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## SPIS TREŚCI

## PRACE ORYGINALNE / ORIGINAL ARTICLES

- Andrey V. Dovzhenko, Vera D. Kuroedova, Lyudmila B. Halych  
THE EVALUATION OF TEETH LOOSENING OF THE UPPER JAW IN ADAPTIVE PERIOD OF ORTHODONTIC TREATMENT BY BRACES  
OCENA ROZLUŻNIENIA UKŁADU ZĘBOWEGO SZCZĘKI W OKRESIE ADAPTACYJNYM LECZENIA PRZY POMOCY APARATU ORTODONTYCZNEGO 123
- Zhanetta A. Chornenka, Elvira Ts. Yasinska, Mariana I. Grytsiuk  
EFFECT OF PROLONGED AND INTERMITTENT TREATMENT ON THE CLINICAL COURSE OF PEPTIC ULCER  
SKUTECZNOŚĆ PRZEDŁUŻONEGO I OKRESOWEGO LECZENIA NA KLINICZNY PRZEBIEG OWRZODZENIA ŻOŁĄDKA 128
- Юрий В. Смянов, Юрий В. Лактин  
ВЛИЯНИЕ НАПРЯЖЕННО-ДЕФОРМАЦИОННЫХ ПРОЦЕССОВ В ЭМАЛИ ЗУБОВ НА МАРГИНАЛЬНУЮ ПРОНИЦАЕМОСТЬ РЕСТАВРАЦИЙ I КЛАССА С РАЗНЫМ ДИЗАЙНОМ КРАЯ КАРИОЗНОЙ ПОЛОСТИ  
THE INFLUENCE OF STRESS-STRAIN PROCESSES IN TOOTH ENAMEL ON THE MARGINAL PERMEABILITY OF CLASS I RESTORATIONS WITH A DIFFERENT DESIGN OF THE EDGE OF THE CARIOUS CAVITY 135
- Maksim Y. Sokolov, Dmytro I. Besh, Olesya M. Besh, Oleg I. Rafaluk, Oleg O. Kapustynskyy  
THE INFLUENCE OF THE MANUAL THROMBUS ASPIRATION ON THE SHORT TERM EFFECTIVENESS OF THE PERCUTANEOUS REVASCLARIZATION IN PATIENTS WITH CORONARY ARTERY THROMBOSIS  
WPŁYW MANUALNEJ TROMBEKTOMII NA KRÓTKOTERMINOWĄ SKUTECZNOŚĆ PRZESKÓRNEJ REWASKULARYZACJI U PACJENTÓW Z ZAKRZEPICĄ TĘTNICY WIĘCOWEJ 140
- Orysa O. Syzon, Marianna O. Dashko  
INDICATORS OF PHAGOCYTOSIS IN WOMEN WITH ACNE DURING COMPREHENSIVE TREATMENT THAT INCLUDED IMMUNOTHERAPY AND PROBIOTICS  
WSKAŹNIKI FAGOCYTOZY U KOBIET Z TRĄDZIKIEM W TRAKCIE KOMPLEKSOWEGO LECZENIA OBLIĘGUJĄCEGO STOSOWANIE IMMUNOTERAPII ORAZ PROBIOTYKÓW 144
- Наталья И. Гасюк, Наталья С. Артемова, Елена М. Ковалева, Валерий И. Похилько, Галина А. Соловьева  
РОЛЬ ГЕНОВ РЕНИН-АНГИОТЕНЗИНОВОЙ СИСТЕМЫ В РАЗВИТИИ НЕБЛАГОПРИЯТНЫХ ИСХОДОВ ЛЕЧЕНИЯ ТЯЖЕЛЫХ ВНУТРИЖЕЛУДОЧКОВЫХ КРОВОИЗЛИЯНИЙ У ПРЕЖДЕВРЕМЕННО РОЖДЕННЫХ ДЕТЕЙ  
THE ROLE OF GENES OF RENIN-ANGIOTENSIN SYSTEM IN THE DEVELOPMENT OF ADVERSE OUTCOMES OF TREATMENT IN SEVERE INTRAVENTRICULAR HEMORRHAGES IN PREMATURE INFANTS 148
- Natalia V. Motorna, Svetlana L. Rybalko, Daria B. Starosyla, Mykhailo M. Guzyk, Iryna G. Strokina, Rostyslav F. Kaminsky, Alina V. Korsak, Sergey I. Savosko, Liudmyla M. Sokurenko, Yurii B. Chaikovskyy  
