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Nutrient and organic substances emissions from diffuse sources to the rivers of Ukrainian Carpathians

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ABSTRACT

Formulation of the problem. The transition from territorial-administrative to basin-based water resources management in Ukraine requires an assessment of the load of water bodies with pollutants from diffuse and point sources. Among the various components of the chemical composition of water entering water bodies, organic matter and nutrients are the most important. The Danube is the second largest river in Europe, with a basin covering the territories of 19 countries and is an important transportation waterway. Within Ukraine, the Danube is divided into the Lower Danube sub-catchment and the Tisza, Prut, and Siret River basins within the Carpathian region. The scientific results presented in this paper were obtained during research within the framework of state budgetary research works of the UHMI, the implementation of which will contribute to the further development of knowledge in the field of hydrometeorology. The results presented in the publication are important for supplementing information when writing the Danube River Basin Management Plan, which is being developed in accordance with Ukraine's obligations under the Association Agreement with the European Union.

Aim of the study. To calculate the supply of nutrients and organic matter from diffuse sources to the rivers of the Danube basin within the Ukrainian Carpathians**.**

Methods. Monitoring data of organic substances and nutrients provided by the Danube Hydrometeorological Observatory (the state surface water monitoring network of the State Emergency Service of Ukraine) for 2018 were used for calculations. To assess the load of water bodies by diffuse sources, a conceptual scheme was developed that allowed to take into account the main pathways of substances supply, i.e precipitation; water runoff from arable land, forests, meadows and pastures, built-up areas, rock outcrops, as well as from the population of rural regions without sewage systems.

Scientific novelty. For the first time:

- the load of nutrients and organic matter in the Danube rivers within the Ukrainian Carpathians by the sources of their income was evaluated;
- it was found that agricultural land is the main source of emissions of inorganic nitrogen and phosphorus compounds.

Practical value. The analysis of the diffuse load by nutrients and organic matter in the Danube rivers can be used as an important part of the River Basin Management Plan. The results of the calculations can also be used to develop measures to achieve certain environmental objectives.

Results. The main source of organic matter in the water of rivers of the Carpathian region ere agricultural enterprises locating within rural settlements that are not equipped with sewage systems. Surface water of Prut and Tisza River basins is the most polluted by organic matter. By source, the nutrient emissions are distributed as follows. For the Tisza and Prut rivers, more than 50% of the total nitrogen compounds emissions come from agricultural land, while for the Siret River the source of nitrogen compounds are forests (46% of the total nitrogen emissions). Accordingly, the total phosphorus runoff for the Tisza and Prut rivers was distributed as follows: the dominant share (up to 45%) comes from agricultural land, the load caused by population not connected to the sewerage systems is 36%. For the Siret River, agricultural land and forested areas are of equal shares (33%) among the sources of phosphorus compounds.

Keywords*: emission, nutrients, organic substances, diffuse sources, pollution, anthropogenic load.*

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Introduction. Economic activity in the catchment area of rivers quite often leads to a negative impact on the overall state of aquatic ecosystems. In the hydrometeorological practice of Ukraine, the assessment of the impact of human activities was most frequently performed based on the sanitary and hygienic principles (MPC, DSTU) and took into account mainly the impact of point pollutants for which monitoring observations were conducted [10, 11]. Point sources (e.g., wastewater outlets) are relatively constant in terms of flow rates, easily identifiable by location, with a constant list and concentrations of pollutants depending on the type of activity and production.

Diffuse pollution combines both anthropogenic and natural components. That is, its level depends not only on the anthropogenic load, but is also determined by climatic, hydrological conditions, properties of the underlying surface and soils [3, 9]. In terms of the nature of substance inflow, the anthropogenic component of diffuse pollution is represented by rural runoff, wash off from agricultural land and built-up areas. Inputs from forested areas, meadows and pastures (covered mainly with grass vegetation), as well as atmospheric deposition on the water surface are associated with the influence of natural factors [10, 13].

Among the various components of the chemical composition of water entering water bodies from both point and distributed sources, organic matter and nutrients are of major concern. Phosphorus and nitrogen compounds play a dominant role among other biogenic elements.

