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Assessment of water pollution with nitrogen and phosphorus along the course of a river: A case study from Northern Bulgaria

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ABSTRACT

The present article seeks to estimate water pollution with nitrogen and phosphorus and to analyse the territorial specifics in the content of those physicochemical variables along the length of a river. As a case area, the watercourse of the Osam River (Northern Bulgaria) is selected. Data concern the values of the following parameters: ammonium nitrogen (N-NH_4), nitrate nitrogen (N-NO_3), nitrite nitrogen (N-NO_2), total nitrogen (N-tot), orthophosphates (P-PO_4), and total phosphorus (P-tot), collected at four sampling points during the period 2015–2020. The assessment is based on the guidelines of Regulation 4/2012 for characterization of the surface waters. The spatial changes in the content of the examined variables are related to land use and for this purpose a map of CORINE Land Cover (2018) is prepared. The resulting information reveals that parameters with the most frequent excesses over the reference norm include N-NO_3, N-tot, P-PO_4, and P-tot, whose highest observed concentrations remain up to four times above the permissible limits for "Good status" recommended in Regulation 4/2012. However, the reported values appear to be lower than those established for previous periods, which shows a positive tendency in water quality. There are also spatial features as moving from upstream to downstream – the upper part is contaminated with P-PO_4 and P-tot due to the release of raw municipal and industrial effluents, while the lower section is loaded with N-NO_3 and N-tot as a result of the excessive fertilization of arable lands. This work enriches past studies with new data for a recent period.

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1. Introduction

The Water Framework Directive (WFD) 2000/60/EC of the European Parliament and the Council of 23 October 2000 aims to protect aquatic bodies from point and diffuse pollution and requires all member states to achieve "good quality" of their water resources. The original target for implementing the set condition was 2015 or at least 2021, but later the deadline was extended to 2027. Despite the considerable efforts made by specialists in this field during the last 20 years, 40% of the European water bodies still do not meet the requirements. Referring to the recent report "European waters: assessment of status and pressures" (2018), published by the European Environment Agency (EEA), one of the most serious environmental issues, causing deterioration in water quality and hindering the European Union (EU) member states to meet the WFD objective, remains nutrient enrichment. This process, originating from increased values of nitrogen and phosphorus, worsens aquatic life, decreases biodiversity, and not least threatens public health (Johnson et al., 2010). As an especially anxious consequence arising from the overloading with nutrient components was stated eutrophication (EEA, 2018). Eutrophication can be both natural and cultural. Natural eutrophication, whose driving forces include erosion, decomposition of biomass, etc., occurs very slowly in aquatic bodies so that maintains native habitats for long terms and not poses harmful effects.

However, as a result of various anthropogenic pressures acting on the environment in the last decades, an accelerated process of degradation of rivers and lakes was established, known as cultural eutrophication (Chislock et al., 2013). Cultural eutrophication is driven by unsustainable practices, such as the excessive use of artificial fertilizers and pesticides in agriculture, the unregulated discharge of raw household and industrial wastewaters, etc., through which too many nutrient components enter into aquatic bodies triggering a range of ecological issues. One such problem includes an overgrowth of algae. In this case, the phytoplankton reproduces rapidly causing a toxic algal bloom, which blocks sunlight under the water surface and so disrupts the ecosystem functions. Then water becomes turbid and looks greenish-brown (Anderson et al., 2002). In addition, the algal bloom uses up all of the available dissolved oxygen. Even when blooms wither, bacterial degradation of their biomass continues to consume oxygen creating a state of hypoxia. When dissolved oxygen declines to hypoxic levels fish and the rest of the aquatic animals suffocate and die (Burkholder, 2001). In short, human-induced eutrophication inflicts an overall form of degradation of aquatic ecosystems, depletes native biodiversity, and facilitates colonization by invasive species (Gallardo et al., 2016).

Consequently, the pollution control and the choice of adequate measures for its mitigation and further prevention are crucial actions facing the EU member states in their collective attempt to cope with the widespread nutrient enrichment. The European region incorporates developed countries with large conurbations, modern industries, intensive farming, etc. However, this anthropogenic influence is a leading source of effluents that enrich streams and lakes with nutrient components, an evidence for which are numerous research articles till now.

Polluted with various forms of nitrogen or phosphorus like nitrates, nitrites, organic phosphorus, and orthophosphates are almost all of the major European rivers, such as Tagus (Cordovil et al., 2018), Seine (Bouleau et al., 2020), Rhine (Kunkel et al., 2020), Elbe (Mayo et al., 2019), Vistula and Oder (Pastuszak et al., 2018), etc. The Danube River, a transboundary watercourse passing through or bordering ten countries, is also one of those hot points not meeting the WFD targets (Radu et al., 2020, Mănoiua & Crăciun, 2021). The river basin is predominantly densely populated, highly industrialized, and occupied by arable lands treated with agrochemical products, which causes contamination with nitrogen and phosphorus all around the catchment area – for critically polluted sections was reported from Austria and Slovakia (Vrana et al., 2014), Hungary (Szabó et al., 2011), Serbia (Takić et al., 2017), and Romania (Zaharia et al., 2022).

