Seasonal variations of nutrients concentration in aquatic ecosystems from Danube Delta Biosphere Reserve

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Abstract. Nutrients (nitrites, nitrates, phosphates) are essentials for organisms life that use photosynthesis. An excessive growth of these aquatic organisms or contrary may cause an imbalance in the entire aquatic ecosystem. Therefore, the present study aims to determine seasonal dynamics of nitrogen concentrations from ammonia, nitrites, nitrates, total nitrogen, phosphorus from orthophosphates, and total phosphorus from 7 deltaic areas that included a number of 37 sampling points, representative for all types of aquatic ecosystems from Danube Delta Biosphere Reserve, during the year 2015. The investigations on nutrients concentrations highlighted significant variations from one season to other and also exceeding's of the maximum allowed concentrations, regulated by existing legislation. The highest exceeding was identified for nitrite concentrations. Concentrations for dissolved forms had minimum and maximum values as follows: nitrogen from ammonia (0.099 mg L^{-1} and 0.267 mg L^{-1}), nitrogen from nitrate (0.164 mg L^{-1} and 1.961 mg L^{-1}), nitrogen from nitrite (0.005 mg L^{-1} and 0.028 mg L^{-1}), dissolved phosphorus (0.005 mg L^{-1} and 0.028 mg L^{-1}). Total forms of nutrients varied in the range of 0.021-0.185 mg L^{-1} for total phosphorus and 3.913-6.359 mg L^{-1} for total nitrogen. Minimum concentrations in maritime area obtained for ammonia, nitrites, orthophosphates and total phosphorus can be attributed to the filter capacity of the Danube Delta.

Key Words: seasonal dynamics, nitrogen, ammonia, nitrites, nitrates, total nitrogen, phosphorus, orthophosphates.

Introduction. Before the period when Danube Delta was declared as Biosphere Reserve, on a background of biogenic elements development in the Danube River, of the flooded areas disappearance, of the agriculture and intensive practiced fishing in the delta but also of the water scarcity in some periods, the lakes in Danube Delta received large amounts of nitrogen and phosphorus that contributed to eutrophication increasing effects in Danube Delta (Gâştescu & Ştiucă 2008).

Nutrients availability is one of the most important factors that adjust macrophyte algae growth and reproduction (Lobban & Harrison 1997). Beside human health risks, nutrients excess lead to an overgrowth development of aquatic organisms through accelerated growth causing in many cases algal blooming. When algal life cycle is ending, large amounts on dissolved oxygen from water is consumed through microbial decomposition, causing death to some fish species and not only. In the same time, deficiency of nutrients limits plants growing capacity.

Nitrogen is found dissolved in water, both in a gas form, originated by diffusion from the atmosphere, or as soluble salt (nitrates and ammonia salts) (Hellawell 1986).

Nitritic form of nitrogen is the most assimilabile; the first step, the oxidation of ammonia to nitrite, is the slowest step. Nitrites come from mineralization of organic substances under the action of bacteria (Mason 2001). Consequently, when nitrite is formed, it is rapidly oxidized to nitrate. Nitrates represent the final form of organic nitrogen oxidation in surface waters, whose presence is determined by water contact with the sediment.

Nitrate concentrations in surface water is dependent on dissolved oxygen concentration, temperature, presence of organic substances and water pH. Under microorganisms action nitrates suffer a reduction process to nitrogen compounds poor or lack of oxygen. Some organic matters that contain nitrogen usually stand to bacterial changes and remain in water or sediments (Riley & Chester 1971). Nitrates concentration in water varies with temperature, concentration of dissolved oxygen from water, presence of organic substances and concentration of hydrogen ions.

Phosphorus is a macroelement with important role in almost all metabolic processes, being, together with nitrogen, the most frequent limitative nutritive factor, in natural biotopes and also in experimental conditions (Neniţescu 1963). It is an important nutrient that can limit algal biomass production, being represented mainly by orthophosphates. Algae take over the phosphorus, as an essential nutrient during photosynthesis.

Some algae are able to ″break″ dissolved organic phosphorus, using alkaline phosphatase and utilize phosphate in the inorganic form. When algae dies or are eaten, organic phosphorus is rapidly transformed in orthophosphate. Phosphorus represents an essential element of organism's growth and can limit phytoplanktonic biomass. Macrophytes, also respond to high amounts of phosphorus in the environment where they live, through high development of biomass.

