

Non-ionized ammonia pollution level of the small rivers water in the Central Development Region of the Republic of Moldova

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Keywords: summary ammonium, non-ionized ammonia, Dniester and Prut rivers, tributaries.

Abstract: In order to maintain fish life, the Council Directive of 18 July 1978 on the quality of freshwater, which must be protected or improved provides that non-ionized ammonia in salmon and cyprinid waters may be in concentrations of $\leq 0,005 \text{ mg/dm}^3$ (indicative) and $\leq 0,025 \text{ mg/dm}^3$ (mandatory). It presents a danger for aquatic biological diversity in concentrations from 0,03 to 2,00 $\text{mg/dm}^3 \text{ NH}_3$ depending on the species and age. This conditions also are specified in Chapter II (Specific substances to be introduced into surface waters, pt. 8) of the Republic of Moldova Government Decision no. 802 of October 9, 2013. The study includes evaluation of non-ionized ammonia in the water of some portions of the Dniester and Prut river tributaries, which crosses the Central Development Region of the Republic of Moldova. In the rivers Răut (upstream of Bălți town and downstream of Varvareuca village), Ciulucul Mare, Ciulucul Mic, Camenca (Gura Camencii village) the ammonia concentration in different years (1981-2019) was low, but in the Răut river water, downstream of the Orhei town at 5°C was of 0,029-0,35 mg/dm^3 and at 15°C - 0,065-0,528 mg/dm^3 . The calculation of the NH_3 content in the water from the left tributaries of the Prut river that crosses the Central Development Region of the Republic of Moldova at a temperature of 5°C and 15°C shows high values in the water of the Gârla Mare river, Medeleni and Catranâc villages (0,394-0,818 mg/dm^3), river Gârla Mică, liman (0,141-0,327 mg/dm^3), Delia river, Pârlița village (0,84-1,78 mg/dm^3), Nârnova river, Nisporeni town and Leușeni village (0,27-0,73 mg/dm^3), Lăpusna river, Voinescu village (0,277-0,65 mg/dm^3). Thus, in the water of different years (1981-2019) on many portions of the Dniester and Prut rivers tributaries, which crosses the Central Development Region of the Republic of Moldova, the concentration of ammonium ($\text{NH}_3 + \text{NH}_4^+$) was high, and depending on the pH and temperature the calculated non-ionized ammonia content exceeded the value of 0,025 mg/dm^3 .

1. Introduction

The Central Development Region (CDR) of the Republic of Moldova is located in the central part of the country with a total area of 1 063 600 thousand ha, 31% of the country total area, and includes 13 rayon's: Anenii Noi, Călărași, Criuleni, Dubăsari, Hâncești, Ialoveni, Nisporeni, Orhei, Rezina, Strășeni, Șoldănești, Telenești, Ungheni with 598 localities organized in 329 administrative-territorial units (Figure 1).

The Central Development Region is crossed by Dniester and Prut rivers and a series of their tributaries (Ciulucul Mare, Ciulucul Mic, Cernea, Cogâlnic, Botna, Răut, Ichel, Ivanos, Ivancea, Brătuleanca, Șoltoiaia, Gârla Mare, Lăpușnița and Nârnova). The main sources of water in the rivers are groundwater, rain and snow. All rivers and streams

constitute a strong hydrographical network, the regime of which depends on climatic conditions.

