

# PESTICIDE RESIDUES IN CONVENTIONALLY AND ORGANICALLY CULTIVATED PLANTATIONS LOCATED IN NORTHEAST HUNGARY

**L. Simon**

College of Nyíregyháza  
Department of Land Management and  
Rural Development, Nyíregyháza,  
Hungary



**S. Barna**

College of Nyíregyháza  
Department of Land Management and  
Rural,  
Nyíregyháza, Hungary



**Cs.Tóth**

College of Nyíregyháza  
Department of Agricultural Science  
Nyíregyháza, Hungary

**Z. Uri**

College of Nyíregyháza, Department of  
Land Management and Rural  
Development  
Nyíregyháza, Hungary



**REZUMAT:** Reziduurile de pesticide au fost investigate în solurile plantațiilor de măr, vișin și broccoli, convenționale și organice, situate în nord-estul Ungariei. Reziduuri de carbohidrați clorurați și triazină au fost, de asemenea, investigate în frunzele de meri și vișini. Reziduuri de carbohidrați clorurați (de exemplu, DDD, DDE) sunt la fel de prezente în solurile plantațiilor convenționale și organice, cu toate acestea, în solurile plantațiilor ecologice concentrația lor este mai mică și au migrat în profunzimea solului. Reziduurile de triazină nu sunt deloc prezente în soluri. Concentrații semnificative de reziduuri de DDT au fost prezente în frunzele cireșilor din plantații convenționale, în comparație cu plantațiile organice. Reziduurile de DDT nu au fost detectate în frunzele merilor, indiferent de cultivarea lor în plantații convenționale sau ecologice.

**Cuvinte cheie:** Reziduuri carbohidrate clorurate și triazină, plantații organice și convenționale, sol, plantă

**ABSTRACT.** Pesticide residues were investigated in the soils of apple, sour cherry and broccoli conventional and organic plantations located in northeast Hungary. Chlorinated carbohydrate and triazine residues were also investigated in the leaves of apple and sour cherry trees. Residues of chlorinated carbohydrates (e.g. DDD, DDE) are equally present in the soils of conventional and organic plantations, however, in the soils of organic plantations their concentration is lower, and they migrated to lower soil deepness. Triazine residues are not present in the soils at all. Significant concentrations of DDT residues were present in the leaves of conventionally cultivated sour cherry trees, as compared to organic plantations. DDT residues were not detected in the leaves of apple trees, regardless of their cultivation in conventional or organic plantations.

**Keywords:** chlorinated carbohydrates and triazine residues, conventional and organic plantations, soil, plant

## 1. INTRODUCTION

Persistent organic pollutants (POPs) (e.g. dichlorodiphenyltrichloroethane - DDT) are organic compounds that are resistant to environmental degradation through chemical, biological, and photolytic processes. Because of this, they have been observed to persist in the environment, to be capable of long-range transport, bioaccumulate in human and animal tissue, biomagnify in food chains, and to have potential significant impacts on human health and the environment. Altogether 17000 tons of DDT-agents were applied to arable soils in Hungary between 1950 and 1969. Since chlorinated carbohydrates are very persistent, their application was forbidden from January 1, 1969 in Hungary. DDT and other pesticides may cause cancer and that their agricultural use was a threat to wildlife,

particularly birds. DDT was subsequently banned for agricultural use worldwide under the Stockholm Convention (2004), but its limited use in disease vector control continues to this day and remains controversial (Simon, 1999; [http://en.wikipedia.org/wiki/Persistent\\_Organic\\_Pollutant](http://en.wikipedia.org/wiki/Persistent_Organic_Pollutant); <http://en.wikipedia.org/wiki/Ddt>).

From 1954 till 1978 the total amount of applied herbicide agents achieved 128000 tons in Hungary. From this amount 35000 tons were chlorophenoxyacetic acid derivatives, and 22000 tons were chloroalkylamine s-triazine derivatives. Because of the persistence of the chloroalkylamine s-triazine derivatives their accumulation was problematic in maize cultures; therefore from 1972 the annually applicable amounts (1.4 kg/ha active atrazine agent/year, in case of monoculture max. 4 kg/ha/3 years) were regulated by triazine-decree in Hungary

(Simon, 1999). Use of atrazine derivatives is controversial due to widespread contamination in drinking water and its associations with birth defects and menstrual problems when consumed by humans at concentrations below government standards. Although it has been banned in the European Union from 2004, it is still one of the most widely used herbicides in the world (<http://en.wikipedia.org/wiki/Atrazine>).

