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Methodology for Assessing the Surface Water Pollution by Nutrients

The preparation of the River Basin Management Plan includes an assessment of the human pressure by polluting substances. This paper suggests a methodology for calculating the total budget of nutrients in the river basin using the balance method. We assessed the supply of nutrients from various sources within the catchment area. Point sources include emissions from urban areas as well as industrial and agricultural enterprises. Diffuse sources are divided into those determined by natural background (emission from territories covered by forests, grass vegetation, direct atmospheric depositions on the water surface) and anthropogenic ones (arable land, rural, built-up areas). The developed method can be applied to basins not provided with monitoring data.

Keywords: *nutrients, nitrogen, phosphorus, diffuse and point sources, load, surface water.*

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Методика оцінки навантаження поверхневих водних об'єктів біогенними елементами

Підготовка Плану управління річковим басейном включає оцінку антропогенного навантаження забруднюючими речовинами. У роботі запропоновано алгоритм розрахунку загального бюджету біогенних елементів у річковому басейні з використанням балансового методу. Оцінюється надходження від різних джерел, розташованих у межах водозбору. Серед точкових джерел розглянуто емісію від міського населення та промислових і сільськогосподарських підприємств. Дифузні джерела розділено на ті, що визначаються природними умовами (емісія з територій, вкритими лісами, під трав'яною рослинністю, атмосферні випадіння на водну поверхню), та антропогенні (орні землі, сільське населення, забудовані території). Розроблений метод може бути застосований для басейнів, не забезпечених даними моніторингу.

Ключові слова: *біогенні елементи, нітроген, фосфор, дифузні та точкові джерела, навантаження, поверхневі води.*

1. The relevance of the research topic

The development of human society goes along with an increase in anthropogenic impact on the environment, and one of its manifestations is a shift in the global cycle of nitrogen (N) and phosphorus (P). Nowadays, the planetary amount of reactive N has tripled the ability of the Earth's ecosystems to dispose of it, and the amount of P is rapidly approaching the limit values ***[1, 2]***. Inevitably,

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nitrogen and phosphorus content has been gradually increasing in water bodies beginning from the industrial period and continuing these days ***[3]***. The European Environment Agency (EEA) has reported that the increased content of nutrients in surface waters is one of the most important manifestations of man-caused pressure, which results in a failure of the "good" ecological status for 60% of surface water bodies in Europe ***[4]***.

Since the 1960s, there has also been a steady increase in concentrations of nitrogen and phosphorus compounds in the surface water of Ukraine, reaching their highest content in the early 1990s ***[5]***. The next decade was a political restructuring in Ukraine, accompanied by a significant economic crisis and a decline in industrial and agricultural production. As a result, the concentrations of N and P in water bodies decreased rapidly, but they still have yet to reach their natural level ***[6]***.

Nitrogen and phosphorus belong to the group of nutrients vital for the development of living organisms. When their concentrations in a water object increase, they stimulate a number of interconnected processes generally described as eutrophication.

Under undisturbed conditions, the nutrients content is usually low, ensuring the balanced development of the ecosystem. However, increased nitrogen and phosphorus concentrations stimulate the rapid reproduction of autotrophic hydrobionts with subsequent accumulation of organic matter ***[7, 8]***. The consequences of this process include the algal blooming, deterioration of water quality, and undesirable imbalance of organisms in the water object.

Today, the process of water eutrophication has reached a global scale ***[9]***, while control over the nutrients content in water is one of the important factors for ensuring the normal functioning of aquatic ecosystems and maintaining water quality. To solve the problem of water pollution in the EU, a legal framework that includes the Directive concerning urban wastewater treatment ***[10]***, the Nitrates Directive ***[11]***, the Water Framework Directive (WFD) ***[12]***, and the Marine Strategy Framework Directive ***[13]*** has been developed. After Ukraine signed the Association Agreement with the EU, these regulatory acts joined the national legislation and are used to manage water resources.

Implementing these Directives in the EU countries helped reduce the nutrients pollution rate in water objects. However, despite tremendous efforts, the problem of nutrient water pollution is still unsolved ***[14–16]***.

The increase in the content of nitrogen and phosphorus compounds in water is associated with various factors, including population growth and its accelerated migration to cities, improper treatment of municipal and industrial wastewater, accumulation of waste, intensive agricultural production, inefficient land use, etc. In different countries, the role of specific factors differs significantly. In EU countries, after the renovation of treatment facilities, 50% of the anthropogenic supply of nutrients is currently associated with agricultural activity ***[4, 17]***. In the so-called Eastern Partnership countries (Ukraine, Moldova, Georgia, Armenia, and Azerbaijan), improper wastewater treatment also plays a significant role ***[18]***.

