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DIETARY INTAKE OF PESTICIDE RESIDUES BY POLISH CONSUMERS DURING 2001

Stanisław Sadło

Institute of Plant Protection, Regional Experimental Station
Langiewicza 28, 35-101 Rzeszów, Poland
e-mail: S.Sadlo@ior.poznan.pl

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Abstract: In 2001, a total of 2125 samples of plant origin were analysed for residues of the most commonly used pesticides in Poland. Detectable residues were found in 18% of the samples, while in about 0.9% of the samples the residues exceeded national or/and EU harmonised Maximum Residue Limits (MRLs). The long-term (chronic) dietary intake, based on monitoring data, was calculated for 7 pesticides in 9 commodities. The intake of 1 of the pesticide analysed (endosulfan/black currant combination) exceeded 1% of Acceptable Daily Intake (ADI) indicating a sufficient margin of safety for the adult consumer. The short-term (acute) intake was estimated for 19 pesticides in 12 commodities according to the European Commission recommendations. The ADI level was exceeded for toddlers in 7 cases, carbendazim in mushroom, linuron in carrot, chlorothalonil in greenhouse tomato, tolylfluanid and EBDC in strawberry and flusilazole and diazinon in apple. In all cases, however, the safety factor of 2 for a toddler still existed.

Key words: fruits, vegetables, pesticide residues, monitoring, dietary intake

INTRODUCTION

A Maximum Residue Level/Limit (MRL) is defined as the highest concentrations of pesticide residues (expressed in mg/kg) likely to occur in or on food commodities after the use of plant protection products according to Good Agricultural Practice (GAP). MRLs are intended primarily as a check that GAP is being followed and to assist international trade of products treated with pesticides. MRLs are not safety limits, and exposure to residues in excess of a MRL does not automatically imply a hazard to health.

The purpose of this work was to assess pesticide residues present in foodstuff of plant origin in relation to their MRLs and to calculate dietary intakes by Polish consumers according to methodology currently recommended by European Commission for the Member States of European Union.

MATERIAL AND METHODS

The Polish national monitoring programme for pesticide residues in commodities of plant origin is carried out by analytical laboratories of the Institute of Plant Protection. Following Commission Recommendation concerning a co-ordinated Community monitoring programme to ensure compliance with maximum levels of pesticide residues in and on certain products of plant origin (OJ L 2, 4.1.2002, p. 8), the laboratories regularly participate in proficiency testing exercises, including European Commission's Proficiency Tests and Food Analysis Performance Assessment Scheme FAPAS (Tab. 1). Accreditation processes are in preparatory phases and the EU Quality Control procedures for analytical laboratories are followed as far as possible.

Table 1. Participation of analytical laboratories of the Institute of Plant Protection in proficiency tests

No. of laboratories	Participation in proficiency tests	
	EU	FAPAS
6	Test II: 3 laboratories Test IV: 5 laboratories	No. 1908: 1 laboratory No. 1913: 2 laboratories No. 1818: 5 laboratories

Samples of fruits, vegetables, potatoes, and cereals taken by trained inspectors from the local Plant Protection Services were analysed unwashed and unpeeled by Multi-Residue Method (MRM) consisting of an extraction of residues with dichloromethane followed by a chromatographic separation and selective EC and NP detection (Ambrus et al. 1981; Luke et al. 1975; Sadło 1998). Along with this MRM method, spectrometric and Thin Layer Chromatographic (TLC) determinations of dithiocarbamate and carbendazim residues were carried out (Chmiel 1979; Murawska 1980).

RESULTS AND CONCLUSIONS

Results of national monitoring programme for pesticide residues

In 2001, a total of 2125 samples of fruits, vegetables, potatoes, and cereals were analysed for residues of the most commonly used pesticides in Poland. Detectable residues were found in 18% of the samples. The most important pesticide/commodity combinations were found for the following cases: endosulfan/black currant, dithiocarbamate/field tomato, tolylfluanid/raspberry or greenhouse tomato, and chlorothalonil/greenhouse tomato (Tab. 2).