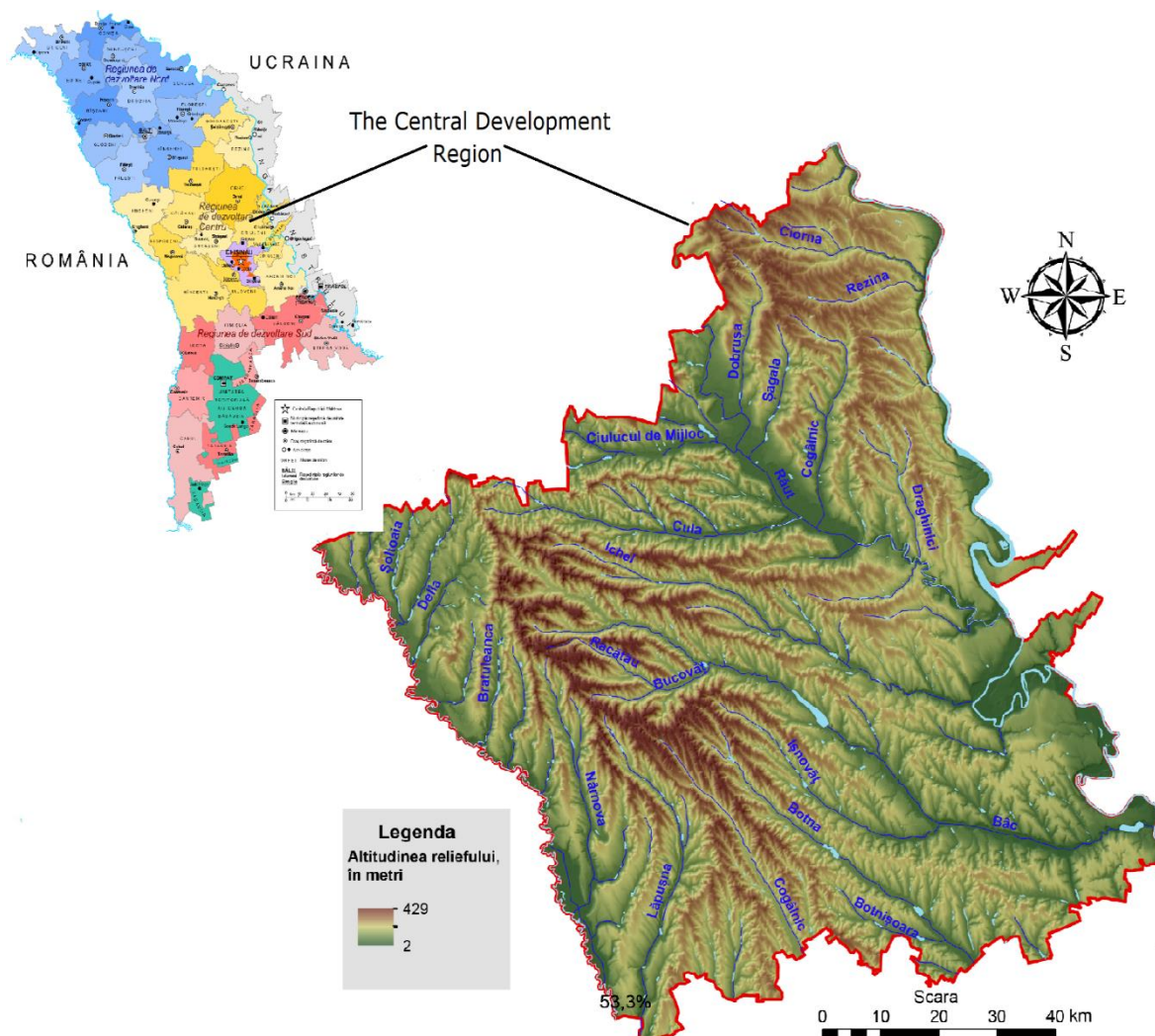


Figure 1. The Central Development Region of the Republic of Moldova (Wikimedia Foundation. (2022, February 11))

Pollution of surface and groundwater in the Republic of Moldova is caused in most cases by rainwater runoff, various landfills, agricultural fields, domestic livestock and discharges of insufficiently treated or untreated wastewater from the domestic and industrial sectors, et al.

From existing sources of pollution ammonium compounds penetrate in the environmental components from atmospheric deposits, following the reduction of nitrogen, by degradation of proteins and nitrogenous organic matter in vegetable and animal waste, from untreated or insufficiently treated industrial wastewater (animal and vegetable processing, chemical, textile, coking industry et al.) and household.

The yearbook of the Environmental Protection Inspectorate specifies that in 2019 year in the CDR of the Republic of Moldova there were 109 (42% of the total in the country) wastewater treatment plants, of which only 6 units fitting with the required treatment normative (about 5,6%), with partial treatment - 2 units (1,8%) and insufficient - 77 units (70,6%), being non-functional 24 (22%) (Yearbook IEP, 2020).

There were 1200 dump sites in 2018, from which 440 (with a total surface about 472 ha) were exploited, as well as illegal - 760 (about 146,6 ha) (Yearbook IEP, 2020).

Our investigations demonstrated the existence of many potential sources of surface water pollution in the CDR of the Republic of Moldova with dangerous pollutants, including ammonium.