Nutrients enter surface water bodies from the quasi-stationary point and non-stationary distributed (diffuse) sources. The point sources include direct wastewater discharges from settlements and industrial and agricultural enterprises. The distributed sources include the nutrients leaching from the catchment area, which quantitatively depends on the combined effect of runoff, landscape characteristics of the basin, and anthropogenic pressure. While point sources are always associated with human activity, the influence of diffuse sources is determined by the joint action of natural and anthropogenic factors.

In order to successfully manage nutrients water pollution, it is important to establish quantitative characteristics of the total anthropogenic load within the catchment area, determine its spatial distribution and primary sources of formation, and impact on the aquatic ecosystem.

In Ukraine, the nutrient release from point sources was traditionally monitored, while researchers mainly neglected the diffuse flows of nutrients.

The purpose of this work is to develop a methodology of quantitative determination of the total nutrients load on the catchment, taking into account point and diffuse sources, to develop methods of river basin management.

2. Research materials and methods

This work is based on the results of long-term observations of hydrological and hydrochemical parameters of the water monitoring network of the State Emergency Service of Ukraine, the chemical composition, and the amount of atmospheric precipitation. Data on the pollutants sewered by individual water users are taken from the State Water Cadaster of Ukraine, the *Water Use* section for 2018 (https://www.davr.gov.ua). The intensity of agricultural production is presented by administrative districts based on statistical data on fertilizer use, manure application, the yield of certain crops, and indicators of animal husbandry for 2018 ***[19]***.

We also used geodata on Ukraine's hydrographic and administrative zoning, population, land use types, and average annual indicators of the runoff depth.

Results and discussion

Until now, only nutrients transportation through the closing cross cross-section of the river basin was evaluated in Ukraine, measured in absolute and relative values: $t \cdot year^{-1}$ and $t \cdot year^{-1} \cdot km^{-2}$, respectively ***[20]***. The indicated data gave an idea of the total amount of transported substances by runoff into the main receiving water body and its interannual variability. However, it did not allow for determining the factors and primary sources of the formation of the nutrients load and its spatial pattern. The transition to the ecological principle of water resources management prompts a detailed analysis of the formation of the general nutrients budget. On its basis, measures aimed at reducing anthropogenic water pollution have been developed.

In world practice, emission flows of nutrients are most often calculated using mathematical models. Among them, the SWAT model stands out in terms of frequency of use and completeness of description of physical processes ***[21]***. At the same time, the model approach requires a large amount of input data and qualified personnel and includes a long process of model calibration and verification. A significant advantage of this approach is the high spatial and temporal discretization of the obtained results and the possibility of forecasting.

Along with the model method, a simpler calculation method is also used. It has successfully proven itself in the water management of large river basins ***[22–26]***. Its main disadvantages include the simplified interpretation of the landscape, the dependence on the available monitoring data, and the use of a large number of empirical coefficients.

Despite the differences in the values of the total nutrient budget, both methods similarly evaluate the relative role of individual load factors ***[27]***, and the choice depends on the set of tasks. Thus, it is a good practice to use the calculation method to carry out general nutrients budgeting and identify local load sources while in critical areas implementing modeling for in-depth study of migration flows of nutrients and development of measures to reduce water pollution is preferable. ***[27]***.

3.1. The primary sources of nutrients supply within the river catchment area

The chemical composition of surface waters reflects the combined effect of the processes of the near-surface moisture cycle, taking into account the influence of natural landscape-geochemical factors and human economic activity.

We determined the total load of nutrients in the closing cross section as the total flow of N and P coming from different sources within the catchment area.

The total impact of point sources depends on water drainage of sewage treatment facilities in settlements and industrial and agricultural enterprises.

Commonly, the diffuse supply of substances into a water body is mainly controlled by the landscape diversity of the catchment ***[28]***, which we reclassified into 5 main classes: forests, meadows and pastures, arable land, built-up areas, and water surface. Atmospheric precipitation on the water surface and wastewater from settlements without sewage networks were also included in the diffuse sources of nutrients supply. A dimensionless retention coefficient characterized the cumulative action of basin transformation processes.