THE STUDY OF LEUKOCYTE PHAGOCYtic ACTIVITY IN THE PRESENCE OF HERPetic INFECTION AND STROKE  
BADANIE AKTYWNOŚCI FAGOCYTARNEJ LEUKOCYTÓW PRZY WSPÓLWYSTANIU ZAKAŻENIA OPRYSZCZKĄ I UDARU 155
- Елена Н. Комар, Наталья Н. Кизлова, Александра Д. Трилевич, Василий В. Кравченко  
ФАКТОРЫ РИСКА ВОЗНИКНОВЕНИЯ НЕБЛАГОПРИЯТНОГО ТЕЧЕНИЯ ЯЗВЕННОЙ БОЛЕЗНИ ЖЕЛУДКА И ДВЕНАДЦАТИПЕРСТНОЙ КИШКИ  
RISK FACTORS FOR ADVERSE COURSE OF GASTRIC AND DUODENAL PEPTIC ULCER 160
- Stanislav G. Saksonov, Tetiana S. Gruzeva, Oksana P. Vitovska  
HEALTH OF OPHTHALMOLOGISTS AS A PREREQUISITE OF QUALITY MEDICAL SERVICES  
STAN ZDROWIA OKULISTÓW JAKO WARUNEK JAKOŚCI USŁUG MEDYCZNYCH 165
- Николай А. Щербина, Людмила А. Выговская  
ОСОБЕННОСТИ МЕСТНОГО ИММУНИТЕТА ПРИ ЛОКАЛЬНОЙ ВОСПАЛИТЕЛЬНОЙ РЕАКЦИИ У БЕРЕМЕННЫХ В ЗАВИСИМОСТИ ОТ РЕАЛИЗАЦИИ ВНУТРИУТРОБНОЙ ИНФЕКЦИИ  
FEATURES OF LOCAL IMMUNITY IN LOCAL INFLAMMATORY REACTIONS IN PREGNANT, DEPENDING ON THE IMPLEMENTATION OF INTRAUTERINE INFECTION 168
- Mykola Kondratiuk, Anna Blagaia, Ihor Pelo  
COMPARATIVE HYGIENIC ASSESSMENT OF ACTIVE INGREDIENTS CONTENT IN THE AIR ENVIRONMENT AFTER TREATMENT OF CEREAL SPIKED CROPS BY COMBINED FUNGICIDES  
PORÓWNAWCZA HIGIENICZNA OCENA ZAWARTOŚCI AKTYWNYCH SKŁADNIKÓW W POWIETRZU PO LECZENIU ZBOŻA KŁOSOWEGO PRZY POMOCY ZŁOŻONYCH FUNGICYDÓW 173
- Vadim G. Krivoschapkin, Sergei I. Semenov, Svetlana S. Maximova, Nadezda N. Tihonova, Anna I. Sivtseva, Snegana S. Sleptsova  
CLINICAL AND LABORATORY CHARACTERISTICS OF HEPATITIS D IN REPUBLIC OF SAKHA (YAKUTIA)  
CHARAKTERYSTYKA KLINICZNA I LABORATORYJNA WIRUSOWEGO ZAPALENIA WĄTROBY TYPU D W REPUBLICIE SACHA (JAKUCJA) 179
- Oleg O. Sherstiuk, Volodymyr H. Hryn, Nataliia I. Vynnyk, Andriy V. Piliuhin, Mykhailo M. Koptev  
STEREOMORPHOLOGY OF THE GLANDULAR PARENCHYMA OF THE INFEROPOSTEROLATERAL AREA OF HUMAN PROSTATE GLAND  
STEREOMORFOLOGIA MIĄSZU POWIERZCHNI DOLNO-TYLNO-BOCZNEJ LUDZKIEGO GRUCZOŁU KROKOWEGO 184
- Valerii O. Novoseletskyy, Mykola A. Stanislavchuk, Volodymyr M. Shkarupa, Liliia V. Shvets  
CHARACTERIZATION OF LEPR GENE Q223R (RS1137101) GENOTYPES IN PATIENTS WITH KNEE JOINT OSTEOARTHRITIS OF DIFFERENT RADIOGRAPHIC STAGES  
CHARAKTERYSTYKA POLIMORFIZMU GENU LEPR Q223R (RS1137101) U PACJENTÓW Z CHOROBA ZWYRODNIENIOWĄ STAWU KOLANOWEGO W RÓŻNYCH STADIACH ZAWANOSOWANIA ZMIAN RADIOLOGICZNYCH 188

