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ОЦІНКА РИЗИКУ ДЛЯ НАСЕЛЕННЯ ПРИ СПОЖИВАННІ ПРОДУКЦІЇ, ВИРОЩЕНОЇ З ЗАСТОСУВАННЯМ ФУНГІЦИДІВ НА ОСНОВІ ДІЮЧИХ РЕЧОВИН КЛАСУ ТРИАЗОЛІВ

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HYGIENIC ASSESSMENT OF THE POPULATION RISK AFTER CONSUMPTION OF AGRICULTURAL PRODUCTS GROWN WITH THE APPLICATION OF TRIAZOLE CLASS-BASED FUNGICIDES

A

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According to the UN forecast, the Earth's population will increase by 2.5 billion by 2050 [1, 2]. Thus, the question of increasing the productivity of agricultural production becomes urgent. In today's realities, it isn't easy to find that the world has backed away from the pesticide's application. The variety and volume of their use increase every year. The advantages of using pesticides are improved agriculture efficiency, protecting of plant crops from diseases and pests, and reducing the price of products. However, there are also disadvantages – this is their negative impact on the environment and human health, which has a cumulative effect [3, 4].

Various key methods at the different stages of registration and monitoring studies of pesticides have been used widely in the countries of the EU and

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Проблема отруєння пестицидами особливо гостро постає серед країн, основним джерелом доходу яких є сільське господарство (США, Індія, Китай, Бразилія, Мексика, Аргентина та ін.). Щорічно повідомляється про близько 3 млн. випадків навмисного та ненавмисного отруєння пестицидами у світі, що призводить до загибелі понад 250000 людей. В Україні за 2020 рік було зареєстровано понад 250 препаратів на основі діючих речовин класу триазолів.

Мета. Оцінка ризику для населення при споживанні продукції, вирощеної з застосуванням фунгіцидів на основі діючих речовин класу триазолів.

Матеріали і методи. Нами проаналізовано параметри стійкості фунгіцидів класу триазолів (дифеноконазол, протіоконазол, ципроконазол і метконазол) та стробілу-

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the USA [5, 6]. One of the working directions of the Bogomolets National Medical University Institute of Hygiene and Ecology (IHE NMU) is the development of hygienic assessment risk models for the population after the consumption of contaminated food products [7, 8]. This is a critical direction in working with pesticides because the population's health depends on it.

According to the World Health Organization (WHO), among agricultural workers, there are known cases of adverse effects of hazardous substances at various stages of working with them: during preparation, dilution, mixing, and application of pesticides [9, 10].

The problem of pesticide poisoning is particularly acute among countries whose primary source of income is agriculture (India, China, Brazil, Mexico, Argentina etc.). About 3 million cases of intentional and unintentional pesticide poisoning are reported in the world annually, resulting in the death of more than 250000 people [11, 12].

Long-term research conducted by IHE of NMU showed that inhalation poi-

рину (азоксистробіну) та їхніх діючих речовин у різних сільськогосподарських культурах. Для визначення вмісту діючих речовин у сільськогосподарських культурах ми застосовували сучасні методи високоефективної рідинної та газової хроматографії.

Для інтегральної оцінки ризику використовували такі критерії: допустима добова доза, період напівруйнування речовин у рослинах та середньодобове споживання продуктів.

Результати роботи. Оцінка ризику для населення, розрахована за методикою, показала, що величини ризику сполук класу триазолів склали 0,0002-0,2239, стробілуринів – 0,0002-0,0016, що не перевищує допустимий рівень (менше 1).

Обчислення, здійснені за методикою, свідчать, що усі досліджувані сполуки класу триазолів належать до помірно небезпечних при споживанні оброблених ріпаку, сої, соняшнику, цукрових буряків, та небезпечні при споживанні зернових культур, стробілуринів – малонебезпечні при споживанні більшості досліджуваних культур. Відмінності у класах небезпечності зумовлені передусім різницею поведінки у різних культурах та тривалістю вегетаційного періоду культур.

Ключові слова: фунгіциди, триазоли, стробілурини, оцінка ризику, харчові продукти.



