A critical ecotoxicological perspective on pesticides used in Hungary

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Abstract

Hungary used 8.75 kg/ha in 1990, but 1.9 kg/ha pesticide in 1997, although because of the increasing rate of the uncultivated fields, we may calculate 3.2 kg pesticide/ha in the cultivated area. During 1998 there was an evaluation of drinking water near Vác, which found 5.7 ppb atrazine, 3.3 ppb prometryn and 0.3 ppb diazinon. Based on at least one pesticide (mostly 2,4-D and MCPA) the drinking water does not meet the EEC-standard in seven places. Between two and three percent of vegetables and fruits had unacceptable pesticide residue levels before 1989. Today, because of the greater number of unskilled farmers, this rate has increased. Evaluations are suggested to deal with many problems with vegetables from greenhouses, especially lettuce and dithiocarbamates relationship need to be carefully regulated. Summarising the environmental (persistency: air, water and soil pollution), acute (in different organisms) and chronic health problems (mutagenicity, carcinogenicity, teratogencity, oestrogen-agonist and immunomodulation effects, bioaccumulation and biomagnification) we concluded 22% of the nearly 400 active ingredients would need to be banned in Hungary. Unfortunately not only the active ingredients, but also contaminants and formulating agents need to be carefully revised.

Background and context

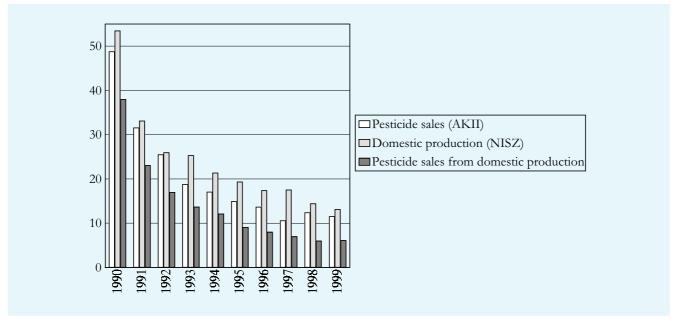
Hungarians working in plant protection like to remember 1968, when we were the first to ban *DDT*, and to forget how we gave up our rigorous pesticide regulation to conform to the European standard. During the sixties, Hungary was in the top four, where *DDT*-contamination was highest in the world in human adipose tissue and milk. In 1966 nearly 70 thousand tonnes of pesticides were used. Today there is no real difference between the plant protection routine in the USA (despite constant efforts for better practice on the part of the Environmental Protection Agency - EPA) and Hungary.

During the seventies in Hungary, as in the other countries in Eastern Europe, agriculture used a vast amount (nearly 60 thousand tonnes) of pesticides (e.g., *atrazine, benomyl, camphechlor, captan, 2,4-D, lindane, mancozeb*, mercuric salts, *methoxychlor*, organophosphorus insecticides, *2,4,5-T* etc.). The only aim was to produce more crops than in the previous year. Chemical-related plant protection displaced our traditional ideas (i.e., agricultural, mechanical, biological methods).

An active ingredient, *klorinol* - manufactured at that period - is the most problematic item in terms of environmental pollution in Hungary today; it is produced by a pesticide factory called BVM (Budapesti Vegyimûvek, now the *atrazine*-interested Oxon is a 40%-owner). *Klorinol* is also known as *fenteracol* or *2,4,5-T-etanol*. Although we may read in one part of a book¹ - which discusses the commercialisation of this herbicide that - the chromatid-type aberrations doubled among workers working with *klorinol* (the effects of *TCDD*-contaminated AGENT ORANGE - *2,4,5-T+2,4-D* - in the Vietnam War were also known)²-³ the factory ignored this knowledge.⁴ Between 1967-1985 BVM manufactured an enormous amount of BUVINOL (*klorinol: atrazine* = 1:1) and collected their 16-28 thousand tonnes of *TCDD*-contaminated highly chlorinated phenolic waste in a small village called Garé in southern Hungary. The first sign of environmental pollution came to light in 1983. The evaluation comprised 50 ha polluted fields and 140,000 m³ contaminated soil. The BVM, with financial support from the government, started the rescue work in 1998, but thus far the highly chlorinated waste has been not eliminated. Today, Garé is a grim reminder of poorly planned chemical production, from an environmental point of view.