Considering these facts it is questionable that derivatives of chlorinated carbohydrates and triazine are still present in conventionally and organically cultivated plantations located in north eastern part of Hungary. Our aim was to investigate the presence of these compounds in the soil of apple, sour cherry, and broccoli conventional and organic plantations. Presence of chlorinated carbohydrate residues were also investigated in the leaves of apple and sour cherry trees.

## 2. MATERIALS AND METHODS

The soil cover in northeast Hungary is diverse. Soil samples were taken during July and August 2010 in conventionally and organically cultivated plantations located in 8 small-regions (Fig.1). In this paper the results of pesticide residue concentrations are presented in soils of Közép-Nyírség (Middle Nyírség) small-region. For this region the characteristic soil cover is brown forest soil with "kovárvány" stripes.



**Fig.1.** Small-regions where soil samples were taken in northeast Hungary

In the sampled plantations of the Közép-Nyírség small-region the Jonathán, Golden, Florina, Idared and Mutsu apple cultivars, the Újfehértói fürtös, Érdi bőtermő, Kántorjánosi 3 and Debreceni bőtermő sour cherry cultivars, and the Verde calabrese, Fiesta, Calabrese and Cruiser broccoli cultivars were grown during 2010. Conventionally cultivated plantations were located in Újfehértó and Nyíregyháza, while organically cultivated plantations were sampled in Nyírpazony-Kabalás and Nyíregyháza settlements.

Combining 30 subsamples 2 parallel soil samples were taken from 0-30 cm depth in every plantations with the help of stick augers (producer: Eikelkamp, The Netherlands). The weight of parallel „A” and „B” combined soil samples exceeded 1.5 kg, respectively. With the help of layer augers

(producer: Eikelkamp, The Netherlands) samples were taken from 0-30 cm, 60-90 cm, or 90-120 cm depth. The total fresh weight of every soil sample from a given depth exceeded 0.35-0.5 kg.

In apple and sour cherry plantations (at four localities) leaf samples were taken during July 2010. From every sampled tree 5 leaves were collected from 1.5-2.0 m height. At least 15 trees were sampled each case. Leaves were thoroughly mixed to form 2 parallel subsamples.

The drying of soil and plant samples and preparation for analysis was described in our former publications (Simon and Barna 2010; Simon et al., 2011). The analysis of chlorinated carbohydrate and triazine residues was done by GC-MS technique (equipments; GC: Agilent 6890, MSD: Agilent 5973) at the laboratory of Bálint Analitika Ltd. (Budapest) with 2 inner replications (Simon and Barna 2010).

## 3. RESULTS AND DISCUSSION

In Table 1 the chlorinated carbohydrate content of conventionally and organically cultivated apple plantations is presented in Közép-Nyírség small-region (near Újfehértó and Nyírpazony-Kabalás settlements, respectively) in upper 0-30 cm and 60-90 cm soil depth. Extremely high is the concentration of p,p'-DDD and p,p'-DDE in lower soil layers, where the formerly applied DDT was leached, and its original form can be still found. The measured concentrations were well-above the 0.1 mg/kg threshold limit determined in Government Joint Decree No. 6/2009. (IV. 14.) (2009) for the total concentration of DDT/DDD/DDE residues.

In the 60-90 cm soil layer the 1.088 mg/kg total concentration of chlorinated carbohydrates is more than 2 times higher than the 0.5 mg/kg threshold limit, which is valid for active agents of pesticides and their decomposition products (Government Joint Decree No. 6/2009. (IV. 14.), 2009). It is very advantageous, however, that in organically cultivated apple plantation, apart from p,p'-DDE traces, chlorinated carbohydrate residues were not detected.