Material and Method. Surface water samples were collected seasonally (spring, summer, autumn) in 2015, from: pre-deltaic area, fluvial delta, maritime delta, Razim-Sinoe, Chilia Branch, Sulina Branch, Sf. Gheorghe Branch. In Tabel 1 are presented the geographic coordinates of the sampling points.

Table 1

Geographic coordinates of sampling points

Sampling area	*Aquatic complex*	*Sampling points*	*Latitude*	*Longitude*
Predeltaic area	Somova Parcheş	Rotund lake	45^{0}14′04.4″	028^{0}30′52.4″
		Parcheş lake	45^{0}13′36.4″	028^{0}35′34.7″
		Somova lake	45^{0}10′59.9″	028^{0}41′00.6″
		Câsla lake	45^{0}10′39.4″	028^{0}44′41.2″
		Water supply channel Somova-Parcheş	45^{0}14′33.5″	028^{0}34′15.1″
		Water discharge channel Alum	45^{0}11′33.1″	028^{0}44′47.6″
		Water discharge channel Somova-Parcheş	45^{0}12′47˝.3″	028^{0}45′43.9″
Fluvial area	Sontea-Furtuna	Fortuna lake	45^{0}13′13.1″	029^{0}07′35.9″
		Nebunu lake	45^{0}14′56.6″	029^{0}00′12.9″
		Şontea channel	45^{0}14′38.2″	029^{0}04′48.8″
	Gorgova-Isac	Cuibul cu Lebede lake	45^{0}07′46.8″	029^{0}20′21.5″
		Uzlina lake	45^{0}05′23.6″	029^{0}15′48.4″
		Isac lake	45^{0}06″35.5″	029^{0}16′22.1″
		Gorgostel lake	45^{0}03′46.2″	029^{0}19′38.7′
		Litcov channel	45^{0}08′19.2″	029^{0}19′09.3″
		Perivolovca channel	45^{0}03′18.2″	029^{0}18′06.1″
	Matita-Merhei	Merhei lake	45^{0}19′09.2″	029^{0}25′55.6″
		Miazazi lake	45^{0}15′55.9″	029^{0}21′36.5″
		Lopatna channel	45^{0}15′26.0″	029^{0}18′26.9″

Sampling area	Aquatic complex	Sampling points	Latitude	Longitude
Maritime area	Roșu-Puiu	Erenciuc lake	$44^0 59'59.1''$	$029^0 25'44.6''$
		Iacub lake	$45^0 08'33.6''$	$029^0 24'09.1''$
		Rosu lake	$45^0 03'28.4''$	$029^0 34'41.3''$
		Rosulet lake	$45^0 04'13.3''$	$029^0 36'44.6''$
		Crisan-Caraorman channel	$45^0 07'17.7''$	$029^0 23'24.6''$
Razim-Sinoe area	Razim - Sinoe	Razim-Bisericuta lake	$44^0 45'46.5''$	$028^0 57'44.4''$
		Goloviţa lake	$44^0 43'36.5''$	$028^0 54'00.6''$
		Goloviţa lake (Gura Portitei)	$44^0 41'53.5''$	$028^0 59'14.3''$
Chilia branch	Chilia branch	Izmail downstream	$45^0 16'44.7''$	$028^0 56'42.7''$
		Ceatal Chilia	$45^0 13'37.8''$	$028^0 44'05.9''$
		Periprava	$45^0 24'09.6''$	$029^0 33'37.2''$
		Bastroe upstream	$45^0 20'36.2''$	$029^0 38'39.0''$
		Bastroe downstream	$45^0 19'59.0''$	$029^0 39'22.6''$
Sulina branch	Sulina branch	Ceatal Sf. Gheorghe	$46^0 11'12.8''$	$029^0 53'24.3''$
		Sulina upstream	$45^0 09'37.1''$	$029^0 36'32.9''$
		Sulina downstream	$45^0 09'39.4''$	$029^0 40'52.6''$
Sf. Gheorghe branch	Sf. Gheorghe branch	Ceatal Sf. Gheorghe	$46^0 11'12.8''$	$029^0 53'24.3''$
		Sf. Gheorghe upstream	$44^0 53'44.3''$	$029^0 34'49.3''$
		Sf. Gheorghe downstream	$44^0 53'22.2''$	$029^0 35'58.6''$

Nutrient analysis. Surface water samples, collected for nutrients determination were stored at 2-5ºC, for maximum 24 hours before analysis. Determination of nutrients dissolved forms was made on filtered samples and total forms on unfiltered samples using UVVIS Lambda 10 PerkinElmer spectrophotometer.