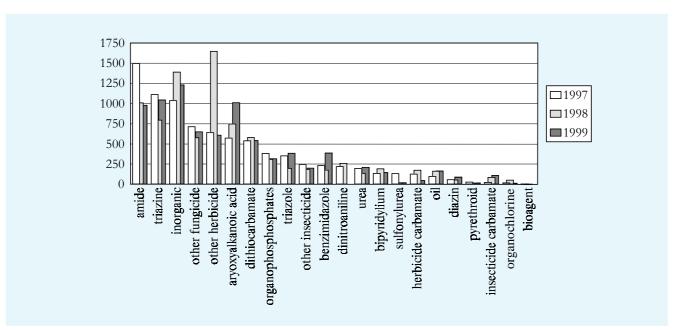
In the seventies, Hungarian agricultural university trained plant protection engineers were technically skilled but lacked a background in ecotoxicology. This is not a great surprise when one considers the level of awareness in this period. During the eighties, the idea of "Integrated Pest Management" (IPM) reached Hungary, yet the result was only a marginal decrease in pesticide consumption. After Hungary's turn to democracy in 1989, privatisation changed the agricultural situation drastically. A huge part of the US-type farming system - once led by skilled engineers - quickly disappeared, and two hundred thousand unskilled farmers started to work on small family farms, using pesticides of course. During the nineties, the most characteristic tendency in Hungarian agriculture was pauperisation. This is the real motivation behind the rapid decrease in pesticide consumption (Figure 1).⁵⁻⁸ Hungary used 8.75 kg pesticides/ha in 1990, but only 1.9 kg/ha in 1997, although because of the increasing amount of uncultivated fields, we may calculate 3.2 kg pesticide/ha in cultivated areas. The impression is that Hungary - like Scandinavian countries⁹⁻¹⁰ - established a highly effective pesticide reduction program, although Hungarian ministries never declared this.

The quantity is only one parameter for measuring or describing pesticide usage. Two or three kg pesticide/ha/year appears to be an optimal value if compared to the rate in the Netherlands (17.5 kg/ha reduced later to 9 kg/ha) or Belgium (10.7 kg/ha) in 1994.¹¹ This statistic may be improved by using more effective pesticides (e.g., pyrethroids, sulfonylurea herbicides etc.), but there is no significant ecotoxicological difference between two pesticides if the same effect is achieved by different doses. The ecotoxicological evaluation of an active ingredient and its commercialised products is very complex, and science remains short on suitable answers.¹² Figure 2 summarises the active ingredient groups, which have been used in the last three years.^{7-8,13} Checking the global sales lists we found similar trends in Hungary: *atrazine*, *metolachlor*, *2,4-D*, *acetochlor*, EBDC, organophosphates, etc.¹⁴



Source: AKII, 1994; 1999; NISZ, 2000

Figure 1. Hungarian pesticide market from 1990 to 1999 (thousand tonnes)



Source: NISZ 1999, 2000

Figure 2. Important active ingredient groups on the Hungarian pesticide market from 1997 to 1999

Below we attempt to summarise the *sensu lato* ecotoxicological problems with pesticides. In the case of the non-living environment we need to keep in mind the persistency of a pesticide that implicates air, water and soil pollution. Methyl bromide is a significant agent in terms of the depletion of ozone in the stratosphere. The goal of discontinuing its use totally in the period 2005-2015 has been declared. ¹⁵ Unfortunately not only the UV-stable gaseous pesticides

arrive by air. One of the main reasons for the Arctic organochlorine pollution (e.g., *camphechlor, chlordane, HCH, PCB*, etc.) is the contaminated rains and snows that arrive by wind. The "off-target" effect of pesticide application widens local problems and in fact globalises them. For water pollution at least relative water solubility and persistency is needed, although dibenzo-dioxin-type, non-water-soluble chemicals accumulate in the sediment and detritus-feeding animals pass them into the aquatic food chain. The "run-off" effect of pesticide application may quickly contaminate the surface water and except for water-soluble products the groundwater as well, over time. There are several examples of highly contaminated rivers (e.g., organochlorines - Ob, Pyasina, etc) and sea (e.g., organochlorines - Kara Sea, Baltic Sea, North Sea, etc.). These were the reasons why the "Red-list" - including surface water contaminant pesticides - was compiled. The second surface water contaminant pesticides - was compiled.