Ammonium compounds in water are found in the form of ammonium ions (NH_4^+) which show low toxicity or as free ammonia (NH_3), which is very toxic. The ionic form (NH_4^+) is 50 times less toxic than the non-ionized form (Şerban, 2011), the content of which varies according to pH and temperature (Novikov et al., 1990; Standard Methods for the Examination of Water and Wastewater, 1995; Wajsbrodt et al., 1993). According to the Council Directive (78/659/CEE, Annex I. List of parameters) on the quality of fresh water to be protected or improved for the preservation of fish life, non-ionized ammonia from salmonic waters and cyprinids may be in concentrations of $\leq 0,005 \text{ mg/dm}^3$ (indicative) and $\leq 0,025 \text{ mg/dm}^3$ (obligatory). It presents a danger for aquatic biological diversity in concentrations from 0,03 and $2 \text{ mg/dm}^3 \text{ NH}_3$ depending on the species and age (78/659/CEE), conditions also specified in Chapter II (Specific substances to be introduced into surface waters, point 8) of the Republic of Moldova Government Decision no. 802 of 9.10.2013 (GD 802/9/2013), (Table 1).

Table 1. Non-ionized ammonia and total ammonia from the list of physical and chemical parameters applied for salmon and cyprinid waters

Parameters	Minimum value (mg/dm^3) and observations			
	salmon waters		cyprinid waters	
	G*	I*	G*	I*
non-ionized ammonia (NH_3)	$\leq 0,005$	$\leq 0,025$	$\leq 0,005$	$\leq 0,025$
ammonium total ($\text{NH}_3 + \text{NH}_4^+$)	$\leq 0,04$	≤ 1	$\leq 0,02$	≤ 1

*G = indicative; I = obligatory

In the national legislation on environmental quality requirements for surface waters, the Regulation on the requirements for the collection, treatment and discharge of wastewater in the sewerage system and/or in water outlets for urban and rural localities (GD 950/2013) and the Environmental Strategy for 2014-2023 years (GD 301/2014) is not specified the ammonia content (NH_3 or N-NH_3), including information only on the high degree of small rivers water pollution in the republic with ammonium ions.

Ammonium compounds are specific pollutants present in wastewater from economic activities in the republic: the industry of yeast, beer, soft drinks, milk, meat and fish processing, leather, animal complexes, et al. The maximum permissible concentration (MPC) of NH_3 in receiving waters is of $0,05 \text{ mg/dm}^3$ (Hazard Index 20) and of the NH_4^+ ion – $0,4 \text{ mg/dm}^3$ (Hazard Index 2,5) (The order 163 / 2003), (Table 2).

Table 2. Relative hazard indices of pollutants in receiving waters

Substance	MPC, mg/dm^3	Hazard index
non-ionized ammonia NH_3	0,05	20
ammonium ion NH_4^+	0,4	2,5

2. Materials and Methods

To assess the concentration of non-ionized ammonia (NH_3) in surface waters of the Republic of Moldova according to pH and temperature was used the National standard (SM 353: 2020/2020). The method is applied for ammonium ($\text{NH}_3 + \text{NH}_4^+$) concentration of $0-4 \text{ mg/dm}^3$ with error of 4,56%; 5,46% and 2,91% for the temperatures of 5°C , 15°C and 25°C respectively, the average value being 4,32% (Standard Methods for the Examination of Water and Wastewater, 1995; Wajsbrodt et al., 1993).

Various methods have been used to determine ammoniacal nitrogen in surface water and wastewater: - photometric method, applicable to lightly contaminated waters, with ammoniacal nitrogen concentrations of 0,1-2,0 mg N/dm³; the error of the method is a maximum of 0,1 mg/dm³; - the method of distillation, applicable to both slightly and severely impure waters, with ammoniacal nitrogen concentrations of 0,1 - 50 mg N/dm³; with subsequent photometry, for ammoniacal nitrogen concentrations from 0,1 to 5,0 mg N/dm³ and method error of not more than 0,05 mg N/dm³; with subsequent titration, for ammoniacal nitrogen concentrations from 5,0 mg to 50 mg N/dm³ and method error of not more than 0,5 mg N/dm³;