Under conditions of excessive concentrations of biogenic and organic substances, aquatic ecosystems are characterized primarily with intensive development of algae and cyanobacteria, which convert nutrients into biomass. Their rapid reproduction causes "blooming" of water, which leads to oxygen deficiency (anaerobic conditions). This creates favorable conditions for the development of pathogenic microflora and pathogens. Organic pollution has a negative impact on the ecosystem as a whole, the use of water for irrigation, fisheries, etc. [14, 23, 27]

Preventing the inflow of organic matter and nutrients into water ecosystem is one of the important factors to minimize eutrophication of aquatic ecosystems and maintaining of surface water quality.

The Danube is the second largest river in Europe, with a basin covering the territory of 19 countries. It is an important European transportation artery with the status of an international transport corridor [4, 5, 16]. The intensive use of the Danube's waters in various sectors of the economy has affected the state of its ecosystem. The action plans of the Danube River Basin protection have significantly reduced the anthropogenic load and pollutant inflows [19]. At the same time, the river ecosystem has not reached a "good" ecological status.

The area of the Danube within the territory of Ukraine is 4.5% and is divided into the 170 km-long Lower Danube sub-catchment (including the river delta and the upstream riparian area) and the basins of the Tisza, Prut, and Siret rivers [1, 4] within the Carpathian region. The sub-basins of the Tisza, Prut, and Siret rivers are represented by the mountainous or folded region and foothills of the Ukrainian Carpathians. Numerous watercourses originating in the Carpathians contribute to the spread of erosive and accumulative landforms. The Tisza sub-basin collects water from the territory of five states; within Ukraine, the catchment area is 11.3 thousand km^2, with a total area of 157.2 thousand km^2 [4, 5, 8]. The main rivers of the region - the Tisza, Prut, and Siret - are located within 4 regions of the country (Zakarpattya, Chernivtsi, Ivano-Frankivsk and Lviv) and account for 3.8% of the total area of the Danube basin or 9.4% of the territory of Ukraine [1, 20, 21].

In terms of water supply, the Carpathian region is a leader in the country. The region's water resources are used in the following areas: 1) drinking and domestic water supply and sewerage; 2) agriculture; 3) industry; 4) fisheries; 5) recreation; 6) environmental water use [1, 4, 22].

Water resources are represented by rivers, lakes, ponds, and reservoirs. The density of the river network ranges from 0.2 to 1.7 km/km^2. The water supply of settlements, industrial enterprises and agricultural farms is provided mainly by groundwater. At the same time, the use of natural water is insignificant due to the complex hydrological regime of rivers, which leads to a shortage in water supply for various industries and population. Almost 70% of the total number of rivers are located in the mountainous areas. [1, 4, 20].

The Carpathian socio-economic region is a part of the Southwestern Economic Region of Ukraine and is characterized with an average level of economic development. The region's characteristic of natural features includes a complex topography and the existence of different zones based on climatic and landscape features, as well as differences in hydrological and hydrochemical regimes in certain river sections [19, 20]. The complex geostructure and the resulting geolithological basement (mountainous, folded region and foothills of the Ukrainian Carpathians) determine the diversity and richness of the region's subsoil, from mineral waters to oil and gas production [4, 5].

The total area of land in the Carpathian region is 11063 thousand hectares (18% of the total in the country) [2]. The main use of land is mainly related to agriculture and forestry, as well as nature reserves and urbanized areas. The Carpathian region ac-

counts for 21% of the total land area of the Ukrainian forests. The Carpathian forests play an important role in climate change mitigation, water protection, water regulation, and soil protection. The Carpathian region ranks second (after Donetsk) in terms of population density: in the plains it is 103.3 people per km², and in the mountainous areas - 38.3 people per km2. The intensive impact of human production activities has significantly affected the natural state of the landscapes [2, 15, 20].

As a result of intensive agricultural development, large areas (especially in the Tisza River floodplain) have been converted into agricultural land and pastures. The intensive use of fertilizers and agrochemicals has led to soil and surface water pollution, and to increased eutrophication of water bodies due to the influx of organic matter and nutrients. The disturbance of soil covers due to plowing and regular deforestation also leads to a significant leaching of these substances. After deforestation, soils remain poorly fertile and are exposed to the adverse effects of direct sunlight and heavy rains. Floods, flash floods, and salt fluxes have become more frequent, leading to the erosion of small river channels and increased nutrient and organic matter removal. Mudflows are observed in the Prut and Siret River basins, especially in the mountainous part of the Prut basin [8, 24, 27].