The residues exceeded national or/and EU harmonised Maximum Residue Limits (MRLs) in 0.9% of the 2125 samples. Table 3 gives detailed information about such cases. In five samples, two pesticides in each sample exceeded the EC-MRL. This combination was endosulfan/carbendazim in black currant.

Out of 2125 samples of plant origin 68 contained residues of two or more pesticides in a single sample of greenhouse and field tomatoes, strawberries, black currant, apples and lettuce. Two samples of greenhouse tomatoes contained four different pesticides. Three different pesticides were found in six tomato and strawberry samples (Tab. 4).

Table 2. Pesticide residues found in commodities of plant origin analysed in the framework of Polish national monitoring programme

Commodity	No. of samples analysed	No. of samples with residues	%	No. of samples with residues>MRL	%
Black currants	42	24	57	15*	36
Strawberries	265	101	38	0	0
Tomatoes (f)	58	22	38	0	0
Tomatoes (g)	295	105	36	0	0
Raspberries	15	5	33	0	0
Peppers	34	8	24	0	0
Mushrooms	58	12	21	1**	2
Lettuce (g)	111	21	19	1**	1
Carrots	127	18	14	1**	1
Apples	287	40	14	0	0
Cucumbers (f)	121	11	9	0	0
Cucumbers (g)	202	13	6	0	0
Onions	66	4	6	0	0
Cherries	102	3	3	0	0
Cabbage	107	1	1	0	0
Potatoes	148	1	1	0	0
Lettuce (f)	3	0	0	0	0
Asparagus	7	0	0	0	0
Beetroot	8	0	0	0	0
Pekingese cabbage	2	0	0	0	0
Cauliflower	37	0	0	0	0
Parsley	1	0	0	0	0
Plums	12	0	0	0	0
Wheat	17	0	0	0	0
Total	2125	389	18	18	0.9

g – greenhouse crop; f – field crop; *exceedance of EC-MRL; ** exceedance of national and EC-MRL

Table 3. Residues exceeding the MRL (national or/and EC-MRL) levels

Commodity	Pesticide	R	Polish / EC-MRL	No. of samples with residues >MRL
Lettuce (g)	procymidone	9.70	5.0 / 5.0	1
Carrot	chlorpyrifos	0.24	0.1 / 0.1	1
Mushroom	carbendazim	1.00	1.0 / 1.0	1
Black currant	endosulfane	0.07–0.50	0.5 / 0.05	10
Black currant	endosulfane + carbendazim	0.07–0.50 0.20–0.40	0.5 / 0.05 1.0 / 0.1	5

Note: see table 2

Chronic intake

Calculation of consumer exposure from any one pesticide. The national estimated daily intake (NEDI) of a pesticide residue from foods of plant origin was calculated by using monitoring data from 2001. A pesticide/food commodity combination was included in this calculation only if the 90th percentile of the pesticide residues were found to be at or above the limit of determination (LOD). Otherwise the intake contribution from that pesticide/commodity combination was assumed to be zero.

Table 4. Samples with residues of more than one pesticide

Commodity	No. of samples analysed	2	3	4	Samples with multiple residues	
					No.	%
Tomatoes (g)	295	23	2	2	27	32
Strawberries	265	19	4	0	23	27
Blackcurrants	41	8	1	0	9	11
Apples	287	3	0	0	3	4
Tomatoes (f)	58	3	0	0	3	4
Lettuce (g)	111	3	0	0	3	4

Note: see table 2

The average consumption data for the commodities were derived from Kubiak et al. (2000) (Tab. 5). However, in order to include a higher percentile of the consumers, the 90th percentile consumption may also be used in calculation by multiplying the average consumption figures with a factor around 2.