БІОЛОГІЧНІ ФАКТОРИ ДОВКІЛЛЯ

soning with pesticides of the triazole class is unlikely if the regulations for their safe usage are followed [13]. However, there are other ways of getting these substances into the body, including food.

In Ukraine, by 2020, more than 250 pesticide preparations containing active sub-

stances of the triazole class and up to 100 fungicides for seed treatment were registered [14].

Aim of the work. Hygienic assessment of the population risk after consumption of agricultural products grown with the application of triazole class-based fungicides.

Table 1

General characteristics of fungicides and conditions of their usage

Name of fungicide	Active substance	Product	Application rates l/kg/ha, times
Camzol Turbo, SC*	Cyproconazole, 80 g/l ⁺ Metconazole, 60 g/l	wheat, rapeseed, soybean, sunflower, winter and spring barley	0,7 l/ha, twice
Split Duo, SC*	Difenoconazole, 125 g/l ⁺ Azoxystrobin, 125 g/l	rapeseed, soybean, sunflower, sugar beet	1,0 l/ha, twice
Meganic, EC**	Difenoconazole, 125 g/l ⁺ Prothioconazole, 175 g/l	wheat, barley, rapeseed, sunflower, sugar beets	1,0 l/ha, twice
Notes: * - Emulsifiable concentrate, ** - Suspension concentrate (flowing)			

Materials and methods.

We analyzed the resistance parameters of triazole fungicides (difenoconazole, prothioconazole, cyproconazole, and metconazole) and strobilurin (azoxystrobin) in various crops (wheat, barley, rapeseed, soybean, sunflower, sugar beet).

Information on the general characteristics of fungicides and the conditions of their application is given in table 1.

To determine the content of active substances in crops, we used modern methods of high-performance liquid chromatography (HPLC) and gas chromatography (GC) (table 2).

To assess the effect of the studied fungicides on the agrocenosis objects, we calculated the degradation half-life (50) of substances in plants. Using the calculation

method of mathematical modelling and reproduction of the degradation processes of pesticides according to actual data, we were able to predict their behaviour [15].

The persistence in plants was evaluated according to the Ukrainian classification of pesticides by hazard degree of SSanN&R 8.8.1.002-98 [16], according to which the own research results were evaluated. The classification divides substances according to their stability in plants into four classes: 1 – highly persistent (50 more than 30 days), 2 – persistent (15-30 days), 3 – moderately persistent (5-14 days), 4 – low-persistent (less than 5 days).

The degradation indicator rates of active substances in the studied crops (decay rate constant (k), half-life periods (50) were calculated using the first-order reaction equation [15].

$$k = \frac{2,303}{t} \lg \frac{C_0}{C_t} \quad DT_{50} = \frac{\ln 2}{k}$$

where C_0 – is the initial content of residual amounts of pesticides (1 hour after pesticide treatment); C_t – is the

content of residual quantities of pesticides after a certain period of time (t) after pesticide treatment.

For the integral assessment of the hazard indicators, while using products, the four-gradation scale proposed by the specialists of IHE NMU was used, the indicators of the allowable daily intake (ADI), the half-life periods (50) in plants and the average daily consumption of the product were evaluated [21, 22]. The basis of the integral assessment is assigning points to each criterion (from 1 to 4 points). After adding all the obtained points, the integral indicator of hazard while using products (IIH) is evaluated as follows: with an IIH value of 3-5 points – substances are slightly hazardous for humans (class 4), 6-8 – moderately hazardous (class 3), 9-11 – hazardous (class 2), >11 – extremely hazardous (class 1).

Statistical processing of the obtained results was carried out using the SPSS Statistics Base v. 22 and MS Excel statistical software package.

Research results and discussion.

The results of in-field studies showed that after the application of Camzlo Turbo, SC, Split Duo, SC, Meganic, EC, the initial concentrations of cyproconazole, metconazole, difenoconazole, prothioconazole and azoxystrobin in the analyzed crops (wheat, barley, rapeseed, soybean, sunflower, sugar beet) were 0.61-5.9 mg/kg; 0.18-4.7 mg/kg; 0.51-1.7 mg/kg; 0.22-0.82 mg/kg; 0.2-2.1 mg/kg, respectively. In the later periods of the study, the residual amounts of the studied active substances gradually decreased in the studied cultures, and their content was not detected in the above-mentioned plant cultures at harvest.

