

Organochlorine insecticide residues in the soils of Romania

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Introduction

Chlorinated hydrocarbon insecticides were used for many years to control agricultural and public health insects before their application was banned in many countries. These insecticides are only very slowly degrading in soils. They may reach fresh water and concentrate in the bodies of some terrestrial and aquatic invertebrates and plants that provide food for vertebrates [1].

The use of such chemicals (products based on DDT and technical mixtures of HCH) was interdicted in Romania since 1985 [2], but due to their high persistence, residues of these substances continued to be found in soil and other environmental compartments.

Since 1992, the Integrated Soil Monitoring System in Romania has been implemented and organised in two subsystems (agricultural soils and forest soils) at three detailed levels [3]. This is the first unitary system for monitoring the soil quality at the national level in our country and it allows establishing certain intervals of different parameter values, including loading with potentially polluting elements and substances.

The present paper summarises the results obtained concerning the contamination of the topsoil with organochlorine insecticide residues (HCH and DDT) in the plots of the 1st level monitoring (16x16-km grid).

Materials and methods

The 1st level monitoring plots were located within a 16x16-km grid, (942 sample plots, of which 670 were on agricultural land, and 272 were in forest). This survey practically covered all the agricultural and forestry use, in a proportion almost similar with that of corresponding lands in the territory. Within each grid plot, a complete characterisation of soils and lands was fulfilled. Soil samples were collected from each soil profile according to the morphogenetical criteria and analysed from the physical and chemical viewpoint. The load with organochlorine insecticide residues was determined only in the samples collected from the first 10-25 cm of soil surface. A total of 912 points were considered, 669 in agricultural land and 243 in forest.

The collected soil samples for the determination of organochlorine insecticide residues were air-dried, ground and 2 mm sieved. The residues were extracted by heating the samples under reflux with a 2:1 mixture of petroleum ether-acetone. After acetone removal, the petroleum ether extracts were dried with anhydrous sodium sulphate and reduced under vacuum to 2-4 ml. In these final extracts the following compounds were determined: alpha-HCH, beta-HCH, gamma-HCH, delta-HCH, o,p'-DDT, p,p'-DDT, o,p'-DDD, p,p'-DDD and p,p'-DDE. If necessary, a further clean-up step on Florisil columns was employed. The method used for determination was gas-liquid chromatography. The separation of compounds was made by using a glass column, packed with Chromosorb WHP 100-120 Mesh coated with 2.5% QF-1 + 1% OV-11 + 0.5% XE-60 stationary phase. For the detection of analytes, an electron capture detector with ⁶³Ni source was used. Blank tests were performed to check for the presence of any possible contamination during the analytical procedure (solvents, glassware, etc.).

Results and discussions

Table 1 presents ranges and average levels of HCH and DDT in soil. It could be observed that relatively small differences in HCH concentration exist between agricultural and forestry uses, while a marked difference between total DDT contents corresponding to the mentioned uses was evidenced. From the total HCH content, the gamma and beta isomers represent the largest fraction. In the case of total DDT content, the most represented were p,p'-DDE and p,p'-DDT, followed by o,p'-DDT.

The intervention values for HCH and DDT concentrations in soil considered within the Environmental Quality Objectives in The Netherlands [4] are 2 mg/kg and 4 mg/kg respectively. These values refer to a standard soil, having 10% organic matter, the most important component of the soil in retaining the organic contaminants. A comparison was made between the concentrations measured within our monitoring system and these values. Thus, in all monitoring plots, both the HCH content and the DDT content were much below the intervention values mentioned, the highest detected concentrations were 0.715 mg/kg for HCH and 2.458 mg/kg for DDT (Table 1).

The reference values for assessing the soil pollution with organochlorine insecticide residues provided by the environmental legislation in Romania [5] are presented in Table 2. It can be observed that the established intervention values for sensitive uses are lower than those from The Netherlands, which is normal, taking into account that all our soils have less than 10% organic matter content. By comparing our data with these values, it was concluded that 0.33% and 0.98% of samples had HCH and DDT contents respectively above the corresponding intervention values, while the alert values were exceeded in 1.1% and 2.2% respectively of studied points (Table 3).

