtained on the territory of the station warehouse of their former storage in the city of Khorol, Poltava Region. (Table 3). The dimensions of this warehouse (length 30 m) make it possible to consider it a linear source of pollution. Most of the warehouse building was destroyed, all pesticides were removed, but the excess of the standard value of OCPs continues to be recorded by field observations.

Sample 5 was used to verify model (5), and the rest to identify its parameters and evaluate adequacy. The obtained adequate model was used to determine the boundaries of the pollution zone for each OCP.

The choice of composition for composting. The data of the experiment to determine the composition of the composting mixture, carried out for seven weeks, are presented in Table 4.

Based on these data we can conclude that the best biostimulator of autochthonous natural microflora for the research is 45% humus in an OCP contaminated mixture.

Thus, a method for determining the volumetric configuration of OCP contaminated soil based on the development of a mathematical model for the case of point and linear surface sources of pollution is proposed and tested in practice. Its use allows to reduce the cost and technically facilitate the process in comparison with the known methods (Ukrainian Research Institute for Environmental Issues, 2013), which provide for sampling at depths up to 6 m.

Table 1. Data from field studies of OCP (village of Dobropolye, Valkovsky District, Kharkiv Region).

Sample number	The distance from the center of the warehouse, m	Depth, m	OCP concentration, mcg/kg		
			α-HCH	β-HCH	γ-HCH
1	7	0.1	285.820	4188.594	142.334
2	7	0.55	216.593	1805.032	103.590
3	15	0.1	-	1383.032	135.505
4	15	0.55	77.873	-	-
5	17	0.1	102.889	1101.135	58.411
6	17	0.55	74.143	163.330	29.013

Table 2. Pollution zones from the former warehouses of OCP (Kharkiv Region).

Site	Pollution zone	Name of OCP					
		DDT	DDD (Derivatives of DDT)	DDE (Derivatives of DDT)	HCH		
					a	b	g
Spodobovka, Shevchenko District	Radius, m	23.59	21.37	19.47	17.48	26.19	13.14
	Depth, m	1.32	2.23	1.35	0.23	0.51	0.37
	Volume, m^3	769	1077	536	74	363	67
Dobropolye, Valkovsky District	Radius, m	30.98	22.77	30.71	16.9	28.3	23.0
	Depth, m	3.77	6.72	2.08	2.9	2	0.7
	Volume, m^3	3790	3648	2054	861	1660	396
Melnikovo, Valkovsky District	Radius, m	47.30	28.28	32.62	21.81	11.12	13.59
	Depth, m	0.63	0.75	0.62	0.36	5.09	0.09
	Volume, m^3	1476	627	694	177	660	18

Table 3. The content of OCP in the soil in the vicinity of the warehouse of their former storage (Khorol City, Poltava Region).

Sample number	The distance from the center of the pollution line, m			The content in the soil, mcg/kg		
	parallel to the source line	perpendicular to the source line	depth	α – HCH	γ- HCH	DDE
1	7.5	5	0.2	1282316.7	446169.8	194140.0
2	0	15	0.2	1838381.1	112432.1	65229.4
3	5	17	0.2	93878.4	3366.9	7031.9
4	0	7.5	0.2	1387507.7	272928.1	522536.1
5	0	21	0.2	24338.7	23921.2	40059.4
6	12	61	0.2	317.9	18.3	152.0
6	12	61	0.55	80.3	7.9	22.3
7	19	52	0.2	5456.8	2364.7	766.2

Table 4. The efficiency of using various types of biological mass in the composting mixture for bioremediation of contaminated OCP land.

The composition of the mixtures, %				OCP mass reduction, %
OCP contaminated soil	Peat	Humus	Straw	
25	1	37	37	91,0
35	0	65	0	56,9
45	0	45	10	96,1
50	0	20	30	96,3
55	0	45	0	98,3
60	0	35	5	93,0
60	0	40	0	98,0
60	0	40	0	98,0

The best available, inexpensive carrier of microflora that feeds on OCP: peat, humus and a straw carrier of natural microflora are defined as 45% humus in an OCP contaminated mixture. This exceeds the cost and technical performance of known compost methods (Chabanyuk et al., 2016.). The developed ex-situ method with isolated bioremediation of contaminated OCP land above their maximum permissible concentrations differs from the existing ones by significantly lower material and time costs and accelerated return of formerly contaminated lands to economic circulation.

Conclusions

A method has been developed for the accelerated, low-cost determination of the volumetric configuration of the land contaminated with OCPs over their normative value, by point and linear sources of pollution. A technique for determining the composition of the compost mixture is proposed. A method of remediation of lands contaminated with OCPs is presented, which is distinguished by its simplicity and quick return to the economic circulation of contaminated lands.

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