The ammonium concentration, expressed as $N\text{-}NH_4^+$, was determined at 655 nm by measuring the absorption of the blue compound formed by the reaction of ammonium ion with salicylate and hypochlorite ions in the presence of sodium nitroprusside, according with SR ISO 7150-1, Water quality, Determination of ammonium, Part 1. Manual spectrometric method at UV-VIS Lambda 10 Perkin Elmer Spectrometer (SR ISO 7150-1 2001).

Nitrite ($N\text{-}NO_2^-$) was determined through formation of reddish purple azo dye produced at pH 2.0 to 2.5 by coupling diazotized sulfanilamide with N-(1-naphthyl)-ethylenediamine dihydrochloride, according with SR EN 26667/ISO 6777/2002, Determination of nitrite, Molecular absorption spectrometric method, using the UVVIS Lambda 10 Perkin Elmer Spectrometer at 540 nm. The analysis was made on filtered water (SR EN 26667/ISO 6777/2002).

The nitrate ($N\text{-}NO_3^-$) was determined according with SR ISO 7890-3:2000 – Water quality, Determination of nitrate, Part 3. Spectrometric method using sulfosalicylic acid, by spectrometric measurement of yellow compound absorbance formed by reaction of sulfosalicylic acid (formed by addition of sodium salicylate in the sample and sulfuric acid) with nitrate followed by treatment with alkaline solution, at UVVIS Lambda 10 Perkin Elmer Spectrometer (415 nm) (SR ISO 7890-3 2000).

Determination of phosphorus content in surface waters was made according with SR EN 6878/2005, Water quality, Determination of phosphorus, Ammonium molybdate spectrometric method using UVVIS Lambda 10 Perkin Elmer Spectrometer at 880 nm. For total phosphorus, the samples were treated on unfiltered water. Ammonium molybdate and potassium antimonyl tartrate react in acid medium with orthophosphate to form a heteropoly acid – phosphomolybdic acid – that is reduced to intensely colored molybdenum blue by ascorbic acid (SR EN 6878 2005).

Data analysis. All nutrients concentrations (nitrogen from ammonia, nitrogen from nitrites, nitrogen from nitrates, total nitrogen, dissolved phosphorus and total phosphorus) are expressed in mg L^{-1}.

Results and Discussion. Physical-chemical characteristics and surface water quality in Danube Delta Biosphere Reserve are determined by natural factors (climate, drainage system) and by diverse economic activities which take place in surrounding area. If in the part of fluvial and maritime Delta, physical-chemical composition of water is generally determined by influence of natural factors, in the pre-deltaic area, modifications can be certainty influenced by effects of industrial activities of Tulcea city.

In water, ammonia results from incomplete degradation of organic substances which contain nitrogen or can also come from soil. It represents the first stage of decomposition of organic substances with nitrogen content in their molecule and that indicates a recent pollution (hours-days) and are consequently very dangerous (Mason 2001).

Concentrations of nitrogen from ammonia presented in Figure 1 are lowest in summer season, comparative with spring and autumn seasons, when vegetative processes are high nitrogen consumers. Minimum concentration was determined in pre-deltaic area, in summer (0.047 mg L^{-1}) and maximum concentration was determined in maritime area, in autumn (0.428 mg L^{-1}), values that do not exceed 0.8 mg L^{-1} nitrogen from ammonia, value corresponding to second quality class, according with Order no. 161/2006 of Environment Ministry and Water Management.