Taking into account the interdiction applied in Romania to use this kind of pesticides in agriculture and forestry as well as the partial data that we have from the second phase of the monitoring system we could draw the conclusion that there is a tendency of decreasing both HCH and DDT contents in soil. The levels of HCH and DDT found in soil, as compared with values cited in the literature [4] do not represent a major environmental risk and the few points where the intervention values given by Romanian legislation were exceeded will be further monitored. A closer mapping around these points will also be carried out.

We haven't established yet links with water quality monitoring programmes but we intend to do it in the nearest future, starting with areas including points with organochlorine levels in topsoil above alert values (10 points for HCH and 20 points for DDT).

Unfortunately, there are no available technologies in our hands in order to reduce the organochlorine residues content in soil below the intervention values established by the Romanian legislation. That is why no decontamination measures have been taken yet.

Levels of the total HCH and DDT contents in the topsoil of 1st level (16x16-km grid) monitoring plots in Romania are presented in Figure 1 and Figure 2. In general, higher concentrations, exceeding the alert values, were found especially in the southeastern part of the country, where these pesticides were intensively used.

Table 1. Content of HCH and DDT in the top soil of the monitoring plots, level I (16x16 km) - Romania

Compound	Agricultural land			Forest land			Total		
	Plots	Range	Average	Plots	Range	Average	Plots	Range	Average
	mg/kg			mg/kg			mg/kg		
alpha-HCH	669	0.001 - 0.033	0.004	243	0.001 - 0.099	0.007	912	0.001 - 0.099	0.004
gamma-HCH	669	0.001 - 0.209	0.017	243	0.001 - 0.085	0.014	912	0.001 - 0.209	0.016
beta-HCH	669	0.001 - 0.682	0.012	243	0.001 - 0.250	0.008	912	0.001 - 0.682	0.011
delta-HCH	669	0.001 - 0.128	0.003	243	0.001 - 0.062	0.002	912	0.001 - 0.128	0.003
Total HCH	669	0.001 - 0.715	0.036	243	0.001 - 0.434	0.030	912	0.001 - 0.715	0.034
pp'DDE	669	0.001 - 0.854	0.031	243	0.001 - 0.052	0.005	912	0.001 - 0.854	0.024
op'DDD + pp'DDD	669	0.001 - 0.192	0.007	243	0.001 - 0.030	0.001	912	0.001 - 0.192	0.006
op'DDT	669	0.001 - 0.406	0.023	243	0.001 - 0.044	0.009	912	0.001 - 0.406	0.019
pp'DDT	669	0.001 - 1.842	0.046	243	0.001 - 0.197	0.018	912	0.001 - 1.842	0.039
Total DDT	669	0.001 - 2.458	0.105	243	0.001 - 0.264	0.032	912	0.001 - 2.458	0.085

Table 2. Reference values used in Romania for organochlorine insecticide residues in soils (mg/kg dm)

Substance	Normal values	Alert values/Type of use		Intervention values/Type of use	
		Sensitive*	Less sensitive**	Sensitive*	Less sensitive**
DDT (total)	< 0.15	0.5	1.5	1	4
DDT	< 0.05	0.25	0.75	0.5	2
DDE	< 0.05	0.25	0.75	0.5	2
DDD	< 0.05	0.25	0.75	0.5	2
HCH (total)	< 0.005	0.25	0.75	0.5	2
alpha-HCH	< 0.002	0.1	0.3	0.2	0.8
beta-HCH	< 0.001	0.05	0.15	0.1	0.4
gamma-HCH	< 0.001	0.02	0.05	0.05	0.2
delta-HCH	< 0.001	0.05	0.15	0.1	0.4
Organochlorine pesticides (total)	< 0.2	1	2	2	5

* = Residential/agricultural land uses, playing fields, parks, protected areas, sanitary zones under restriction regime

** = Commercial/industrial land uses

Table 3. Distribution of soil samples within HCH and DDT concentration intervals

HCH			DDT		
Interval of concentration (mg/kg)	Number of samples	%	Interval of concentration (mg/kg)	Number of samples	%
< 0.011	146	16.01	< 0.011	85	9.32
0.011 - 0.25*	756	82.89	0.011 - 0.5*	807	88.49
0.25* - 0.5**	7	0.77	0.5* - 1**	11	1.21
> 0.5**	3	0.33	> 1**	9	0.98
	912	100		912	100

* Alert value

** Intervention value

References

- Alexander, M., Biodegradation and Bioremediation, Academic Press, 1994, p 272-275.
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- Romanian Ministry of Water Management, Forestry and Environmental Protection, Order for Approving the Regulation for Assessing the Environmental Pollution, No 756/1997.

