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MIGRATION OF STABLE ORGANIC SOIL CONTAMINANTS
IN A LINK OF TROPHIC CHAINS

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Abstract: *The materials, concerning the relevant problem of agricultural areas where the storages with toxic chemicals are situated, were presented. The results of the eco-toxicological condition of pesticide load on arable lands were given. The results of the phytocoenosis evaluation (belonging to various agro-biological groups), which was formed within edaphotope of the toxic chemical storages in the conditions of poly-component soil contamination with the residues of chlorine-organic pesticides, were presented.*

Key words: *unsuitable pesticides, migration, contamination, toxic load, chlorine-organic pesticides, soil.*

INTRODUCTION

The storages with toxic chemicals and adjacent areas are a large source of stable organic contaminants to enter the environment, which were included to Stockholm convention "On stable organic contaminants", the party of which is Ukraine. Despite the fact that DDT and HCCH (hexachlorocyclohexane) are forbidden in our country, remaining amounts of the mentioned pesticides are still recorded in the environment due to their high stability and ability to accumulate in natural objects.

When residues of pesticides together with foodstuffs come to a human organism and together with fodder to an animal organism continuously, toxic substances accumulate there and have a negative effect on various functional systems of organisms causing disorders in their work.

Cleaning from harmful substances is done through metabolic transformation of toxicants into more labile compounds in tissues, capable of being more easily removed from an organism. The removal process of harmful substances, including pesticide metabolites, from endotherms with biological liquids and fecal masses is time-consuming, and its intensity, to a great extent, is connected with a degree of toxic load of xenobiotics on man and animal [1–3]. The indicator of a degree and quality of toxic load of pesticides on man and domestic animals, who/which live in a certain agricultural region, can be the content levels of these pesticides in milk of man and endothermic animals. However, cattle milk, and first of all cow milk, can be considered to be the most suitable and available indicator of pesticide effect on organisms [4].

The great advantage of this toxic load indicator is the unlimited feasibility to sample biomaterial and deficit-free scope, required for conducting sanitary-hygienic studies, as cattle herds are almost in every household of different agricultural regions.

ANALYSIS OF THE LATEST RESEARCH AND PUBLICATIONS

Scientific principles of the evaluation of a negative effect of old pesticides, which belong to SOC (stable organic contaminants), for man and environment, and the remediation measures of the soils contaminated with them, were worked out in the previous works of domestic and foreign scientists: M.H. Prodanchuk, M.A. Klisenko, S.D. Melnychuk, L.I. Mokliachuk, T.L. Makarchuk, V.F. Demchenko, J. White, B. Zib, A. Nurzhanova in the years of 1995-2015.

MATERIALS AND METHODS

Soil sampling was done with the method of a square grid by the methodology [1], the way of a single envelope in four directions (south, north, east and west) at distance 1, 5, 15, 25 and 50 m from the inactive storage of mineral fertilizers and toxic chemicals.

The chlorine-organic content of pesticides was measured on chromatography "Crystal 2000" with an electron absorbing detector and a glass 1-m column, diameter 3 m, filled with a carrier - chromatone N-AW DMS (0.16–0.20mm), with fixed phases: SE-30 (5 %), gas-carrier nitrogen, brand "Osch".

A specific analysis of phytocoenosis composition was made with help of a square frame with area 0.25 м^2 (50x50 cm), in triple replication diagonally from each of 16 experimental plots (3 record plots for each of them) [1, 5, 6]. An atlas-determinant was used for plant identification [7, 8]. The research was carried out in the laboratory of quality and safety of agricultural produce of the National University of bio-resources and nature use of Ukraine. The content of DDT (dichlorodiphenyltrichloroethane) and its derivatives was determined with gas-liquid chromatography on the example of Crystal Luxe 4000, using capillary column Zebron ZB-1 (the U.S.A.). The technique, described in reference books, was used to prepare tests and analysis samples [4].

The content of mass lipid part was determined with an acidic method (ГОСТ 5867-90), that of mass protein part - refractometric method (ГОСТ 25179-90) with help of a protein analyzer in milk АБМ-1 [9, 10]. The content determination of chlorine-organic pesticides was done on chromatography "Crystal 2000" with an electron absorbing detector and a glass 1-m column, diameter 3 m, filled with a carrier - chromatone N-AW DMS (0.16–0.20mm), with fixed phases: SE-30 (5 %), gas-carrier nitrogen, brand "Osch ".