In Table 2 the concentrations of chlorinated carbohydrate derivatives are shown in the soil of conventionally and organically cultivated sour cherry plantations. In conventionally cultivated plantation the DDD, DDE and DDT isomers were detected not only in deeper soil layer, but also in upper soil. In 0-30 cm layer their total concentration was more than only in deeper soil layer, but also in upper soil. In 0-30 cm layer their total concentration was more than 3 times higher, while in 30-60 cm layer their total concentration was more than 2 times higher, than the 0.5 mg/kg threshold limit value is. Only traces of DDT residues are present in the soil of Nyírpazony - Kabalás organic sour cherry plantation, and their total concentration is significantly lower, than that of conventional plantation.

**Table 1.** Chlorinated carbohydrate residues in the soils of conventionally and organically cultivated apple plantations in Közép-Nyírség small-region (August 2010, Hungary).

Compounds	Újfehértó conventional plantation (0-30 cm soil layer)	Újfehértó conventional plantation (60-90 cm soil layer)	Nyírpazony - Kabalás organic plantation (0-30 cm soil layer)	Nyírpazony - Kabalás organic plantation (60-90 cm soil layer)
mg/kg*				
α,β,δ-HCH	udl	0.005	udl	udl
γ-HCH/lindan	udl	udl	udl	udl
Hexachlorbenzol	udl	udl	udl	udl
Heptachlor	udl	udl	udl	udl
Heptachlorepoxid	udl	udl	udl	udl
o,p'-DDD	udl	0.077	udl	udl
p,p'-DDD	0.002	<b>0.268</b>	udl	udl
cis-chlordan	udl	udl	udl	udl
Endosulfan-I	udl	udl	udl	udl
trans-Chlordan	udl	udl	udl	udl
o,p'-DDE	udl	0.010	udl	udl
p,p'-DDE	0.025	<b>0.436</b>	0.001	0.001
Endrin	udl	udl	udl	udl
Endosulfan-II	udl	udl	udl	udl
o,p'-DDT	udl	0.079	udl	udl
p,p'-DDT	0.001	0.213	udl	udl
Endrin-aldehyde	udl	udl	udl	udl
Aldrin	udl	udl	udl	udl
Dieldrin	udl	udl	udl	udl
Endosulfan-sulfat	0.003	udl	udl	udl
Endrin-ke-ton	udl	udl	udl	udl
Metoxichlor	udl	udl	udl	udl
<b>Total:</b>	<b>0.031</b>	<b>1.088</b>	<b>0.001</b>	<b>0.001</b>

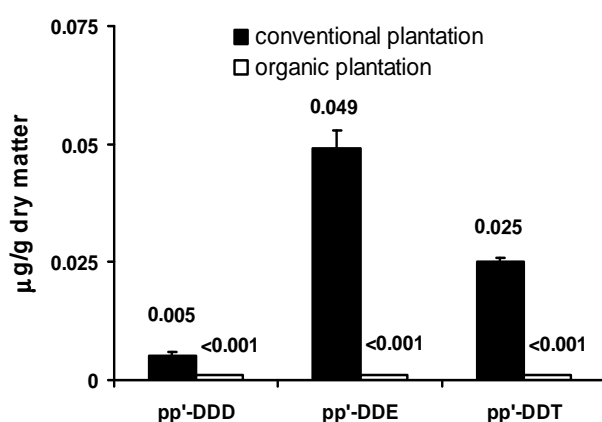
\* All data concerns to dry soil. udl = under the detection limit (<0.001 mg/kg per compound). Measurement accuracy± 10%. n=2.

**Table 2.** Chlorinated carbohydrate residues in the soils of conventionally and organically cultivated sour cherry plantations in Közép-Nyírség small-region (August 2010, Hungary).