The photometric method can be used for water with ammoniacal nitrogen content greater than 2,0 mg N/dm³, by diluting the water samples accordingly. The distillation method can be used in the case of water with an ammoniacal nitrogen content lower or higher than that specified above by distilling volumes of water samples respectively greater than or less than 500 cm³. The water samples are collected in glass or plastic containers, in compliance with the regulations in force regarding the taking of samples of surface water and wastewater. Before taking the water samples, the vessels are washed with 1:1 hydrochloric acid after volume, with distilled water and then with the test sample. The water samples are analyzed as soon as possible after collection and if this is not possible the samples are fixed by acidification with 1:1 sulfuric acid by volume to pH=4. Before the determination is carried out by the photometric method, the water samples are filtered through filter paper or through a filter crucible.

3. Results

Based on the information that the water quality of the small rivers in the Republic of Moldova is characterized by a high degree of ammonium ion pollution (Bulat, 2017), the content of non-ionized ammonia (NH₃), which is more toxic, was calculated in the waters of some right tributaries of the Dniester and left ones of the Prut rivers, which crosses the central region of the country, on existing information of the total concentration of ammonium (NH₃ + NH₄⁺) and the pH of water in different years (Yearbook, 2015; Yearbook, 2016, Yearbook IEP, 2019; Bulat, 2017; Gladchi, 2019; GD 301/2014; Lozan et al., 2015; Lupașcu et al., 2001; Ropot et al. 2002; Sandu et al., 2019).

The study was conducted according to pH (7,0-10,0) and water temperature, which varies by seasons, but also by day and night, selecting the temperature of 5°C and 15°C.

3.1. The content of non-ionized ammonia in the water of the Prut River left tributaries.

The calculation of the NH₃ content in the water from the Prut River left tributaries which crosses the central region of the Republic of Moldova, at a temperature of 5°C and 15°C shows high values in the water of the Gârla Mare river, Medeleni and Catranac villages (0,394-0,818 mg/dm³), Gârla Mică river, liman (0,141-0,327 mg/dm³), Delia river, Pârlița village (0,84-1,78 mg/dm³), Nârnova river, Nisporeni town and Leușeni village (0,27-0,73 mg/dm³), Lăpușna river, Voinescu village (0,277-0,65 mg/dm³), Brătuleanca river, Brătuleni village (0,361-0,75) (Figure 2). Thus, the ascendances of the 0,025 mg/dm³ NH₃ value from the requirements allowed in the national norm (GD 802/2013; 78/659/CEE) reach tell about 71 times (Delia river, Pârlița village), the reason being the presence of pollution sources, especially wastewater discharges.

There is a fairly high correlation between NH₃ and pH at both 5°C (R² = 0,6931) and 15°C, which indicates the dependence of NH₃ concentration on water pH (Figure 2).

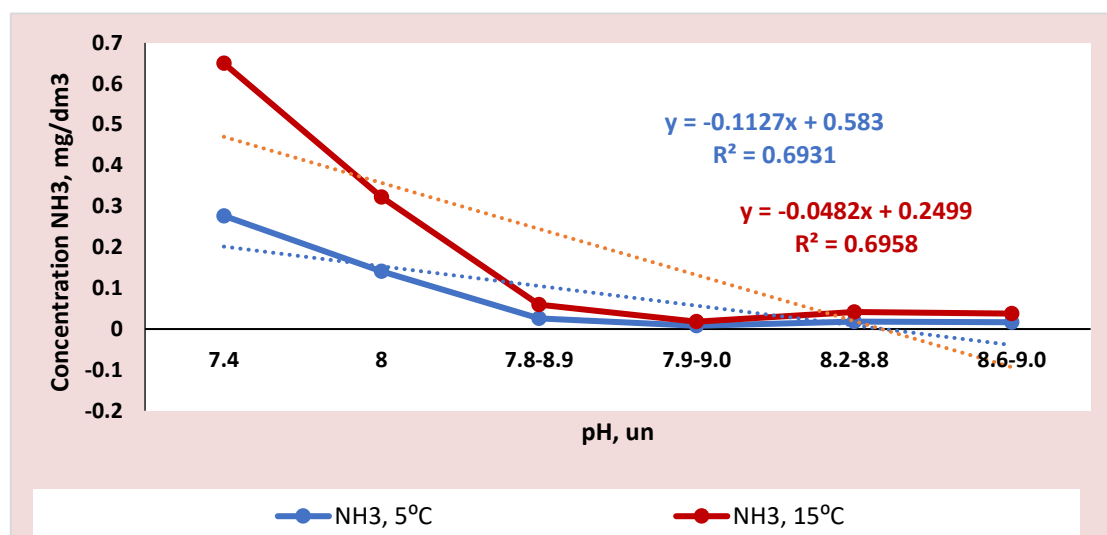


Figure 2. Non-ionized ammonia (NH_3) content as a function of pH and temperature content in the water of the Prut river left tributaries.