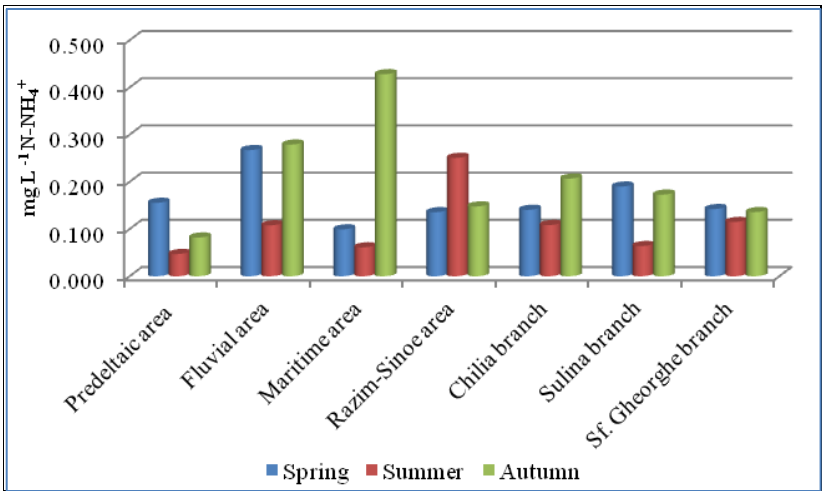

Figure 1. Average concentrations of nitrogen from ammonia determined seasonally, in surface waters from deltaic areas in 2015.

Low values of nitrites concentrations presented in Figure 2, are attributed to oxidative processes in aquatic environment, when ammonia ions transforms in nitrites and nitrates, ranging between 0.008 mg L^{-1} (summer season in fluvial area) and 0.047 mg L^{-1} (autumn season in pre-deltaic area), maximum value obtain, being the only that exceed the quality standard of 0.030 mg L^{-1}, according to good ecological status from Order no. 161/2006 of Environment Ministry and Water Management.

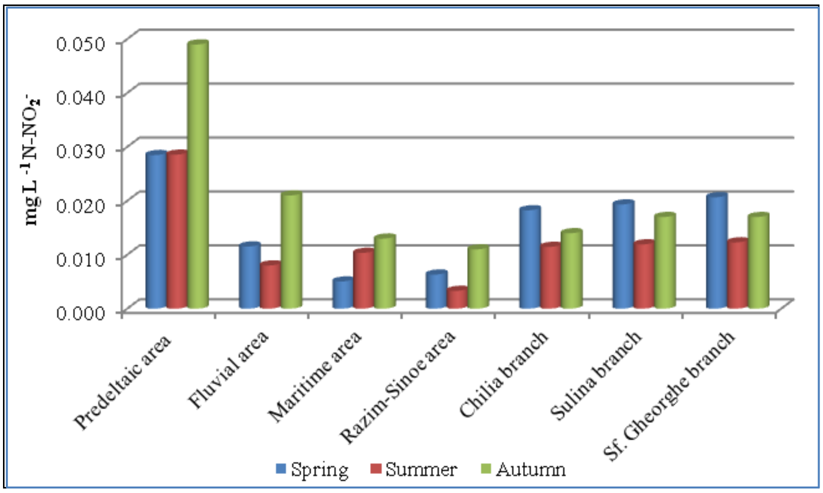

Figure 2. Average concentrations of nitrogen from nitrites determined seasonally, in surface waters from deltaic areas in 2015.

Nitrates, the final form of nitrogen from ammonia oxidation, have higher concentrations than ammonium and nitrites (Laane 1992). Minimum concentrations of nitrates presented in Figure 3, were determined in summer season 2015 and maximum concentrations in the spring of 2015, values decreasing from 2.120 mg L^{-1} (Chilia branch in spring season) to 0.030 mg L^{-1} (Razim-Sinoe area in autumn season), values that do not exceed quality standard of 3 mg L^{-1}.

Nitrogen inorganic forms generally, have higher values on Danube branches, compared to those obtain in pre-deltaic, fluvial, maritime areas and Razim-Sinoe complex. Total nitrogen loads in surface waters come both from natural and anthropogenic activities. Industrial wastes from residual, agricultural and domestic waters represent important sources in total nitrogen concentrations (organic and inorganic nitrogen) increasing (Galloway et al 1995; Galloway & Cowling 2002).