Ten percent of the Hungarian pesticide assortment belongs to the water contaminants (Figure 3)¹⁹: alachlor, atrazine, azinphos-methyl, dichlorvos, endosulfan, fenitrothion, lindane, malathion, simazine, trifluralin, etc. In these cases, there is a strong correlation between the annual sale and the surface water contamination (i.e., atrazine, 2,4-D, diazinon, dichlorprop, MCPA, phorate, prometryn, terbutryn were measured in the mid-nineties - László Győrfi, pers. comm.).

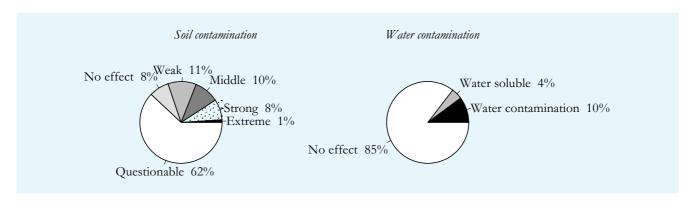


Figure 3. Pesticides with problematic environmental properties in Hungary (1998)

Groundwater contamination is a bit more complicated. Sometimes a long time is needed for an active ingredient to reach it (e.g., DBCP in California). Internationally (e.g., in Germany, Italy, Japan, UK, USA etc.), the most problematic are *atrazine* and *simazine* (but *cyanazine*, *prometryn*, *terbuthylazine*, *terbutryn* too), as when they reach the oxygen-less soil layer, they stop being catabolised. This was the main reason why Germany banned *atrazine* (Peter Seel, pers. comm.).

In Hungary in 1998, an evaluation of drinking water before purification was undertaken near Vác, finding 5.7 ppb *atrazine*, 3.3 ppb *prometryn* and 0.3 ppb *diazinon*.²⁰ Unfortunately, only 1-5% of the *atrazine* is catabolised during traditional methods of water purification. Based on at least one pesticide (mostly *2,4-D* and *MCPA*) the drinking water does not meet the EEC-standard (0.1 ppb) at Verõce, Surány, Tököl, Gödöllő, Sárvár, Szolnok and Eger.²⁰ The *atrazine*-contamination was higher in Vác than in lowa (2.2 ppb), where negative health effects have been shown in babies.²¹

Soil contamination is more local, although it may cause significant problems for the people leaving in the affected area. In the cotton-growing area of Moldova, where 40 kg pesticide/ha/year (including organochlorines, mostly *DDT* and *dicofol*) was used, the immunomodulation effects among children resulted in a deterioration in their general health condition.²² In Hungary, we use some extremely persistent pesticides (e.g., *fenarimol, tebuconazole, triadimefon, triadimenol*, etc.) (Figure 3).¹⁹

The effects of pesticides on living organisms are very diverse, and can be divided into acute and chronic effects. Most of those working in plant protection study only the acute effects and we cannot find even the most basic information on chronic toxicity in "The Pesticide Manual"²³, which is probably the most frequently used handbook.

The description of acute effects (direct toxicity) is usually based on animal model studies where LC_{50} or LD_{50} values are investigated. Rats are used as a model for mammals. There are no accepted models for birds and fish, and to compare the different active ingredients is often highly complex.²⁴ There is a huge "black hole" in the reactions of predatory and parasitoid insects and worms. Without detailed knowledge of "non-target" effects, IPM-type plant protection is merely a good idea.²⁴

The acute toxicity of pesticides in mammals and birds usually correspond to one another, as between fish and aquatic animals (e.g., amphibians, arthropods, etc.), although these two groups react in very different ways to many types of pesticides (Figure 4). High acute toxicity means increasing risks from accidents. In the Hungarian assortment we found some pesticides (e.g., *aldicarb, terbufos, phorate, oxamyl, parathion-methyl*), which are more toxic for mammals than potassium cyanide. Pyrethroids are extremely dangerous for aquatic organisms. In 1998, there was an accident where the Chinoin manufactured *cypermethrin* reached the Danube and nearly 25 tonnes of fish died over the next few days.