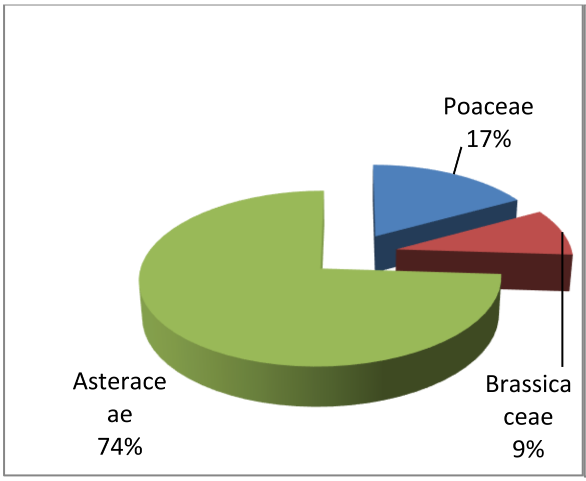
Phytocoenosis composition near chemical storages was made using a square frame with area 0.25 м^2 (50x50 cm), in fourfold replication [1, 6]. Botanical diversity of plants was studied at distance 0–1, 5–10, 15–25 and 25–50 m from the territory of chemical storages. An atlas-determinant was used for plant identification [3, 5, 8].

RESULTS AND DISCUSSIONS

To study the migration processes of stable chlorine-organic compounds in a soil-plant system, samples of wild plants were taken, those which were mostly suitable for phyto-extraction of ChOP from contaminated soil and agricultural crops, grown on arable lands, situated within a sanitary-protective zone of the toxic chemical storage in Torchytsia village, Stavyshche district, Kyiv region, where the highest contamination at level 304 MAC was recorded earlier.

The researchers of the Institute of agro-ecology and nature use have classified a group of wild plants which are the best for phyto-stabilization and phyto-degradation of xenobiotics in the conditions of soil phyto-toxicity and polycomponent pesticide contamination, namely: taraxacum officinale – *Taraxacumofficinale*Wigg. (Asteraceae); common xanthium – *Xanthiumstrumarium*L. (Asteraceae); bitter sagebrush – *Artemisiaabsinthium*L. (Asteraceae); common sagebrush – *Artemisiavulgaris*L. (Asteraceae); Canadian erigeron – *Erigeroncanadensis*L. (Asteraceae); common yarrow – *Achilleamillefolium*L. (Asteraceae); couch grass – *Elytrigiarepens*L. (Poaceae); wild carrot – *Daucuscarota*L. (Apiaceae); biennial oenothera – *Oenotherabiennis*L. (Onagraceae); calamagrostis – *Calamagrostisepigeioes*(L.) Roth. (Poaceae) [11–14]. Most of the mentioned species belong to Asteraceae or Poaceae families, they are capable of vegetative propagation or they form a great number of seeds, have an extended life cycle. Most of them are explerents. Characteristic substances, common for the mentioned species (organic acids, phenol compounds, ether oils, flavonoids, tannic substances), can be involved in the resistance mechanisms to high pesticide load [15].

A botanical structure by families was analyzed within the experimental plots. The results of the research are given in Fig. 1–4. At a 1-m distance vegetation was presented with only three Asteraceae families – 74% diversity, Poaceae –17 % and Brassicaceae – 9%.



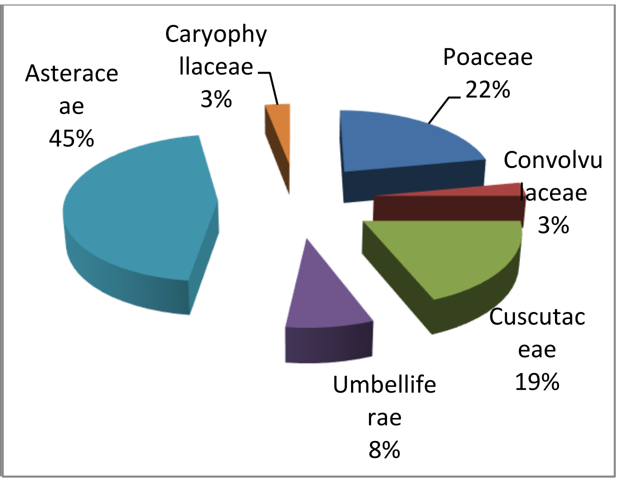


Fig 1: Botanical structure of vegetation at 0–1 м distance of the composition

Fig 2: Botanical structure of vegetation at 5–10 м distance of the composition

At a 5-10 m distance from the storages with toxic chemicals Coryophyllaceae and Convolvulaceae were identified in a range of 3% of the total number. Totally, 6 families were recorded at this distance. The dominant ones were Asteracea and Poaceae. Their share was 67% of the total number.

