# FOOD CONTAMINATION WITH CHLOROORGANIC COMPOUNDS

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**Summary.** Contamination of food with persistent chloroorganic pesticides is a serious hazard to human health. The aim of this study was to determine students' exposure to chloroorganic insecticides ingested with food. The content of lindane, as well as 1,1,1-trichlor-2,2-di(4-chlorphenyl)ethane (DDT) and its metabolites, were determined by gas chromatography in duplicates of 30 daily food rations of the students of the Faculty of Food Sciences of the University of Warmia and Mazury in Olsztyn, collected during three winter months of 2005. Only in one ration 1,1,1-trichlor-2,2-di(4-chlorphenyl)ethane (DDT) was detected at level of 0.0564 mg/kg fat; 1,1-dichlor-2,2-di-(p-chlorphenyl)ethene (DDE) was detected in all samples (0.0105 mg/kg on average, ranging from 0.0017 mg/kg to 0.0546 mg/kg), and 1,1-dichlor-2,2-di-(p-chlorphenyl)ethane (DDD) – in five samples (0.0325 mg/kg on average, ranging from 0.0022 mg/kg to 0.1433 mg/kg), respectively. The examined diets (except for the one containing DDT) supplied a daily average of 0.0004 mg/kg ΣDDT, which accounts for 0.06 % acceptable daily intake (ADI).
**Key words:** diet, DDT, lindane, food safety

# MAISTO TARŠA CHLORORGANINIAIS JUNGINIAIS

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**Santrauka.** Maisto tarša chlororganiniais pesticidais kelia didelį pavojų žmonių sveikatai. Šio darbo tikslas buvo nustatyti chlororganinių insekticidų kiekį Olštino Varmijos ir Mozūrijos universiteto studentų kasdieniame maisto racione. Dujų chromatografijos metodu buvo nustatyti lindano, 1,1,1-trichlor-2,2-di(4-chlorfenil)etano (DDT) bei jo metabolitų kiekiai. Bandiniai rinkti tris 2005 metų žiemos mėnesius, 30 dienų. Tik viename dienos racione buvo rasta DDT – 0,0564 mg/kg riebalų; visuose bandiniuose rasta 1,1-dichlor-2,2-di-(p-chlorfenil)eteno (DDE) (vidutiniškai 0,0105 mg/kg, kintant nuo 0,0017 mg/kg iki 0,0546 mg/kg), o 1,1-dichlor-2,2-di-(p-chlorfenil)etano (DDD) – penkiuose bandiniuose (vidutiniškai 0,0325 mg/kg, kintant nuo 0,0022 mg/kg iki 0,1433 mg/kg). Nustatytas bendras chlororganinių junginių kiekis dienos racione siekia 0,0004 mg/kg, t. y. 0,06 proc. didžiausios leistinos chlororganinių junginių koncentracijos.
**Raktažodžiai**: maistas, DDT, lindanas, maisto sauga.

**Introduction.** Safe food and correct nutrition are among the most important environmental factors which affect human health, i.e. the physical development, good mental well-being, productivity at a proper level and ability to absorb information. Adequate nutrition is therefore an environmental factor, which significantly affects the health of an individual and, consequently, ensures the continuity of the human population (Alcock, 2000; Schlatter, 1994).

The chemical compounds that people may be exposed to both due to their profession or to the environment they are in, differ in terms of their physicochemical properties and toxicity, their emission, range of influence and persistence in the environment (Wille et al. 1998, Wasiela, Dutkiewicz 1994). Great concern was caused by chloroorganic compounds, which proved to be extremely persistent in the environment and accumulative in the food chain (Rogan, Chen, 2005, Shi at al. 2006, Weiss, 2005).

The application of DDT-containing pesticides has been banned in most countries since the 1960s. The hopes

that it would be possible to clear the environment of the residues of the compound have proved to be futile; despite the passage of time, the monitoring of the environment and food in many countries has confirmed the ubiquity of the compound. Bettinetti et al. (2006) mentions four reasons why the level of DDT residues remains constant: 1. manufacturing and application of DDT containing preparations in controlling malaria and other tropical diseases, 2. manufacturing of DDT for the production of acaricide by the name of Dicofol, 3. transport through the air over considerable distances, and 4. waste dumped in areas where application of DDT is no longer allowed.