The purpose of study. The unsatisfactory ecological condition of the Danube requires further research of the Carpathian region rivers of the Danube River Basin District to assess the load of water with nutrients and organic matter from various sources and their impact on the overall condition of the rivers. Given such a feature of diffuse sources as dispersion within the river basin (as opposed to point sources) and the dependence of the pollution level on not only anthropogenic impact, the issue of diffuse pollution of surface waters of the basin by inorganic nitrogen, phosphorus, and organic matter is important and relevant and determines the purpose of our study.

Analysis of the latest research and publications. The Danube River Basin is one of the most studied in Ukraine. The results of studies of the hydrological regime and chemical composition are presented in numerous textbooks and monographs, as well as in specific publications. Thus, the hydrological characteristics of the basin are widely presented in scientific papers by V. Vyshnevsky, Zh. Shakirzanova, and V. Hrebiny. The study of the anthropogenic load on the European part of the Danube is presented in Ottavia Zoboli‘s publications. The characteristics of hydrochemical regime formation and runoff of chemical elements in the Danube basin within the Ukrainian Carpathians and the lower part of the Danube are presented in publications by N. Osadcha, D. Klebanov [3, 4, 8, 22]. The study of the impact of diffuse sources and the characteristics of pollutant load of surface water bodies in general are presented in [6-7, 11, 12, 17].

One of the significant results of studying the flow of nutrients to surface water bodies was the development of methodology for assessing their load is presented in [6].

As of today, studies of the quantitative and qualitative diffuse load characteristics of river basins in Ukraine are extremely insufficient. In particular, for Ukraine within the Danube sub-basin, which covers the Carpathian region, diffuse load calculations have not been carried out before. That is why the main objectives of this work are:

- to research the main ways of mentioned substances entering the rivers of the Carpathian region (within Ukraine), according to the developed conceptual scheme;
- to identify the dominant sources of nutrients and organic matter emissions to the surface water of rivers.

Materials and methods. The organic matter and nutrients monitoring data provided by the Danube Hydrometeorological Observatory of the State Emergency Service of Ukraine for 2018 were used for calculations. Chosen year was close to the average water content in the long-term context and represented the largest amount of data on hydrochemical parameters for emission calculations. It should also be noted that since 2018, 29 more cities have been equipped with primary and secondary (microbiological) treatment facilities in the Tisza, Prut, and Siret sub-basins, which, in our opinion, could change the share of nutrients and organic matter in the emission sources.

To analyze the load of water bodies by diffuse sources, a model was used that took into account the main pathways of substance income, i.e., from atmospheric precipitation, with water runoff from arable land, forests, meadows and pastures, built-up areas, as well as from the population of rural regions not connected to sewage systems. The total supply of nutrients within a particular catchment was determined according to the methodology presented in [9, 12]. Taking into account that the purpose of the study is to evaluate the diffuse load, the emission fluxes of nutrients from wastewater treatment plants of urban agglomerations, industrial and agricultural enterprises were not considered in calculations.

To obtain the daily concentrations of nutrients and organic substances, we applied a graphical interpolation of the available monitoring data using an approximating polynomial.

According to [9, 18, 25], an important factor in the formation of the anthropogenic component of the diffuse load on surface water bodies is the type

of land cover. To determine the main landscape units within the Danube River basin, a digital map was developed based on Landsat8 multispectral images obtained from (USGS EarthExplorer/ https://earthexplorer.usgs.gov/) for 2018. The results are presented in Table 1.

Table 1

Danube River basin main land cover types, km^2

Land cover types	Tisza	Siret	Prut	Lower Danube
Arable land	3821,7	699,2	4178,7	4427,8
Grass vegetation (meadows and pastures)	3263,5	269,5	1504,4	717,9
Forests	5634,3	1093,8	3566,8	17,7
Waterlogged areas	0,34			440,9
Built-up areas	8,07		7,8	29,1
Opened areas	0,82		14,7	212,5
Water surface	8,42	0,20	12,5	569,9

The organic matter load from the rural population was assessed by the calculation method using the adjusted coefficients of organic matter intake due to the vital activity of 1 person. In line with the recommendation of the Intergovernmental Panel on Climate Change (IPCC), the level of organic matter generation expressed via indirect indicator BOD5 in urban wastewater was proposed to be 50 g/day per person [2, 19, 21]. The level of organic matter content expressed by COD indicator was calculated using the conversion factor of BOD5 to COD (1.7). Accordingly, the value of 85 g/day per person was used to calculate the organic matter load.

