ПИТАННЯ ГІГІЄНИ ТА ЕКОЛОГІЇ

UDC 504.5:632.95.024:614.77]:634.7/635.6]]-043.2-047.77
DOI https://doi.org/10.32782/2226-2008-2025-1-14

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ASSESSING THE POTENTIAL HEALTH RISKS OF PESTICIDES USED FOR BERRY AND MELON CROPS IN CONTAMINATION OF GROUND AND SURFACE WATERS

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The aim: to assess the health risks of pesticides used for the berry and melon crops in case of the ground and surface waters contamination.

Materials and methods: natural hygienic experiments, during which chemical-analytical (high-performance liquid chromatography (HPLC), gas-liquid chromatography (GLC), mathematical and statistical methods were used.

Results: the research showed that according to the integral groundwater hazard contamination index (IHCI) fludioxonil, pyraclostrobin, and pendimethalin are slightly hazardous for humans (class 4); phenhexamide, boscalid, fluxapiroxad, azoxystrobin, spirodiclofen, and abamectin are moderately hazardous (class 3); cyprodinil, difenoconazole, metalaxyl-M, and glyphosate are hazardous (class 2); and within the private farming: penconazole, and abamectin refer to class 3 (moderately hazardous), difenoconazole, azoxystrobin, and metalaxyl-M to class 2 (hazardous).

Conclusions: the study revealed acceptable degree of the health risk of pesticides used for berry and melon crops, associated with consumption of contaminated water.

Keywords: herbicides, insecticides, fungicides, chemical water pollutants, risk assessment.

УДК 504.5:632.95.024:614.77]:634.7/635.6]]-043.2-047.77
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ОЦІНКА ПОТЕНЦІЙНОЇ НЕБЕЗПЕКИ ВПЛИВУ ПЕСТИЦИДІВ, РЕКОМЕНДОВАНИХ ДЛЯ ЗАХИСТУ ЯГІДНИХ ТА БАШТАННИХ КУЛЬТУР, НА ОРГАНІЗМ ЛЮДИНИ ПРИ ПОТРАПЛЯННІ В ҐРУНТОВІ ТА ПОВЕРХНЕВІ ВОДИ
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Метою дослідження було оцінити потенційну небезпеку впливу пестицидів для захисту ягідних та баштанних культур на організм людини при потраплянні в ґрунтові та поверхневі води. Використовували методи натурного гігієнічного експерименту, розрахунковий, статистичні.

Виявили, що за інтегральним показником небезпечності пестицидів при надходженні досліджуваних діючих речовин у воду, при їх застосуванні в промисловому секторі, діючі речовини флудіоксоніл, піраклостробін, пендиметалін є малонебезпечними (4 клас); фенгексамід, боскалід, флуксапіроксад, азоксистробін, спіродиклофен, абамектин – помірно небезпечні (3 клас); ципродиніл, дифеноконазол, металаксил-М, гліфосат – небезпечні (2 клас); в приватному секторі: пенконазол, абамектин – 3 клас, дифеноконазол, азоксистробін, металаксил-М – 2 клас.

Висновки: ризик негативного впливу на здоров'я людей досліджуваних пестицидів при споживанні води є допустимим.

Ключові слова: гербіциди, інсектициди, фунгіциди, хімічні забруднювачі води, оцінка ризику.

Introduction. Currently, the main source of drinking water supply for the population in Ukraine is centralized systems, which utilize both surface water bodies and underground (interstratal) waters from deep aquifers.

However, centralized water supply systems cover only 22.1% of rural settlements and 86.7% of urban-type settlements in the country. Decentralized water supply remains the most problematic in Ukraine, particularly due to anthropogenic pollution, including organo-mineral and phosphate-potassium fertilizers, as well as chemical plant protection products. This issue is exacerbated by the fact that sources for decentralized water supply often lack reliable protection [1].

Drinking water can be contaminated by pesticides, as these chemicals can enter surface or groundwater, which are commonly regarded as the main sources of drinking water. The presence of pesticides in drinking water can pose potential health risks to humans, depending on the amount and toxicity of the pesticides, as well as the frequency and duration of human exposure to the contaminated water [2].

Specialists from the U.S. Environmental Protection Agency (EPA) recommend assessing the risk of pesticide exposure to both human health and environmental health [3]. They advise mathematical models for predicting pesticide concentrations in food products, water, residential, and occupational environments [4]. Despite society's aspiration for zero risk, achieving this is impossible in the real world. Recognizing the risks associated with pesticide use leads to justified decision-making regarding acceptable levels of risk for society [5].