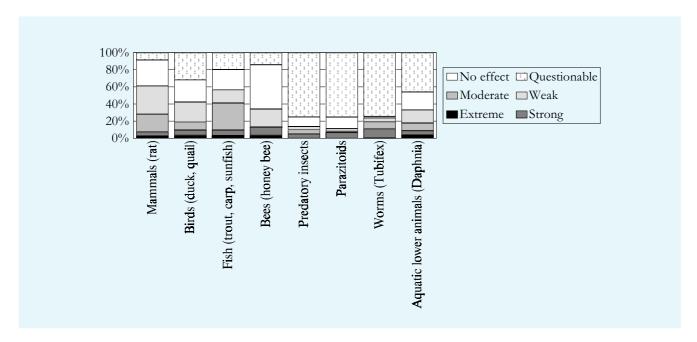


Figure 4. Acute toxicity of pesticides in Hungary (1999)

Chronic toxicity is seemingly much more problematic in terms of practical consequences, but there is no consensus among scientists to validate results (Figure 5).¹⁹ Nearly a quarter of the pesticides used in Hungary is regarded as mutagens.²⁵⁻²⁶ *Captan, carbaryl, chlorothalonil, 2,4-D, dichlorvos*, EBDC [i.e., ethylenebis (dithiocarbamates)], *malathion, phosphamidon* are the active ingredients for which several positive results have been collected. Scientists agree that 60-90% of mutagenic chemicals is carcinogenic.²⁷ In this respect (see Figure 4), the international qualification has not reached the minimum. There is no more uncertain part of our judgement than the carcinogenicity of a single chemical. The International Agency of Cancer (IARC)²⁸ and the EPA²⁹ have their own and differing lists about animal carcinogens which possibly or probably human carcinogens.

The EPA²⁹ qualified **B2** category (= probably human carcinogens): *acifluorfen, alachlor, captan, chlorothalonil, folpet, mancozeb, maneb, metiram, procymidone, propargite, zineb*; **C** category (= possibly human carcinogens): *acephate, amitraz, asulam, atrazine, benomyl, bifenthrin, bromoxynil, clofentezine, cyanazine, cypermethrin, dichlobenil, dichlorvos, diclofop, dimepithin, dimethoate, fomesafen, fosetyl, hexazinone, lindane, linuron, methidathion, methomyl, metolachlor, oxadiazon, oxadixyl, oxyfluorfen, permethrin, phosmet, phosphamidon, propyzamide, propiconazole, hexythiazox, simazine, terbutryn, thiophanate-methyl, triadimefon, triadimenol, trifluralin.*

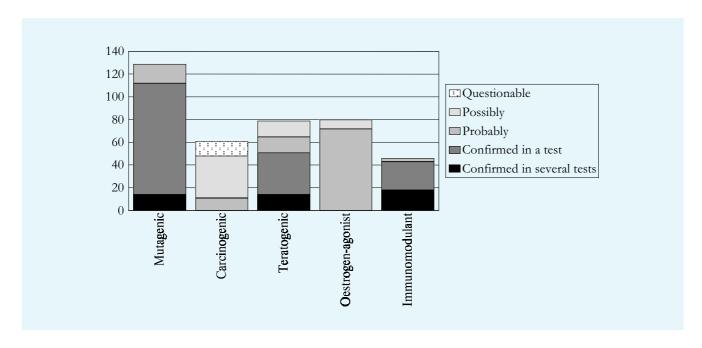


Figure 5. Chronic toxicity of pesticides in Hungary (1999)

IARC²⁸ found aryloxyalcanoic acids, *dichlorvos*, *HCH*-group, *nitrofen* are possibly carcinogens (Group **2B** - the agent is possibly carcinogenic to humans; there is sufficient evidence of carcinogenicity in experimental animals), but there is only limited evidence (Group **3** - the agent, mixture or exposure circumstances are not classifiable as regards its carcinogenicity to humans) about the carcinogenicity of *atrazine*, *chlorothalonil*, *lindane* and methyl bromide. IARC qualified in the **2A** group (i.e., the agent is probably carcinogenic to humans; a positive association has been observed between exposure and human cancer for which a casual interpretation is credible, but chance, bias or confounding could not be ruled out with reasonable confidence) the situation of people who work constantly with pesticides, but cannot name the individual reason.³⁰

US National Cancer Institute (NCI) and National Toxicology Program (NTP) between 1976 and 1992 evaluated pesticides:³¹ **CE** (= clear evidence between chemical and carcinogenicity): *captan (duodenum polypoid carcinoma* in mice), *nitrofen* (liver cancer in mice and pancreas cancer in female rats), *trifluralin* (liver-, lung- and stomach cancers in female mice), *chlorothalonil* (kidney cancer in rats), *daminozide* (ovarian cancers in rats), *ziram* (thyroid cancer in male rats), *dichlorvos* (stomach cancer in female mice); SE (= several evidence between chemical and carcinogenicity): dichlorvos (leukaemia and pancreatic *adenoma* in male rats, stomach cancer in male mice).