Pesticide residues at harvest were compared with the national maximum allowable levels (MALs) [17], and approved in the EU MRLs [18], and it was established that there is no exceedance of the norms established in Ukraine and the EU.

The results obtained during field studies and using the mathematical modelling method made it possible to calculate the stability parameters of the studied fungicides of the triazole class (cyproconazole, metconazole, difenoconazole, prothioconazole) and strobilurin (azoxystrobin) in various crops (degradation rate constant (K) and half-life period (50) (table 3).

Mathematical processing of the obtained results showed that in the soil and climatic conditions of Ukraine, the process of decomposition of the studied compounds of triazoles and strobilurins class in crops obeyed an exponential dependence, which can be observed on the corresponding graphs (figs. 1-3). The values of the coefficient of determination (R^2) were in the range of 0.5548-0.9983, which indicates a significant relationship between the studied variables and the reliability of the selected model when simulating the results of field studies of pesticides [15].

Table 2

Analysis Methods of the studied fungicides

Active substance	Product	Method [№]	Limit of quantification, mg/kg	Limit of detection, mg/kg
Triazoles				
Cyproconazole	wheat, barley	GC [938-2009]	0,05	0,02
	rapeseed	GC [938-2009]	0,2	0,06
	soybean	GC [1013-2010]	0,2	0,06
	sunflower	GC [1062-2011]	0,2	0,06
Metconazole	wheat, barley	GC [1197-2012]	0,1	0,03
	rapeseed	GC [676-2006]	0,2	0,06
	soybean	GC [pending]	0,1	0,03
	sunflower	GC [pending]	0,05	0,02
Difenoconazole	rapeseed	GC 994-2010]	0,1	0,03
	soybean	GC [1468-2018]	0,05	0,02
	sunflower	GC [1439-2015]	0,05	0,02
	sugar beets	GC [6147-91]	0,1	0,03
	wheat, barley	GC [69-97]	0,05	0,02
Prothioconazole (Prothioconazole destio)	wheat, barley	GC [664-2006]	0,1	0,03
	rapeseed	GC [931-2009]	0,1	0,03
	sunflower	GC [1094-2011]	0,1	0,03
	sugar beets	GC [1449-2015]	0,3	0,1
Strobilurins				
Azoxystrobin	rapeseed	HPLC [873-2009]	0,2	0,06
	soybean	HPLC [989-2010]	0,2	0,06
	sunflower	HPLC [828-2008]	0,2	0,06
	sugar beets	HPLC [829-2008]	0,1	0,03

HYGIENIC ASSESSMENT OF THE POPULATION RISK AFTER CONSUMPTION OF AGRICULTURAL PRODUCTS GROWN WITH THE APPLICATION OF TRIAZOLE CLASS-BASED FUNGICIDES

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The problem of pesticide poisoning is particularly acute among countries whose primary source of income is agriculture (India, China, Brazil, Mexico, Argentina etc.).

About 3 million cases of intentional and unintentional pesticide poisoning are reported in the world annually, resulting in the death of more than 250000 people. In Ukraine, by 2020, more than 250 pesticide preparations containing active substances of the triazole class.

Aim of the work: Hygienic assessment of the population risk after consumption of agricultural products grown with the application of triazole class-based fungicides.

Materials and methods: We analyzed the resistance parameters of triazole fungicides (difenoconazole, prothioconazole, cyproconazole, and metconazole) and strobilurin (azoxystrobin) in various crops. To determine

the content of active substances in crops, we used modern methods of high-performance liquid and gas chromatography.

For the integral assessment of the hazard indicators was used, the indicators of the allowable daily intake, the half-life periods in plants and the average daily consumption of the product were evaluated.

Research results: The risk assessment for the population, calculated according to the methodology showed that the risk values of triazoles were 0.0002-0.2239, strobilurins – 0.0002-0.0016, which does not exceed the acceptable (less than 1).