Also, to estimate the input of nutrients and to compare possible changes we used the semi-empirical conceptional model MONERIS [26, 27] using observation data from the Transboundary National Monitoring Network (TNMN), established under the program of the International Commission for the Protection of the Danube River for certain years (2012, 2018).

Results and discussion. Based on the analysis of the main types of land use within the studied river basins, we can conclude a certain disproportion both between them and for the river basins. For the Tisza and Siret rivers, almost half of the basin area (>45%) is occupied by forests, while for the Prut River, the same percentage of the area is cultivated for agricultural production.

Organic substances. A comparative analysis of the average annual concentrations has indicated that during the period 2012-2018, the content of organic substances estimated via COD values decreased from 17.1 to 14.8 mg O_2/l for the Tisza River; from 14.2 to 12.6 mg O_2/l for the Prut River. Similar trends were observed for the BOD_5 indicator - its values did not exceed the critical limits (5-7 mg O/l).

The significance of diffuse sources for organic pollution of the Danube basin rivers was assessed in terms of their sources of supply. Given that precipitation has virtually no effect on the migration of organic matter in surface waters, this source can be neglected. It is known from previous studies [11, 12] that the main source of organic compounds is the households of the predominantly rural population that are not connected to sewage systems. Within the Tisza, Prut, and Siret River basins, there are a total of about 1,200 villages and towns. Drainage in such households is carried out on the relief by accumulation in sedimentation pits. According to the calculations, 11853 tons of organic substances per year (by BOD_5 indicator) and 20149 tons of organic substances per year by COD indicator) are discharged to the surface water of the studied rivers from distributed sources. The spatial distribution of organic substances load in the rivers of the study basin from rural settlements is shown in Fig. 1.

The Prut and Tisza River basins play a key role in organic pollution. Up to 50% of the total organic substances load among the studied rivers of the Carpathian region is formed within their boundaries. Most likely, this distribution of the load can be explained by the fact that the ratio of the number of settlements without sewage systems to the area of a that particular sub-basins is the highest.

Nutrients. As of 2018, the average annual content of nitrogen compounds in the water of the Tisza River, Prut River, and Siret River equaled to 0.58, 0.62, and 0.78 mg N/l respectively. The emission flow of nitrogen compounds from the territory of these river basins was equal to 12878 t/year, 5807 t/year and 3193 t/year, respectively. The distribution of the main sources of nitrogen is shown in Fig. 2.

The sources of nitrogen emissions are distributed as follows: more than 50% came from agricultural land (for the Prut and Tisza rivers), and 46% from forested areas (for the Siret River). Forests consume a large amount of nutrients for their growth, and it is the forested areas that make the main contribution to nitrogen migration within the watershed. According to the calculations, the supply of nitrogen compounds with atmospheric precipitation and from the

