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## Occurrence and distribution of the cyclodiene-type organochlorine pesticides in soils of Vojvodina Province, Serbia

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**Abstract:** Aldrin, endrin and dieldrin are highly toxic and persistent cyclodiene-type organochlorine pesticides. Although these compounds have been banned over the last few decades in many countries, they can still be found in the environment, especially in the agricultural soil. In order to determine the residue levels of cyclodiene-type organochlorine pesticides in soils of Vojvodina Province, and to obtain detailed information on their spatial distribution, 1370 soil samples under different land uses were collected from Vojvodina Province and analyzed for the presence of aldrin, dieldrin and endrin. The residues of those pesticides were detected in the soils of Vojvodina in measurable concentrations and dieldrin was the most dominant compound in the soil. However, the concentrations of analysed cyclodiene-type organochlorine pesticides in the most soil samples from Vojvodina province were lower than Dutch target values for soil quality. Soil organic matter and clay content were found to be positively associated with aldrin content while clay content was found to be negatively associated with endrin.

**Key words:** soil; aldrin; endrin; dieldrin; Vojvodina Province.

### INTRODUCTION

Dieldrin, aldrin and endrin are highly toxic cyclodiene-type organochlorine insecticides that are extremely persistent in the environment. Originally developed in the 1940s as an alternative to DDT, aldrin, dieldrin and endrin proved to be highly effective insecticides and were widely used during the 1950s to early 1970s. In spite of their high efficiency in insect control, their use have been prohibited in many countries since the 1970s due to the fact that their accumulation in crops poses a potential threat to human health.<sup>1</sup> They were prohibited in Serbia in 1991, but before that were extensively used on arable soils for insect control especially in Vojvodina since it is an agricultural region with an intensive

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agricultural production. However, these pesticides continue to be detected in different environments, especially agricultural fields where these pesticides were previously used.<sup>2-8</sup>

Despite some differences in reports concerning the half-lives of aldrin, dieldrin and endrin in soil, most studies have shown that these pesticides are highly persistent in soil. According to the results of the experiment conducted by Meijer *et al.*<sup>9</sup> the half-life of dieldrin in soil is about 25 years. Donoso *et al.*<sup>10</sup> reported that the half-life of endrin in soil ranged up to 12 years.

Also, some studies have indicated contamination of the water environment around agricultural fields by cyclodiene-type organochlorine pesticides.<sup>11-13</sup> Aldrin, dieldrin and endrin residues in agricultural fields cause contamination of not only the water environment, but also of crops grown in contaminated soil. High levels of these pesticides have been detected in a variety of crops around the world.<sup>14</sup>

In Serbia, a dieldrin residue level of 5–73 ng/g and aldrin residue level of 1–49 ng/g were reported in wheat grown in Central Bačka and Central Banat regions.<sup>15</sup> In some of the wheat samples concentration of dieldrin and aldrin was greater than the maximum permissible level of 10 ng/g according to European Commission,<sup>16</sup> which indicates possibility of high concentrations of dieldrin and aldrin in soils where the wheat was grown.

The aim of this study was to determine the residue levels of cyclodiene-type organochlorine pesticides aldrin, dieldrin and endrin in soils of the Vojvodina Province, and to obtain detailed information on their spatial distribution.

#### EXPERIMENTAL

Details about soil sampling are given in Supplementary material to this paper.

After sampling, soil was air-dried, homogenised and sieved through a 2-mm stainless sieve before analysis. The water and potential soil pH, organic matter content and free CaCO<sub>3</sub> content in soil were determined in accordance with ISO methods for soil quality.<sup>17-19</sup> Particle size distribution was determined in the < 2 mm fraction by the internationally recognized pipette method.

##### *Sample extraction and clean-up*

Ten grams of the soil sample was weighed precisely and mixed with 5 g of anhydrous sodium sulfate. The mixture was spiked with surrogate compound 2,4,5,6-tetrachloro-*m*-xylene (TCMX), and then Soxhlet-extracted for 24 h with 150 mL mixture of hexane/acetone (1:1 vol. ratio). The crude extracts were concentrated by rotary vacuum evaporation, and then solvent-exchanged by re-dissolving in 2 mL of hexane. A solid phase extraction tubes containing 1 g of silica sorbent were pre-eluted by 4mL of hexane. After introduction of 2 mL of the extract, the column was eluted with 5mL of *n*-hexane and 5mL of *n*-hexane/diethyl ether (1:1 vol. ratio). The two fractions were combined as a single fraction and then reduced to 1 mL under a gentle stream of nitrogen before analysis.