In animal tests several pesticides have teratogenic effects.³² *Benomyl, captan, carbaryl, 2,4-D, diazinon, dimethoate*, EBDC and *malathion* have been shown to have teratogenic effects in different animal species.³³ After the CONTERGAN report most scientists believe there is no clear evidence linking animal and a human teratogens.

Immunomodulation and endocrinological (mostly oestrogenic) effects are not a significant part of the pesticide qualification, although most scientists suggest the opposite. *Carbaryl* and *parathion-methyl* have strong immunomodulation effects and reduce the protective properties of the organisms against pathogens and transformed, canceric cells.²² Lots of pesticides (*atrazine*, organochlorines, etc.) are regarded as having oestrogenic-agonist effects.³⁴⁻³⁵

There are two further effects, which deserve mention. Bioaccumulation means a chemical builds up in a tissue of a single organism. Usually lipid-rich tissues (e.g., adipose tissue, marrow, gonads, mamma, etc.) are the targets, and these types of chemicals are usually secreted by mamma. For a longer period "persistent organic pollutants" (*POP*-chemicals): organochlorine insecticides (*DDT*, *HCH*, *camphechlor*, *aldrin*, *dieldrin*, *endrin*, *chlordane*, *heptachlor*, *mirex*), industrial chemicals (*PCB*, *PBB*) and organotin fungicides (*TBTO*), and for a shorter time further pesticides (*atrazine*, *chlorpyrifos*, *chlorothalonil*, *methoxychlor*, *metolachlor*, *terbufos*, *trifluralin*) are built up. Biomagnification is an accumulation via the food chain, and *POP*-chemicals have a bad reputation in this respect.¹⁶

The problems with active ingredients are summarised in Table 1, where we recognise how many reasons are not sufficient for us to ban a single pesticide. We concluded that 22% of the near 400 active ingredients would need to be banned in Hungary (Fig. 6). 19,36 Pesticide revision resulted in a similar situation in Sweden in 1990 when they reduced 230 active ingredients to 150.9 This would not shock the main parts of Hungarian practice, because there are as many as 40 parallel technologies for an important pest. In such cases there are no acceptable reasons for a risk analysis, 37 the potential danger should be enough.

Table 1. Suggestion to ban pesticides in Hungary for different reasons

9 reasons: gamma-HCH (lindane)

8 reasons: dichlorvos, endosulfan, parathion-methyl 7 reasons: chlorpyrifos, cypermethrin, dimethoate

6 reasons: 2,4-D, aldicarb, atrazine, carbofuran, diuron, linuron, malathion, methomyl, permethrin, trifluralin

5 reasons: carbaryl, cyanazine, diazinon, fenitrothion, fenthion, maneb, zineb

4 reasons: alachlor, benomyl, bifenthrin, copper-oxychlorid, mancozeb, metiram, phosmet, pirimicarb, simazine, thiram, triadimefon, ziram

Pesticide residue in vegetables and fruits is an important parameter of chemical safety. In Hungary, 2-3% of produce having unacceptable residue levels was common before 1989. Today, because of the greater number of unskilled farmers, this rate is higher. High residue levels in vegetables from greenhouses is one of the most problematic fields here as it is everywhere in the world (Figure 7).8 The presented results suggest many problems with vegetables from greenhouses in Hungary; especially lettuce and dithiocarbamates relationship need to be regulated carefully. Some lettuce samples contained a massive amount of residue (Figure 8).8

Beyond the active ingredients we need to remember two further parameters related to a commercial pesticide product: contamination with its metabolites and solvents that were used for commercialisation. Dibenzo-dioxintype contamination is known with regards to aryloxyalcanoic acid-type herbicides (especially 2,4,5-T), nitroso-derivatives in nitrogen-containing pesticides (e.g., nitroso-atrazine, nitroso-simazine, nitroso-carbaryl, nitroso-carbofuran, etc.), ETU/PTU (i.e., ethylenethiourea/propylenethiourea) in EBDC-type fungicides (e.g., mancozeb, maneb, zineb, metiram, propineb, etc.), and lead in copper products. Sometimes these very dangerous contaminants are responsible for chronic effects.^{8,38}