Therefore, the total nutrients balance within an individual catchment is determined by the following equation (1):

$$E_{\mathrm{N,P}} = (E_{\mathrm{N,P\,popul.}} + E_{\mathrm{N,P\,industr.}} + E_{\mathrm{N,P\,forest}} + E_{\mathrm{N,P\,grass\,veg.}} + E_{\mathrm{N,P\,arable}} + E_{\mathrm{N,P\,build\text{-}up}} + E_{\mathrm{N,P\,atm.}} + E_{\mathrm{N,P\,households}}) \cdot k_{\mathrm{reten.}}, \quad (1)$$

where $E_{\mathrm{N,P\,popul.}}$—is the emission flow of nutrients through treatment facilities of urban agglomerations; $E_{\mathrm{N,P\,industr}}$—is the flow of wastewater through treatment facilities of industrial and agricultural enterprises; $E_{\mathrm{N,P\,forest}}$—is the emission flow of nutrients formed on the territories covered by forests; $E_{\mathrm{N,P\,grass\,veg.}}$—is the flow of nutrients formed on the territories under grass vegetation (meadows and pastures); $E_{\mathrm{N,P\,arable}}$—is the nutrients flow formed on the territories under arable land;

$E_{\mathrm{N,P\,build\text{-}up}}$—is the nutrients flow, which is formed on built-up areas; $E_{\mathrm{N,P\,atm.}}$—is the nutrients flow from atmospheric precipitation depositing on the water surface; $E_{\mathrm{N,P\,households}}$—is the flow formed from households that are not connected to sewerage facilities and drain wastewater into individual water-permeable sumps; $E_{\mathrm{N,P\,erosion}}$—the emission flow of nutrients resulting from the erosion process; $k_{\mathrm{reten.}}$—is the integral indicator of basin retention capability.

3.2. Calculation of point sources load

Studies ***[29, 30]*** note a correlation between nitrogen load and population density and establish a regression equation between these parameters. According to the obtained results, an average load formed by 1 person is 3.1–3.8 $\mathrm{kg} \cdot N \cdot \mathrm{year}^{-1}$. At the same time, Stålnacke highlights the significant dispersion of points around the linear dependence within different catchments ***[31]***.

In this regard, to calculate the $E_{\mathrm{N,P\,popul.}}$ indicator, it is worth using the direct calculation of the nutrients supply from the urban residents who live within the studied catchment and discharge wastewater to treatment facilities according to the formula (2):

$$E_{\mathrm{N,P\,popul.}}, [\mathrm{kg} \cdot \mathrm{year}^{-1}] = N \cdot r \cdot Exc_{\mathrm{N,P}} \cdot j, \qquad (2)$$

where $E_{\mathrm{N,P\,popul.}}$—is the nutrient emission from the urban residents that discharge wastewater to treatment facilities, $\mathrm{kg} \cdot \mathrm{year}^{-1}$; N—the total number of the urban population within the river catchment area, ppl; r—is the share of the urban population served by centralized sewage; $Exc_{\mathrm{N,P}}$—is the indicator of N and P excretion from one person, $\mathrm{g} \cdot \mathrm{day}^{-1} \cdot \mathrm{ppl}^{-1}$; j—degree of nutrients removal by treatment facilities, %.

The number of urban population is calculated based on the geodata of administrative zoning and river basin boundaries. By the criteria of Directive 91/271/EEC concerning urban wastewater treatment ***[10]***, the population was reclassified into urban residents, who live in settlements with a population of > 2,000 ppl, and rural — < 2,000 ppl. The number of urban residents that use sewage networks with subsequent treatment of wastewater at treatment facilities is determined based on the data from local water utilities. The share of the urban residents, who are not served by sewage, is added to the number of rural residents.

The physiological coefficients of one person's excretion of nitrogen and phosphorus compounds are stable and provided in reference sources ***[4]***. Considering the smaller number of protein-containing products in the food basket of the Ukrainian population compared to the general European population, we adjusted these coefficients based on the chemical analysis of wastewater to the following values: $\mathrm{N_{gen.}} = 7.3\ \mathrm{g\,N} \cdot \mathrm{day}^{-1} \cdot \mathrm{ppl}^{-1}$; $\mathrm{P_{gen.}} = 1.5\ \mathrm{g\,P} \cdot \mathrm{day}^{-1} \cdot \mathrm{ppl}^{-1}$.

The degree of nutrient removal by treatment facilities depends on the treatment technology. In Ukraine, mechanical (primary) and biological (secondary) treatments are used. By mechanical treatment, 10% of $\mathrm{N_{gen.}}$ and $\mathrm{P_{gen.}}$ are removed, while biological wastewater treatment technology allows disposing of 20% of $\mathrm{N_{gen.}}$ and 30% of $\mathrm{P_{gen.}}$.

The load from industrial and agricultural enterprises is based on the data from the *Water Use* section of the State Water Cadastre (https://www.davr.gov.ua).

The total load from point sources is the sum of the urban population's contributions and industrial and agricultural enterprises.