The national estimated daily intake (NEDI) of any one pesticide has been calculated using the following formula:

$$Intake = \frac{F}{bw} \times \frac{R}{ADI} \times 100\%,$$

where:

Intake = from any one pesticide [% of ADI],

F = the average food consumption of the relevant commodity [kg/day],

R = the 90th percentile of the residue level found in composite samples [mg/kg],

bw = the average bodyweight of an adult [60 kg]

ADI = the acceptable daily intake [mg/kg b.w/day]. The ADI is the estimate of the amount of a substance in food, expressed on a body-weight basis, that can be ingested daily over a lifetime without appreciable health risk to the consumer. The ADI is based on the no observed adverse effect level (NOAEL) and, therefore, reflects chronic toxicity.

Table 5. Average consumption figures for fresh fruit, vegetables and potatoes included in Polish national monitoring programme of pesticide residues (Kubiak et al. 2000)

Commodity	Consumption		
	Total	Included in national monitoring	
	kg/day	kg/day	%
Fruit	0.149	0.072	48
– Polish Origin	0.085	0.072	85
– Imported	0.064	0.000	0
Vegetables	0.351	0.298	85
– Polish Origin	0.342	0.298	87
– Imported	0.018	0.000	0
Potatoes (Polish Origin)	0.340	0.340	100
Total	0.841	0.710	84

Calculation of consumer exposure from any one commodity. Assuming additive impact of different pesticides on a consumer of fruit and vegetables, the national estimated daily intakes (NEDI) from any one commodity have been calculated using the following formula:

$$Intake = \frac{F_n}{bw} \times \sum_{i=1}^{n} \frac{R_i}{ADI_i} \times 100\%,$$

where:

Intake = from any one commodity [% of ADI]; the long-term consumer exposure was expressed by their 90th percentile level.

F_n = the average food consumption of the relevant commodity [kg/day],

R_i = residue level of an "*i*" pesticide of the "n" different ones found in a composite sample [mg/kg].

Results of the assessment of the national estimated long-term intake. The estimated short-term intake has been calculated for 7 different pesticides in 9 commodities due to assumptions made about the residue levels. The intake of endosulfan reached the highest level compared with ADI and exceeded 1% of the ADI. The intakes of the other pesticides were well below 1% of the ADI (Tab. 6). Table 7 presents the intake of pesticide residues for commodities included in the Polish national monitoring programme. The highest intake was found to be in the case of black currant. The total intake of pesticide residues from 10 commodities reached the total level of 5.3% of ADI.

Table 6. The estimated chronic intake of certain pesticides based on their 90th percentile residues found in composite samples in 2001

Pesticide	Commodity	R	F	Intake	
				[mg/kg bw]	[% of ADI]
Endosulfan	Black currants	0.15	0.044	0.018	1.826
Linuron	Carrots	0.02	0.050	0.008	0.826
Carbendazim	Black currants	0.10	0.044	0.007	0.731
EBDC	Tomatoes (f)	0.30	0.042	0.007	0.708
Chlorothalonil	Tomatoes (f)	0.10	0.042	0.002	0.236
Chlorothalonil	Tomatoes (g)	0.20	0.016	0.002	0.183
Carbendazim	Mushrooms	0.30	0.003	0.002	0.164
Procymidone	Tomatoes (g)	0.10	0.016	0.000	0.027
Tolylfluanid	Strawberries	0.26	0.003	0.000	0.006
Procymidone	Raspberries	0.06	0.001	0.000	0.003
Chlorothalonil	Peppers	0.01	0.003	0.000	0.002
Tolylfluanid	Raspberries	0.19	0.001	0.000	0.001

Note: see table 2

Short-term intake

Calculation of short-term consumer exposure from any one pesticide. The acute dietary intake (short-term exposure) has to be considered for those pesticides that are classified as acutely toxic. Acute Reference Doses (ARfD), used for such calculations, is

Table 7. The estimated 90th percentile of chronic pesticide intake from any one compound in 2001

Commodity	Intake of pesticide residue [% of ADI]
Black currants	3.848
Carrots	0.826
Tomatoes (f)	0.708
Tomatoes (g)	0.274
Mushrooms	0.164
Apples	0.044
Lettuce (g)	0.037
Strawberries	0.023
Raspberries	0.004
Peppers	0.002
Total	5.330

Note: see table 2

the estimate of the amount of a substance in food, expressed on a body-weight basis, that can be ingested over a short period of time, usually during one meal or one day, without appreciable health risk to the consumer. It therefore reflects the acute toxicity. At present, the ARfDs have been fixed for certain pesticides. The calculations of short-term exposures have been estimated in relation to the ADIs taken from "The Pesticide Manual" 10th edition (Tomlin 1994). The results obtained can be easily recalculated using actual ARfD values.