### *Analysis*

The determination of cyclodiene-type organochlorine pesticides was performed on an Agilent 7890A gas chromatograph equipped with a  $^{63}\text{Ni}$  micro-electron capture detector (GC- $\mu\text{ECD}$ ). The separation was performed on a fused silica capillary column (HP-5MS, 30 m $\times$ 0.25 mm i.d., and 0.25  $\mu\text{m}$  film thickness). Helium was used as carrier gas at a constant flow of 1.4 mL/min and high purity nitrogen was used as make-up gas (30 mL/min). The temperature program was as follows: initial temperature of 70 °C was held for 2 min, increased to 150 °C at a rate of 25 °C/min, then increased to 200 °C at a rate of 3 °C/min, and finally increased to 280 °C at rate of 8 °C/min and held for 10 min. The injector and detector temperatures were set at 300 °C. One microliter of each sample was injected in the splitless mode. Identification of aldrin, dieldrin and endrin in the samples was based on the comparison of retention time between samples and the standard solution. Quantification was performed by five-point calibration method using standard solutions in hexane (from 2.5 to 100 ng/mL,  $r^2 > 0.992$ ). To confirm the results, typical samples selected were confirmed with an Agilent 6890 GC-5975MSD system. The GC parameters were the same as described above. The MS was operated in electron impact ionization mode with electron energy of 70 eV. The ion source, quadruple and transfer line temperatures were held at 230, 150 and 280 °C, respectively. Target compounds were monitored in the selected ion monitoring (SIM) mode.

### *Statistical analysis*

The GIS (Geographic Information System) mapping technique was used to produce spatial distribution maps of the aldrin, endrin, and dieldrin concentrations. Data were plotted with the ArcView software 10.0 (ESRI) using a 16 km $\times$ 16 km output grid. Thus, one grid point represents in general one sampling point.

Descriptive statistical parameters, such as mean value, median, minimum, maximum values and relative standard deviation, were calculated to describe main physical and chemical characteristics of analyzed soils and concentrations of aldrin, endrin and dieldrin in all the samples. The correlation of analyzed pesticides with various soil parameters was analyzed by the Karl Pearson correlation coefficient. A *P*-value less than 0.05 (two tail) was considered to be statistically significant. All statistical analyses were carried out using the Statistica 12 (StatSoft Inc., Tulsa, OK, USA).

## RESULTS AND DISCUSSION

### *Physical and chemical characteristics of analyzed soils*

The main physical and chemical characteristics of analyzed soils are given in Table I. All soil properties ranged in wide intervals, indicating high heterogeneity of soils in Vojvodina Province. The pH value in KCl ranged from 3.42 to 9.27 indicating that all analyzed soils were acid to alkaline. The  $\text{CaCO}_3$  content ranged from 0.00 to 33.9 %, and organic matter content ranged from 0.31 to 9.66 %. The particle size distribution was also variable with the greatest variability in sand and silt content (0.20–50.07 % and 1.30–49.4 %, respectively).

### *Status of cyclodiene-type organochlorine pesticide residues in soil*

The mean, minimum, maximum concentrations, and detection frequencies of aldrin, endrin and dieldrin in the soils of Vojvodina Province are shown in Table

II. Dutch target values<sup>20</sup> for cyclodiene-type pesticides in soils which indicate the level of sustainable soil quality are also given in Table II.