One third of the world's population relies on groundwater for drinking purposes. Groundwater contamination is a global issue with serious implications for human health and the environment. This necessitates deep understanding, as access to safe drinking water is a fundamental human right. However, contaminants such as hydrocarbons, toxic metals, pesticides, microplastics, and nanoparticles represent risks to human health and sustainable socio-economic development. Controlling and managing groundwater pollution and its associated health risks require the development of vulnerability, hazard, and risk maps [6].

Since the pesticides under study have not previously been used on berry and melon crops and are currently in the pre-registration testing phase for these crops, it is crucial to assess the potential health risks to the population from the contamination of surface and groundwater.

Objective. To assess the risk to the population from a water safety perspective after the contamination of groundwater and surface water by pesticides, specifically fungicides, herbicides, and insecticides, used for the protection of berry and melon crops.

Materials and Methods. Methods of natural hygiene experiments were used, involving chemical-analytical techniques such as high-performance liquid chromatography (HPLC) and gas-liquid chromatography (GLC). The obtained data were analysed using the custom software MedStat v. 5.2 [7].

The authors studied the behavior of active ingredients (AIs) of pesticides used for the protection of berry and melon crops in the agro-industrial sector (AIS) and private personal farms (PPFs). The pesticides studied included Kitch, Trinol, Amistar Gold, Switch, Signum, Topaz, Apron, Maxim, Uniform, Sercadis Plus, Ridomil Gold, Stomp Aqua, Dual Gold, Herbolex, Protect, Vertimec, and others.

The potential migration of pesticides into groundwater was predicted using the SCI-GROW indicator (Screening Concentration in Groundwater) [8]. Substances were classified due to their stability and migration capacity in the soil, according to the State Sanitary Rules and Standards 8.8.1.002-98 [9].

The potential migration of pesticides into groundwater was also assessed using the GUS index (Groundwater Ubiquity Score), calculated according to the authors according to the formula [10, 11]: $GUS = \log DT_{50} \times [4 - \log K_{oc}]$, where DT_{50} is stability in soil, day; K_{os} is the organic carbon sorption coefficient (organic carbon), ml/g organic carbon.

In the next stage, we calculated the following indicators: the maximum possible daily intake of pesticide from water (MPDIPW), the acceptable daily intake of pesticide from water (ADIW), and the health risk from the leaching of the active ingredients into water (R). These calculations were performed according to the defined methods [12]: MPDIPW = SCI-GROW × N × V V (μg/day); PDIP = ADD × M × 1000 (μg/day); ADIW = PDIP × 0.2 (μg/day), P = MPDIPW/ADIW (≤ 1 risk is permissible, > 1 risk is not permissible), where N is the maximum rate of pesticide consumption, taking into account the frequency of treatments, kg (l)/ha; V – daily rate of water consumption by a person, l (3 l – in a temperate climate, 5 – 10 l – in a hot climate); ADD – acceptable daily dose, mg/kg; M is the average weight of a person (60 kg); PDIP – permissible daily intake of pesticide. The results of these studies are detailed in [13].

To assess the risk to the population from consuming contaminated groundwater and surface water, we used the following method [14]. We calculated the potential groundwater and surface water contamination index (LEACHmod).

The LEACHmod indicator assesses the solubility of the substance in water (Sw, mg/L), the field half-life of the compound in soil (T_{50} field, days), and K_{oc} (mL/g) and calculated by the formula: $LEACH_{mod.} = \frac{S_w \times DT_{50\,field}}{K_{oc}}$, where Sw – solubility in water, mg/ L; DT_{50} field – period of half-life of substances in the base in natural conditions, day; K_{oc} – coefficient of sorption by organic carbon, ml/g of water. The T_{50} was agreed as that from literature sources [15], and we also considered the ADD (Acceptable Daily Dose) (mg/kg). These indicators (LEACHmod, T_{50} in water, ADD) were converted into scores according to the methodology, and an integrated hazard index for the pesticides in the water (IPW) according to the formula: IHCI = LEACHmod + T_{50} + ADD (points) was calculated. After summing all the scores, we determined the hazard class [16].