3.3. Calculation of diffuse sources load

The supply from diffuse sources is determined by the amounts of nutrients in the landscape elements, the intensity of anthropogenic activity, and the volume of runoff that transports substances from the subsurface to the river network. The total contribution from areas covered by forests, meadows and pastures covered by grass vegetation, as well as atmospheric precipitation on the water surface, is associated with the action of natural factors and can be used to calculate the background concentrations of nutrients.

To calculate the runoff of nutrients from forest areas, we use the following formula (3):

$$E_{\mathrm{N,P\,forest}}, [\mathrm{kg} \cdot \mathrm{year}^{-1}] = F_{\mathrm{forest}} \cdot h \cdot C, \qquad (3)$$

where F_{forest}—is the area of catchment covered by forests, km^2; h—is the annual average layer of runoff, mm; C— is the concentration of nutrients ($\mathrm{N_{gen.}}$ and $\mathrm{P_{gen.}}$) from the forest areas in water runoff, $\mathrm{mg\ N,P\ dm^{-3}}$.

Data on the concentration of nitrogen ($\mathrm{N_{gen.}}$) and phosphorus ($\mathrm{P_{gen.}}$) compounds in the runoff formed within forest landscapes are taken from scientific publications ***[24, 25, 26, 32, 33]***. To perform the calculations, we determined the following concentrations: 0.7 mg $\mathrm{N \cdot dm^{-3}}$ and 0.05 mg $\mathrm{P \cdot dm^{-3}}$.

To calculate the runoff from areas covered with grass vegetation, we used the formula (4):

$$E_{\mathrm{N,P\,grass\,veg.}}, [\mathrm{kg \cdot year^{-1}}] = F_{\mathrm{grass\,veg.}} \cdot h \cdot C, \qquad (4)$$

where $F_{\mathrm{grass\ veg.}}$—is the area of the catchment covered by meadows and pastures, km^2; h—is the annual average layer of runoff, mm; C—is the concentration of nutrients in runoff formed on meadows and pastures, mg $\mathrm{N,P \cdot dm^{-3}}$. Concentrations of elements were taken from the scientific publications mentioned for forest areas: 1.58 mg $\mathrm{N \cdot dm^{-3}}$ and 0.13 mg $\mathrm{P \cdot dm^{-3}}$ for $\mathrm{N_{gen.}}$ and $\mathrm{P_{gen.}}$, respectively.

Nutrients supply to the water surface with atmospheric precipitation is recommended to calculate by the formula (5):

$$E_{\mathrm{N,P\,atm.}}, [\mathrm{kg \cdot year^{-1}}] = F_{\mathrm{wtr\,surf.}} \cdot P \cdot C, \qquad (5)$$

where $F_{\mathrm{wtr\ surf.}}$—is the water surface area, km^2; P—is the annual amount of precipitation, mm; C—is the annual average concentration of BE in precipitation, mg $\mathrm{N,P \cdot dm^{-3}}$.

We used adjusted-for-wind data on the amount of precipitation, obtained from the corresponding map, and brought to the basin boundaries by the method of interpolation. The content of nutrients in precipitation was estimated according to the data of the state precipitation monitoring system.

The water surface area of lakes is taken from reference literature or geodata. In contrast, the water surface area of rivers is calculated based on the data on their length and width. The length of the rivers was determined from geodata, and the width is most often available only for tributaries of the 1st order. To determine the width of the tributaries of the 2nd and 3rd order, we have assumed that the complex of water-erosion processes within the catchment is closely related to the structure and morphometry of the river network ***[34, 35]***. Using the Horton-Strahler theory, we have studied the regularities of connection between the main geomorphological characteristics of river systems and the structure of river basins. This theory states that the river's width increases proportionally with the increase of its order ***[36]***.

To calculate the average width (B_i) of the 2nd-order watercourse, we used the formula (6):

$$B_i = \frac{B_\Omega \cdot \sum\limits_{l=1}^{i} \overline{L_{ci}}}{\sum\limits_{l=1}^{\Omega} \overline{L_{ci}}}, \qquad (6)$$

where L_{ci}—is the average length of the watercourse of the 2nd order determined from the digital map of the hydrographic network; B_Ω—is the width of the river of the 1st order in the closing cross section according to the reference literature.

Also, the width of the river can be calculated using the dependencies obtained ***[37]*** for the main river (7) and its tributaries (8).

$$B_{\mathrm{main\,river}}, [\mathrm{m}] = 0.26 \cdot S^{0.49} \cdot M^{0.45} \cdot I^{-0.025}, \qquad (7)$$

where $B_{\mathrm{main\ river}}$—is the width of the main river, m; S—is the catchment area of the river, km^2; M—the runoff modulus, l $\mathrm{s^{-1} \cdot km^{-2}}$, I—is the average slope of the catchment, %.

$$B_{\mathrm{tributary}}, [\mathrm{m}] = 0.082 \cdot S^{0.0395} \cdot M^{1.545} \cdot I^{-0.025}, \quad (8)$$

The keys for the formula (8) are the same as for (7).