TABLE I. Physical and chemical characteristics of analyzed soils; *SD* – standard deviation

Data	Parameter				Particle size distribution, %			
	pH (in KCl)	pH (in H <sub>2</sub> O)	CaCO <sub>3</sub> content %	Organic matter content, %	Sand (200– 2000 μm)	Fine sand (20–200 μm)	Silt (<20 μm)	Clay (<2 μm)
Minimum	3.42	4.68	0.00	0.31	0.20	14.80	5.30	1.30
Maximum	9.27	10.3	33.9	9.66	50.7	70.10	56.5	49.4
Mean	7.07	7.87	7.58	3.05	4.37	39.94	33.6	21.9
<i>SD</i>	0.79	0.67	6.89	0.95	8.29	11.48	7.77	9.75
Median	7.37	8.09	6.40	3.07	1.50	40.05	34.6	21.8
Variance	0.63	0.45	47.4	0.91	68.8	131.9	60.3	95.2
Skewness	-1.85	-1.48	0.73	0.61	3.87	0.245	-0.76	0.59
Kurtosis	3.14	3.14	-0.19	2.36	16.9	-0.17	1.69	0.47

TABLE II. The concentration of aldrin, endrin and dieldrin in soils under different land uses collected from Vojvodina Province and Dutch target values for soil;<sup>20</sup> *SD* – standard deviation, nd – not detected

Component	Mean ng/g	<i>SD</i>	Minimum ng/g	Maximum ng/g	Detection frequencies, %	Target value ng/g
Agricultural fields ( <i>N</i> = 1086)						
Aldrin	0.12	0.37	nd	4.29	13.2	0.06
Endrin	0.22	2.71	nd	85.33	8.7	0.04
Dieldrin	0.33	1.22	nd	24.55	22.2	0.50
Forestlands and grasslands ( <i>N</i> = 214)						
Aldrin	0.10	0.33	nd	3.15	11.9	0.06
Endrin	0.04	0.23	nd	2.09	4.3	0.04
Dieldrin	0.31	1.12	nd	11.39	17.6	0.50
Orchards and vineyards ( <i>N</i> = 70)						
Aldrin	0.19	0.72	nd	4.94	11.4	0.06
Endrin	0.25	1.10	nd	8.38	11.4	0.04
Dieldrin	0.41	0.88	nd	5.74	32.8	0.50

The concentration of aldrin in all the samples varied from not detected to 4.94 ng/g with the highest detection frequency in soil samples taken at agricultural fields. Aldrin concentrations higher than target value according to the Dutch standard (0.06 ng/g)<sup>20</sup> were measured in 176 soil samples which represent 12.8 % of all soil samples taken in the Vojvodina. Compared with the concentration of aldrin in soils from other countries and regions, the concentration of aldrin in agricultural field soils in this study was comparable with aldrin concentrations detected in agricultural soils in Shanghai, China<sup>8</sup> which varied from not detected to 6.62 ng/g with the detection frequency of 36 %, but it was much

lower than the mean aldrin concentration in agricultural soils from Southern Sonora, Mexico<sup>21</sup> which was reported to be 1600 ng/g.

Concentration of aldrin detected in the forest soils of Taurus Mountains in Turkey<sup>22</sup> ranged from 0.002 to 0.185 ng/g, which is much lower than the concentration of aldrin in forestland and grassland soils within this study.

Dieldrin concentration in all the samples varied from not detected to 24.55 ng/g with the highest detection frequency in soil samples taken at orchards and vineyards. Dieldrin concentrations higher than target value, according to the Dutch standard (0.50 ng/g), were detected in 311 soil samples which represents 22.7 % of all soil samples in our study. In comparison with aldrin and endrin, dieldrin exhibited a significantly higher mean concentration and detection frequencies in all types of soils. This is probably due to the fact that aldrin is quickly transformed in the environment into dieldrin,<sup>23</sup> and dieldrin is extremely persistent in soil.<sup>9,24</sup> According to Seike *et al.*,<sup>25</sup> soils remain contaminated with dieldrin even when aldrin and dieldrin have not been used for the past 40 years. Similar ratio between dieldrin and aldrin was reported in forest soils of Taurus Mountains in Turkey,<sup>22</sup> implying that aldrin residues in Turkey have already been degraded.