Compounds	Újfehértó conventional plantation (0-30 cm soil layer)	Újfehértó conventional plantation (60-90 cm soil layer)	Nyírpazony-Kabalás organic plantation (0-30 cm soil layer)	Nyírpazony - Kabalás organic plantation (60-90 cm soil layer)
mg/kg*				
α,β,δ-HCH	udl	0.008	udl	udl
γ-HCH/lindan	udl	udl	udl	udl
Hexachlorbenzol	udl	udl	udl	udl
Heptachlor	udl	udl	udl	udl
Heptachlorepoxid	udl	udl	udl	udl
o,p'-DDD	0.072	0.097	udl	udl
p,p'-DDD	<b>0.284</b>	<b>0.346</b>	udl	udl
cis-chlordan	udl	udl	udl	udl
Endosulfan-I	udl	udl	udl	udl
trans-Chlordan	udl	udl	udl	udl
o,p'-DDE	0.017	0.012	udl	udl
p,p'-DDE	<b>1.090</b>	0.552	0.003	udl
Endrin	udl	udl	udl	udl
Endosulfan-II	udl	udl	udl	udl
o,p'-DDT	0.085	0.091	udl	udl
p,p'-DDT	<b>0.263</b>	<b>0.250</b>	0.002	udl
Endrin-aldehyde	udl	udl	udl	udl
Aldrin	udl	udl	udl	udl
Dieldrin	udl	udl	udl	udl
Endosulfan-sulfat	0.003	udl	udl	udl
Endrin-ke-ton	udl	udl	udl	udl
Metoxichlor	udl	udl	udl	udl
<b>Total:</b>	<b>1.814</b>	<b>1.356</b>	<b>0.005</b>	udl

\* All data concerns to dry soil. udl = under the detection limit (<0.001 mg/kg per compound). Measurement accuracy± 10%. n=2.

Concerning the DDT derivatives in the soil of conventionally and organically cultivated broccoli plantations the total concentration of chlorinated carbohydrates was definitively less than in apple or sour cherry orchards. In 0-30 cm soil layer of conventional broccoli altogether 0.024 mg/kg, while in organic broccoli plantations altogether 0.033 mg/kg chlorinated carbohydrate derivative was found. In both plantations 0.002 mg/kg p,p'-DDD was detected in 0-30 cm soil layer. In 0-30 cm soil layer of conventional plantation 0.022 p,p'-DDE, and in organic plantation 0.013 mg/kg p,p'-DDE was found. In organic plantation the dieldrin concentration was 0.018 mg/kg, while in conventional plantation the concentration of this compound was below the <0.001 mg/kg detection limit.



**Fig. 2.** DDT-residues in the leaves of sour cherry trees cultivated in conventional (Újfehértó, Hungary) or organic (Nyírpazony-Kabalás, Hungary) plantations (July 2010, n=2).

DDT residues were present in the leaves of conventionally cultivated sour cherry trees (Fig. 2),

**Table 3.** Chlorinated carbohydrate or triazine derivatives in various layers of the soils of conventionally or organically cultivated plantations (July and August 2010, Hungary).

Conventional plantations				Organic plantations			
<i>Chlorinated carbohydrate derivatives*</i>							
0-30 cm	30-60 cm	60-90 cm	90-120 cm	0-30 cm	30-60 cm	60-90 cm	90-120 cm
+	x	+	+	+	x	+	-
From 13 soil samples in 11 are present (85%)				From 21 soil samples in 14 are present (66%)			
<i>Triazine derivatives**</i>							
-	x	-	-	-	x	-	-

\*22 residues were determined, \*\*17 residues were determined, + present; - not present; x not investigated

## ACKNOWLEDGEMENTS

This research was supported by EA\_NORVEGALAP-BIOBEL09 project, realized during 2010 in the College of Nyíregyháza. Valuable assistance of Ms Valéria Darvasiné Tasi, Ms Zsófia Nemesné Száva, Ms Marianna Pelachné Erdős, Dr. Béla Szabó, Dr. Sándor Vágvölgyi and Mr János Veisz is appreciated.

while in the leaves of apple plantations they were not detected (data not shown). Concentration of DDT residues was significantly higher in the leaves of conventionally cultivated sour cherry trees, while in organic plantation their concentration was below the detection limit. Our results are in agreement with Dinya et al. (2010).

Seventeen triazine derivatives (atrazine-desizopropil, atrazine-desethyl, atraton, prometon, simazine, atrazine, propazine, terbumeton, terbuthylazine, secbumeton, sebuthylazine, metribuzin, simetryn, ametryne, prometryne, terbutyne and hexazinone) were investigated in 0-30 cm and 60-90 cm soil depths of apple, sour cherry and broccoli plantations. Regardless of conventional or organic cultivation neither derivative was detected in the soils even in deeper soil layers.

In Table 3 is summarized the occurrence of chlorinated carbohydrate or triazine derivatives in various layers of the soils. Altogether 34 soil samples were taken in 28 locations of 8 small-regions (Fig. 1).