The total budget of nutrients from forest areas, meadows, and pastures, and supply with atmospheric precipitation represents the emission flow of nutrients as of natural origin.

The anthropogenic influence is reflected in the emission flows of nutrients from the following sources: supply from arable lands and built-up areas, as well as from the residents of rural settlements.

Supply from the built-up areas $E_{\mathrm{N,P}}$ build-up represents nutrients emissions from urban areas covered with a mostly waterproof surface (asphalt, concrete, restrained urban conditions). The amount of pollutants in such areas is determined by the accumulated dust, dry deposits from the atmosphere, and debris washed away during the formation of showers and is calculated by the formula (9).

$$E_{\mathrm{N,P\,build\text{-}up}}, [\mathrm{kg \cdot year^{-1}}] = F_{\mathrm{build\text{-}up}} \cdot h \cdot C, \qquad (9)$$

where $F_{\mathrm{build\text{-}up}}$—is the built-up area, km^2; the rest of the keys are similar to those for formulas (3) and (4). The concentrations of $\mathrm{N_{gen.}}$ and $\mathrm{P_{gen.}}$ in the sewage runoff of urban agglomerations are 2.3 mg $\mathrm{N \cdot dm^{-3}}$ and 0.2 mg $\mathrm{P \cdot dm^{-3}}$, respectively ***[24, 38]***.

An important source of diffuse water pollution are the rural residents and part of the urban residents, who do not have access to sewage networks and drain wastewater into individual permeable septic tanks. The total supply of nutrients from this source is calculated according to the formula (10).

$$E_{\mathrm{N,P\,households}}, [\mathrm{kg}\cdot\mathrm{year}^{-1}] = N \cdot Exc_{\mathrm{N,P}} \cdot (1-s), \quad (10)$$

where $E_{\mathrm{N,P\ households}}$ — is the total supply from households without sewage network; N—is the number of the population that discharges wastewater into individual septic tanks, ppl; $Exc_{\mathrm{N,\,P}}$—is the rate of N and P excretion from one person, $\mathrm{g}\cdot\mathrm{day}^{-1}\cdot\mathrm{ppl}^{-1}$; $\mathrm{N_{gen.}} = 7.3\ \mathrm{g\ N}\cdot\mathrm{ppl}^{-1}\ \mathrm{day}^{-1}$; $\mathrm{P_{gen.}} = 1.5\ \mathrm{g\ P}\ \cdot \mathrm{ppl}^{-1} \times \mathrm{day}^{-1}$; s—is the dimensionless soil retention coefficient, which represents the total effect of the processes of BE transformation in soils. BE in soils is contained in the absorption complex, soil solution, and organic matter and can be presented in various species. The speed and transformation of individual species of BE depends on several factors, the total effect of which is considered by applying the appropriate coefficients ***[39]***. Thus, according to ***[37, 38]***, the soil retention coefficients for nitrogen and phosphorus are 0.8 for $\mathrm{N_{gen.}}$ and 0.9 for $\mathrm{P_{gen.}}$, respectively.

According to numerous publications, arable lands are the main factor in surface water pollution by BE. Population growth encourages producers to increase agricultural production and implement intensive technologies, which include a high-dose mineral fertilizer application. In the last 30–40 years, there has been an increased loss of nutrients in agricultural areas via surface runoff and infiltration into groundwater. Many studies indicate that more than half of anthropogenic changes in the nitrogen cycle are caused by synthetic inorganic fertilizers application ***[40–42]***. Reducing the diffuse pollution by nutrients from the agricultural activity is one of the most important challenges in the field of water resources management now. In EU countries, unlinking the increase in agricultural production and the growth of surface water pollution is a new and ambitious task ***[43]***.

$E_{\mathrm{N,P}}$ arable is the emission flow of BE, which is formed on the arable land. It is calculated according to the same logic as other land use types (11).

$$E_{\mathrm{N,P\,arable}}, [\mathrm{kg}\cdot\mathrm{year}^{-1}] = F_{\mathrm{arable}} \cdot \mathrm{NB(PB)} \cdot R, \quad (11)$$

where F_{arable}—is the area of arable land, km^2; NB(PB)—is the nitrogen (phosphorus) balance in the agricultural system, $\mathrm{kg}\cdot\mathrm{ha}^{-1}$, R—is the export coefficient by runoff. The value of this coefficient depends on such parameters as the size of the river basin, the origin and amount of precipitation, soil texture, landscape indicators, and agricultural techniques. Depending on the set of conditions, it can vary from 0.08 to 0.8 for nitrogen and from 0.0001 to 0.1 for phosphorus. The general methodology for its calculation is outlined in ***[44]*** and with improvements in ***[45]***. Export coefficients adjusted to the conditions of Ukraine are presented in ***Table 1***.