The use of DDT nowadays raises various controversies. Rogan and Chen (2005) have recently reviewed the abundant literature on health hazards and benefits resulting from the use of the compound. They warned that the success in controlling malaria may be offset over time by an increased number of early births, reduced time of natural breastfeeding, increased infant death rate as well as neurological and reproductive defects. DDT belongs to a group of compounds called endocrine disruptors that influence the biological function of hormones (Weiss, 2005). Scholars are not unanimous in their opinion on the carcinogenic properties of the compound. On the one hand, Rogan and Chen (2005) question the value of the evidence of the epidemiological studies conducted so far but, on the other hand, Darbre (2006) published a literature review which confirmed the functional role of the combined interactions of cosmetic chemicals with environmental estrogens in supporting breast cancer development.

In countries where chloroorganic insecticides have not been applied for over 30 years, DDT in the environment may come from long forgotten dumpsites of unused preparations. In Poland, DDT-containing preparations have been banned since 1974, and those that have not been used were disposed in so-called "tombs", often located at random sites without prior geological examinations, built carelessly and without appropriate insulation (Grzybowski at al. 2005).

The aim of this study was to determine the students' exposure to chloroorganic insecticides ingested with food.

**Material and methods.** Materials taken for the study were the daily food rations of 30 students (27 female and 3 male) in the 3rd and 5th year of the Faculty of Food Sciences of the University of Warmia and Mazury in Olsztyn, collected during the period from November 2005 to February 2006. Diets were collected in plastic containers, certified for contact with food. The rations were duplicates of the food and drink consumed during a day. Each ration was weighed, inedible parts were removed and the rest was frozen until an analysis was performed.

After being defrosted at the room temperature, the samples were re-homogenised and fat was extracted by the method of Schmidt-Bondzyński, modified by Retzalffa (Polish norm PN – 73A – 86232) in which a given sample, subjected to acidic hydrolysis, is extracted with a mixture of petroleum benzine and diethyl ether.

The residues of chloroorganic insecticides (DDT and its metabolites and lindane) were determined by the method developed by Amarowicz et al. (1986), which consists in simultaneous hydrolysis of fat with sulphuric acid and extraction with n-hexane. The analysis was performed with a Unicam PU 4600 apparatus with an electron capture detector.

The results were analysed with Statistica 7.0 Software.

**Results and discussion.** One common feature of the examined diets was their low weight (1670 g on average, ranging from 802 g to 3196 g). In all the samples the presence of DDE was detected; lindane was found in ten of them. DDD was detected in five of them, whereas the original DDT was found in only one of them. The unique diet, whose weight was equal to 2735 g, was accumulated by one of the three male students taking part in the experiment (Table 1).

Table 1. **Chloroorganic insecticides concentration in students' diets** (mg/kg of fat)

| | γ-HCH | DDE | DDD | DDT | ΣDDT |
|---|---|---|---|---|---|
| Average | 0.0017 | 0.0105 | 0.0325 | 0.0564* | 0.01777 |
| Standard deviation | 0.0026 | 0.0122 | 0.0619 | - | 0.03744 |
| Variation coefficient | 1.52 | 1.16 | 1.90 | - | 2.10 |
| Range | 0.0001-0.0075 | 0.0017-0.0546 | 0.0022-0.1433 | - | 0.0017-0.2046 |
| Median | 0.0006 | 0.0067 | 0.0044 | - | 0.0073 |

* determined in only one diet

The results achieved in this study are lower than those from the studies performed in previous years. The average content of hexachlorcyclichexane (γ-HCH) in the examined food rations was equal to 0.0017 mg/kg, whereas ΣDDT was 0.0177 mg/kg. Daily food rations of students at the Olsztyn University were analysed back in 1983 by Amarowicz and Smoczyński (1984) where the mean lindane and ΣDDT content in fat of the students' daily rations amounted to 0.0054 mg/kg and 0.0413 mg/kg respectively. Earlier studies conducted at the University of Warmia and Mazury in Olsztyn showed the content of chlorinated hydrocarbons to be decreasing. The determined levels of lindane were about 0.0034 mg/kg of fat, whereas ΣDDT was 0.052 mg/kg (Skibniewska, Smoczyński, 2000) and 0.0029 mg/kg fat for γ-HCH and 0.0463 mg/kg for ΣDDT (Radzymińska et al. 2005) and were lower than those determined earlier by those authors. Diets of workers in Poznań (Przysławski et al. 1991) contained the examined compounds at higher concentrations than those determined in this study (γ-HCH –

0.0953 mg/kg, ΣDDT – 0.2290 mg/kg).

Whole day food rations collected in hospitals in four Polish towns (Skibniewska 2001) contained γ-HCH in concentrations of 0.0031 mg/kg of fat and ΣDDT – 0.0395 mg/kg. Wilhelm et al. (2002) analyzed 98 duplicate diets of German children aged from1.5 to 5.3 years. They found 0.0053 $mg \cdot kg^{-1}$ of γ-HCH, 0.0007 $mg \cdot kg^{-1}$ of DDE and 0.0020 $mg \cdot kg^{-1}$ of DDT.