The Bulgarian part of the river basin is not an exception. The Danube's tributaries, sourcing from the territory of the country, concentrate a lot of urban and rural settlements, industrial factories, as well as spacious croplands. As a result of this human presence, their waters appear to be seriously loaded with nutrients from decades (Varbanov & Gartsiyanova, 2015).

The Osam River is the second longest tributary of the Danube River in Bulgaria. The major ecological issues in its drainage area arise from the lack of public sewage infrastructure in almost all of the settlements, because of which both domestic and industrial wastewater emitted from towns and villages continues to be directly discharged into the river channel. The diffuse release of agricultural runoff from the surrounding farmlands remains an unsolved problem as well. Those human-related activities contaminate the river waters with ammonia, nitrates, nitrites, and orthophosphates (Gartsiyanova, 2015, 2017). The mentioned works provide valuable data, but refer to previous times and do not allow obtaining up-to-date information concerning the current state of water quality.

In this context, the present paper aims to assess water pollution with regards to nitrogen and phosphorus compounds and to analyze spatial distribution of those variables along the length of the river. The realization of the stated objective is expected to update past studies with findings for a contemporary period.

2. Materials and methods

2.1. Study area

The Osam River is a right tributary of the Danube River with a length of 314 km and an area of the drainage basin of 2824 km^2 (Hristova, 2012). The main watercourse is formed after the confluence of two tributaries – the Beli Osam River and the Cherni Osam River not far from the town of Troyan. The river initially flows north to the town of Lovech, later turns northeast running like this until the town of Levski, at last changing its direction heading northwest and maintaining it to its emptying into the Danube River near the village of Cherkovitsa (Fig. 1).

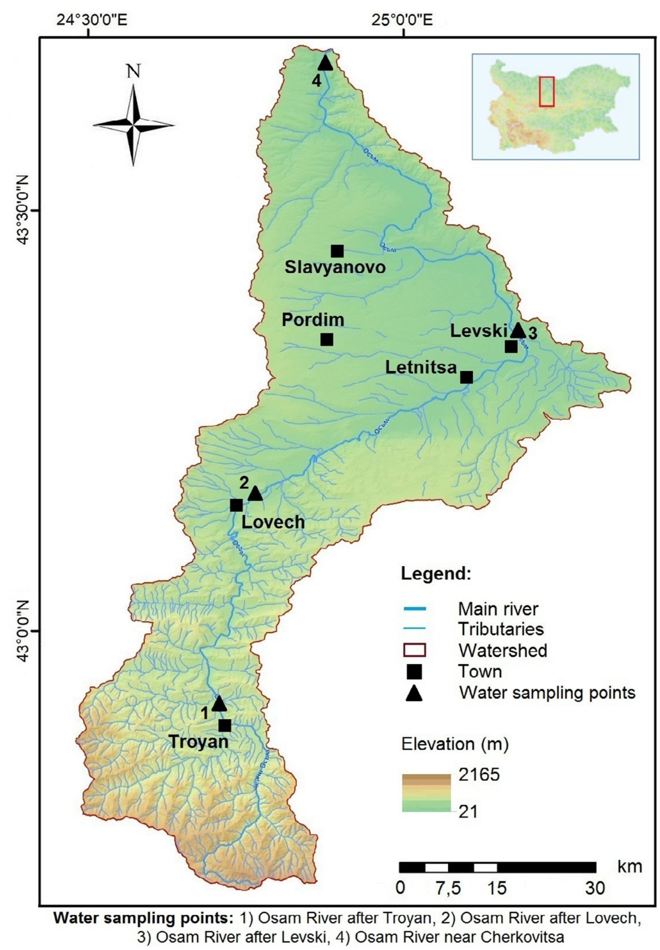

Figure 1. Map of the relief and drainage network in the Osam River basin showing the location of water sampling points

The Osam River basin occupies the central parts of three geomorphological units – the Balkan range mountains (Troyanska Stara planina), the Fore-Balkans (Devetashko Plateau, Mikrenski and Lovchanski hills), and the Danubian Plain. The relief from south to north is mountainous, hilly, and flat. In the same direction are changing the climate conditions. The mean annual air temperature increases from 4–5°C up to 10–11°C, while the amount of precipitation decreases from 1100–1200 mm to 550–650 mm. The hydrological features also vary over territory. The average annual streamflow ranges between 3.42 m^3/s (the Beli Osam River at the hydrometric station in Troyan) and 14.10 m^3/s (the Osam River at the stream gauge near Sanadinovo) with highest discharge volumes from April to June and lowest runoff between August and October (Hristova, 2012). The upper part of the catchment area is covered by centuries-old deciduous forests protected in the Central Balkan National Park, while the flat unit is occupied by arable farmlands with cereal and technical crops. The Osam River basin concentrates 84 settlements, including the towns of Troyan, Lovech, Letnitsa, Levski, Pordim, and Slavyanovo (Fig. 1).