In the present study, soils under different land use exhibited mean concentrations of dieldrin which varied from 0.31. to 0.41 ng/g. These mean concentrations were higher than dieldrin concentrations detected in agricultural soils in Shanghai (0.09 ng/g)<sup>8</sup> but comparable with dieldrin concentrations found in the forest soils at Taurus mountain in Turkey (0.26–1.87 ng/g).<sup>22</sup> Dieldrin concentrations in our study were significantly lower than mean dieldrin concentrations in agricultural soils in Alabama, USA (5.19 ng/g),<sup>26</sup> North Portugal (286 ng/g),<sup>5</sup> Lower Fraser Valley, Canada (450 ng/g)<sup>4</sup> and Agra, India (780 ng/g).<sup>11</sup>

Endrin concentration in all the samples varied from not detected to 85.33 ng/g with the highest detection frequency in soil samples taken at orchards and vineyards. Endrin concentrations higher than target value according to the Dutch standard (0.04 ng/g) were detected in 113 soil samples which represents 8.25 % of all soil samples in our study. Mean concentration of endrin in forestlands and grasslands soils in our study was 0.04 ng/g which is comparable with endrin concentrations found in soils of Taurus Mountain in Turkey (0.03–0.44 ng/g). Mean concentration of endrin in agricultural soils was 0.22 ng/g which is much lower than the one found in agricultural soils in Shanghai (nd – 4.32 ng/g) and Lower Fraser Valley, Canada (70 ng/g), thus indicating that endrin was less used as insecticide on field crops in Vojvodina than elsewhere in the world.

Distribution pattern of aldrin, dieldrin and endrin in soils under different land use was also investigated. Comparison of three different types of land use showed the highest aldrin, dieldrin and endrin mean concentrations in orchard and vineyard soils, while the lowest were in the forestland and grasslands soils.

The highest aldrin detection frequency was in agricultural soils, while endrin and dieldrin detection frequencies were highest in orchard and vineyard soils.

The occurrence of pesticide residues (retention, mobility and degradation) in soil depends on properties of compounds themselves and various environmental factors such as pH, soil moisture, soil organic carbon, clay content, etc.<sup>27,28</sup> All these parameters are responsible for organochlorine pesticide (OCP) levels in soil system as well as for the possible adsorption on various soil components such as organic matter, clay and other minerals.

Soil organic matter is one of the most important factors affecting pesticide behavior in soils. Organochlorine pesticides tend to bind with soil organic matter due to their hydrophobic nature which means that the increase in soil organic matter content may lead to the increase of the pesticide residue levels in soil.<sup>29,30</sup> On the other hand, increased pesticide residue levels in soils can prohibit its further microbial degradation<sup>31</sup>. In addition, soil pH can affect the concentrations of organochlorine pesticides through modification of structure of soil organic matter and reduction of microbial activity at the lower pH values, which leads to the lower pesticide degradation in soil.<sup>32</sup> Also, it has been well established that soil types, especially clay content, affect the persistence and retention of organochlorine pesticide residues.<sup>33,34</sup>

Correlations have been examined between aldrin, endrin and dieldrin concentrations in soil and various soil parameters. In this study, soil organic matter was found to be positively associated only with aldrin ( $r = 0.1694$ ,  $p < 0.05$ ), indicating that adsorption of this compound is enhanced by soil organic matter, while no significant correlation was observed between organic matter and endrin or dieldrin. The results did not show any definite pattern of soil pH. Clay content in soil was found to be positively associated with aldrin ( $r = 0.3178$ ,  $p < 0.05$ ) and negatively associated with endrin ( $r = -0.236$ ,  $p < 0.05$ ). This is in good correlation with findings of Baskaran *et al.*,<sup>35</sup> who indicated that the role of clay minerals as pesticide sorbents is of higher importance where organic matter contents are relatively low.

#### *Spatial distribution of cyclodiene-type organochlorine pesticide residues*

Distribution pattern of aldrin, dieldrin and endrin in soils from different districts in Vojvodina was investigated. The mean, minimum, maximum concentrations, and detection frequencies of aldrin, endrin and dieldrin in soils of Vojvodina districts Bačka, Banat and Srem, are shown in Table III.

The maximum aldrin concentration (4.94 ng/g) was found in soil sample collected from the orchard in West Bačka, while maximum dieldrin concentration (24.55 ng/g) was found in the soil sample collected from the agricultural field in North Banat. The highest endrin level (85.33 ng/g) was found in a soil sample collected from the agricultural field in West Bačka. This extremely high

level of endrin in the soil is probably due to its excessive use in the past, since this concentration was detected in soil used for crop production, and endrin was frequently used for insect control in field crops such as grains.