Within a 15-25 m distance, botanical diversity increased up to 10 families. Most of the plants belonged to Asteracea and Poaceae. And only 2% belonged to Plantaginaceae.

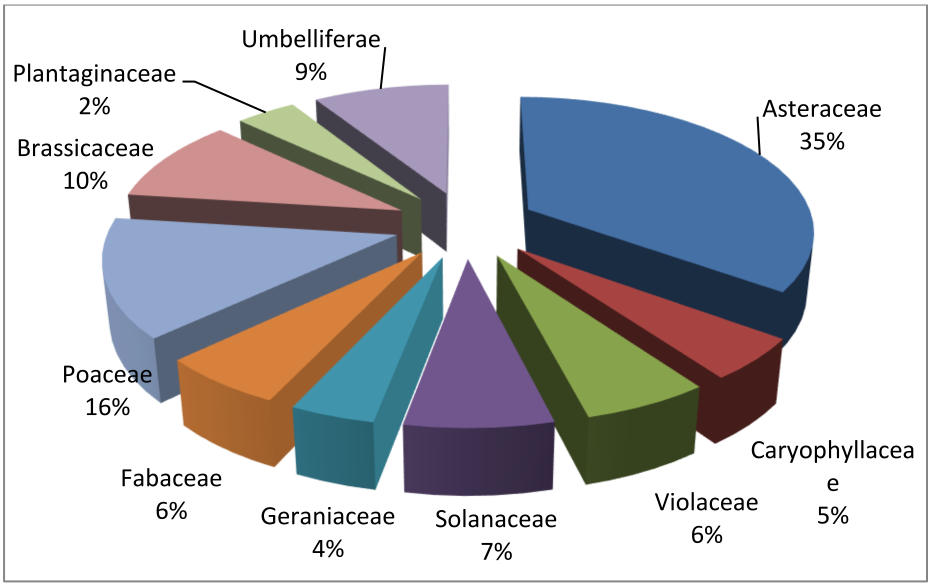


Fig 3: Botanical structure of vegetation at 15–25 m distance from the storage.

At 25–50 m 16 families were identified. Asteracea amounted for 34 % of the total quantity of plants. Poaceae took the second place and their number was 12 %. Polygonaceae, Brassicaceae, Fabaceae and Umbelliferae ranged within 6 %.