2.2. Methodology

Statistical information, concerning the measured content of six physicochemical variables has been used. Time-series include data about the following water quality parameters: ammonium nitrogen ($N\text{-}NH_4$), nitrate nitrogen ($N\text{-}NO_3$), nitrite nitrogen ($N\text{-}NO_2$), total nitrogen (N-tot), orthophosphates ($P\text{-}PO_4$), and total phosphorus (P-tot). The measurements were carried out by the Executive environment agency at four sampling sites during the period 2015–2020 with a frequency of observations four times per year. The monitoring points, falling within surface water bodies of types R4: Semi-mountain rivers in a Pontic province (measuring sites after the towns of Troyan and Lovech) and R7: Large tributaries of the Danube (sampling points after the town of Levski and near the village of Cherkovitsa), have been selected so that cover parts of upstream, midstream, and downstream reaches of the Osam River (Fig. 1). Water quality has been analyzed following the guidelines of the National regulatory standard – Regulation H-4/2012 for characterization of the surface waters (Table 1). The term “water pollution” will be used in this work when the value of a given physicochemical parameter does not meet the reference norms for “good” and “excellent” status, recommended in Regulation H-4/2012.

Descriptive statistics, i.e. minima, maxima, and means, calculated for each variable have been initially presented and then the spatio-temporal variability of the parameters has been shown. Later the territorial changes in water quality along the length of the river have been related to land use. For this purpose, a map of the CORINE land cover was prepared using the database of COPERNICUS Land Monitoring Services (2018). Finally, the relative share of each one of the land cover classes as a ratio between its area and total catchment area has been computed.

3. Results and discussion

The obtained statistical data from the physicochemical monitoring of the surface waters in the Osam River basin demonstrate significant differences in the quantitative and spatio-temporal features of the examined variables (Table 2). The subsequent analysis confirms this statement.

Results show increased values of P-tot and $P\text{-}PO_4$ in water sampling points of the Osam River after Troyan and after Lovech – up to four times above the reference norms for “good status” recommended for surface water bodies of type R4 (Semi-mountain streams) in Regulation H-4/2012. Frequency analysis indicates that between 25.0% and 70.8% of the recorded samples in those monitoring sites do not meet the requirements, so water quality could be assessed as “moderate” regarding P-tot and $P\text{-}PO_4$. The content of the rest studied variables remain mostly within the reference range for “excellent” or “good status” (Tables 2, 3). Different results are established for downstream. The waters of the Osam River after Levski and near Cherkovitsa are contaminated with $N\text{-}NO_3$ and N-tot, whose highest observed concentrations exceed up to two times the maximum permissible limits for “good status” recommended for surface water bodies of type R7 (Large tributaries of the Danube River) in Regulation H-4/2012. Frequency analysis shows that from 62.5% up to 79.2% of all samples in those monitoring points do not meet the norms, so water quality could be categorized as “moderate” respecting $N\text{-}NO_3$ and N-tot. The concentrations of the rest examined variables almost constantly fall within the range for “excellent” or “good” status (Tables 2, 3).

This result is confirmed by evaluating the territorial distribution of examined variables along the length of the river. The analysis of the spatial changes of the parameters finds the content of $N\text{-}NO_3$ and N-tot obviously rises from upper to lower reaches, while the values of the rest variables increase/decrease from one sampling site to another. Taking into consideration the location of sampling points, the watercourse could be divided into three sectors. The first unit is locked between measuring sites after Troyan and after Lovech. This part is characterized by increasing content of $N\text{-}NO_3$, N-tot, $P\text{-}PO_4$, and P-tot, and decreasing values of $N\text{-}NH_4$ and $N\text{-}NO_2$. The second sector encompasses that segment of the river situated between monitoring points after Lovech and after Levski. There are rising concentrations of $N\text{-}NH_4$, $N\text{-}NO_3$, $N\text{-}NO_2$, and N-tot, and declining content of $P\text{-}PO_4$ and P-tot. The third unit occupies downstream section between sampling sites after Levski and near Cherkovitsa with growing values of $N\text{-}NO_3$, N-tot, and P-tot, and falling content of

Table 1. Reference values of physicochemical parameters at two different water body types as pointed out in Regulation H-4/2012 for characterization of the surface waters

Water body types	Water quality status	Water quality parameters					
		$N\text{-}NH_4$, mg/l	$N\text{-}NO_3$, mg/l	$N\text{-}NO_2$, mg/l	N-tot, mg/l	$P\text{-}PO_4$, mg/l	P-tot, mg/l
R4	Excellent	<0.04	<0.5	<0.01	<0.5	<0.02	<0.025
	Good	0.04–0.4	0.5–1.5	0.01–0.03	0.5–1.5	0.02–0.04	0.025–0.075
	Moderate	>0.4	>1.5	>0.03	>1.5	>0.04	>0.075
R7	Excellent	<0.1	<0.7	<0.03	<0.7	<0.07	<0.15
	Good	0.1–0.3	0.7–2.0	0.03–0.06	0.7–2.5	0.07–0.15	0.15–0.3
	Moderate	>0.3	>2.0	>0.06	>2.5	>0.15	>0.3

$N\text{-}NH_4$, $N\text{-}NO_2$, and $P\text{-}PO_4$ (Fig. 2, Table 2).