Calculations carried out according to the method show that all studied compounds of triazoles class belong to the moderately hazardous when consuming treated products of rapeseed, soybean, sunflower, sugar beet and hazardous when consuming cereals; strobilurin – low hazard when consumed the most of the studied cultures.

Differences in hazard classes are primarily due to differences in the behaviour of pesticides in different crops and the length of the vegetative season.

Keywords: fungicides, triazole, strobilurin, risk assessment, food products.

Statistical processing of degradation rate indicators in investigated triazoles and strobilurin (table 3) in wheat, barley, soybeans, rapeseed, sunflower and sugar beets allowed us to calculate the average values of 50 of fungicides of these classes in crops. The obtained results showed that 50 for the drug Camzol Turbo, SC (cyproconazole – (15.4±2.4) days, metconazole – (12.2±0.8) days). Split Duo, SC (difenoconazole – (14.8±2.3) days, azoxystrobin – (19.2±4.8) days). Meganic, EC (difenoconazole – (14.6±5.1) days, prothioconazole – (16.7±3.4) days).

According to SSanN&R 8.8.1.002-98, all studied substances of the triazoles class belong to class II, except metconazole – class III, strobilurin – azoxystrobin – to class II [15].

The risk assessment for the population, calculated according to the methodology [7] showed that the risk values of triazoles were 0.0002-

0.2239, strobilurins – 0.0002-0.0016, which does not exceed the acceptable (less than 1) (table 4).

Calculations carried out according to the method [8] show that all studied compounds of triazoles class (cyproconazole, metconazole, difenoconazole and prothioconazole) belong to the 3-rd class of hazard in terms of IHH – moderately hazardous when consuming treated products of rapeseed, soybean, sunflower, sugar beet and to 2-nd class of hazard – hazardous when consuming cereals; azoxystrobin (active substance of the strobilurin class) – pertains to the 4-th class of hazard – low hazard when consumed the most of the studied cultures (table 4). Differences in hazard classes are primarily due to differences in the behaviour of pesticides in different crops and the length of the vegetative season.

Scientific studies carried out in China showed that triazoles are quickly absorbed

into agricultural products and are pretty stable, which caused a significant increase in their residual amounts in final consumer products [20]. Research that was previously conducted in Ukraine also indicates that the studied class of pesticides is persistent in growing crops and requires control [21].

For comparison, we analyzed half-life values (50) of substances of the triazole class in plants grown in the soil and climatic conditions of EU countries. So, as an example, for the compounds we studied, the indicators were: cyproconazole – 3.5-16.0, metconazole – 26.6-368.5, difenoconazole – 0.9-31.9, prothioconazole – 0.49-1.4 days. The range of values among the described fungicides was from 0.49 to 368.5 days. The results we got correlated with similar data obtained in other EU countries. In some cases, the degradation occurred faster [18].

The obtained results indicate that after the usage of

fungicides containing active substances of the triazole class, it is necessary to control the content of the specified class in grain crops.

Conclusions

1. We established that, according to SSanN&R 8.8.1.002-98, in terms of per-

sistency in vegetative crops, all studied substances of triazole class pertain to class II (persistent), except for metconazole – class III (moderately persistent), strobilurin (azoxystrobin) – class II (persistent compounds).

2. It has been proven that

compounds of triazole class (cyproconazole, metconazole, difenoconazole and prothioconazole) pertain to the 3-rd class of hazard in terms of the value of the integral indicator of hazard when consuming products (IIH) – moderately hazardous when