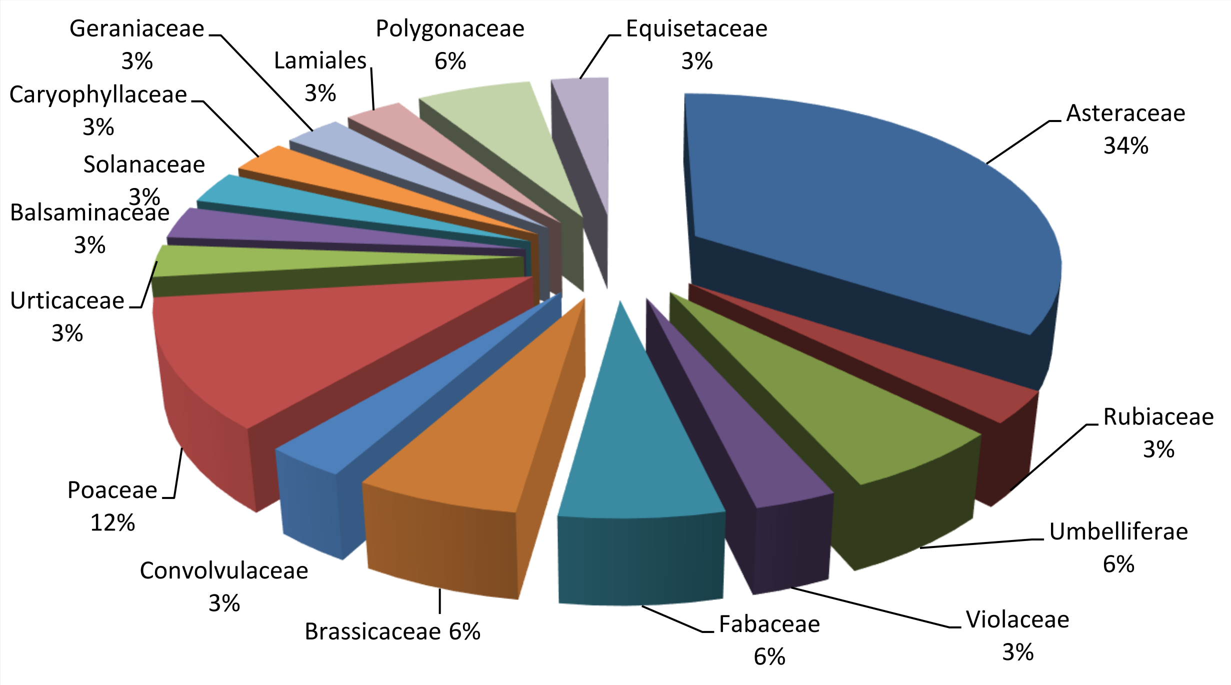


Fig 4: Botanical structure of vegetation at 25–50 m distance from the storage.

Equisetaceae, Lamiales, Geraniaceae, Caryphyllaceae, Solanaceae, Balsaminaceae, Urticaeae, Convolvalaceae and Rubiaceae did not exceed 3%.

To study the process of ChOP (chlorine-organic pesticide) accumulation by wild plants from sanitary-protective zones of the toxic chemical (Stavyshche district, Torchytsia village) at a 5-km distance from the storage, wild plants, tolerant to available pesticides in the soil and capable of bio-accumulation of chlorine-organic pesticides, were sampled. Residual amount of ChOP in plant tissues and a rhizosphere soil was determined (Table 1).

Table 1: Content of chlorine-organic pesticides in plants

Type of plant	Content of total isomers and metabolites DDT, mkg/kg		
	in rhizosphere soil	in plants	Accumulation coefficient, %
Artemisiaabsinthium	3562,6± 231,2	534,1±17,1	14,9
Taraxacumofficinale	2763,0± 25,4	324,7±19,8	11,7
Achilleamillefolium	322,5±1,4	211,7±3,5	6,5
Artemisiavulgaris	1834,9± 145,2	187,5±6,7	10,2
Elymusrepens	1567,0± 22,7	97,3±1,5	6,2
Calamagrostisepigejos	935,4±2,6	83,2±0,7	8,8
Poapratensis	756,6± 5,2	76,3±4,6	10,0

It has been established that chlorine-organic pesticides are most intensively accumulated by the representatives of Asteraceae: Artemisia absinthium, Taraxacum officinale, Achilleamille folium; Poaceae crops show lower accumulation intensity.

It is to be stated that most of the presented species are perennials, which can grow in the conditions of soil phyto-toxicity and accumulate DDT and its metabolites in root tissues in large amounts [16, 17]. This confirms the feasibility to use these species as phyto-stabilizers of SOC in the soil to prevent migration of toxicants into adjacent areas.

Thus, it has been found out that as the distance from the storages increases, plant diversity increases as well as the number of classes. Besides it has to be mentioned that Asteraceae and Poaceae are the most resistant to toxic chemicals, including chlorine-organic compounds.

A serious violation of sanitary requirements is a disregard of the boundaries of the protective zones around the toxic chemical storages. Quite frequently sanitary-protective zones are included in general agricultural land use without previous examination which is a threat of the contamination of agricultural produce with stable toxicants, including ChGP.

Taking into consideration a significant soil contamination of the studied areas with residual pesticides, their possibility to migrate and to accumulate toxicants in a chain “soil-plant”, the content of residual pesticides in the samples of agricultural crops, grown in the soils, contaminated with stable organic contaminants, was defined.

The research was carried out in the household of “Interagroinvest Ltd.”, a branch of Torchytsia, Sravyshche district, where previous trials showed soil contamination at level 304 MAC. These objects were studied: alfalfa, winter wheat, sugar beets, corn and grain, spring barley. Plants were taken at a 15-25 m distance from the contamination source. The research results are given in Table 2.