The analysis of temporal variability of the parameters does not find clearly expressed seasonal cyclicality in the values (Fig. 3A, B). Exceptions are $N\text{-}NO_2$, $P\text{-}PO_4$, and P-tot (after Troyan), as well as $P\text{-}PO_4$ and P-tot (after Lovech) with higher concentrations in summer, autumn, and winter, and lower ones in spring. Although hydrometric data are not used, knowing the intra-annual runoff distribution of the Osam River, we could summarize that the described seasonal changes in the content of the physicochemical variables are inversely related to flow regime.

Table 2. Descriptive statistics of the values of nitrogen and phosphorus compounds

		Water quality parameters					
Water sampling points	**Values**	**$N\text{-}NH_4$, mg/l**	**$N\text{-}NO_3$, mg/l**	**$N\text{-}NO_2$, mg/l**	**N-tot, mg/l**	**$P\text{-}PO_4$, mg/l**	**P-tot, mg/l**
	Minimum	0.070	0.236	0.004	0.530	0.011	0.017
Osam River after Troyan	Average	0.136	0.685	0.014	0.987	0.031	0.039
	Maximum	0.331	1.570	0.052	2.000	0.080	0.087
	Minimum	0.060	0.430	0.007	0.750	0.018	0.028
Osam River after Lovech	Average	0.125	1.043	0.011	1.582	0.075	0.091
	Maximum	0.273	3.390	0.021	4.300	0.160	0.160
	Minimum	0.050	1.470	0.016	1.670	0.014	0.020
Osam River after Levski	Average	0.187	2.311	0.037	3.150	0.057	0.065
	Maximum	0.376	4.020	0.075	5.160	0.104	0.110
	Minimum	0.040	1.290	0.007	2.000	0.019	0.042
Osam River near Cherkovitsa	Average	0.166	2.534	0.018	3.356	0.056	0.074
	Maximum	0.435	4.380	0.042	5.500	0.099	0.145

Table 3. Frequency (% of all samples) of water quality parameters in a given physicochemical status according to Regulation H-4/2012 for characterization of the surface waters

		Water quality parameters					
Water sampling points	**Water quality status**	**$N\text{-}NH_4$, mg/l**	**$N\text{-}NO_3$, mg/l**	**$N\text{-}NO_2$, mg/l**	**N-total, mg/l**	**$P\text{-}PO_4$, mg/l**	**P-total, mg/l**
	Excellent	–	41.7	37.5	–	45.8	25.0
Osam River after Troyan	Good	100.0	54.1	58.3	95.8	29.2	70.8
	Moderate	–	4.2	4.2	4.2	25.0	4.2
	Excellent	–	8.3	50.0	–	12.5	–
Osam River after Lovech	Good	100.0	83.4	50.0	66.7	16.7	41.7
	Moderate	–	8.3	–	33.3	70.8	58.3
	Excellent	12.5	–	37.5	–	75.0	100.0
Osam River after Levski	Good	70.8	29.2	50.0	37.5	25.0	–
	Moderate	16.7	70.8	12.5	62.5	–	–
	Excellent	29.2	–	8.3	–	83.3	100.0
Osam River near Cherkovitsa	Good	62.5	25.0	91.7	20.8	16.7	–
	Moderate	8.3	75.0	–	79.2	–	–