At conventionally cultivated plantations from 13 soil samples in 11 are present chlorinated carbohydrate derivatives, this is 85%. In organically cultivated plantations from 21 soil samples in 14 are present chlorinated carbohydrate derivatives, this represents 66%. As a general observation, in the soils of organic plantations the concentration of chlorinated carbohydrate derivatives is definitively lower and they migrated to lower soil deepness, than in conventionally cultivated plantations. In 90-120 cm soil layer of organic plantations, in contrast to conventionally cultivated plantations, chlorinated carbohydrate derivatives are not present at all.

## REFERENCES

- [1] **Dinya, Z.** 2010. Bio/nem bio agrártermékek beltartalmi értékeinek összehasonlítása (Comparison of chemical composition of bio or non bio agricultural products). In: Tóth, Cs. (ed.). Organikus gazdálkodás biológiai alapjainak komplex agronómiai, bioanalitikai vizsgálata az Észak-Alföldi Régiót jól reprezentáló fajták tekintetében (Complex agronomical,

- bioanalytical investigation of the biological bases of organic farming regarding the well representing cultivars of Észak-Alföld Region of Hungary). Conference proceedings. November 2011, College of Nyíregyháza, Nyíregyháza, Hungary. Bessenyei György Book Publisher, Nyíregyháza. pp. 27-99. (ISBN 978-615-5097-11-9). (in Hungarian)
- [2] **Government Joint Decree** No. 6/2009. (IV. 14.) KvVM-EüM-FVM együttes rendelet a földtani közeg és a felszín alatti víz szennyezéssel szembeni védelméhez szükséges határértékekről és a szennyezések méréséről. (KvVM-EüM-FVM on limit values established for the protection of groundwater and the geological medium), 2009. (in Hungarian)
- [3] **Simon, L.** (ed.) 1999. Talajszennyeződés, talajtisztítás (Soil pollution, soil remediation). Környezetügyi Műszaki Gazdasági Tájékoztató. 5. kötet. Környezetgazdálkodási Intézet. Budapest. pp.1-221. (in Hungarian)
- [4] **Simon, L., Barna, S.** 2010. Toxikus elemek és növényvédőszer-maradványok a konvencionális és ökológiai termesztésű ültetvények talajában és talajvizében (Toxic elements and pesticide residues in the soils and soil water of conventionally and organically cultivated plantations). In: Tóth, Cs. (ed.). Proceedings of the conference “Organikus gazdálkodás biológiai alapjainak komplex agronómiai, bioanalitikai vizsgálata az Észak-Alföldi Régiót jól reprezentáló fajták tekintetében” (Complex agronomical, bioanalytical investigation of the biological bases of organic farming regarding the well representing cultivars of Észak-Alföld Region of Hungary). Conference proceedings. November 2011, College of Nyíregyháza, Nyíregyháza, Hungary. Bessenyei György Book Publisher, Nyíregyháza. pp. 100-124. (ISBN 978-615-5097-11-9). (in Hungarian)
- [5] **Simon, L. Barna, S., Uri, Zs., Koncz J.** 2011. Toxikus elemek és növényvédőszer-maradványok a konvencionális és ökológiai termesztésű ültetvények talajában és a gyümölcsfák leveleiben (Toxic elements and pesticide residues in the soils and orchard leaves of conventionally and organically cultivated plantations). In: Mócsy, I., Szacsvai, K., Urák, I., Zsigmond, A. R., Szikszai, A. (eds.). VII. Kárpát-medencei Környezettudományi Konferencia (7<sup>th</sup> Environmental Science Conference of the Carpathian Basin). Cluj-Napoca, March 24-27, 2011. Vol. 2. Ábel Publisher, Cluj-Napoca, pp. 566-572. (ISSN 1842-9815) (in Hungarian)
- [6] <http://en.wikipedia.org/wiki/Atrazine> (downloaded by October 16, 2011)
- [7] <http://en.wikipedia.org/wiki/Ddt> (downloaded by October 16, 2011)
- [8] [http://en.wikipedia.org/wiki/Persistent\\_Organic\\_Pollutant](http://en.wikipedia.org/wiki/Persistent_Organic_Pollutant) (downloaded by October 16, 2011)