SOIL MONITORING SYSTEM IN ROMANIA - Level I (16 x 16 km grid)
Scale 1:3,500,000

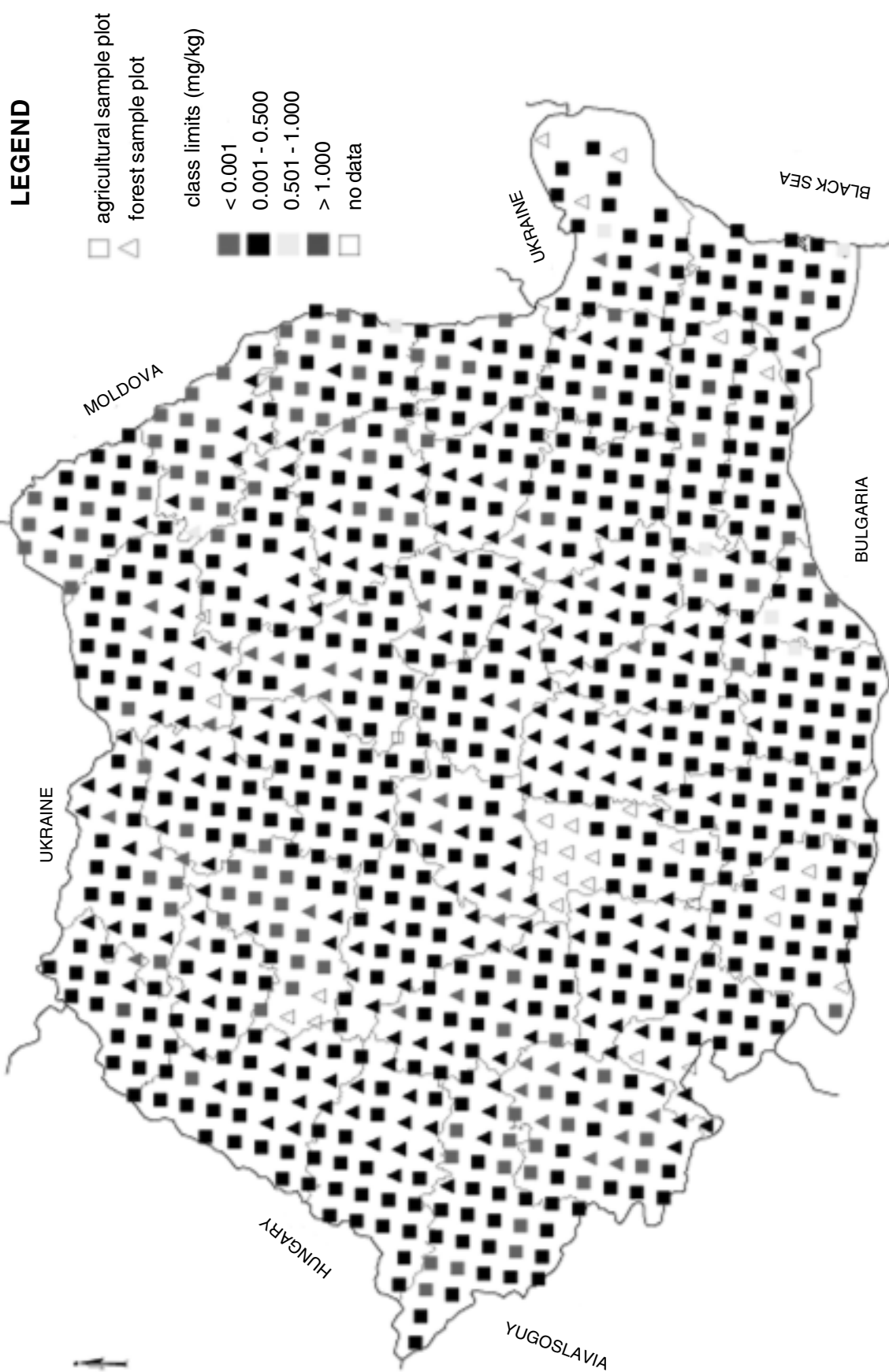


Figure 1. Levels of total HCH contents in the topsoil of 1st level (16x16 km-grid)
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SOIL MONITORING