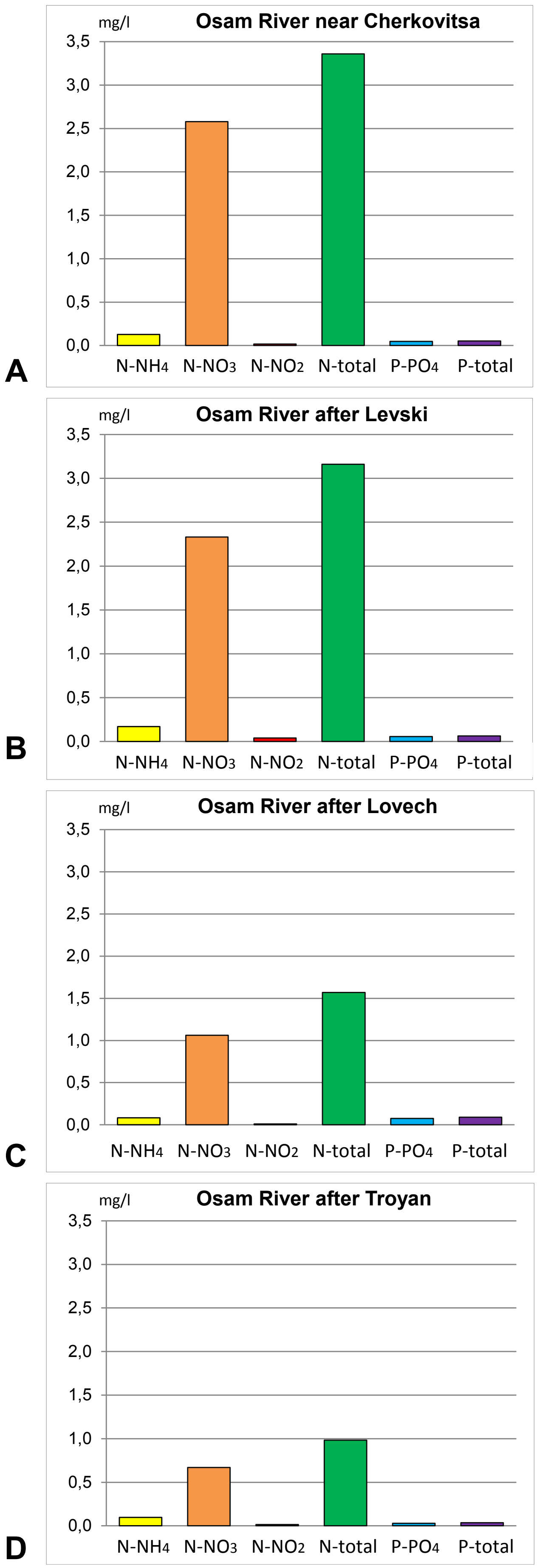

Figure 2. Schematic map of the Osam River basin showing the spatial changes in the content of nitrogen and phosphorus compounds along the length of the river (based on average values)

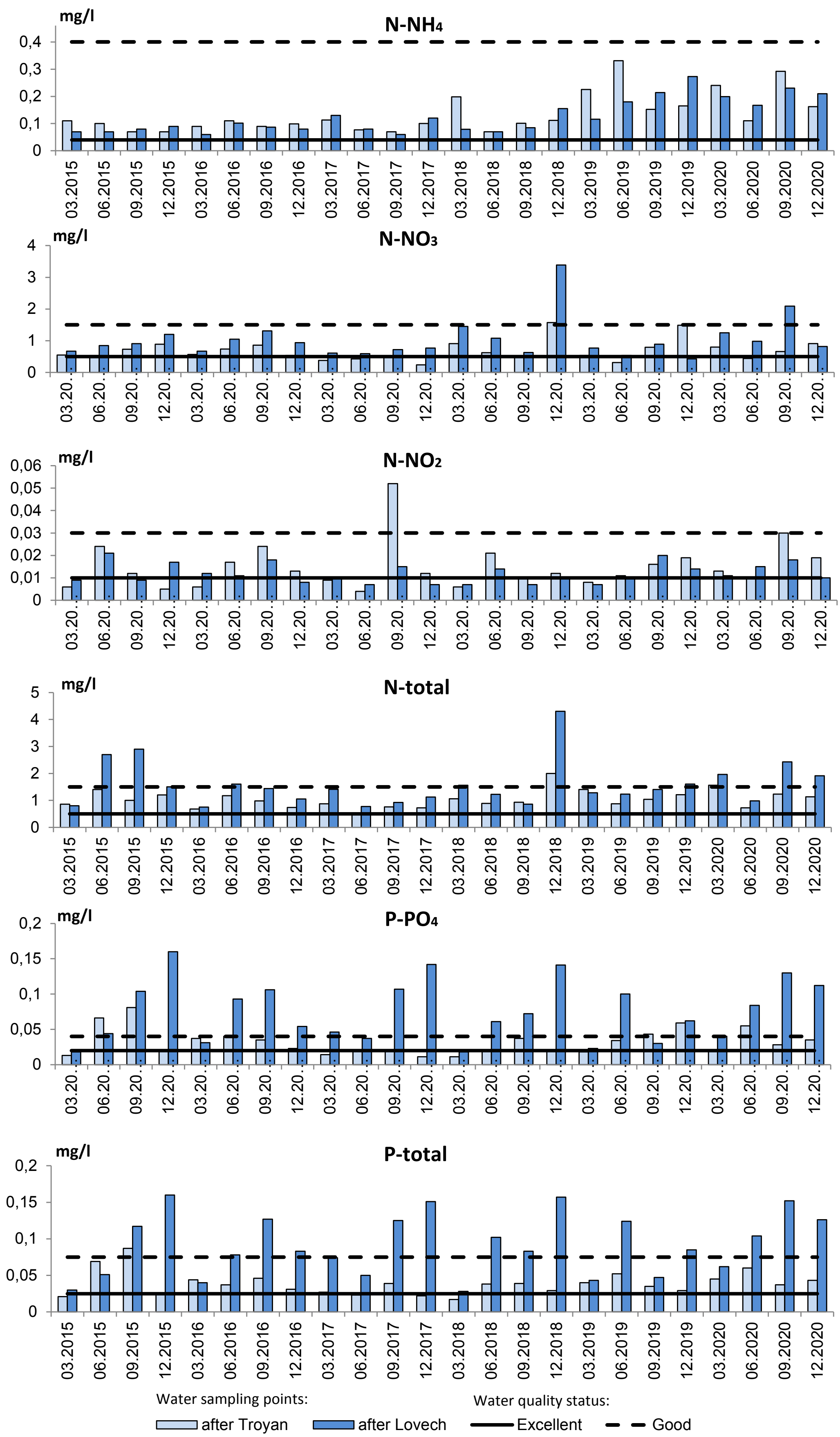

Figure 3A. Dynamics in the values of nitrogen and phosphorus compounds compared to the reference norms for surface water bodies of type R4 as recommended in Regulation 4/2012