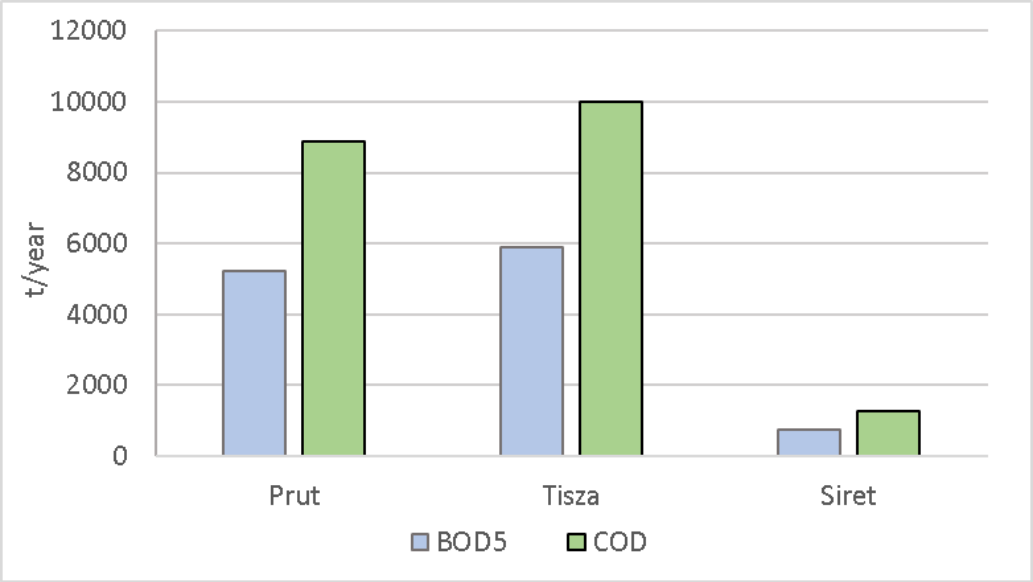

Fig. 1. Spatial distribution of the Danube River basin water pollution by organic substances from the rural population

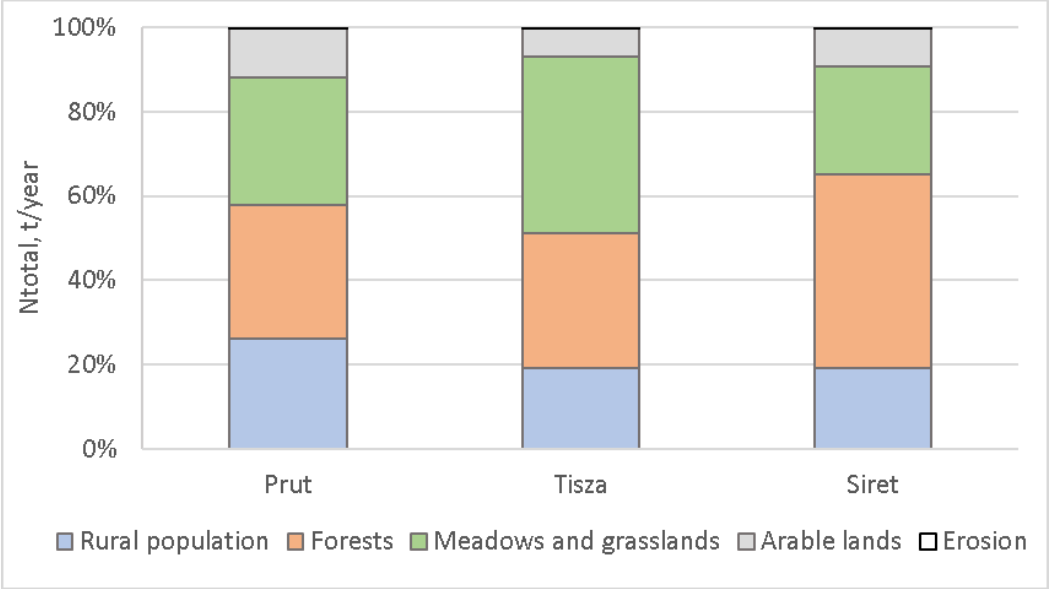

Fig. 2. The main sources of nitrogen compounds emission to the rivers of the Danube basin

built-up area for these basins is insignificant (<1%), therefore these data were neglected on the chart (Fig. 2).

The significant influence of agricultural sources on the runoff of nitrogen compounds is manifested in the ratio of the main forms of nitrogen in aqueous solution. For the Tisza, Prut, and Siret rivers, the nitrate form of nitrogen accounted for 78%, 42%, and 32% of the total, respectively; ammonium - 21%, 55%, and 66%; nitrite - 0.5%, 3.2%, and 3%, respectively. The dominance of the nitrate form of nitrogen, in our opinion, indicates the influence of mainly diffuse sources, primarily of agricultural origin. The predominance of the ammonium form of nitrogen in the water of the Prut and Siret rivers is most likely due to the influence of point sources - industrial and household wastewater. It should be noted that, according to previous studies [5, 9, 11], the dominant share of the total amount of wastewater inflows belongs to municipal wastewater.

The average annual content of mineral phosphorus for the Tisza, Prut, and Siret rivers was in the range of 0.02 - 0.04 mg P/l. According to calculations, the total load of phosphorus compounds for the rivers listed above is 71.6 t/year, 61.2 t/year and 12.6 t/year, respectively. By source, the total phosphorus runoff for the Tisza and Prut rivers was distributed as follows: the dominant share (up to 45%) comes from agricultural land, the load from the population without sewerage systems is 36%. For the Siret River, agricultural land and forested areas account for equal shares (33%) among the pathways of phosphorus compounds (Fig. 3).