## COMPARATIVE HYGIENIC ASSESSMENT OF ACTIVE INGREDIENTS CONTENT IN THE AIR ENVIRONMENT AFTER TREATMENT OF CEREAL SPIKED CROPS BY COMBINED FUNGICIDES

### ПОРÓWNAWCZA HIGIENICZNA OCENA ZAWARTOŚCI AKTYWNYCH SKŁADNIKÓW W POWIETRZU PO LECZENIU ZBOŻA KŁOSOWEGO PRZY POMOCY ZŁOŻONYCH FUNGICYDÓW

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#### ABSTRACT

**Introduction:** The quality of the air environment significantly affects the health of the population. Chemical plant protection products in the spring and summer time may be the main pollutants of the air environment in rural areas. Chemical plant protection products are dangerous substances of anthropogenic origin. If applying pesticides in high concentrations, the risk of poisoning by active ingredients of pesticide preparations in workers directly contacting with it increases.

**The aim:** Comparative hygienic assessment of active ingredients content in the air environment after treatment of cereal spiked crops by combined fungicides was the aim of the work.

**Materials and methods:** Active ingredients of the studied combined fungicides, samples of air, and swabs from workers' skin and stripes from overalls were materials of the research. Methods of full-scale in-field hygienic experiment, gas-liquid chromatography, high-performance liquid chromatography, as well as statistical and bibliographic methods were used in the research.

**Results and conclusions:** Active ingredients of the studied combined fungicides were not detected in the working zone air and atmospheric air at the levels exceeding the limits of its detection by appropriate chromatography methods. Findings confirmed the air environment safety for agricultural workers and rural population if studied combined fungicides are applied following the hygienically approved suggested application rates and in accordance of good agricultural practice rules. However the possible complex risk for workers after certain studied fungicides application may be higher than acceptable due to the elevated values for dermal effects. The complex risk was higher than acceptable in epy case of aerial spraying of both studied fungicides, meanwhile only one combination of active ingredients revealed possible risk for workers applying fungicides by rod method of cereal spiked crops treatment.

**KEY WORDS:** combined fungicides, air pollution, risk assessment

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#### INTRODUCTION

The quality of the air environment significantly affects the health of the population. At the same time, the impact of pollutants on health is studied more fully in urban and industrial areas, with regular monitoring [1, 2], and monitoring is not carried out on continuing basis in rural areas [3]. Chemical plant protection products in the spring and summer time may be the main pollutants of the air environment in rural areas.

Chemical plant protection products are dangerous substances of anthropogenic origin and bear not only the benefits for mankind (as a way to reduce crop losses), but can also do significant harm to the environment and human health. Its application is accompanied by inevitable air pollution, both the working zone area and the atmospheric one. If applying pesticides in high concentrations, the risk of poisoning by active ingredients (a.i.) of pesticide preparations in workers directly contacting with it increases, and air drift with airflow over long distances is also possible

carrying these substances from the crop fields to adjacent areas, which in turn can lead to air pollution in populated areas and a deterioration in public health. Taking this into account, when allowing new chemical molecules and combinations of already known a.i. in fungicides to be applied on crops detailed research should be carried out to determine the residual quantities of the a.i. in the air environment, both the working zone area and in the atmospheric air.