TABLE III. The concentration of aldrin, endrin and dieldrin in soils from different districts of Vojvodina Province; *SD* – standard deviation, nd – not detected; *N* – number of samples

Component	Mean ng/g	<i>SD</i>	Minimum ng/g	Maximum ng/g	Detection frequencies, %
Bačka ( <i>N</i> = 523)					
Aldrin	0.15	0.43	nd	4.94	16.6
Endrin	0.41	3.87	nd	85.33	14.3
Dieldrin	0.35	1.08	nd	14.68	22.9
Banat ( <i>N</i> = 606)					
Aldrin	0.11	0.39	nd	4.29	11.5
Endrin	0.07	0.45	nd	7.57	4.8
Dieldrin	0.26	1.42	nd	24.55	15.3
Srem ( <i>N</i> = 240)					
Aldrin	0.08	0.29	nd	2.13	8.3
Endrin	0.05	0.29	nd	2.19	3.7
Dieldrin	0.43	0.61	nd	3.02	41.2

Spatial distribution maps of the aldrin, endrin, and dieldrin concentrations are shown in Figs. S-2–S-4 of the Supplementary material.

In comparison with other two districts, the highest mean values of aldrin and endrin were detected in soil samples from Bačka, together with the highest detection frequencies of 16.6 and 14.3 % respectively, which is in good correlation with the fact that more than 80 % of soil samples taken in Bačka originated from agricultural fields, and that aldrin was frequently used in the past against soil insects. This is also supported with the findings of Škrbić<sup>15</sup> who detected aldrin in all wheat samples grown in Central Bačka and Central Banat in 2004.

The highest mean value of dieldrin was found in Srem district along with the highest detection frequency of 41 %, while maximum dieldrin concentration was detected in soils of Banat district, which is in good correlation with the high concentrations of dieldrin in wheat samples grown in Central Banat in 2004 thus indicating excessive use of aldrin or dieldrin in the past.<sup>15</sup>

#### CONCLUSIONS

The present study examined the occurrence and spatial distribution of cyclo-diene-type organochlorine pesticides in soils of Vojvodina Province, Serbia. Although the use of aldrin, endrin and dieldrin was prohibited in 1991, residues of those pesticides still exist in the soils of Vojvodina in measurable concentrations. Dieldrin was the most dominant compound in the soil. However, the concentrations of analysed cyclo-diene-type organochlorine pesticides in the most soil samples from Vojvodina province were lower than Dutch target values for



soil quality. Soil organic matter and clay content were found to be positively associated with aldrin content while clay content was found to be negatively associated with endrin. Other physical and chemical properties of the soils demonstrated no effect on loading the pollution in the soils of Vojvodina.

#### SUPPLEMENTARY MATERIAL

Details about soil sampling and spatial distribution maps of the aldrin, endrin and dieldrin concentrations are available electronically from <http://www.shd.org.rs/JSCS/>, or from the corresponding author on request.

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#### ИЗВОД

#### ПРИСУСТВО И ДИСТРИБУЦИЈА ОРГАНОХЛОРНИХ ПЕСТИЦИДА ЦИКЛОДИЕНСКОГ ТИПА У ЗЕМЉИШТУ АУТОНОМНЕ ПОКРАЈИНЕ ВОЈВОДИНЕ

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Алдрин, ендрин и диелдрин су органохлорни пестициди циклодиенског типа са израженом токсичношћу и дуготрајношћу. Иако је у многим земљама у последњих неколико декада употреба ових једињења забрањена, она се још увек могу наћи у животној средини, нарочито у пољопривредном земљишту. Како би се утврдио садржај остатака органохлорних пестицида циклодиенског типа у земљишту Војводине и добили детаљни подаци о њиховој просторној дистрибуцији, прикупљено је 1370 узорака земљишта различите намене на територији целе Војводине, а затим је у њима анализирано присуство алдрина, диелдрина и ендрина. Остаци ових пестицида су у земљиштима Војводине детектовани у мерљивим концентрацијама а најзаступљеније једињење је био диелдрин. Међутим, у већини узорака земљишта из Војводине концентрације анализираних пестицида циклодиенског типа биле су ниже од холандских граничних вредности за квалитет земљишта. Утврђена је позитивна корелација између садржаја органске материје у земљишту концентрације алдрина као и позитивна корелација између алдрина и садржаја глине док је између концентрације ендрина и садржаја глине утврђена негативна корелација.

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