Table 2: Content of residual amount of DDT in agricultural crops, mg/kg

Place	Years of research	Crop	Pesticide	Content, mg/kg	МДР, мг/кг	Control, mg/kg (over 300 m from the storage)
Torchytsia village	2011	Medicagosativa	4,4′-DDE	0,034	0,05	0,0062
			4,4′-DDD	0,022		not found
			4,4′- DDT	0,028		not found
			Σ DDT	0,084		0,0062
	2012	Winterwheat (grain)	4,4′- DDE	0,0039	0,02	not found
			4,4′- DDD	н.в.		not found
			4,4′- DDT	н.в.		not found
			Σ DDT	0,0039		not found
	2013	Sugarbeets (rootcrops)	4,4′- DDE	0,15	0,1	0,048
			4,4′- DDD	0,10		not found
			4,4′- DDT	0,12		not found
			Σ DDT	0,37		0,048
	2014	Corn (grain)	4,4′- DDE	0,0058	0,02	not found
			4,4′- DDD	н.в.		not found
			4,4′- DDT	н.в.		not found
			Σ DDT	0,0058		not found
	2015	Springbarley (grain)	4,4′- DDE	0,0031	0,02	not found
			4,4′- DDD	н.в.		not found
			4,4′- DDT	н.в.		not found
			Σ DDT	0,0031		not found

The content of total isomers and DDT metabolites in the biomass of alfalfa and sugar beet roots, grown on arable lands which are adjacent to the inactive toxic chemical storages, exceeds maximum admissible concentrations (MAC) by 1.7 and 3.7 times, respectively. Small concentrations (0.0031–0.0058 mg/kg) were found in grain of winter wheat, corn and spring barley, due to poor ability to accumulate chlorine-organic pesticides by grain crops.

Thus, having analyzed the received research results, one can state that inactive toxic chemical storages are a powerful source of the environmental pollution, in particular the agricultural soils, agricultural crops which are grown in immediate proximity of the storages. A serious threat to man's health occurs when xenobiotics enter trophic chains.

An important component in human nutrition is good-quality cow milk and the products made of it. Which is why, the amount of DDT and its derivatives in the concentrations which exceed maximum admissible ones, can be one of the worsening factors of human health [18].

It is a well-known fact that ChOP are extremely stable in the environmental objects and are not damaged under the effect of any other factors of the environment, in particular humidity and temperature. Chlorine-organic pesticides have a high cumulative ability in fatty tissues of man and animals. DDT pesticide is known to decompose when metabolites of DDE (dichlordifenildichlorethylene) and DDD (dichlordifenildichlorethane) are formed, which are more toxic and stable in the environmental objects [19–21].

When animals consume contaminated fodder which contain residual chlorine-organic pesticides, the accumulation of toxicants take place in fatty tissues. Under continuous consumption of contaminated products, gradual accumulation of toxicants in a human organism takes place which leads to a negative effect on various functional systems and causes problems in their work [21]. Having entered cow organism through fodder, DDT and its derivatives are removed for a long time, including with milk.

A serious violation of sanitary standards is cattle pasturing near the sources of negative anthropogenic pressure, which are toxic chemical storages. During the time when the research was done, frequent facts of cattle pasturing by the residents of Torchytsia village near the half-ruined storage of toxic chemicals were recorded. To find out possible cow milk contamination with residual chlorine-organic pesticides, samples were taken from 5 households of Torchytsia village in 2014 in summer (July) and winter (December) periods. Taking into account the fact that chlorine-organic pesticides are more intensively accumulated in fatty tissues and fatty milk balls, we did research to determine mass share of fat and protein in milk of the cows, which were kept in Torchytsia village, Stavyshche district. The previous research confirmed significant levels of soil contamination of the sanitary-protective zone near the toxic chemical storage, the contamination of wild plants and agricultural crops with residual chlorine-organic pesticides.

It was established that in all milk samples the lowest mass fat share was in cow milk in a summer period (July). The indicators of fat content ranged within 3.7–5.2%.

In a winter period (December) mass fat share in milk increased compared with a summer period and it was 4.8–6.0 %. It can be explained by the decrease of milk yields in winter and the ration change (substitution of green mass for hay, haylage and silage) (рис. 5).