#### THE AIM

Aim of the work: comparative hygienic assessment of active ingredients content in the air environment after treatment of cereal spiked crops by combined fungicides.

Tasks of the research: 1) determine the actual levels of azoxystrobin, benzovindiflupyr, epoxiconazole, kresoxim-methyl, metalaxyl-M, propiconazole, prochloraz, tebuconazole, fludioxonil and cyproconazole in the air of the working zone, 50 m, 300 m or

**Table I.** Content of active ingredients in combined fungicides and the terms of pesticides application

| Formulation                             | Active ingredient | Content of a.i. in formulation, g/l (g/kg) | Application rates for formulation, l/ha (l/t) | Application rates for treatment solution, l/ha (l/t) | Treated area, ha (amount of treated grain, t) | Number of treatments | Crops                                   | Location of the site   |
|---|-------------------|--|---|--|---|----------------------|---|------------------------|
| Treatment of the grain and its planting |                   |  |   |  |   |                      |   |                        |
| F. №1                                   | fludioxonil       | 18,75                                      | 2   | 10   | 1   | 1                    | grain of spring barley                  | 50°13'40"N, 30°39'00"E |
|   | cyproconazole     | 6,25                                       |   |  |   |                      |   |                        |
| F. №2                                   | metalaxyl-M       | 20   | 1,5   | 10   | 1   | 1                    | grain of spring barley and spring wheat | 49°21'7"N, 27°21'15"E  |
|   | tebuconazole      | 30   |   |  |   |                      |   |                        |
| Rod treatment                           |                   |  |   |  |   |                      |   |                        |
| F. №3                                   | azoxystrobin      | 80   | 0,75  | 300  | 1   | 1                    | spring barley, spring and winter wheat  | 49°21'7"N, 27°21'15"E  |
|   | cyproconazole     | 200  |   |  |   |                      |   |                        |
| F. №4                                   | epoxiconazole     | 75   | 1,5   | 300  | 1   | 2                    | spring and winter wheat                 | 49°47'59"N, 30°00'04"E |
|   | prochloraz        | 300  |   |  |   |                      |   |                        |
| F. №5                                   | kresoxim-methyl   | 240  | 1,0   | 200  | 1   | 1                    | spring and winter wheat                 | 49°36'29"N, 28°05'51"E |
|   | tebuconazole      | 125  |   |  |   |                      |   |                        |
| F. №6                                   | benzovindiflupyr  | 83,33                                      | 0,6   | 300  | 1   | 2                    | spring barley, spring and winter wheat  | 49°21'7"N, 27°21'15"E  |
|   | propiconazole     | 208,33                                     |   |  |   |                      |   |                        |
|   | cyproconazole     | 66,67                                      |   |  |   |                      |   |                        |
| F. №7                                   | azoxystrobin      | 120  | 1,0   | 300  | 1   | 2                    | spring barley, spring wheat             | 50°15'33"N, 31°09'31"E |
|   | tebuconazole      | 200  |   |  |   |                      |   |                        |
| F. №8                                   | azoxystrobin      | 80   | 2,0   | 300  | 1   | 2                    | spring barley, spring and winter wheat  | 50°20'24"N, 30°25'22"E |
|   | tebuconazole      | 160  |   |  |   |                      |   |                        |
| Aerial spraying                         |                   |  |   |  |   |                      |   |                        |
| F. №3                                   | azoxystrobin      | 80   | 0,75  | 100  | 3   | 1                    | winter barley                           | 48°37'15"N, 25°44'15"E |
|   | cyproconazole     | 200  |   |  |   |                      |   |                        |
| F. №6                                   | benzovindiflupyr  | 83,33                                      | 0,6   | 100  | 3   | 2                    | winter barley                           | 48°37'15"N, 25°44'15"E |
|   | propiconazole     | 208,33                                     |   |  |   |                      |   |                        |
|   | cyproconazole     | 66,67                                      |   |  |   |                      |   |                        |

1000 m from the edge of the field in the treatment zone on day 0, and also in the treatment zone and at a distance of 100 m from the treatment site on days 3 and 7; 2) give a hygienic assessment of the content of azoxystrobin, benzovindiflupyr, epoxiconazole, kresoxim-methyl, metalaxyl-M, propiconazole, prochloraz, tebuconazole, fludioxonil and cyproconazole in atmospheric air and working zone air after the studied combined fungicides application.