The short-term intake of pesticides has been calculated for eleven commodities (Tab. 7). The consumption figures used for calculation of the national estimated intake are based on the 97.5th percentile consumption of consumes only, which reflects the largest portion consumed during one meal or during one day. The United Kingdom consumption figures have been used. Following the Commission recommendations the residue level is taken from monitoring data without any correction for reduction in the level after preparation of the food. The level is multiplied with a variability factor defined as the quotient between the maximum and the mean residue of individual units in a sample.

The national estimated short-term intake (NESTI) was calculated using the following formulae:

$$Intake = \frac{F}{bw} \times \frac{HR \times V}{ADI} \times 100\%,$$

where:

Intake = from any one pesticide/commodity combination [% of ADI],

HR = the highest residue found in composite samples of the crop [mg/kg],

F = 97.5th percentile of the food consumption [kg/day],

V = a default variability/homogeneity factor [V=1.00 for cherries and black currant while for the other crops V=3.44].

Results of the assessment of the national estimated short-term intake. Toddlers (children weighing 14.5 kg), with low body weight relative to their consumption, compose a risk group of approaching the acute reference dose when consuming large amount of fruits or vegetables if these products contain quite high levels of pesticide residues. Table 8 shows that the national estimated short-term intake (calculated according to the European Commission recommendations) for toddlers exceeded the ADI level for linuron in carrots, flusilazole and diazinon in apples, tolylfluanid and ethylenedithiocarbamates in strawberry, carbendazim in mushroom and chlorothalonil in greenhouse. For majority of cases, including exceedances of national and

Table 8. The estimation short-term intake of any one pesticide based on its highest residue found in composite samples in 2001