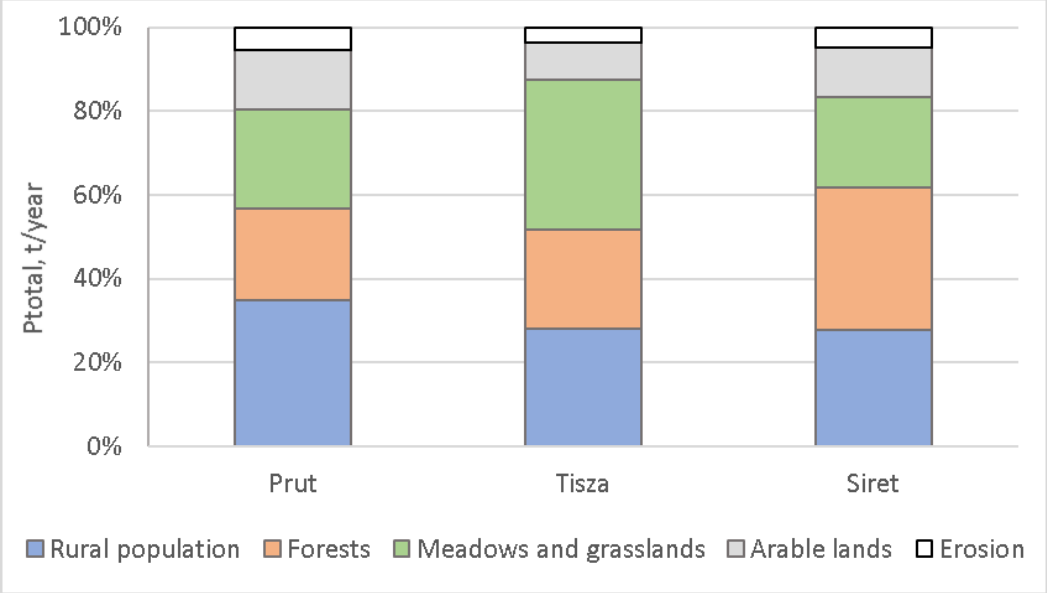

Fig. 3. The main sources of phosphorus compounds emission to the rivers of the Danube basin

It should be noted that for phosphorus compounds (as opposed to nitrogen compounds), a slightly higher percentage (up to 11%) of its emissions is observed due to erosion processes. This process is affected by the peculiarities of phosphorus behavior in soils: once it gets into the soil with mineral fertilizers, it is quickly adsorbed by the minerals of the soil-forming rocks and retained by them for a long time. The high level of land plowing and the significant development of erosion processes contribute to the migration of phosphorus compounds as a part of suspended particles [14, 15, 23, 27].

Along with the direct calculation of nutrient runoff, we modeled the flow of nutrients from the territory of the Ukrainian part of the Tisza, Prut, and Siret rivers using the semi-empirical conceptional model MONERIS. The model is based on the balance principle, which takes into account the main pathways of nutrient inputs and outputs within the Danube watershed [25, 26]. The MONERIS model allows estimating the supply of common forms of nutrients. The total nitrogen (N_{total}l) and phosphorus (P_{total}) were defined as the total amount of their inorganic and organic forms According to TNMN data, the relative content of inorganic nitrogen forms in the studied water bodies is 55%, organic - 45%; for phosphorus - 36% and 64%, respectively.

According to the modeling results, in 2012 the water of the Tisza and Prut rivers received 14.1 thousand tons of total nitrogen and 1.1 thousand tons of total phosphorus. In 2018, the total input of N_{total} to surface waters from the Tisza, Prut, and Siret rivers was 20.9 thousand tons/year (with a runoff rate of 0.08 t/km^2 *year), and phosphorus - 0.33 thousand tons/year (0.9 kg/km^2 *year) (Table 2).