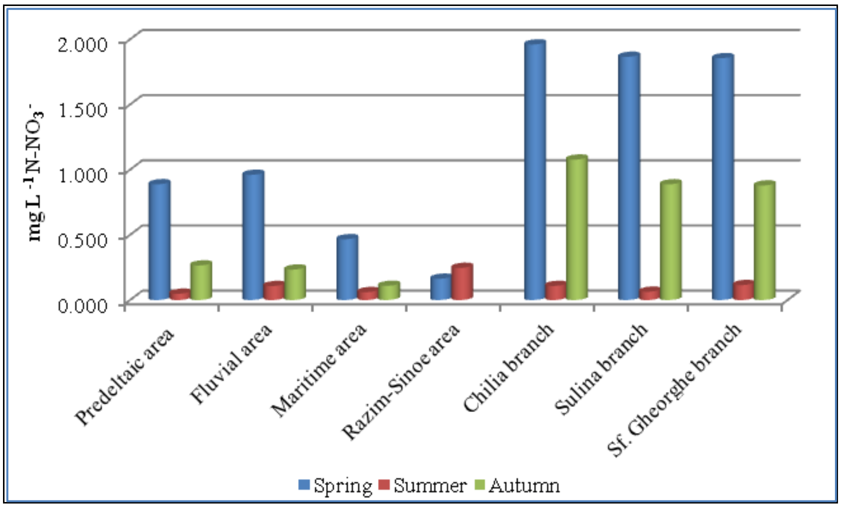

Figure 3. Average concentrations of nitrogen from nitrates determined seasonally, in surface waters from deltaic areas in 2015.

Concentrations of total nitrogen (Figure 4) in spring season have a relative uniform trend, with values in the range of 3.913 mg L^{-1} (Razim-Sinoe area) and 6.359 mg L^{-1} (fluvial area). In summer season, concentration values of total nitrogen have a higher variation, because of the higher temperatures and low water levels (1.754 mg L^{-1} in pre-deltaic area and 12.897 mg L^{-1} in Chilia branch). In autumn, total nitrogen concentrations varied between 4.367 mg L^{-1} (Chilia branch) and 9.469 mg L^{-1} (pre-deltaic area).

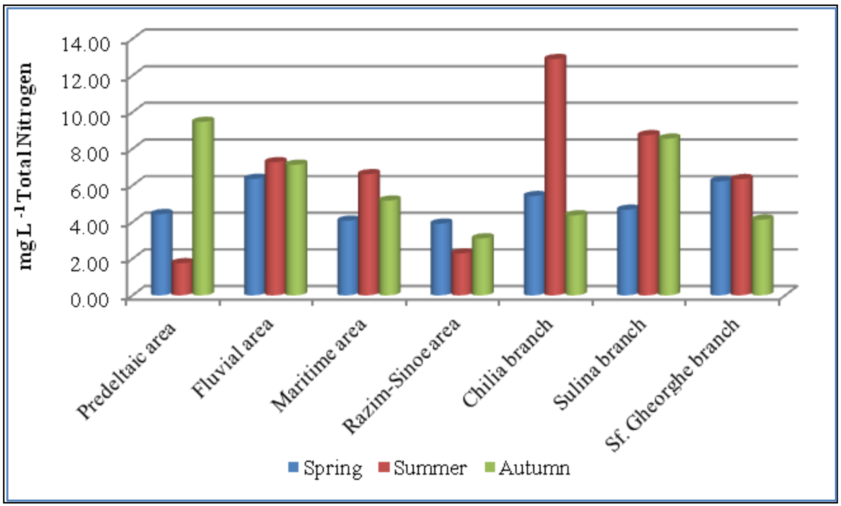

Figure 4. Average concentrations of total nitrogen determined seasonally, in surface waters from deltaic areas in 2015.

In Figures 5-10 are represented seasonally, linear dependences between concentrations of nitrogen from nitrite and concentrations of nitrogen from nitrate, in Danube lakes and branches. In all three seasons, taking into account the determined concentration values, a directly proportional relationship was observed, between concentrations determined from water lakes (Pearson coefficients 0.766 in spring, 0.467 in summer and 0.797 in autumn), while on the Danube branches an inverse relationship was observed (-0,447 in spring, -0.496 in summer and -0.692 in autumn). This indicates that when nitrites concentration increase, nitrates concentration in water lakes also increases and nitrates concentrations from branches water decreases, because of the complex processes of nitrification/denitrification that occur in aquatic complexes.