Using the information on daily consumption, the daily intake of chloroorganic insecticide by students of the University of Warmia and Mazury was determined (Table 2). The average values (excluding the ration containing high concentrations of the parent DDT) were the following: γ-HCH – 0.0311 mg/day, DDE – 0.1707 mg/day and DDD – 0.1729 mg/day. It has been estimated in European countries that an individual consumes daily 0.3 mg DDE and 0.372 mg DDE with food (Urieta, 1996; Wilhelm, 2002; Vaz, 1995), which accounts for several percent of ADI. Lazaro (1996) determined 0.1432 mg/kg of ΣDDT in meals containing fish which were the main source of the compounds. Larger amounts of DDT are consumed by inhabitants of Asia, where the content of DDT and its metabolites is close to ADI (Sudaryanto et al. 2007).

Table 2. **Daily intake of chloroorganic insecticides with students' diet ***(mg/day)

| | γ-HCH | DDE | DDD | ΣDDT |
|---|---|---|---|---|
| Average | 0.0311 | 0.1707 | 0.1729 | 0.1946 |
| Standard deviation | 0.0427 | 0.2219 | 0.2006 | 0.2480 |
| Variation coefficient | 1.37 | 1.29 | 1,16 | 1.27 |
| Range | 0.0016-0.1326 | 0.0282-0.9334 | 0.0164-0.4187 | 0.0293-0.9334 |
| Median | 0.0126 | 0.1012 | 0.1061 | 0.1022 |

* the diet containing DDT excluded

Skibniewska (2001) determined the lindane content in the daily rations of hospital food as 0.0035mg/day and that of ΣDDT as 0.055 mg. Sudaryanto et al. (2007) examined the level of the daily consumption of fish contaminated with chloroorganic compounds and determined the ΣDDT content to be lower than that determined in this study: 0.0011 mg/day. Darenrud (2006) also determined higher amounts of the discussed compounds than those determined in this study. The total DDT in the selected products (fish, meat, dairy products, eggs, etc.) amounted to 0.523 mg/day, whereas HCH was 0.081 mg/day.

DDT (0.3684 mg/day) was detected in one ration, which consisted mainly of products of plant origin, i.e. bread, vegetables and jam, also 30 g of mortadella, 45 g of hard cheese, 20 g of butter, 100 g of rice, 30 g of peanuts, 6 mugs of tea. Due to the lipophilic properties of chloroorganic compounds (www.codexalimentarius.net/), higher levels of their residues are determined in food of animal origin, which was in small amounts and which should supply mainly DDE. Therefore, it should be concluded that the source of residue of maternal DDT were products imported from the area where DDT-based preparations are still in use (rice, peanuts or tea).

A study conducted by Góralczyk et al. (2000) into the residue of chloroorganic compounds in tea showed that the mean total DDT content in black tea was 0.003 mg/kg of the product, and that of lindane was 0.002 mg/kg. The content of the discussed compounds in green tea and in fruit teas was similar – 0.003 mg/kg for ΣDDT and 0.001mg/kg for HCH. In none of the examined teas was the highest acceptable residue values found to exceed Polish norms: ΣDDT – 0.2 mg/kg, γ-HCH – 0.1 mg/kg.

An adult should not ingest more than 600 μg Σ DDT/day and 60 μg γ-HCH/day (www.codex alimentarius.net/). The examined diets (except for the ration containing DDT) supplied 0.1946 mg/kg ΣDDT on average, which accounts for 0.06 % ADI. The results for lindane (γ-HCH) are also lower than the ones established by the World Health Organization (WHO).

However, it is justified to monitor the content of hazardous compounds with a persistent presence in food – these include chloroorganic compounds – and to take actions aimed at reducing the content of the compounds, e.g. by composing an appropriate daily diet for humans.

**Conclusions**

1. 1,1-Dichlor-2,2-di-(p-chlorphenyl)ethene (DDE) was detected in all of the examined food rations and lindane in ten of the samples. In the other rations, only traces were found. 1,1,1-Trichlor-2,2-di(4-chlorphenyl)ethane (DDT) was found only in one ration.
2. The levels of the other chlorinated hydrocarbons in daily rations are much lower than the acceptable daily intake. It results from the slow degradation of the discussed compounds, both in the environment and in food.

**Acknowledgements**

Research granted from scientific sources as a research project in 2006-2009 MNiSW No: N305 006 31/0326.

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Received 9 December 2008
Accepted 24 April 2009