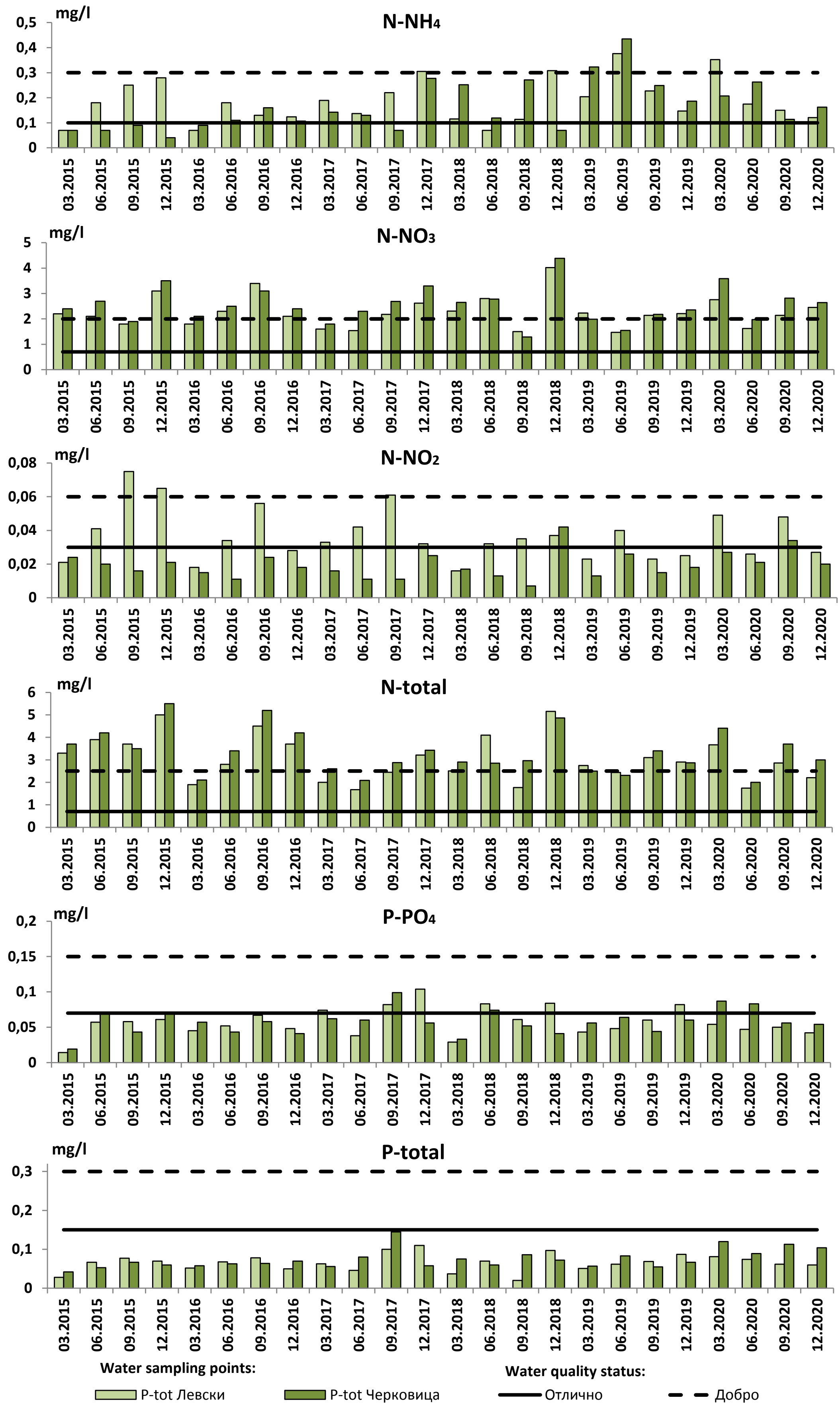

Figure 3B. Dynamics in the values of nitrogen and phosphorus compounds compared to the reference norms for surface water bodies of type R7 as recommended in Regulation 4/2012

Compared to previous studies, the results of this paper outline some similarities, but highlight a lot of differences as well. Gartsiyanova (2015, 2017) and Varbanov & Gartsiyanova (2015) explored the water quality of the Osam River based on input data for the period 1990–2014. Gartsiyanova (2015, 2017) informed for critically elevated values of N-NH_4 and N-NO_3 from 1990 to 1993, for increased content of N-NO_2 between 1994 and 2007, and for contamination with P-PO_4 from 1998 to 2009 at sampling point after Lovech. The cited author established continuous pollution with N-NH_4 and P-PO_4 between 1996 and 2005 at measuring sites after Levski and near Cherkovitsa and pointed out that the maximal recorded values of those parameters exceeded from 10 up to 25 times the highest permissible limits for "good status" recommended in Regulation H-4/2012. Varbanov & Gartsiyanova (2015), using the Canadian Water Quality Index (CWQI), reported the following results.