## MATERIALS AND METHODS

A.i. of the studied combined fungicides, samples of air, swabs from workers' skin and stripes from overalls were materials of the research.

Methods of full-scale in-field hygienic experiment, gas-liquid chromatography (GLC), high-performance liquid chromatography (HPLC), as well as statistical and bibliographic methods were used in the research. The studies for preparations No. 1 and 2 were carried out during the grain treatment and its planting using a PS-10 seed protectant, a disk seeder equipped with MTZ-82 tractor; for formulations (f.) No. 3, 4, 6-8 – at the rod treatment method of cereal crops spraying using MTZ-82 tractor equipped with trailed boomless sprayer OPS-2000 (Landini-2000 – for preparation No. 5); for preparations No.3 and 6 – at aviation method of crops spraying using AEROS-2 trike, equipped with a small-drop sprayer.

Table II. Meteorological conditions during the treatments

| Formulation                             | Air temperature at the time of treatment, °C | Atmospheric pressure, mm Hg. | Relative humidity, % | Air movement speed, m/s |
|---|--|------------------------------|----------------------|-------------------------|
| Treatment of the grain and its planting |  |                              |                      |                         |
| F. №1                                   | 10   | 755                          | 55                   | 1,0-2,0                 |
| F. №2                                   | 14   | 750                          | 70                   | 1,5-2,5                 |
| Rod treatment                           |  |                              |                      |                         |
| F. №3                                   | 19   | 745                          | 70                   | 1,0-2,0                 |
| F. №4                                   | 25   | 745                          | 60                   | 2,0                     |
| F. №5                                   | 17   | 750                          | 60                   | 1,0-1,5                 |
| F. №6                                   | 19   | 742                          | 70                   | 1,0-2,0                 |
| F. №7                                   | 20   | 745                          | 60                   | 1,0-2,0                 |
| F. №8                                   | 18   | 745                          | 60                   | 1,0-2,0                 |
| Aerial spraying                         |  |                              |                      |                         |
| F. №3                                   | 21   | 745                          | 60                   | 1,0-2,0                 |
| F. №6                                   | 21   | 745                          | 60                   | 1,0-2,0                 |

Content of a.i. in the studied combined fungicides as well as the terms of pesticides application is given in the table I.

Preparation of treatment solutions and filling-in of the seed protectant, as well as sprayer tanks was carried out on specially equipped sites (solution sites) in the immediate vicinity of the treatment site, the spray tank was pre-filled with 1/3 water, then the required amount of the preparation was added, the contents of the tank were mixed with a hydraulic stirrer (rod treatment), then water was added to the required volume. The duration of the operation for the preparation of the treatment solution was 10 minutes (for all types of treatments), the duration of grain treatment and packing was 20 minutes each, the planting of the treated grain lasted 30 minutes, the processing time for cereal spiked crops treatment was 40 minutes in the rod spraying and 20 minutes at aerial spraying. All treatment works with the abovementioned preparations were carried out under the allowable meteorological conditions given in Table II.

Determination of a.i. content in air samples after grain and cereal spiked crops treatment were conducted in accordance with the «Methodological Guidelines for the Hygienic Assessment of New Pesticides», approved in Ukraine.