But the concentration of pH-dependent forms of (NH_4^+ + and NH_3) deviates greatly from one river to another (Table 3).

Table 3. Total concentration of ammonium compounds ($\text{NH}_3 + \text{NH}_4^+$) and pH in the waters of the left tributaries of the Prut river.

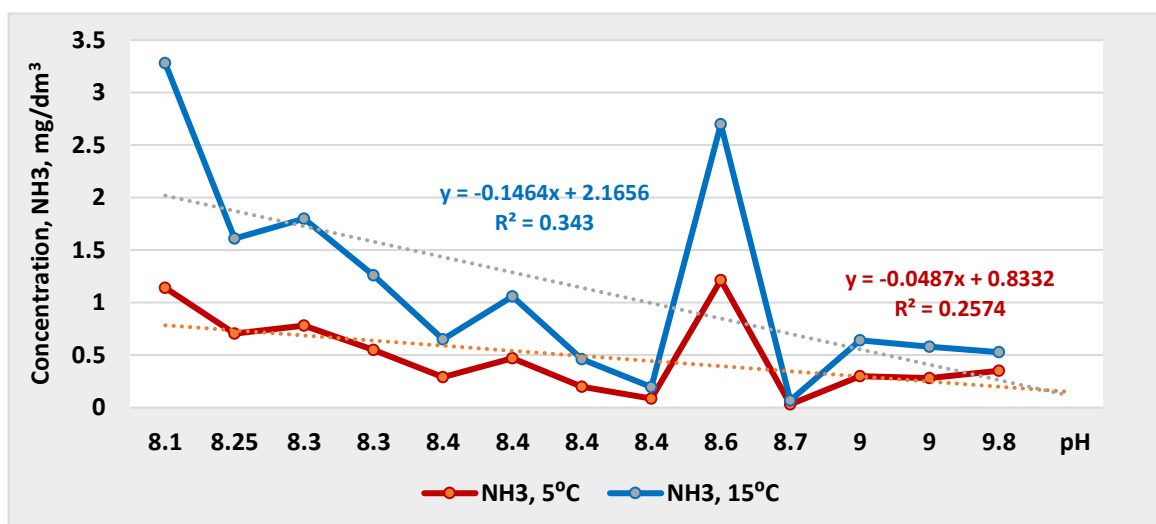
River	Water sample collection sections	pH, un	($\text{NH}_3 + \text{NH}_4^+$), mg/dm^3
Gârla Mare	Medeleni and Catranâc villages	7,9-9,0	0,81-0,96
Gârla Mică	liman	8,0	12,9
Delia	Parlita village	7,8-8,9	3,8-10,4
Brătuleanca	Bratuleni village	8,6-9,0	0,41-3,63
Nârnova	Nisporeni town, Leuşeni village	8,2-8,8	1,06-6,4
Lăpuşna	Voinescu village	7,4	10,3

3.2. The ammonia content in the water of the Dniester River right tributaries

The content of ammonium in the surface waters of the country varies annually (Yearbook SEI, 2015; Bulat, 2017). According to the data presented by the State Ecological Inspectorate, for example, the concentration (years 2001-2014) in the water from Ghidighici lake, dam, was decreasing and varied from $4,4 \text{ mg/dm}^3$ to $0,11 \text{ mg/dm}^3$, and in Bâc river downstream of Chişinău municipality (Sângera village) being increasing - from $16,4 \text{ mg/dm}^3$ (2001 year) to $77,0 \text{ mg/dm}^3$ (2012 year) and subsequently decreasing until at $25,4 \text{ mg/dm}^3$ (2014 year) (Yearbook SEI, 2015).