SYSTEM IN ROMANIA - Level I (16 x 16 km grid)
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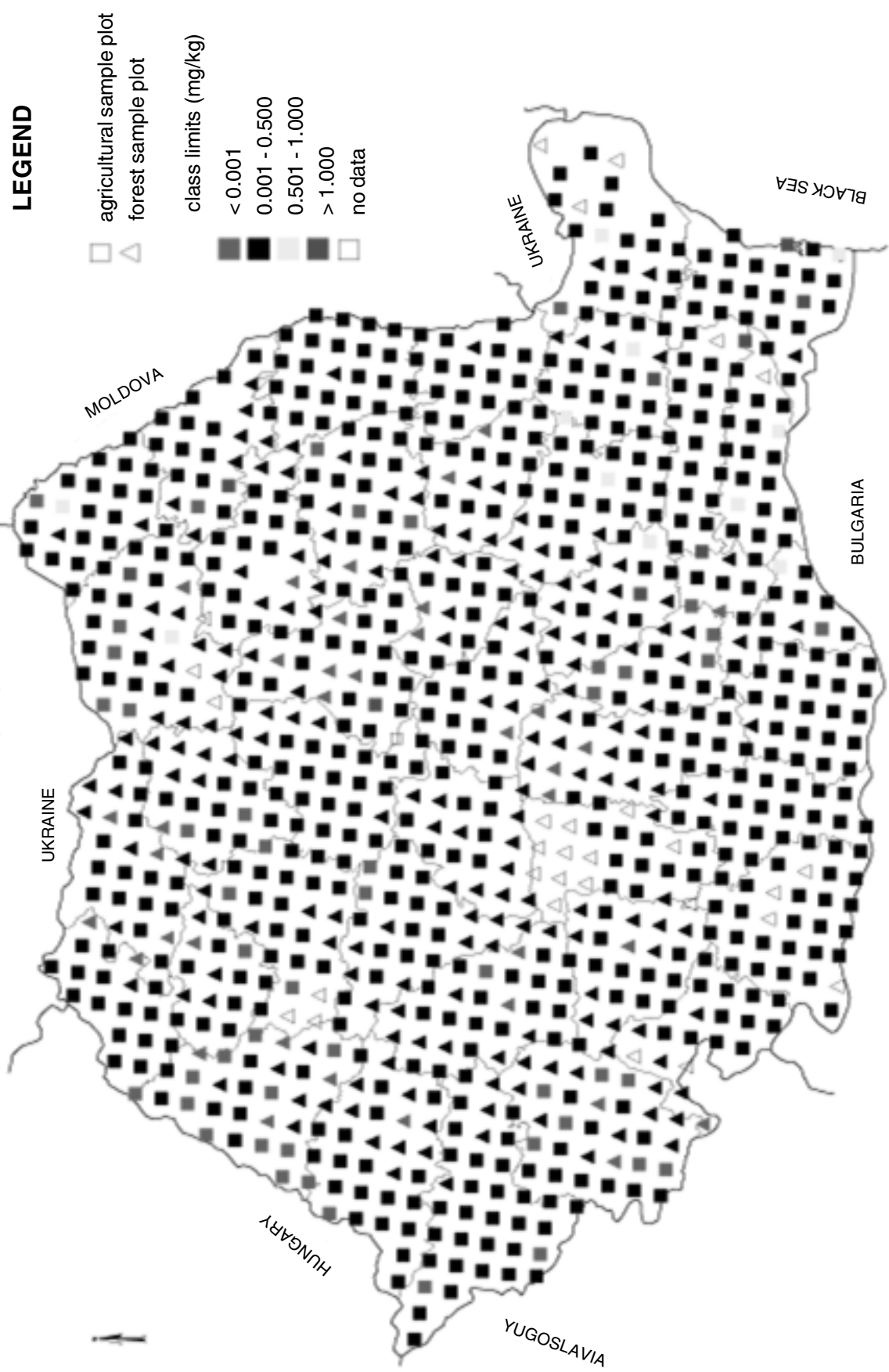


Figure 2. Levels of total DDT contents in the topsoil of 1st level (16x16 km-grid)
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