Pesticide	Commodity	R	ADI	Consumption		Intake			
				Adult	Toddler	Adult	Toddler	Adult	Toddler
				[kg]		[mg/kg bw]		[% of ADI]	
Procymidone	Lettuce (g)	9.70*	0.100	0.093	0.025	0.044	0.058	44*	58*
EBDC		2.60	0.030			0.012	0.015	40	51
Iprodione		3.00	0.200			0.014	0.003	7	9
Chlorotalhonil		0.30	0.030			0.001	0.002	5	6
Pirimicarb		0.06	0.020			0.000	0.000	1	2
Dichlofluanid		0.50	0.300			0.002	0.003	1	1
Carbendazim	Cucumber (g)	0.30	0.010	0.084	0.072	0.001	0.005	12	51
EBDC		0.20	0.030			0.001	0.003	3	11
Chlorothalonil		0.10	0.030			0.000	0.002	1	6
Procymidone		0.20	0.100			0.001	0.003	1	3
Carbendazim	Mushroom	1.10*	0.010	0.153	0.046	0.008	0.012	83*	120*
Procymidon	Pepper	0.20	0.100	0.09	0.05	0.001	0.002	1	2
EBDC	Cucumber (f)	0.30	0.030	0.084	0.072	0.001	0.005	4	17
Chlorothalonil		0.20	0.030			0.001	0.003	3	11
Metalaxyl		0.08	0.030			0.000	0.001	1	5
Linuron	Carrot	0.19	0.002	0.226	0.104	0.002	0.005	105	234
Chlorpyrifos		0.24*	0.010			0.003	0.006	27*	59*
DDT		0.02	0.020			0.000	0.000	1	2
Chlorothalonil	Tomato (g)	1.80	0.030	0.157	0.093	0.014	0.040	46	132
Procymidone		4.00	0.100			0.031	0.088	31	88
EBDC		0.50	0.030			0.004	0.011	13	37
Iprodione		1.30	0.200			0.010	0.029	5	14
Pyrimethanil		0.48	0.200			0.004	0.011	2	5
Dichlofluanid		0.60	0.300			0.005	0.013	2	4
EBDC	Tomato (f)	1.00	0.030	0.157	0.093	0.008	0.022	26	74
Chlorothalonil		0.20	0.030			0.002	0.004	5	15
Tolylfluanid	Strawberry	5.74	0.100	0.203	0.111	0.057	0.151	57	151
EBDC		1.40	0.030			0.014	0.037	46	123
Procymidone		0.50	0.100			0.005	0.013	5	13
Dichlofluanid		0.30	0.300			0.003	0.008	1	3
Pyrimethanil		0.21	0.200			0.002	0.006	1	3
Endosulfan	Black currant	0.50*	0.006	0.036	0.016	0.000	0.001	4*	9*
Flusilazole		0.08	0.001			0.000	0.000	4	9
Carbendazim		0.40*	0.010			0.000	0.000	2	4*
Fenitrothion		0.20	0.005			0.000	0.000	2	4
EBDC		0.20	0.030			0.000	0.000	0	1
Triadimefon		0.06	0.030			0.000	0.000	0	0
Flusilazole	Apple	0.05	0.001	0.308	0.199	0.001	0.002	76	236
Diazinon		0.07	0.002			0.001	0.003	53	165
Carbendazim		0.20	0.010			0.003	0.009	30	94
Captan		0.67	0.100			0.010	0.032	10	32
Pirimicarb		0.10	0.020			0.002	0.005	8	24
Tolylfluanid		0.48	0.100			0.007	0.023	7	23
Pirimethanil		0.32	0.200			0.005	0.015	2	8
EBDC	Cherry	0.30	0.030	0.247	0.04	0.001	0.001	4	3
Captan		0.60	0.100			0.002	0.002	2	2

* Intakes estimated for exceedances of national or/and EC-MRLs. Note: see table 2

EC-MRLs estimations, short-term intakes were well below the levels of ADIs. Taking into account that the ARfD values now available are 2-10-fold (on average 5-fold) higher than those fixed for the ADI (Pesticide Safety Directorate; SANCO/397/01-Final), the average safety factor of 5 for an adult consumer still existed.

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POLISH SUMMARY

POBIERANIE POZOSTAŁOŚCI ŚRODKÓW OCHRONY ROŚLIN PRZEZ POLSKIEGO KONSUMENTA W 2001 ROKU

W 2001 roku łącznie przebadano 2125 próbek pochodzenia roślinnego. Pozostałości środków ochrony roślin znaleziono w 18% próbek a tylko w 18 z nich przekroczyły najwyższy dopuszczalny poziomu (NDP).

Na podstawie uzyskanych wyników obliczono następnie dzienne pobranie pozostałości przez polskiego konsumenta, które tylko w przypadku endosulfanu w czarnej porzeczce przekroczyło 1% Acceptable Daily Intake (ADI).

Zgodnie z rekomendacją Komisji Europejskiej dla 19 różnych substancji znalezionych w 12 produktach pochodzenia roślinnego oszacowano również jednorazowe ich pobranie, które dla karbendazymu w pieczarkach, linuronu w marchwi, chlorotalonilu w pomidorach szklarniowych, tolilfluanidu i ditiokarbaminianów w truskawkach oraz flusilazolu i diazynonu w jabłkach nieznacznie przekroczyło poziom ADI. Ustalone do chwili obecne dopuszczalne jednorazowe pobranie (Acute Reference Dose; ARfD), kształtujące się średnio na poziomie 5-krotnie wyższym od ADI, dowodzą, że nawet dla dzieci w wieku szkolnym istniał jeszcze znaczny margines bezpieczeństwa.