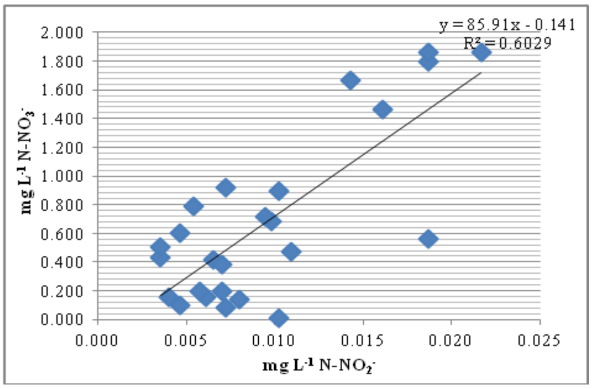

Figure 5. Linear dependence of nitrogen from nitrites and nitrates concentrations in RBDD lakes waters, in spring.

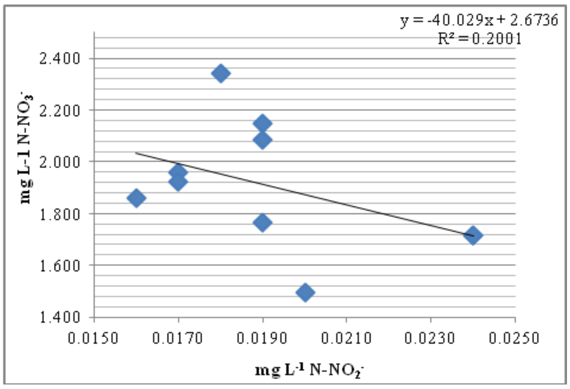

Figure 6. Linear dependence of nitrogen from nitrites and nitrates concentrations from Danube branches waters, in spring.

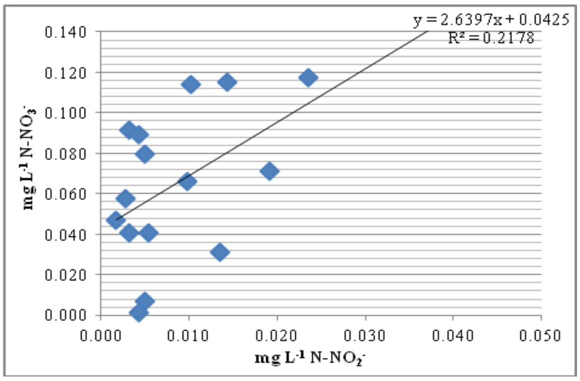

Figure 7. Linear dependence of nitrogen from nitrites and nitrates concentrations in RBDD lakes water, in summer.

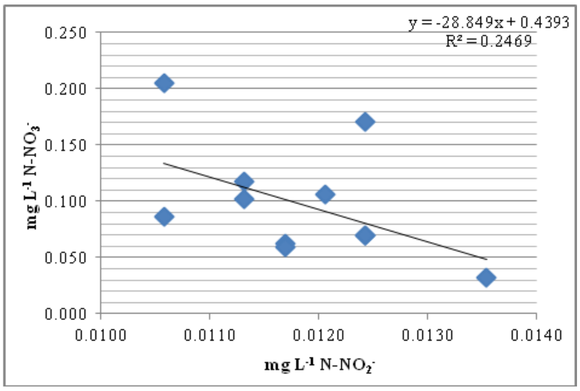

Figure 8. Linear dependence of nitrogen from nitrites and nitrates concentrations from Danube branches waters, in summer.

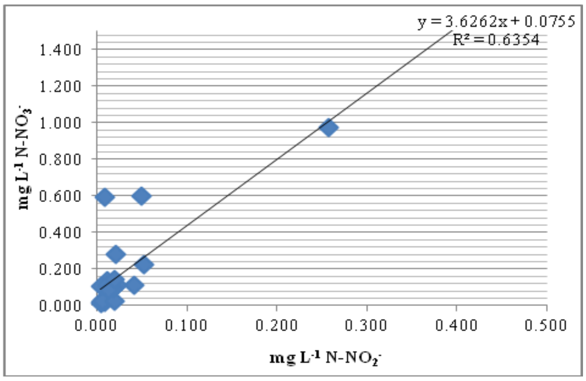

Figure 9. Linear dependence of nitrogen from nitrites and nitrates concentrations in RBDD lakes water, in autumn.