Table 1. **Export coefficient of nutrients from arable lands for the conditions of Ukraine**

Precipitation, mm · year^{-1}	Soil texture			
	Sand	Loam	Silt	Clay
Nitrogen				
0–600	0.35	0.32	0.30	0.26
600–1200	0.44	0.41	0.38	0.35
Phosphorus				
0–600	0.0375	0.033	0.029	0.025
600–1200	0.0499	0.045	0.0415	0.0374

If calculation is necessary within the farms, R is determined considering additional factors that reflect local conditions.

At the initial stage, researchers tried to separately estimate the emission flow of nutrients from areas under different crops, which significantly complicated the calculation process ***[24]***. Later, they came up with the integrated parameter of Nitrogen Balance (NB), which represents the total ratio between nitrogen supply from various sources (mineral fertilizers, manure) and its absorption by crops. From an ecological point of view, NB differs from the agricultural balance. A positive NB value is a surplus formed within the river catchment, which determines the quantitative nitrogen loss to the environment.

The EEA and the Organization for Economic Cooperation and Development (OECD) use NB as the main criterion for analyzing the risk of water pollution by agricultural lands ***[46, 47]***. Phosphorous Balance (PB) is also evaluated, aiming for water resources management.

The input information for calculating NB and PB is the state statistics data in the crop and livestock industries. The input components include mineral and organic fertilizers application, atmospheric precipitation, and symbiotic and non-symbiotic fixation.

The total balance of nitrogen and phosphorus compounds in the soil is calculated by the formula (12):

$$\begin{aligned}&\mathrm{NB(PB)}, [\mathrm{kg}\cdot\mathrm{ha}^{-1}] = \\ &\quad ((Mf_{\mathrm{N,P_2O_5}} + Of_{\mathrm{N,P_2O_5}} + N_{\mathrm{atm.}} + N_n + F_n) - \\ &\qquad\qquad Y_{\mathrm{N,P_2O_5}})/F_{\mathrm{arable}} \quad (12)\end{aligned}$$

where NB(PB)—is the balance of N and $\mathrm{P_2O_5}$ formed in the soil system during the year, $\mathrm{kg}\cdot\mathrm{ha}^{-1}$; $Mf_{\mathrm{N,\ P_2O_5}}$—is the amount of N and $\mathrm{P_2O_5}$ supplied by mineral fertilizers, $\mathrm{kg}\cdot\mathrm{ha}^{-1}$; $Of_{\mathrm{N,\ P_2O_5}}$— is the amount of N and $\mathrm{P_2O_5}$ supplied by organic fertilizers, kg · ha^{-1}; N$_{\text{atm.}}$—is the supply of nitrogen compounds on the soil surface with atmospheric precipitation, kg N · ha^{-1}, phosphorus is not taken into account; N_{n}—is the non-symbiotic supply of nitrogen, kg · ha^{-1}; F_{n}—is the symbiotic nitrogen fixation by legumes or perennial grasses, kg · ha^{-1}; $Y_{\text{N, P}_2\text{O}_5}$—removal of nitrogen and phosphorus compounds with the crop yield. Consumption coefficients of different crops are taken from ***[48]***, and yield indicators are obtained from official statistical data.

The calculation of nitrogen and phosphorus supply and removal is carried out by individual crops and grouped by administrative districts, and then the obtained results are associated to river catchments. Since phosphorus is used in the form of P_2O_5 compound in agriculture, the calculated PB is associated to P by applying a coefficient of 0.436.

The contribution of mineral fertilizers is calculated according to available statistical data on crop production ***[19]***. Data on the population of individual groups of animals are used to calculate the supply of nutrients with organic fertilizers; as a result, it helps to calculate manure production. Using reference data on the content of nitrogen and phosphorus compounds in manure ***[49]***, we calculate the total supply of nutrients with organic fertilizers by the formula (13):

$$Of_{\text{N, P}_2\text{O}_5} = \frac{\text{LP}_{(\text{cows, calves, pigs, sheep \& goats, poultry})} \cdot X}{F_{\text{arable}}}, \qquad (13)$$

where $Of_{\text{N, P}_2\text{O}_5}$—is the amount of N and P_2O_5 supplied by organic fertilizers, kg/ha; LP—is the livestock heads by main groups (cows, calves, pigs, sheep and goats, poultry), number of heads; X—is the content of N, P_2O_5 in the manure of individual groups of animals, kg · t^{-1}; F_{arable} —total sown area, ha.