Samples of the materials were sampled and delivered to the laboratory in accordance with the "Uniform Rules for sampling agricultural products, food and environmental objects for the determination of micro-quantities of pesticides". Air samples during the preparation of treatment solutions, during the treatment of grain and its packing, filling the seed protectant with treated grain and sprayers with treatment solutions, in the tractor cab and in the cockpit of the airplane, and also in the treated sites were sampled by portable two-channel electroaspirator EA-2-20. Control samples were taken from areas where the treatment was not performed.

Also, to estimate the risks of dermal and inhalation impact on workers contacting with plant protection products, while they were performing work operations, cotton fabric stripes attached to the work clothes during the period of work were sampled.

Each value of the content level in the samples was estimated as the average of the 3 samples obtained.

Three workers were involved in the process of grain treatment; when sowing grain – 2 persons; during the treatment of cereal spiked crops – 2 workers (rod treatment) and 3 workers (aerial spraying). In the course of all production operations, the workers were provided with overalls, which included: an overall of synthetic fabric and boots, gloves and a respirator (when filling the tanks or seed protectant) were used as personal protective equipment.

Determination of azoxystrobin, benzovindiflupyr, kresoxim-methyl and fludioxonil content was performed by HPLC; metalaxyl-M, propiconazole, prochloraz, tebuconazole, cyproconazole, and epoxiconazole were performed by GLC.

Limits of detection (LOD) by the above methods for the studied a.i. content in the air of the working zone and atmospheric air, as well as hygienic standards of tentatively safe exposure levels (TSEL) are given in table III.

## RESULTS AND DISCUSSION

In the course of the studies it was found that the residual quantities of the a.i. of preparations Nos. 1-2 in breathing zone (b.z.) of tank loaders (the site for preparation of solutions and filling of seed protectant), in the b.z. of the seed protectant operator did not exceed the limits of detection of the methods, whereas in the b.z. of the grain bag sewing machine operator during f. No.1 application

Table III. Hygienic norms and limits of detection of the studied pesticides in the working zone air and atmospheric air

| Active ingredient | Limit of detection, mg/m <sup>3</sup> |                  | tentatively safe exposure level, mg/m <sup>3</sup> |                  |
|-------------------|---------------------------------------|------------------|--|------------------|
|                   | Atmospheric air                       | Working zone air | Atmospheric air                                    | Working zone air |
| azoxystrobin      | 0.001                                 | 0.001            | 0.01   | 1                |
| benzovindiflupyr  | 0.0005                                | 0.016            | 0.002  | 0.1              |
| cyproconazole     | 0.008                                 | 0.05             | 0.01   | 0.1              |
| epoxiconazole     | 0.0004                                | 0.005            | 0.0005   | 0.01             |
| fludioxonil       | 0.002                                 | 0.002            | 0.002  | 1                |
| kresoxim-methyl   | 0.025                                 | 0.5              | 0.05   | 1                |
| metalaxyl-M       | 0.008                                 | 0.25             | 0.01   | 0.5              |
| prochloraz        | 0.0008                                | 0.05             | 0.001  | 0.1              |
| propiconazole     | 0.0008                                | 0.004            | 0.001  | 0.5              |
| tebuconazole      | 0.002                                 | 0.005            | 0.02   | 0.4              |

the concentration of fludioxonil was 0.2 mg/m<sup>3</sup>, cyproconazole – 0.05 mg/m<sup>3</sup>, f. No. 2 application – the a.i. were not detected. Concentrations of a.i. in the b.z. of seeders operators and tractor drivers did not exceed the limits of detection for the methods of determination. The level of a.i. content in the air of possible drift at a distance of 50 meters from the treated site, as well as at the leeward side edge of the field scattered by the treated grain (during the treatment/planting, on days 3 and 7 after treatment) was below the LOD.

During the studies of the air environment at the treatment of cereal spiked crops by the ground (rod) method, it was found that the a.i. residual amounts of preparations Nos. 3-8 in the b.z. of the tank loaders (the sites for the preparation of solutions and filling of rod sprayers), the b.z. of the tractor drivers, and also in the air at the zones for making the crops treatment after 1 and 3 hours, and 3 days were less than the limits of the detection of the methods. The level of a.i. content in the air of possible drift at a distance of 300 meters from the leeward side edge of the treated fields during treatment and at a distance of 100 meters on days 3 and 7 the concentration of the studied preparations a.i. was below the limits of detection of the methods.