In the water of some portions of the Dniester river right tributaries, which crosses the Central Development Region of the Republic of Moldova, the total ammonium concentration was low in different years (1981-2019) and the ammonia content was less than $0,025 \text{ mg/dm}^3$: Răut river (upstream of Bălţi municipality and downstream Varvareuca village), Căinar river (Gura Căinarului village), Ciulucul Mare and Ciulucul Mic rivers, Camenca river (Gura Camencii village), Cogâlnic river (Cişmea village), et al.

Thus we have an average correlation between the ammonia concentration at the temperature of 5°C (0,2574), at 15°C (0,343) and the pH of the water of the Dniester river (Figure 3).



8.1-Bâc / Călărași town, downstream; 8.25 - Bâc/Gura Bâcului village;
 8.3-Bâc/Sângera and Bulboaca villages; 8.3-Bâc/Strășeni town, downstream;
 8.4-Ichel / Goieni village; 8.4-Răut/Bălți town, downstream;
 8.4-Camenca/ Gvozdova village; 8.4-Cubolta/Cubolta village;
 8.6-Bâc/Gura Bâcului village; 8.7-Botna/Ulmu – Chircăiești villages;
 9-Ciulucul Mare/river overflow section; 9-Ciulucul Mijlociu/river overflow section
 9.8-Răut/Ustia village, downstream.

Figure 3. Non-ionized ammonia (NH_3) as a function of pH and temperature content in the water of the Dniester river right tributaries.

The total concentration of nitrogen forms ($\text{NH}_3 + \text{NH}_4^+$) in the water of the right tributaries of the Dniester river, does not correlate with the pH and has quite high values compared to the pH, such as river Bâc (Călărași, Sângera and Gura Bâcului) (Table 4).

Table 4. Total concentration of ammonium compounds ($\text{NH}_3 + \text{NH}_4^+$) and pH in the waters of the right tributaries of the Dniester river.

River/Section	pH, un	($\text{NH}_3 + \text{NH}_4^+$), mg/dm ³
Răut / Bălți town, downstream	7,95-8,4	4,7-17,3
Răut / Ustia village, downstream	8,8-9,8	0,72-3,4
Camenca / Gvozdova village	8,4	7,6
Cubolta / Cubolta village	8,4	3,2
Ciulucul Mare /river overflow section	9,0	3,1
Ciulucul Mijlociu /river overflow section	7,9-9,0	1,6-2,8
Bâc / Călărași town, downstream	8,1	83,0
Bâc / Strășeni town, downstream	7,75-8,3	2,9-25,6
Bâc / Sângera and Bulboaca villages	7,6-8,3	14,8-89,0
Bâc / Gura Bâcului village	7,4-8,25	11,5-36,3
	8,6	28,8
Ichel / Goieni village	7,6-8,4	6,7-10,7
Botna / Ulmu – Chircăiești villages	8,0-8,7	0,59-1,26

4. Discussion

Specific to the properties of non-ionized ammonia is that its percentage at temperatures from 0°C to 30°C and the pH 7,0–10,0 is increasing of 2,2–10,8 times. With the change of pH at the same temperature, the percentage of non-ionized ammonia in water increases hundreds of times. For example, at 10°C the percentage of NH_3 from pH 7,0 to pH 10,0 increases of 395 times, and at temperature 0°C being an increase of 594 times, so the NH_3 content (mg/dm^3) will also increase in water as temperature and pH value increase (Figure 4).

The concentration of total ammonium in the Răut river water, downstream Orhei town in 2016 year was of 0,07–0,56 mg/dm^3 (Gladchi, 2019), and calculated non-ionized ammonia was of 0,0057–0,013 mg/dm^3 .