Water quality after Troyan could be evaluated as "marginal" from 1991 to 2005, later improves its properties achieving "good" condition between 2006 and 2014. According to their physicochemical state, river waters after Lovech almost constantly fall within "poor" and "marginal" categories, as only in 2011, 2013, and 2014 have "good" status. Water quality near Cherkovitsa appears to be "poor" from 1990 to 2002, later achieves "marginal" condition from 2002 to 2010, after 2010 could be assessed as "good", reaching "very good" state in 2014 (Varbanov & Gartsiyanova, 2015). The current work shows different results – the waters after Lovech are contaminated with P-PO_4 and P-tot, while the concentrations of N-NH_4, N-NO_3, and N-NO_2 remain mostly in the reference range for "excellent" or "good" status. In contrast, the waters after Levski and near Cherkovitsa are polluted with N-NO_3 and N-tot, while the values of N-NH_4 and P-PO_4 fall below the highest permissible limits for "good status" (Fig. 3A, B). There is one more discrepancy as well – the maximum content of the physicochemical variables during the period 2015–2020 exceeds by no more than four times the reference standards in Regulation H-4/2012, while Gartsiyanova (2017), working with data for the period 1990–2014, detects excesses of up to 25 times above the same reference norms. This contradiction confirms the already established by Varbanov & Gartsiyanova (2015) a positive trend in water quality and gives us an argument to claim that the Osam River has continued to significantly improve its physicochemical condition between 2015 and 2020, which is probably a result of the deepening demographic crisis in the region.

Water quality is a reflection of both natural processes and anthropogenic activities, occurring in a catchment area. Gartsiyanova (2017) recognizes land use as a key factor determining the qualitative status of surface waters in the Osam River basin. Because of this reason, the results obtained in the current work will be interpreted through the prism of land cover classes (Fig. 4).

The CORINE Land Cover map illustrates that the predominant land cover classes in the Osam River basin are "Arable lands, incl.

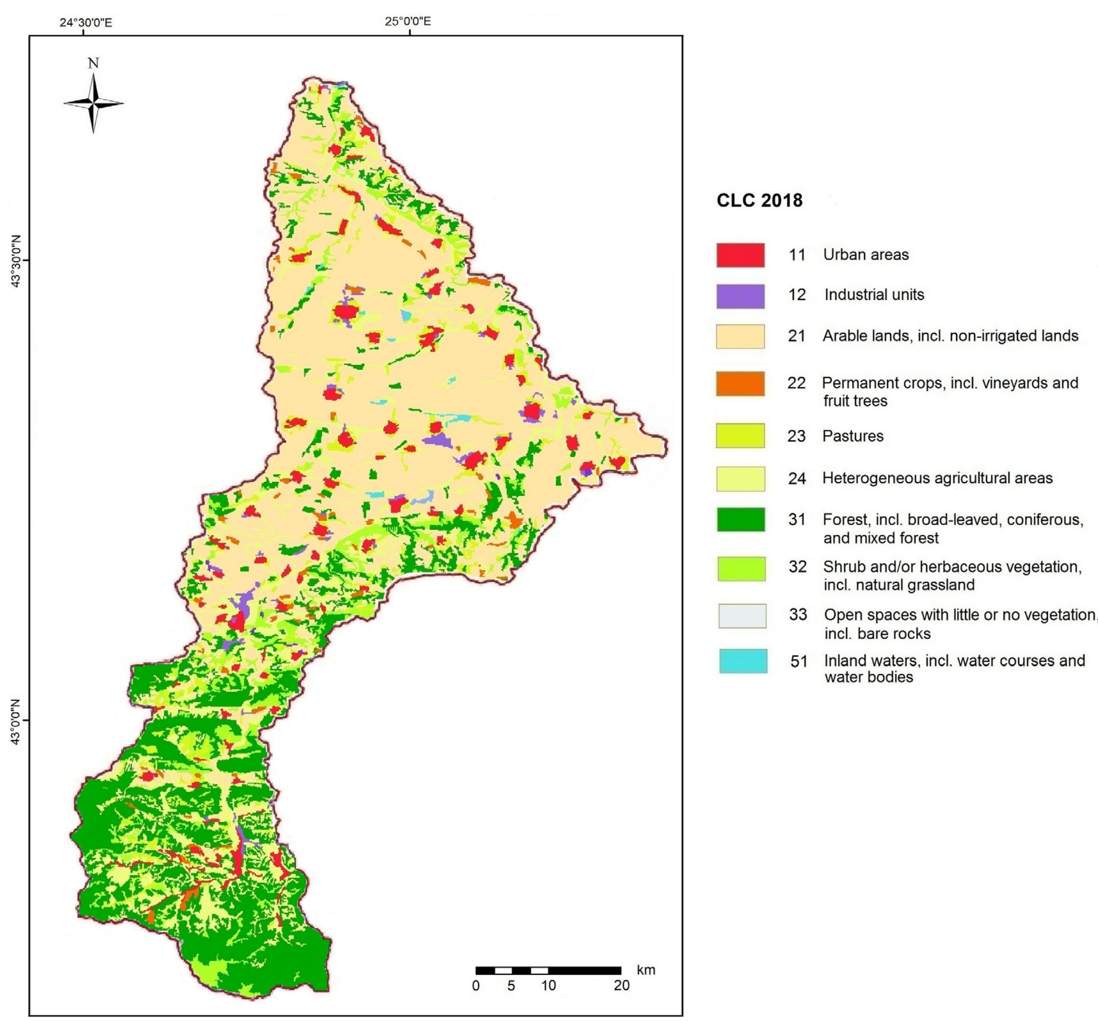

Figure 4. Map of the CORINE Land Cover (2018) in the Osam River basin

Table 4. Relative shares of the CORINE Land Cover classes (% of total catchment area)