In the course of the study of the air environment, spraying preparations on cereal crops by an aerial method, it was found that azoxystrobin (component of f. No. 3) was detected in the air of the tank loader b.z. (the treatment solution preparation site and filling) at 0.005±0.001 mg/m<sup>3</sup>. It was also found in the b.z. of the flagman at a level of 0.006±0.002 mg/m<sup>3</sup> and in the area of the crops treatment after 1 hour at the level of 0.005±0.003 mg/m<sup>3</sup>, whereas in the pilot b.z. and in the area of the crops treatment in 3 hours ca and 3 days after was no longer detected. The second component of f. No. 3 tebuconazole at all stages of treatment and the following hours after was not detected in the air in quantities exceeding LOD. When assessing the air environment during the cereal crops treatment with the f. No. 6 the component propiconazole was found in the b.z. of the flagman at a level of 0.04±0.006 mg/m<sup>3</sup>, and also after 1 hour in the treatment zone at

0.009±0.001 mg/m<sup>3</sup>. The level of a.i. content in the air of possible drift at a distance of 1000 meters from the leeward side edge of the treated fields during treatment, after 1 and 3 hours, and at a distance of 100 meters after 3 and 7 days, the concentration of the studied preparations a.i. was below the limits of detection of the methods.

The study also determined the level of the f. No. 1-8 a.i. content in swabs from bare skin areas and stripes from the overalls of workers, since pesticides can affect the body of workers by dermal exposure together with inhalation one [4].

The following results were obtained: the f. No. 1 component - fludioxonil was found in swabs from gloves of loader and the bag sewing machine operator at 0.003 and 0.004 mg/swab, respectively, as well as in stripes from overalls in the breast region of the bag sewing machine operator at 0.002 mg/dm<sup>2</sup>. The f. No.2 a.i. – tebuconazole and metalaxyl-M were detected in swabs from gloves of the loader at the level of 0.003 and 0.002 mg/swab, respectively; the f. No.3 a.i. (azoxystrobin and cyproconazole) were found in swabs from loader gloves at the level of 0.043 mg/swab and 0.01 mg/swab, as well as in the stripes from the breast and forearm at the level of 0.012 mg/dm<sup>2</sup> and 0.0057 mg/dm<sup>2</sup>, respectively. Component of f. No.4 - prochloraz was found in swabs from the gloves of loader at the level of 0.007 mg/swab, as well as in stripes from the region of the chest and forearms – 0.002 mg/dm<sup>2</sup>; the f. No. 5 a.i. (kresoxim-methyl and tebuconazole) were found in swabs from the loader gloves at 0.003 mg/swab and 0.0045 mg/swab respectively, the concentration of tebuconazole was detected in the stripes from the chest and forearms region, as well as from the back and thighs of the loader at the level of 0.003 mg/dm<sup>2</sup> and 0.002 mg/dm<sup>2</sup>, respectively. Component of f. No.6 – propiconazole was found in swabs from loader gloves at the level of 0.005 mg/swab; the f. No. 7 a.i. (azoxystrobin and tebuconazole) were found in swabs from loader gloves at a level of 0.013 mg/swab and 0.02 mg/swab, respectively, and also found in the stripes from the chest and forearms region of the loader at the level of 0.0011 mg/dm<sup>2</sup> and 0.002 mg/dm<sup>2</sup>, respectively.