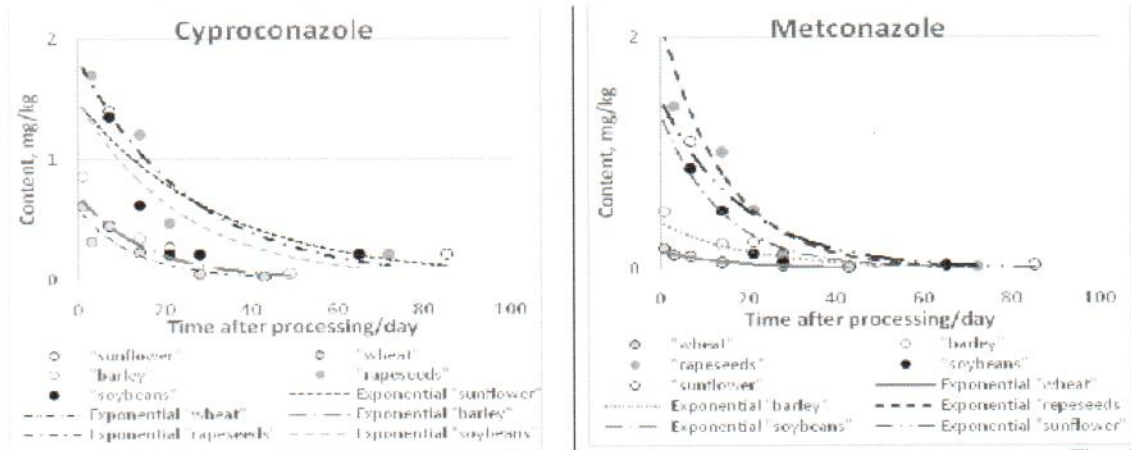


Fig. 1

Dynamics of the content of cyproconazole and metconazole in agricultural products (the active substances of Camzol Turbo, CS)

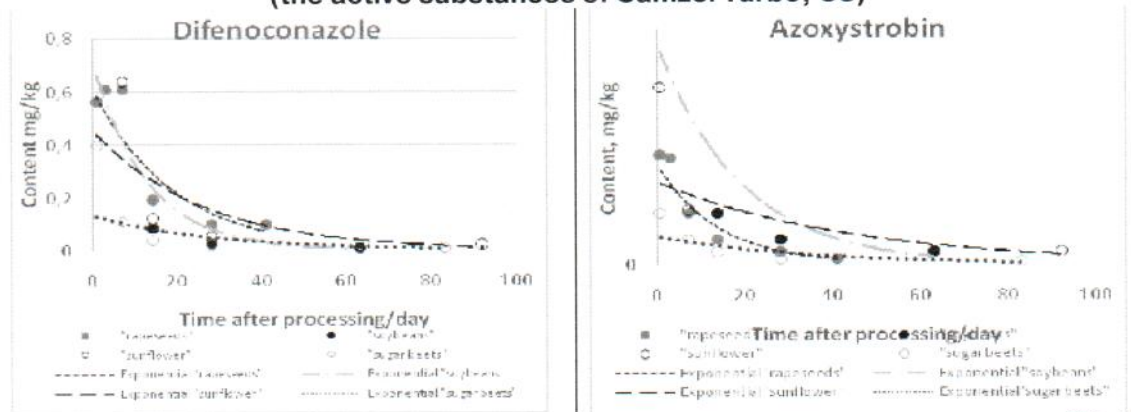


Fig. 2

Dynamics of the content of difenoconazole and azoxystrobin in agricultural products (the active substances of Split Duo, CS)

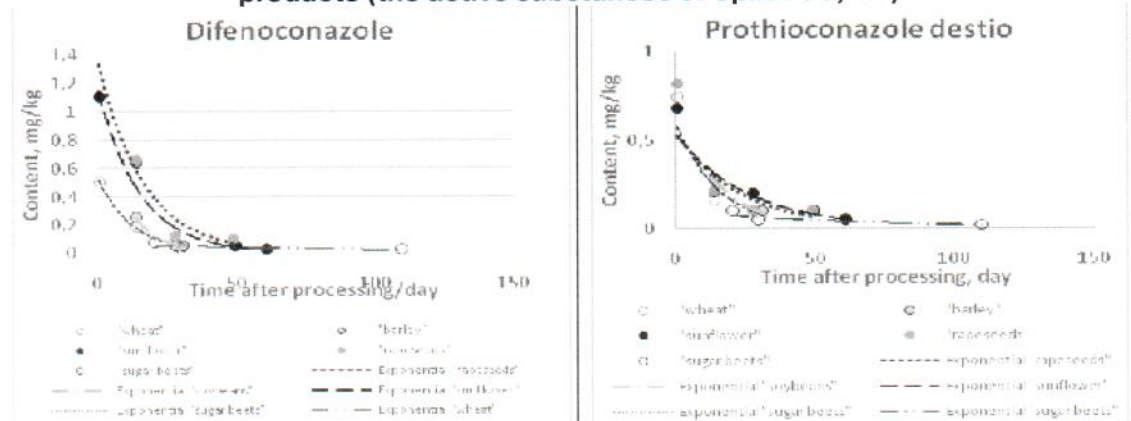


Fig. 3

Dynamics of the content of difenoconazole and prothioconazole-destio in agricultural products (the active substances of Meganic, EC)