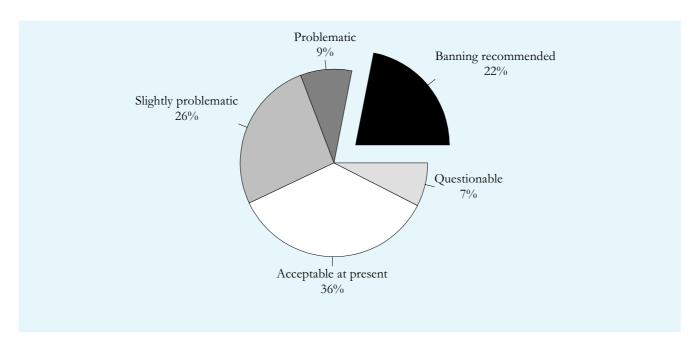


Figure 6. Ecotoxicological view of pesticides in Hungary (1999)

On several occasions it was found that an active ingredient is not mutagenic alone, despite the results with its commercialised products. Sometimes solvents are quite a few times more hazardous. Benzol (in *cypermethrin*), benzol+xylol (in *alachlor*), benzol+xylol+cyclohexanol (in *dimethoate*), chloroform (in *dichlorvos*), 1,4-dioxane+2-nitropropane (in *metaldehyde*) were found to be formulating agents.³⁹

Pesticide re-registration/revision is a very important procedure in agriculture. We need to check periodically whether the "innocent" designation (FDA rule) is appropriate for our pesticides. Prevention is the real goal of chemical safety procedures. The task is not very easy because of its economic consequences. Farmers prefer to buy a cheaper product, and nearly 30% of what pesticide producers sell is ecotoxicologically unacceptable, but cheap. Scandinavian efforts to declare the parameters of an unacceptable pesticide are one of the most important steps towards a real breakthrough, 40 which would be highly appreciated, in the European Union regulation.

Data originated from: Bács-Kiskun, Borsod-Abaúj-Zemplén, Fejér, Fovárosi, Jász-Nagykun-Szolnok, Somogy, Szabolcs-Szatmár-Bereg, Vas, Veszprém counties

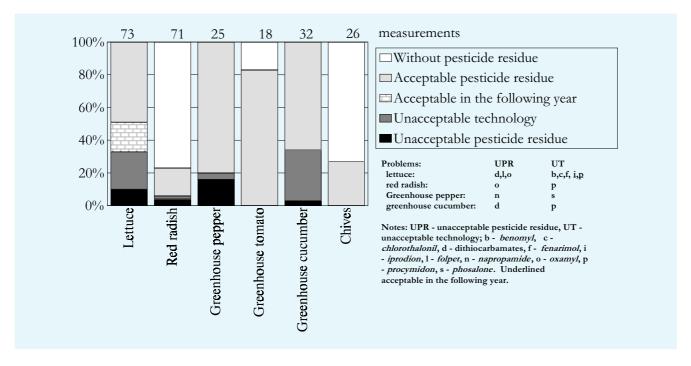


Figure 7. Pesticide residue in vegetable in early spring in Hungary (based on Rózsavölgyi ed., 1997)

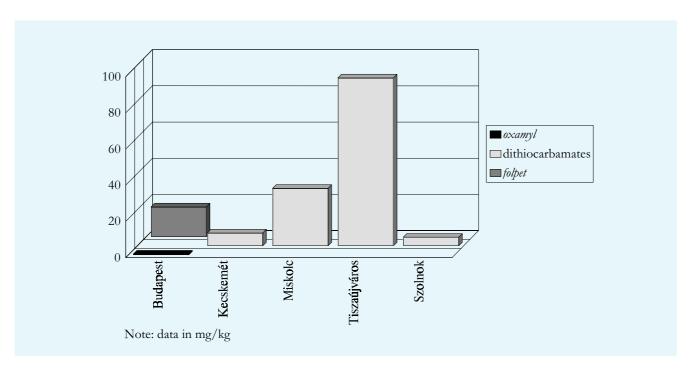


Figure 8. Pesticide residue in lettuce in Hungary (based on Rózsavölgyi ed., 1997)

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