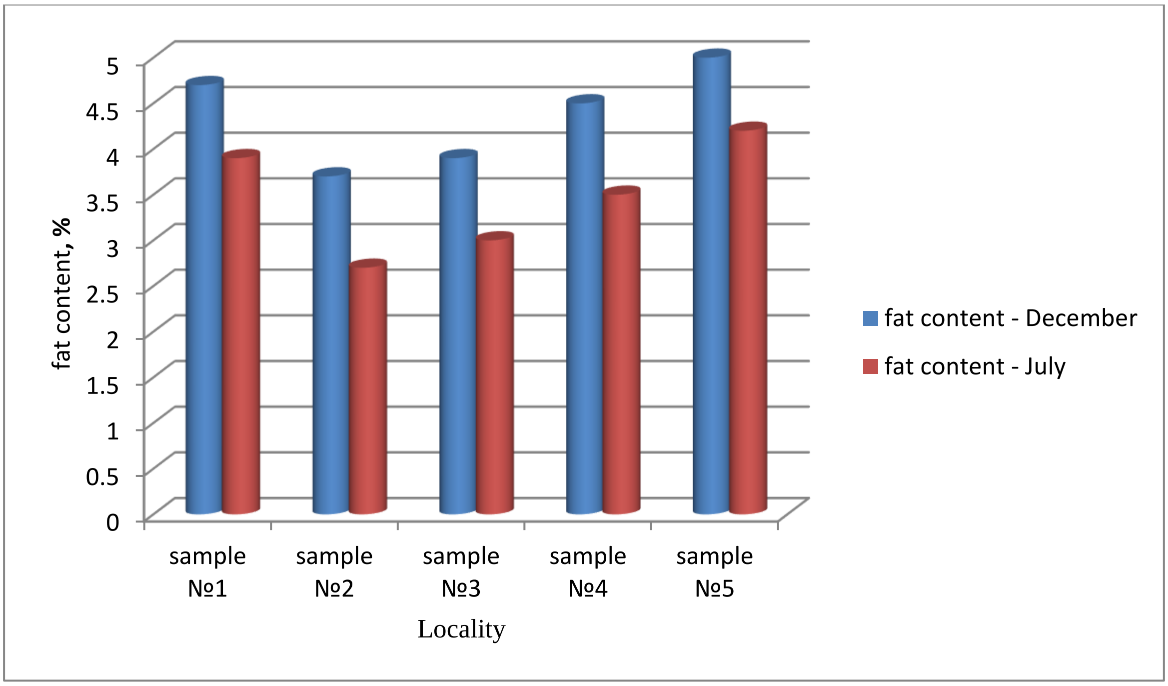


Fig 5: Fat content in cow milk, Torchytsia village, Stavyshche district

The content of total isomers and DDT metabolites in milk samples was defined. The results of the analytical findings are given in Tables 3 and 4. A direct correlation between fat content in milk and its contamination with residual chlorine-organic pesticides was found out.

Table 3: Content of DDT and its derivatives in cow milk,

Torchytsia village, Stavyshche district (July)

Households Sample, №	Content				∑ DDT, mg/kg
	fat, %	DDE, mg/kg	DDT, mg/kg	DDD, mg/kg	
1	3.9	0.030	0.021	0.015	0.066±0.007
2	2.7	0.021	0.017	0.013	0.051±0.004
3	3.0	0.025	0.018	0.014	0.057±0.005
4	3.5	0.028	0.020	0.015	0.063±0.006
5	4.2	0.031	0.024	0.020	0.075±0.005

MAL 0.05 mg/kg – adults, 0.01 mg/kg – children

The highest value of total isomers and DDT metabolites (0.075 mg/kg), which exceeded MAL (maximum admissible level) by 1.5 times for adults and 7.5 times for children, was found in a milk sample from household № 5 (fat content 4.2 %). The lowest content of total isomers and DDT metabolites (0.051 mg/kg), which corresponded a MAL level for adults, but exceeded a fixed rate for children by 5 times, was found in sample № 2 with fat content 2.7 %.

Milk samples, taken in a winter period, regardless of the fat content increase, contained residual chlorine-organic pesticides – 0.0024–0.0063 mg/kg, which did not exceed established standards (Table 4).