Results. A comprehensive hygienic assessment was conducted on the persistence of pesticides in soil, specifically those used for the berry and melon crops in both the agro-industrial and private sectors. The authors predicted the potential risk of groundwater contamination by these pesticides. Field studies to detect the presence of the investigated pesticides were held across different soil-climatic zones in Ukraine in the period from 2021 to 2023. In the agro-industrial sector (Kyiv, Odesa and Cherkasy regions), boom spraying and drip irrigation methods (Kyiv region) were utilized, while in private farms (Kyiv, Vinnytsia and Poltava regions), backpack spraying was employed. In order to assess the risk to the population when drinking water from underground and surface sources, under the conditions of the use of pesticides to protect berry and melon crops, 486 soil samples were studied.

Based on the results of the conducted field studies, we reproduced a mathematical model of the behavior of pesticides in soil, and calculated indices of their persistence and human health hazards (Table 1). The analysis revealed that among the fungicide formulations used in the agro-industrial sector, difenoconazole was identified as the most hazardous (class 1A – extremely hazardous*, class 2 – hazardous**).

The hazard classification depends on whether the T_{50} parameter is derived from the water-sediment phase or the water phase alone. Pyraclostrobin was found to be the least hazardous, classified as class 3* (4**) in terms of hazard.

Among the herbicides, pendimethalin was classified under the 3* (4**) hazard class, while glyphosate was classified as 1B* (2**). For insecticides in the industrial sector, the moderately hazardous active ingredients included spirodiclofen and abamectin (classified as 1B* (3**)), azoxystrobin (classified as 2* (3**)), and metalaxyl-M (classified as 1B* (2**)).

Active ingredients recommended for use in the private farms were predominantly classified under the 1B* hazard class (2-3**, as detailed in Table 1).

Discussion. Based on the criterion of soil persistence (T_{50}) under the EU soil and climate conditions, fenhexamid, copper, glyphosate, spirodiclofen, and abamectin are classified as low-persistence substances (hazard class IV). Metalaxyl-M, fludioxonil, and S-metolachlor are classified as moderately persistent (hazard class III). Cyprodinil and pyraclostrobin are considered persistent (hazard class II). Azoxystrobin, fluxapyroxad, boscalid, difenoconazole, pendimethalin, and penconazole are classified as extremely persistent (hazard class I). These classifications indicate, in most cases, a similarity in hazard levels based on soil persistence.

The result analysis (Tables 2) indicated that, according to the GUS index, most of the studied pesticides are unlikely to leach from the soil or have a low likelihood of leaching when used for berry and melon crops in both the agro-industrial sector and private farms. The exception is metalaxyl-M, which is likely to leach from the soil.

A comparative analysis of the values with similar indices obtained under same conditions in EU countries showed that, in most cases, the SCI-GROW index is significantly lower in EU countries than in Ukraine.

The comprehensive health risk assessment of the studied pesticides used for the berry and melon crops showed the water consumption health risk (P) to be within acceptable standards. Similar results were obtained applying other pesticides in the Ukrainian conditions. Thus, the risk to humans using pesticides with basic substance amicarbazone ranges between 7.4×10^{-4}–1.2×10^{-3} (acceptable risk), with bicyclopyrone – 8.3×10^{-5}–3.2×10^{-1} (acceptable risk), with pydiflumetofen – 3.4×10^{-5}–9.8×10^{-4} (acceptable risk) [16, 17].

The studies of the integral pesticide hazard indicator have shown that when active ingredients, used in industry, migrate into water, the following: fludioxonil, pyraclostrobin, and pendimethalin are of low hazard (class 4); fenhexamid, boscalid, fluxapyroxad, azoxystrobin, spirodiclofen, and abamectin are moderately hazardous (class 3); and cyprodinil, difenoconazole, metalaxyl-M, and glyphosate are hazardous (class 2). In the private farming: penconazole and abamectin refer to class 3 (moderately hazardous), and difenoconazole, azoxystrobin, and metalaxyl-M – class 2 (hazardous).