consuming rapeseed, soybean, sunflower, sugar beet processing products and to 2-nd class of hazard – hazardous when consuming cereals; azoxystrobin (active substance of strobilurin class) – pertains to the 4-th class of hazard – low hazardous when consumed with the most of the studied crops.

3. Estimated risk of adverse effects of triazole and strobilurin fungicides on human health when consuming agricultural products grown during their application was 0.0002-0.2239 for triazole, and 0.0002-0.0016 for strobilurins, respectively, which does not exceed the acceptable level (less than 1).

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Table 3

Studies results of active substances content of triazoles and strobilurins in crops and their speed of degradation

Name of fungicide	Active ingredient	Product	Kinetic equation	R ²	K, day	τ ₅₀ , day	M±m τ ₅₀ , day
Carazol Turbo, SC	Cyproconazole *	Wheat	$y = 0,5783e^{-0,076x}$	0,8568	0,076	9,0562	15,4±2,4
		Rape seeds	$y = 0,6915e^{-0,063x}$	0,9833	0,039	17,894	
		Soy beans	$y = 0,6915e^{-0,063x}$	0,9833	0,042	16,336	
		Barley	$y = 0,6915e^{-0,063x}$	0,9833	0,063	11,004	
	Metconazole *	Wheat	$y = 0,1557e^{-0,065x}$	0,9534	0,049	13,972	12,2±0,8
		Rape seeds	$y = 2,1663e^{-0,069x}$	0,8117	0,069	10,007	
		Soy	$y = 1,3529e^{-0,073x}$	0,8186	0,066	10,512	
		Barley	$y = 0,3967e^{-0,05x}$	0,9671	0,05	13,871	
Split Duo, CS	Difenoconazole *	Rape seeds	$y = 0,6103e^{-0,052x}$	0,851	0,052	13,288	14,8±2,3
		Soy beans	$y = 0,7052e^{-0,078x}$	0,9062	0,078	8,8651	
		Sugar beets	$y = 0,1334e^{-0,036x}$	0,6199	0,036	19,137	
		Sunflower	$y = 0,4563e^{-0,038x}$	0,7659	0,038	17,935	
	Azoxystrobin *	Rape seeds	$y = 0,3865e^{-0,071x}$	0,9369	0,071	9,7322	19,2±4,8
		Soy beans	$y = 0,8493e^{-0,052x}$	0,839	0,052	13,246	
		Sugar beets	$y = 0,1094e^{-0,03x}$	0,7465	0,03	23,163	
		Sunflower	$y = 0,3215e^{-0,022x}$	0,569	0,022	30,763	
Meganic, EC	Difenoconazole *	Wheat	$y = 0,5559e^{-0,081x}$	0,9983	0,081	8,5226	14,6±5,1
		Barley	$y = 0,9983e^{-0,101x}$	0,9952	0,101	6,8319	
		Rape seeds	$y = 0,6915e^{-0,063x}$	0,9833	0,062	11,19	
		Sunflower	$y = 1,3713e^{-0,058x}$	0,9463	0,058	11,898	
	Prothioconazole *	Sugar beets	$y = 0,0726e^{-0,012x}$	0,7757	0,020	34,37	16,7±3,4
		Wheat	$y = 0,6104e^{-0,065x}$	0,9451	0,065	10,691	
		Barley	$y = 0,7248e^{-0,067x}$	0,9909	0,067	10,327	
		Rape seeds	$y = 0,3967e^{-0,05x}$	0,9671	0,042	16,389	
		Sunflower	$y = 0,3402e^{-0,04x}$	0,5548	0,04	17,147	
		Sugar beets	$y = 0,0996e^{-0,013x}$	0,7143	0,024	29,23	

Notes: * – differences in 50 values are not significant according to the Student's test at $p > 0.05$ when pesticides are used on different crops; M is the average value; m is the error of the arithmetic mean.