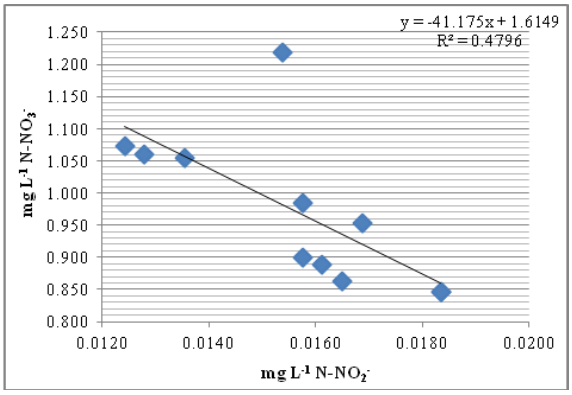

Figure 10. Linear dependence of nitrogen from nitrites and nitrates concentrations from Danube branches waters, in autumn.

In Table 2 are presented percentage variations of organic and inorganic nitrogen forms in all three seasons (spring, summer, autumn), in 2015.

Table 2

Seasonal percentage variation of inorganic and organic nitrogen forms

Sampling area	*Spring*		*Summer*		*Autumn*	
	Inorganic nitrogen (%)	*Organic nitrogen (%)*	*Inorganic nitrogen (%)*	*Organic nitrogen (%)*	*Inorganic nitrogen (%)*	*Organic nitrogen (%)*
Pre-deltaic area	24.197	75.803	7.087	92.913	4.193	95.807
Fluvial area	19.496	80.504	3.079	96.921	7.490	92.510
Maritime area	14.035	85.965	2.007	97.993	10.632	89.368
Razim-Sinoe area	7.812	92.188	21.916	78.084	-	-
Chilia branch	39.117	60.883	1.772	98.228	29.746	70.254
Sulina branc	44.415	55.585	1.600	98.400	12.602	87.398
Sf. Gheorghe branch	32.458	67.542	3.824	96.176	24.988	75.012

Organic nitrogen is one of the most important sources of nitrogen for phytoplankton growth (Otsuki et al 1993).

In summer and autumn periods, high content of organic nitrogen (a range of 78.084-98.400% in summer and 70.254-98.807% in autumn), compared with low inorganic nitrogen content (a range of 1.600-21.916% in summer and 4.193-29.746% in autumn), is due to the fact that organic nitrogen seems to be assimilated by aquatic organisms with a much lower rate than inorganic forms of nitrogen, in those periods (Fahmy et al 1996).

It has to be stressed out that organic matter containing nitrogen, usually resist to bacterial changes and remain in water or sediments (Riley & Chester 1971).

Inorganic phosphorus is indispensable for biological process, because it carries the energy taken from the sun in photosynthesis and stored in macroergic phosphate groups.

In accordance with the Order 161/2006 of Environment Ministry and Water Management, it can be noted that in the studied period it were no exceedings of the maximum admitted values of 0.200 mg L^{-1}, according to second class on quality (good ecological status). Maximum concentrations of ortophosphat, shown in the Figure 11, were determined in autumn season (0.074 mg L^{-1} in Sulina branch) and minimum in summer season (0.003 mg L^{-1} in maritime area).

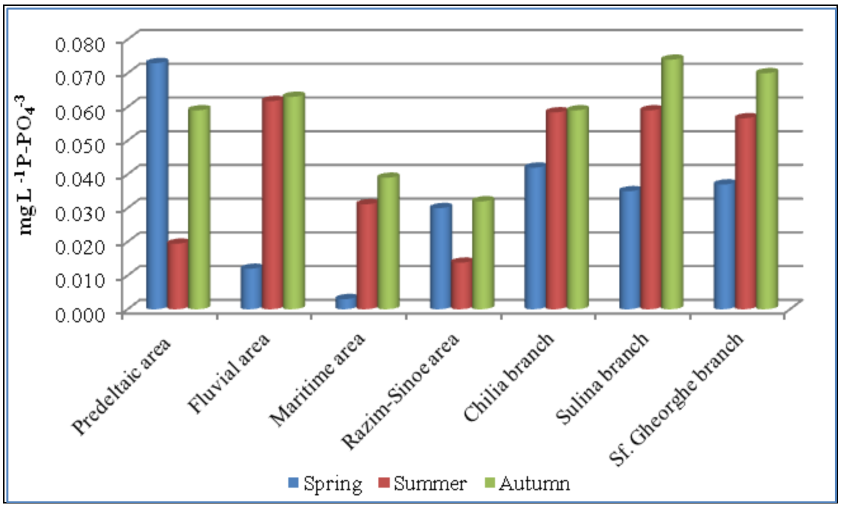

Figure 11. Average concentrations of inorganic phosphorus determined seasonally, in surface waters from deltaic areas in 2015.