Table 4: Content of DDT and its derivatives in cow milk, Torchytsia village, Stavyshche district (December)

Households Sample, №	Content				∑ DDT. mg/kg
	fat, %	DDE, mg/kg	DDT, mg/kg	DDD, mg/kg	
1	4.7	0.0020	0.0016	0.0012	0.0048±0.0004
2	3.7	0.0011	0.0009	0.0004	0.0024±0.0003
3	3.9	0.0024	0.0016	0.0012	0.0052±0.0006
4	4.5	0.0014	0.0010	0.0007	0.0031±0.0003
5	5.0	0.0029	0.0018	0.0016	0.0063±0.0007

MAL 0.05 gm/kg – adults, 0.01 mg/kg – children

This is explained by stable conditions of keeping animals and feeding them with the fodder which do not contain ChOP. However, residual isomers and DDT metabolites were found in all the milk samples, which can be explained by slow removal of toxicants with fat milk balls, accumulated in cow organisms in a summer period. A direct correlation between fat content and content of chlorine-organic pesticides was found in winter milk samples.

CONCLUSIONS

1. A serious violation of sanitary requirements is a disregard of the boundaries of the protective zones around the toxic chemical storages. Quite frequently sanitary-protective zones are included in general agricultural land use without previous examination which is a threat of the contamination of agricultural produce with stable toxicants, including ChOP. Taking into consideration a significant soil contamination of the studied areas with residual pesticides, their possibility to migrate and to accumulate toxicants in a chain "soil-plant", the content of residual pesticides in the samples of agricultural crops, grown in the soils, contaminated with stable organic contaminants, was defined.

2. So, the analysis of the received research results prove that inactive toxic chemical storages are a powerful source of the environmental pollution, in particular the agricultural soils, agricultural crops which are grown in immediate proximity of the storages. A serious threat to man's health occurs when xenobiotics enter trophic chains.

3. Agro-ecological estimation of the contamination of the soil and trophic chain components with stable organic contaminants was scientifically grounded and carried out based on the research done.

4. It has been established that sanitary zones of the toxic chemical storages in the territory of Kyiv region are dangerous sources of ChOP which enter trophic chains; scientific-methodological approaches to the determination of the area of heavily contaminated land were explained. The total area of the land contaminated with unacceptable pesticides in the territory of Kyiv region was identified, it is 2298.5 ha.

5. Based on the quantitative evaluation of the territories contaminated with unacceptable pesticides in Skvyra and Stavyshche districts of Kyiv region, it was found out that the zones of critical contamination occupied not less than 12.6 ha near each of the storage.

6. The estimation of ChOP spread from the source of contamination to arable lands was made; the example was the toxic chemical storage, highly contaminated with pesticides in Torchytsia village, Stavyshche district. DDT was recorded in agricultural soils in the concentrations which very much exceeded hygienic standards by 304 times.

7. Wild plants proved to be able to extract DDT from the contaminated soil. The coefficients of the accumulation of non-acceptable pesticides and their metabolites in the system "soil-wild plant" were defined. Chlorine-organic pesticides are most intensively accumulated by the representatives of Asteraceae: Artemisia absinthium, Taraxacum officinale (534.1 and 324.7 mkg/kg).

8. It has been established that anaerobic decomposition is an effective way to utilize plants which contain DDT in their tissues. The utilization processes of wild plants, contaminated with DDT, were studied. It has been shown that one of the ways of the contaminated mass utilization is the technology which makes it possible to get high efficient organic fertilizers and energy (biogas).

9. The DDT contamination degree of cultivated crops, grown in the contaminated soil, was determined. The content of total isomers and DDT metabolites in the biomass of alfalfa and sugar beet roots, grown on arable lands which are adjacent to the inactive toxic chemical storages, exceeds maximum admissible concentrations (MAC) by 1.7 and 3.7 times respectively. Small concentrations (0.0031–0.0058 mg/kg) were found in grain of winter wheat, corn and spring barley, due to poor ability to accumulate chlorine-organic pesticides by grain crops.

10. Milk samples, taken in a winter period, contained residual chlorine-organic pesticides – 0.0024–0.0063 mg/kg, which did not exceed established standards. However, milk samples, taken in a summer period, contained residual DDT: the highest value exceeded MAL by 1.5 times for adults and by 7.5 times for children; the lowest value (0.051 mg/kg) corresponded to MAL for adults, but it exceeded the established standards for children by 5 times.

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