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Table 4

Risk assessment of adverse effects on human health when consuming agricultural products treated by studied triazole and strobilurin-containing fungicides

Name of fungicide	Active ingredient	ADI, mg/kg	ADIPP, mg/day	Product	MAL (JA) ¹ , mg/kg	MRL (EU) ² , mg/kg	Average product consumption per day, g/day [19]	Risks, a.u.	IID		
									value	class	
Camzol Turbo, SC	Cyproconazole	0,002	0,084	Wheat, barley	0,05	0,1	313,4	0,0746	0,1062	2+4+4=10	2
				Rape seeds	0,2	0,4	19,5*	0,0139		3+4+1=8	3
				Soy beans	0,2	0,07	5,2	0,0037		3+4+1=8	3
				Sunflower	0,2	0,05	19,5*	0,0139		3+4+1=8	3
	Metconazole	0,001	0,042	Wheat, barley	0,1	0,15	313,4	0,2239	0,2647	2+4+4=10	2
				Rape seeds	0,2	0,2	19,5*	0,0279		2+4+1=7	3
				Soy beans	0,05	0,05	5,2	0,0037		2+4+1=7	3
				Sunflower	0,05	0,7	19,5*	0,0093		2+4+1=7	3
Split Duo, CS	Difenoconazole	0,002	0,084	Rape seeds	0,2	0,5	19,5*	0,0070	0,0363	2+4+1=7	3
				Soy beans	0,05	0,1	5,2	0,0012		2+4+1=7	3
				Sugar beets	0,1	0,2	65,8**	0,0235		3+4+1=8	3
				Sunflower	0,05	0,05	19,5*	0,0046		3+4+1=8	3
	Azoxystrobin	0,03	1,260	Rape seeds	0,2	0,5	19,5*	0,0009	0,0037	2+1+1=4	4
				Soy beans	0,2	0,5	5,2	0,0002		2+1+1=4	4
				Sugar beets	0,1	5	65,8**	0,0016		3+1+1=5	4
				Sunflower	0,2	0,5	19,5*	0,0009		4+1+1=6	3
Meganic, EC	Difenoconazole	0,002	0,084	Wheat, barley	0,05	0,1	313,4	0,0746	0,1097	2+4+4=10	2
				Rape seeds	0,1	0,5	19,5*	0,0070		2+4+1=7	3
				Sunflower	0,05	0,05	19,5*	0,0046		2+4+1=7	3
				Sugar beets	0,1	0,2	65,8**	0,0235		4+3+1=8	3
	Prothioconazole	0,001	0,042	Wheat, barley	0,1	0,1	313,4	0,2239	0,4084	2+4+4=10	2
				Rape seeds	0,1	0,15	19,5*	0,0139		3+4+1=8	3
				Sunflower	0,1	0,2	19,5*	0,0139		3+4+1=8	3
				Sugar beets	0,1	0,01	65,8**	0,1567		4+3+1=8	3

Notes: ADI – allowable daily intake, ADIPP – allowable daily intake of pesticide with particular products, MAL – maximum allowable levels, MRL – maximum residue levels, IIH – integrated indicator of hazard when using products.

* – the possible intake of substances is calculated by oil,

** – the possible intake of substances – calculation based on sugar; a.u. – arbitrary units.

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Table 4
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IID	
value	class
4+4=10	2
4+1=8	3
4+1=8	3
4+1=8	3
4+4=10	2
4+1=7	3
4+1=7	3
4+1=7	3
4+1=7	3
4+1=8	3
4+1=8	3
4+1=4	4
4+1=4	4
4+1=5	4
4+1=6	3
4+4=10	2
4+1=7	3
4+1=7	3
4+3=8	3
4+4=10	2
4+1=8	3
4+1=8	3
4+3=8	3

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