The supply of nitrogen compounds with atmospheric precipitation is calculated based on information about the annual amount of precipitation and the content of nitrogen compounds in it (14).

$$N_{\text{atm.}}, [\text{kg N} \cdot \text{ha}^{-1}] = A_{\text{N}} \cdot P \cdot 1\,\text{ha}, \qquad (14)$$

where N$_{\text{atm.}}$—is the supply of nitrogen compounds with atmospheric precipitation, kg N · ha^{-1}; P—the annual amount of atmospheric precipitation, mm; A_{N}—is the average annual content of $(\text{N–NO}_3^-)+(\text{N–NH}_4^+)$ nitrogen compounds in atmospheric precipitation, mg N · dm^{-3}.

Quantitative indicators of precipitation were calculated with correction for wind ***[50]***. Taking into account climate changes, the average multi-year amount of precipitation was calculated from 1990 through 2021 and clustered by physical and geographical zones.

Non-symbiotic nitrogen fixation (N$_{\text{N}}$) is 22.5 kg N · ha^{-1} for sod-podzolic soils and 31.2 kg N · ha^{-1} and 35 kg N · ha^{-1} for gray forest soils and chornozem, respectively. Symbiotic fixation (F$_{\text{N}}$) under leguminous crops is 0.94 kg N · c^{-1}, and under perennial grasses — 0.33 kg N · c^{-1} ***[48]***.

The influence of erosion processes on nutrients emission is estimated using the soil erosion index and the content of the corresponding elements in the solid phase of the soil by the formula (15):

$$E_{\text{N, P eros}}, [\text{kg} \cdot \text{year}^{-1}] = SE \cdot C_{(\text{N, P})\,\text{soil}}, \qquad (15)$$

where *SE* — is the indicator of erosion processes, t · year^{-1}; $C_{(\text{N, P})\ \text{soil}}$—is the content of nitrogen and phosphorus compounds in soils, mg · kg^{-1}.

The soil erosion index (*SE*) is calculated by the formula (16):

$$SE, [\,\text{t} \cdot \text{year}^{-1}] = SEL \cdot g, \qquad (16)$$

where *SE* — is the indicator of erosion processes, t · year^{-1}; *SEL*—is the soil erosion loss, t · year^{-1}; *g*—is the share of sedimentary rocks reaching the channel system, %.

Soil erosion loss was calculated according to the map published in ***[51]***, while the share of sedimentary rocks reaching the channel system *g*, %—by formula (17) ***[37]***.

$$g, [\%] = 0.006684 \cdot (I_{\text{avg.}}, [\%] - 0.25)^{0.3} \times (20 + F_{\text{arable}}, [\%])^{1.5} \qquad (17)$$

where $I_{\text{avg.}}$—is the average slope of the catchment, %.

The concentration of nitrogen and phosphorus compounds in the soil is calculated experimentally. Based on the analysis of published data, we used an average content of 100 mg · kg^{-1} for phosphorus and 20 mg · kg^{-1} for nitrogen ***[48]***.

3.4. Basin retention of nutrients

The nutrients emission flow formed within the catchment undergoes several biological, chemical, and physicochemical processes in soils, floodplains, wetlands, canal networks, and groundwater. As a result, the calculated emission flow consistently exceeds the values observed during monitoring. As some individual processes are hardly manifested, they are hard to parameterize. Consequently, the integral indicator of basin retention (kret) is most often used to calculate the actual removal of nitrogen and phosphorus.

Basin retention quantitatively characterizes the difference between nutrients emission from various sources and nutrients runoff through the outlet into the main water receiver.

First of all, the nutrients removal is determined by bioassimilation, nitrification, and sedimentation processes. The amount of basin retention has high spatial heterogeneity and temporal variability and is largely controlled by the hydrological and geomorphological parameters of the basin. It is also sensitive to changes in climate, land use and management ***[52, 53]***. In particular, for nitrogen and phosphorus compounds, the fluctuation range of k_{ret}. is 23–84% and 39–72%, respectively ***[54]***. Small watercourses play the most important role in nitrogen retention. These watercourses can rapidly absorb and transform nitrogen due to the high ratio of catchment area to water volume ***[55]***.