Total phosphorus concentrations dynamic, presented in Figure 12, may grow temporary after abundant rains due to suspended particles and soluble forms that are washed from shore and transported in lakes water or due to re-suspended in windy days (Lombardo 2006). Also, the total phosphorus concentration can increase in summer period due to his release from sediment this situation can occur even in areas with good ecological status (Wang 2010).

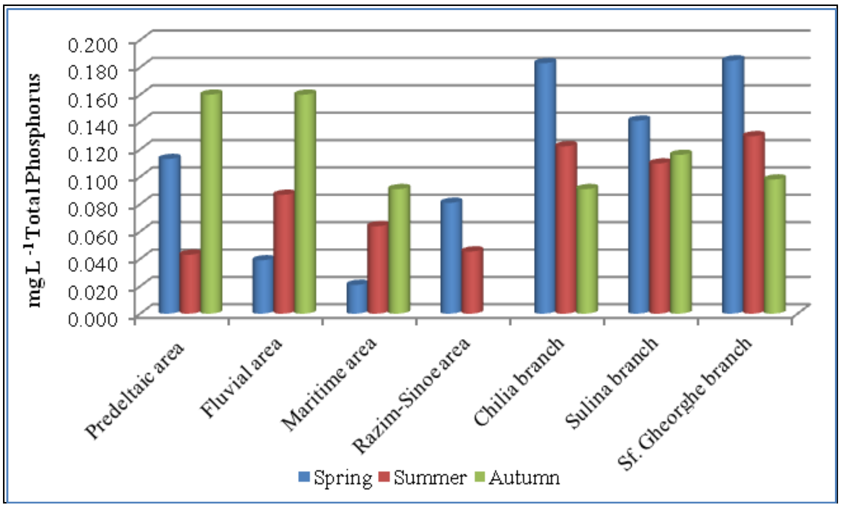

Figure 12. Average concentrations of total phosphorus determined seasonally, in surface waters from deltaic areas in 2015.

Analyzing the total phosphorus concentration results, determined in the 7 areas, in spring, summer and autumn, reveals significant higher amounts in branches area (0.185 mg L^{-1} in Sf. Gheorghe branch in spring) where anthropogenic inputs are more significant and the lowest level in maritime area (0.021 mg L^{-1}, in spring) values that do not exceed maximum admitted concentration according to second quality class (0.400 mg L^{-1}) of Order no. 161/2006.

In the Table 3 are presented percentage variation of organic and inorganic phosphorus forms, in three seasons (spring, summer and autumn), in 2015. Changes in bacterial population during the seasons can cause variations in organic and inorganic phosphorus concentrations.

Table 3

Seasonal percentage variation of organic and inorganic phosphorus forms

Sampling area	*Spring*		*Summer*		*Autumn*	
	Inorganic P (%)	*Organic P (%)*	*Inorganic P (%)*	*Organic P (%)*	*Inorganic P (%)*	*Organic P (%)*
Pre-deltaic area	64.602	35.398	45.147	54.853	36.875	63.125
Fluvial area	30.769	69.231	71.129	28.871	39.375	60.625
Maritime area	14.286	85.714	48.699	51.301	42.857	57.143
Razim-Sinoe area	37.037	62.963	30.414	69.586	83.117	16.883
Chilia branch	22.951	77.049	47.751	52.249	64.835	35.165
Sulina branch	24.823	75.177	53.749	46.251	63.793	36.207
Sf. Gheorghe branch	20.000	80.000	43.690	56.310	71.429	28.571

Conclusions. Based on the results obtained for spring season, we conclude that the values of nutrients concentrations, for deltaic ecosystems, are the result of their relative low consumption by phytoplankton and aquatic plants that inhabit the surface water in spring season, this results being in accordance with what Abdel-Moati et al (1988) found in their study. In the summer season, exceedings of maximum admitted concentrations were identified in fluvial area and Chilia branch, maximum values being identified in autumn season with exceedings for pre-deltaic area, fluvial area and Sulina branch.

Also, it was demonstrated that in the case of deltaic ecosystems, differences in nutrients concentrations are due to anthropogenic activities, characteristics of the area, amount of organic matter, mineralization degree and water flow speed.

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