It should be noted that for the surface waters of the studied rivers, there was an increase in the content of N_{total}l compounds (on average by 27%) compared to the data of 2012. On the other hand, the content of P_{total}l compounds decreased (by 30% on average). Earlier calculations in [4, 11] show that the dominant part of nitrogen is of agriculture origin (41%), while for phosphorus the origin was the influence of municipal wastewater (54%). Consequently, decrease of P_{total} can be explained by the implementation in Ukraine measures aimed at reducing the flow of nutrients and organic matter directly from municipal wastewater as it was done by EU countries. Among the studied watersheds, a significant increase in total nitrogen compounds was observed for the Prut River, which could be due to the impact of municipal wastewater. The Chernivtsi

Table 2

Nitrogen and phosphorus compounds inflow to the Tisza and Prut rivers (thousand tons per year)

River	N_{total}		P_{total}	
	2012	2018	2012	2018
Tisza	8,1	9,9	0,47	0,13
Prut	5,9	9,2	0,57	0,17
Siret	-	1,6	-	0,03
Sum		**20,9**		**0,33**

wastewater treatment plant operates inefficiently due to outdated equipment, which leads to discharges of untreated or insufficiently treated wastewater.

Conclusions. The main source of organic substances in the surface water of the Carpathian region rivers is agricultural households locating within rural settlements that are not equipped with sewage systems. The impact of other factors is insignificant. The Prut and Tisa River basins play a key role in organic pollution (up to 50% of the total organic substances load).

Nutrient emissions are distributed by source as follows. For the Tisza and Prut rivers, more than 50% of the total nitrogen compounds emissions come from agricultural land, and for the Siret River the source of nitrogen compounds are forests (46% of the total nitrogen emissions).

Accordingly, the total phosphorus runoff for the Tisza and Prut rivers was distributed as follows: the dominant share (up to 45%) comes from agricultural land, the load from the population without sewerage systems is 36%. For the Siret River, agricultural land and forested areas account for equal shares (33%) among the sources of phosphorus compounds. The influence of other factors is much smaller.

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Надходження біогенних елементів та органічних речовин від дифузних джерел до річок Українських Карпат

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Перехід від територіально-адміністративного до басейнового управління водними ресурсами в Україні вимагає оцінки навантаження водних об'єктів забруднюючими речовинами комплексно від дифузних та точкових джерел. Характерною особливістю дифузного забруднення є те, що його рівень у багатьох випадках визначається кліматичними, гідрологічними умовами, властивостями підстильної поверхні і ґрунтів. Серед різноманітних компонентів хімічного складу вод, які надходять до водних об'єктів найбільшу увагу привертають органічні речовини та біогенні елементи. В роботі представлено результати системних досліджень щодо кількісної оцінки сучасного стану та виносу біогенних елементів та органічних речовин поверхневими водами р. Дунай у межах Української частини Карпат - річок Тиса, Прут та Сірет. Карпатський соціально-економічний регіон має власні природні особливості та вирізняється широким спектром використання водних ресурсів у багатьох сферах господарської діяльності. Незадовільний екологічний стан Дунаю вимагає додаткових досліджень змін хімічного складу води під впливом природних та антропогенних чинників у межах його української частини, що й зумовило актуальність даної роботи. Для оцінки навантаження водних об'єктів від дифузних джерел розроблено концептуальну схему, яка дозволяла врахувати основні шляхи надходження речовин - від атмосферних опадів, з водним стоком з орних земель, лісів, луків і пасовищ, забудованих територій, виходів порід, а також від населення сільських регіонів не облаштованих каналізацією. Основним джерелом надходження органічних речовин до поверхневих вод річок Карпатського регіону є господарства в межах сільських поселень, не обладнаних каналізацією. Ключову роль у забрудненні органічними речовинами відіграють басейни річок Прут та Тиса. За джерелами надходження емісія біогенних елементів розподіляється наступним чином. Для річок Тиса та Прут більше 50% загальної емісії сполук нітрогену припадає на території сільськогосподарських угідь, для р. Сірет – 46% - для лісових масивів. За джерелами надходження загальний стік фосфору для річок Тиса та Прут розподілявся наступним чином: домінуюча частка – до 45% - надходить за рахунок внеску сільськогосподарських земель, навантаження від населення, необладнаного каналізаційними системами становить 36%. Для р. Сірет у рівних долях - 33% - серед шляхів надходження сполук фосфору вирізняються сільськогосподарські угіддя та території, вкриті лісами. Вплив решти чинників є значно меншим.

Ключові слова*: емісія, біогенні елементи, органічні речовини, дифузні джерела, забруднення, антропогенне навантаження*

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