Studying 100 river basins different in size, Behrendt and Opitz ***[38]*** developed a statistical model for estimating the nutrient retention, which takes into account water temperature and hydrological parameters among the main factors of the transformation of the nutrients emission flow. The specified model with improvements ***[37]*** is recommended for calculation of k_{ret}.

$$k_{ret\text{TN}} = \left(1 - \frac{1}{1 + 4.74 \cdot e^{0.067T} \cdot HL^{-1}}\right) \qquad (18)$$

where T—is the water temperature, °C; HL—is the hydraulic load, m · year $^{-1}$. This parameter is calculated by the formulas (19) for tributaries and (20) for the main river:

$$HL_{\text{trib}_i} = \frac{Q_i}{F_{\text{water surf.}}}, \qquad (19)$$

$$HL_{\text{main river}} = \frac{Q_{\text{main river}}}{F_{\text{water surf.}}}, \qquad (20)$$

where $F_{\text{water surf.}}$—is the area of the water surface of the i-th tributary or the main river in the outlet, m^2; Q_i—is the annual water flow of the i-th tributary or main river, m^3.

The resulting value of the annual total nitrogen (TN) flow is calculated by the formula (21):

$$LoadTN = EmissionTN(1 - k_{ret}) \qquad (21)$$

The annual flow of total phosphorus (TP) compounds is calculated using formulas (22) for main river and (23) for tributaries, respectively.

$$LoadTP = \frac{1}{1 + 13.3 \cdot HL^{-0.93}} \cdot EmissionTP \qquad (22)$$

$$LoadTP = \frac{1}{1 + 26.6 \cdot q^{-1.71}} \cdot EmissionTP \qquad (23)$$

3.5. Calculation accuracy

Evaluation of the accuracy of the nutrients runoff calculation is carried out based on water monitoring data (24).

$$Runoff_{\text{N, P}}, [\text{kg} \cdot \text{year}^{-1}] = \frac{1}{n} \sum_{i=1}^{n} C_i \cdot Q_i, \qquad (24)$$

It should be noted that the accuracy of such a calculation depends on the completeness of these observations on water's chemical composition. If wastage of water has a discretization of 1 day, then sampling for the chemical composition of water is carried out much less often. Typically, monthly sampling properly reflects changes in water's chemical composition ***[12]***. Otherwise, an acceptable method of filling data gaps (approximating polynomial, regression dependence of substance concentrations on water flow, etc.) should be applied. To overcome this shortcoming, the OSPAR Commission suggests the following normalization procedure ***[56]***:

$$Runoff_{\text{N, P}}, [\text{kg} \cdot \text{year}^{-1}] = \frac{\underline{Q}}{Q_h} \left(\frac{1}{n} \sum_{i=1}^{n} C_i \cdot Q_i\right), \qquad (25)$$

where $\underline{Q}$—the annual average daily water consumption, m$^3 \cdot$ s^{-1}; Q_{h}—the annual average water consumption for the dates of the sampling of water chemical composition.

According to the general practice of evaluating the effectiveness of modeling, the calculation results are considered satisfactory if the deviation is < 25%.

In the case of a significant difference between the calculated value of the nutrients runoff and the one established based on monitoring, adjustments are made to the export coefficient of nitrogen and phosphorus from arable lands, applying additional factors that affect the migration capacity of water within the catchment area (atmospheric precipitation, agricultural approaches, the slope of the basin, catchment area, etc.).

3.6. The scale of the nutrients load calculation

Preparation of the River Basin Management Plan requires the calculation of the total load of the polluting substance within the basin. However, for diffuse pollution, it is usually not possible to find out a clear correlation between indicators in a factor-load dependence since its significance is largely influenced by local landscape conditions and the precipitation origin, which can notably neutralize the effect of such a factor. Therefore, to determine the spatial differentiation of the load within the river basin, the smallest scale should be selected. Normally, the main load of nutrients is determined by catchments of low order (no more than 5 according to the Strahler system), in which the content of nutrients is mainly determined by lateral inflow, and they often have no point sources ***[53]***. At the same time, monitoring of such small watersheds is an exceptional event. To carry out quantitative calculations, it is necessary to focus on the lowest-rank basins, for which monitoring data is available.

4. Conclusions

This methodology was developed to calculate a total load of nutrients for the River Basin Management Plans preparation. It is based on the mass balance of the main supply sources. Among the point sources, we considered the water drainage of nutrients as a part of the wastewater drainage systems of municipal, industrial, and agricultural enterprises. Supply from diffuse sources included both natural and anthropogenic sources of emission flow. The influence of nutrients transformation processes within the catchment was taken into account by using a dimensionless retention coefficient. Based on the obtained results, we can evaluate the role ratio of point and diffuse sources in the formation of nutrients load, establish the natural background values for nutrients concentrations, and also determine the quantitative impact of anthropogenic activity.

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