Table 1

Assessment of the Hazard of Pesticides Used for Berry and Melon Crops in Groundwater and Surface Water According to [16]

Acting substance	Sw	T_{50} soil, days	K_{oc}	LEACH	Points	T_{50} Water-sediment, days	Points	T_{50} Water phase, days	Points	ADD	Points	∑ IHCI*	∑ IHCI**	HC*	HC**
Agro-industrial sector															
Cyprodinil	13	29.4	2277	0.17	3	142	4	12.5	3	0.03	1	8	7	2	2
Fludioxonil	1.8	17.74	145600	0.0002	1	575	4	2.0	1	0.015	2	7	4	2	4
Fenhexamid	24	7.62	475	0.39	3	10.9	3	4.92	1	0.18	1	7	5	2	3
Cyprodinil	13	21.19	2277	0.12	3	142	4	12.5	3	0.03	1	8	7	2	2
Fludioxonil	1.8	10.8	145600	0.0001	1	575	4	2.0	1	0.015	2	7	4	2	4
Boscalid	4.6	38.1	772	0.23	3	545	4	5.0	2	0.04	1	8	6	2	3
Pyraclostrobin	1.9	33.26	9304	0.007	1	28	3	2.0	1	0.03	1	5	3	3	4
Fluxapyroxad	3.44	53.92	728	0.25	3	847	4	4.4	1	0.02	2	9	6	1B	3
Difenoconazole	15	30.08	3760	0.12	3	1053	4	3.0	1	0.002	4	11	8	1A	2
Metalaxyl-M	26000	35.18	78.9	11592.9	4	32.1	4	24.8	3	0.03	1	9	8	1B	2
Azoxystrobin	6.7	32.85	589	0.37	3	205	4	6.1	2	0.03	1	8	6	2	3
Metalaxyl-M	26000	31.14	78.9	10261.60	4	32.1	4	24.8	3	0.03	1	9	8	1B	2
Pendimethalin	0.33	37.12	17491	0.0007	1	16	3	4.0	1	0.008	2	6	4	3	4
Glyphosate	100000	7.96	1424	558.99	4	20.8	3	9.9	2	0.01	2	9	8	1B	2
Spirodiclofen	0.05	7.87	31037	0.000013	1	3.2	1	0.7	1	0.001	4	6	6	3	3
Abamectin	0.02	9.59	6631	0.000029	1	89	4	2.4	1	0.0002	4	9	6	1B	3
Private farming															
Difenoconazole	15	24.77	3760	0.099	2	1053	4	3.0	1	0.002	4	10	7	1B	2
Azoxystrobin	6.7	32.85	589	0.37	3	205	4	6.1	2	0.03	3	10	8	1B	2
Penconazole	73	30.94	2205	1.02	4	853	4	2.0	1	0.007	2	10	7	1B	3
Metalaxyl-M	26000	31.14	78.9	10261.60	4	32.1	4	24.8	3	0.03	1	9	8	1B	2
S-metolachlor	480	12.7	200.2	30.45	4	43.3	4	9.0	2	0.01	2	10	8	1B	2
Abamectin	0.02	5.89	6631	0.000018	1	89	4	2.4	1	0.0002	4	9	6	1B	3

Notes: 1. Sw – water solubility under 20°C (mg/l^{-1}); 2. IHCI – IHCI points (regarding water-sediment DT_{50} (days); ∑ IHCI** – IHCI points (regarding only water phase T_{50} (days); 3. HC* – Hazard Class* (regarding water-sediment T_{50} (days); HC** – Hazard Class **regarding only water phase T_{50} (days).*

Table 2

The results of the comparison of the resistance of the studied pesticides in the soil and the probability of their leaching into the groundwater when they are used to protect berry and melon crops