CLC Level 1	CLC Level 2	%
1. Artificial surfaces	1.1 Urban areas	3.27
	1.2 Industrial units	0.84
2. Agricultural areas	2.1 Arable lands, incl. non-irrigated lands	57.34
	2.2 Permanent crops, incl. vineyards and fruit trees	0.78
	2.3 Pastures	3.64
	2.4 Heterogeneous agricultural areas	2.92
3. Forests and semi-natural areas	3.1 Forest, incl. broad-leaved, coniferous, and mixed forest	26.51
	3.2 Shrub and/or herbaceous vegetation, incl. natural grassland	4.18
	3.3 Open spaces with little or no vegetation, incl. bare rocks	0.07
5. Water bodies	5.1 Inland waters, incl. water courses and water bodies	0.45

non-irrigated lands", representing 57.34% of the drainage area, and "Forest, incl. broad-leaved, coniferous, and mixed forest", comprising 26.51% of the region, while the rest of the extracted classes have a smaller relative share (Fig. 4, Table 4).

The mountainous part of the Osam River basin is occupied by forest, shrub and/or herbaceous vegetation, incl. natural grassland, pastures, and small-scale heterogeneous arable lands mixed with areas of primary vegetation (Fig. 4). Water pollution in this section arises from natural processes like soil erosion, but most of all it is a result of the pastoral farming practices, which are a source of animal manure and that to some extent explains the increased content of total phosphorus and orthophosphates in the waters of Osam River after Troyan and after Lovech. Additional issues deteriorating upstream water quality include the raw domestic and industrial wastewater released from settlements with lacking or not fully built public sewage networks. Noteworthy examples are the towns of Troyan and Lovech (agglomerations with a population equivalent of greater than 10 000 people) that despite being economic centers with operating chemical and pharmaceutical enterprises, wood-processing manufactories, milk- and meat-processing plants, etc., continue to be served by obsoleted or undeveloped urban sewerage systems and wastewater treatment facilities. In the vicinity there are also the balneology resort villages of Shipkovo and Chiflik, where a plethora of hotel complexes was built in recent years. Consequently, the major sources worsening water quality in this part of the river basin remain the untreated or partly treated effluents emitted from pastures, households, hotels, and industries. Such wastewaters contain both organic matter and synthetic materials, which enrich aquatic streams with phosphorus and its compounds. Downstream water pollution has another origin. The flat part of the catchment area is an agricultural region covered by spacious arable fields (Fig. 4). Water quality declines due to the release of soil runoff saturated with artificial fertilizers and pesticides that are widely applied onto farmlands. Those products are used to be achieved an accelerated yield of crops or the same crops to be protected from pests, but their excessive dispersal causes adverse ecological effects and explains the elevated concentrations of nitrates and total nitrogen in the waters of Osam River after Levski and near Cherkovitsa. In addition, almost all of the settlements, including the town of Levski (an agglomeration with a population equivalent of more than 2000 people), still are not connected to urban sewers and deposit their wastes into illegal rubbish dumps, which also worsens downstream water quality.

4. Conclusion

The conducted research, dedicated to water pollution with nitrogen and phosphorus along the course of a selected river during the period 2015–2020, showed that among six variables, four of them (N-NO_3, N-tot, P-PO_4, and P-tot) most frequently exceed the reference standards for "Good status" outlined in Regulation 4/2012. Compared to past works, the reported values in this study were lower than those stated for previous times, which marked an improvement in water quality. The temporal analysis did not find clearly pronounced seasonal cyclicality in the concentrations. Evaluating the spatial changes of the parameters was established that the content of N-NO_3 and N-tot increased from upstream to downstream, while the loading with P-PO_4 and P-tot decreased following the same direction. Water contamination in upper and middle reaches emerged due to the discharges of inadequately treated effluents emitted from domestic sources, hotel complexes, and industrial factories, while in the lower part pollution was a result of the releasing soil runoff from croplands treated with agrochemical products. In order to limit the inflow of nutrients into the river, it is necessary to expand the public sewage system and wastewater treatment facilities in the agglomerations with a population equivalent of greater than 2000 people as required the Directive 91/271/EEC concerning the collection, treatment, and discharge of the wastewaters released from urban areas and certain industrial sectors.

Efforts should be focused not only on the construction of modern sewerage networks but also on renewing the existing ones by installing trickling filters for extraction and removal of pollutants. Regarding agricultural pollution, stricter compliance with the rules for best farming practices is strongly recommended. The implementation of those advices is expected to improve the physicochemical condition along the watercourse, which would protect aquatic life and human health providing safe access to clean water resources.

This work complements previous articles with knowledge for a contemporary period. Water quality indices for further extension of the obtained results could be calculated.

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