**Table IV.** Values of calculated risk for workers (in arbitrary units)

| Formulation | combined percutaneous effect | combined inhalation effect | complex effect |
|-------------|------------------------------|----------------------------|----------------|
| Nº1         | 0,4503                       | 0,2459                     | 0,6962         |
| Nº2         | 0,1617                       | 0,2592                     | 0,4209         |
| Nº3         | 0,8044                       | 0,2558                     | <b>1,0602</b>  |
| Nº4         | 0,9885                       | 0,6375                     | <b>1,6260</b>  |
| Nº5         | 0,3849                       | 0,2922                     | 0,6771         |
| Nº6         | 0,5036                       | 0,2557                     | 0,7593         |
| Nº7         | 0,0471                       | 0,0077                     | 0,0548         |
| Nº8         | 0,0628                       | 0,1446                     | 0,2074         |
| Nº3         | <b>2,5133</b>                | 0,5767                     | <b>3,0900</b>  |
| Nº6         | <b>3,1871</b>                | 0,9362                     | <b>4,1233</b>  |

The f. No.8 a.i. (azoxystrobin and tebuconazole) were found in swabs from loader gloves at levels of 0.009 mg/swab and 0.025 mg/swab, respectively, the component of the f. - tebuconazole was found in the stripes from the chest and forearms region of the loader at 0.003 mg/dm<sup>2</sup>.

In the aerial spraying of cereal spiked crops by the f. No. 3, its a.i. (azoxystrobin and cyproconazole) were detected in swabs from the tank loader gloves at the level of 0.03 mg/swab and 0.007 mg/swab respectively, as well as these substances were found in the stripes from chest and forearms region of the loader and the pilot at the 0.001 mg/dm<sup>2</sup> and 0.009 mg/dm<sup>2</sup>, respectively. Estimating the f. No.6 application by the aerial method, its component - propiconazole was found in swabs from loader gloves at 0.004 mg/swab, as well as in stripes from the chest and forearms region, and from the back and thighs at the level of 0.003 mg/dm<sup>2</sup> and 0.005 mg/dm<sup>2</sup>, respectively.

On the basis of the obtained data on the f. Nos. 1-8 a.i. concentration in the air of the working zone, as well as in swabs from bare skin areas and stripes from overalls, we also calculated the combined and complex risks of dermal and inhalation effects, in conditions of the all work operations performance by one person, as such conditions can arise in small farms (table IV).

It was found that the combined risk of percutaneous and inhalation effects, in conditions when all the operations were performed alone, applying these combined fungicides, in the case of f. No. 3 (in rod and aerial spray treatment), f. No.4 and 6 application, the calculated risk of possible harm to workers via dermal and inhalation routes of exposure exceeded 1 arbitrary unit, but the combined effect of active substances (both the dermal and inhalation influence separately) in the conditions of the all stages of treatment work performance, was lower than 1 arbitrary unit. It should be noted that both f. No.3 applications, as by rod spraying and aerial spray treatment, were accompanied with elevated possible risk for workers.

In the case of f. No. 1, 2, 5, 7, 8 applications, the calculated risk was not exceeding 1. But it should be noted that the performance of work on the preparation of treatment

solutions of combined fungicides and its loading to the sprayers is accompanied by the highest degree of risk of harmful effects (mainly by the dermal route of exposure).

## CONCLUSIONS

Based on the results of the studied fungicides content dynamics determination (azoxystrobin, benzovindiflupyr, cyproconazole, fludioxonil, epoxiconazole, kresoxim-methyl, metalaxyl-M, propiconazole, prochloraz and tebuconazole) in the air of working zone and atmospheric air during formulations application with the maximum proposed rates it was found that there is no pollution of air above developed and approved hygienic standards and the air quality is not deteriorating at time and after applications.

But the findings of above mentioned dynamics together with the detected content of active ingredients in the swabs and stripes during formulations application with the maximum proposed rates suggested that particular combinations of fungicides have possible risk of effects on the agriculture workers who applying combined fungicides for cereal spiked crops treatment. Instructions and guidelines for these combined fungicides safe application were developed by us and approved by Ministry of Health of Ukraine and accepted for the use in work of State Service of Ukraine on Food Safety and Consumers Protection. The findings should be taken into account in the programs of monitoring the labour conditions of agriculture workers using combined fungicides on cereal spiked crops.

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**INTERNATIONAL SCIENTIFIC AND PRACTICAL CONFERENCE,  
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