Acting Substance	T_{50}^{1}	T_{50}^{2}	Reliability of Differences (t)	Hazard Class $^{1}/^{2}$	GUS¹	GUS²
Agroindustrial sector						
Fludioxonil	14.27±1.56	17.0±4.36$^{F\ S}$	-0.58*	III/III	-1.33±0.06	-1.35
Cyprodinil	25.29±20.02	51.33±25.31$^{F\ B}$	-1.02*	II/II	0.89±0.02	1.06
Fenhexamid	7.62±0.26	2.89±2.62$^{UK.\ S}$	1.79*	IV/IV	1.17±0.02	-0.42
Boscalid	38.10±0.32	254.00±33.49$^{S\ F}$	-6.44**	II/I	1.76±0.004	2.68
Pyraclostrobin	33.26±0.99	44.30±26.89$^{G\ H}$	-0.41*	II/II	0.05±0.0004	0.05
Fluxapyroxad	53.92±0.63	196.8±95.89$^{H\ F}$	-1.49*	II/I	1.97±0.005	2.57
Difenoconazole	30.08±5.32	125.60±72.72$^{S\ UK}$	-1.31*	II/I	0.62±0.03	0.83
Metalaxyl-M	3.33±1.36	18.10±6.55$^{B\ Gr}$	2.13*	IV/III	3.19±0.03	2.42
Copper Oxychloride	10.64±0.05	3.40±3.30$^{F\ H}$	2.19*	IV/IV	1.03±0.002	-1.00
Azoxystrobin	37.17±1.29	154.5±70.79$^{UK.Nw}$	-1.66*	II/I	1.93±0.01	3.10
Pendimethalin	37.12±1.62	109.13±42.71$^{Nd\ S}$	-1.68*	II/I	-0.38±0.005	-0.28
Glyphosate	7.96±0.05	7.08±3.65$^{G.S}$	0.24*	IV/IV	0.76±0.002	0.21
Spirodiclofen	7.87±0.69	7.03±3.44$^{A.Nd}$	0.24*	IV/IV	-0.44±0.02	-0.42
Abamectin	9.59±0.01	1.27±0.27$^{A\ M}$	31.2**	IV/IV	0.18±0.001	0.015
Private Farming						
Difenoconazole	24.77±1.65	125.60±72.72$^{S\ UK}$	-1.39*	II-III/I	0.59±0.01	0.83
Azoxystrobin	32.85±1.75	154.5±70.79$^{UK\ Nw}$	-1.72*	II/I	1.86±0.03	3.10
Penconazole	30.94±2.58	90.57±13.86$^{L\ P}$	-4.22*	II/I	0.96±0.03	0.49
Metalaxyl-M	31.14±0.76	18.10±6.55$^{B\ Gr}$	1.98*	IV/III	3.14±0.02	2.42
Copper Oxychloride	12.94±1.17	3.40±3.30$^{G\ F}$	2.42*	IV/IV	1.10±0.07	-1.00
S-metolachlor	12.70±0.19	27.47±15.2$^{F\ G}$	0.97*	III/III	1.87±0.01	2.32
Abamectin	5.89±0.03	1.27±0.27$^{A\ M}$	17.24**	IV/IV	0.14±0.001	0.015

*Notes: 1. * – difference is not reliable by Student criterion (t), $p>0.05$; 2. ** – difference is reliable by Student criterion, $p<0,05$. 3. T_{50}^{1} – period of half-destruction of pesticides in soil by authors' data (soil and climate of Ukraine). 4. T_{50}^{2} – period of half-destruction of pesticides in soil by the EU authors [17–]: ^{F} – France, ^{S} – Spain, ^{B} – Belgium, ^{UK} – UK, ^{C} – Slovakia, ^{G} – Germany, ^{H} – Hungary, ^{Gr} – Greece, ^{Nw} – Norway, Nd – Netherlands, ^{A} – Austria, ^{M} – Malta, ^{L} – Latvia, ^{P} – Poland*

The comparative analysis of the results regarding the risk assessment for the population [18, 19] consuming water showed that the use of fungicides of the strobilurin, anilide, phenylpyrrole, and triazole classes presents a similar risk to that found in analogous studies with other preparative forms. Triazole fungicides (difenoconazole), amide herbicides, and the analyzed insecticides of the avermectin and tetronic acid derivative classes are less hazardous compared to pyrimidine compounds.

Risk assessment for the population in other countries showed that residual amounts of persistent pesticides are detected in water. However, the potential risk to public health did not exceed one, indicating a minimal health risk [20].

Conclusions

1. The comprehensive assessment of the health risks posed by the pesticides recommended for berry and melon crops determined that the risk of their negative human health effect through water consumption is acceptable.

2. The hazard classes of pesticides have been established, based on the integral groundwater hazard contamination index (IHCI), when these pesticides are used in agricultural production. Fludioxonil, pyraclostrobin, and pendimethalin are classified as slightly hazardous (Class IV). Fenhexamid, boscalid, fluxapyroxad, azoxystrobin, spirodiclofen, nd abamectin are moderately hazardous (Class III). Cyprodinil, difenoconazole, metalaxyl-M, and glyphosate are hazardous (Class II). In household plots (HHP): penconazole and abamectin are Class III (moderately hazardous), while difenoconazole, azoxystrobin, and metalaxyl-M are Class II (hazardous).

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Надійшла до редакції 27.11.2024 р.
Прийнята до друку 27.03.2025 р.
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