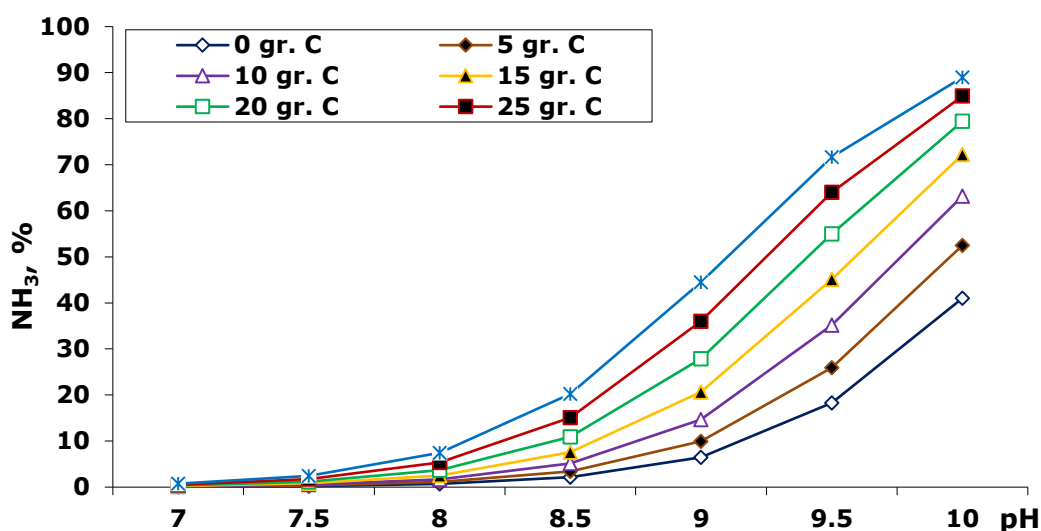


Figure 4. Dynamic of NH_3 percentage (%) in dependence of water pH and temperature.

In the information from 2015–2018 years (Gladchi, 2019) is mentioned the ammonium concentration at Ustia village of 0,72–3,4 mg/dm^3 , ammonia being calculated at the temperature of 5°C of 0,029–0,35 mg/dm^3 and at that of 15°C - of 0,065–0,528 mg/dm^3 . In the water from some parts of the Răut, Ciulucul Mare and Ciulucul Mijlociu, Camenca, Cubolta, Bâc, Ichel and Botna rivers the ammonia content calculated for the temperature of 5°C was from 0,011 (Ichel river, Ratuș village) to 1,14 mg/dm^3 (Bâc river, downstream of Călarăși town), and at the one of 15°C it was from 0,026 (Ichel river, Ratuș village) to 1,8 mg/dm^3 (Bâc river, downstream Chișinău municipality, Sângera village), 2,7 mg/dm^3 – Bâc river, Gura Bâcului village, and 3,28 mg/dm^3 – Bâc river, downstream Călarăși town, being exceeded the concentration of 0,025 mg/dm^3 NH_3 in most cases at both temperatures by 1,04 – 131,2 times (Figure 3).

Thus in different years (1981–2019) in the water on many portions of the Dniester and Prut rivers tributaries, which crosses the CDR of the Republic of Moldova, the concentration of ammonium ($\text{NH}_3 + \text{NH}_4^+$) was high in depending on the pH and temperature and calculated non-ionized ammonia content exceeded the value of 0,025 mg/dm^3 .

Currently it is observed that, in a comparative aspect, the ichthyofauna of all natural aquatic ecosystems in the Republic of Moldova has undergone cardinal changes (mostly in small rivers, natural lakes), the successional character of ichthyocenosis being as a sure model in elucidating the well-being of aquatic ecosystems and impact factors (Bulat, 2017). The indications in the Regulation on the conditions for the discharge of wastewater into bodies of water include not only the means of determining the degree of water pollution from rivers, lakes, springs, wells, et al, but also the measures, activities to be undertaken for waters sanitation and adjacent territories, which directly contributes to the protection of our health, of all living things in the Republic of Moldova.

5. Conclusions

1. Based on the studies carried out, it was established that the content of ionized ammonia can be determined by the proposed process only in natural waters, the ionic strength of which falls within the limits (0,01-0,075 mg/dm³).

2. The concentration of ionized ammonia as a function of temperature and pH is not strictly straightforward, therefore no constant coefficient has been established for its determination in water.

3. The calculation procedure of the ionized ammonia content can be applied for the evaluation of the damage caused to the environment by violating the normative legislation in the field of water.

4. In the waters of small rivers (river basins of the Dniester and Prut rivers) the summary concentration (NH₃⁺ NH₄⁺) of ammonium is high, and the content of non-ionized ammonia calculated according to pH and temperature, exceeds the value of 0,025 mg/dm³, specified in the requirements of Directive 78/659/EEC and GD no. 802 of 9.10.2013.

5. Monitoring the content of non-ionized ammonia in the waters of water basins, especially those for fishing, is very important and requires strict compliance with the requirements of various regulations.

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