

# The 3rd AAPG/SEG/EAGE International Geosciences Student Conference

29-31 May 2012, Belgrade, Serbia

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***Publisher:*** Association of Geophysicists and Environmentalists of Serbia (AGES)

***For Publisher:*** Snežana Komatina-Petrović, Association of Geophysicists and Environmentalists of Serbia (AGES)

***Printed by:*** PROOF, Belgrade

Copies: 500

**ISBN**

***All papers in the Proceedings are reviewed***

*The Proceedings are published with the financial support of the Ministry of Science and Education of Serbia*

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CIP - Каталогизација у публикацији  
Народна библиотека Србије, Београд

55(082)  
624.13(082)  
620.91:550.36(082)  
502/504(082)

AAPG/SEG/EAGE International Geosciences  
Student Conference (3 ; 2012 ; Beograd)  
[Proceedings] / The 3rd AAPG/SEG/EAGE  
International Geosciences Student Conference,  
29-31 May 2012, Belgrade, Serbia ; [organizer  
Association of Geophysicists and  
Environmentalists of Serbia (AGES) ; editor  
Saša Smiljanić]. - Belgrade : #Association of  
Geophysicists and Environmentalists of Serbia  
(#AGES), 2012 (Belgrade : Proof). - [248]  
str. : ilustr. ; 30 cm

Tiraž 500. - Bibliografija uz pojedine  
radove.

ISBN 978-86-913953-5-3  
1. Association of Geophysicists and  
Environmentalists of Serbia (Beograd)  
a) Геологија - Зборници b) Инжењерска  
геологија - Зборници c) Геотермална  
енергија - Зборници d) Животна средина -  
Заштита - Зборници  
COBISS.SR-ID 191120908

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## Nitrates in groundwater of Serbia

Maja Todorović\*, Marina Ćuk, Jovana Milosavljević

### Summary

Nitrogen occurs in water environments in the form of nitrate, nitrite and ammonium ions. When nitrogen in the form of nitrate appears in groundwater, it becomes highly mobile due to its solubility and the anion shape. The sources of nitrate ions in groundwater can be both natural and artificial. The main artificial sources are agriculture and both communal and industrial waste waters. The presence of nitrate in groundwater, especially in that used for drinking has extremely harmful effect on human health and impacts animals and plants as well. The most significant harmful impact of nitrates on human health is nitrate reduction into nitrite which contributes to the development of the disease of methemoglobinemia, and they also have mutagenic and carcinogenic effect on a large number of organs. Maximal permissible nitrate concentration in drinking water is 50 mg/L. In order to define the quality of groundwater for public supply on the territory of Serbia from the aspect of nitrate presence, groundwater samples have been taken from 100 sources of regional and local significance. The results have shown that the nitrate concentration in 77% samples is below MPC of 50 mg/L. The data analysis has been carried out by lithological environments and results have shown that water from alluvial aquifers is the most polluted and from the regional aspect, the most endangered is groundwater of the Velika Morava River alluvion. By isotopic methods, it has been determined that the source of nitrate in groundwater in part of this alluvion originates from organic land nitrate and waste water of rural population living in the recharge zone of the alluvial aquifer.

### Introduction

Nitrates occur naturally as part of a nitrogen cycle. The presence of this ion in groundwater points to complete oxidation of nitrogen containing compounds. Nitrates formed exclusively in natural way can be found in nature in insignificantly low concentrations and as such do not harm the health of living organisms. Nitrate concentrations up to 10 mg/L, appear in groundwater as a natural process of precipitation, infiltration and the mineralization of organic substances in soil. The value of nitrates over this limit reflects, most frequently, the impact of an antropogenic factor. Regulations from the field of drinking water quality have adopted the boundary value of 50 mg/L as the maximal permissible nitrate concentration in drinking water (Official Gazette, FRY, Vol. 42/98 and 44/99).

There are various ways of water polluting and they most frequently appear individually. The sources of water pollution by nitrates can be natural and artificial. Significantly higher nitrate concentrations into groundwater are emitted by artificial sources, among which the most significant are: industrial and communal waste waters, production of oil and oil products, the use of mineral fertilizers, the use of pesticides, collection of mineral salts owing to watering, disposal of manure, the extension of agricultural land causing soil erosion etc. The relevant factor of anthropogenic water pollution is the atmosphere contaminated by gaseous, liquid, and solid matters. From this point of view, acid rains developed by dissolution of nitrogen oxide and sulphur in rain drops are especially significant.

### Theory

Nitrogen appears in water systems in the shape of nitrate, nitrite and ammonium ions. Nitrate is a more stable shape of oxidised nitrogen, but it can be reduced by microbial activity to nitrite. Nitrites appear by the activity of nitrification bacteria as intermediar products of the nitrate formation but they appear in low concentrations. The natural content of nitrate and nitrite in the environment is a consequence of the nitrogen cycle in nature, but they appear in low concentrations in normal conditions. Due to the stability of nitrates their concentration in surface and groundwater mostly does not exceed 10 mg/L, while the presence of nitrites is about 1 mg/L. However, owing to anthropogenic activities nitrate content in numerous water systems worldwide is increasing significantly.

The transformation of nitrogen compounds in the nitrogen cycle includes several mechanism: ammonification, volatilization, nitrification and denitrification. The disturbance of these processes results in nitrate appearance in groundwater and drinking water. The movement of nitrate through a life environment is conditioned by transport mechanism. Transport mechanism embraces at the same

time percolating, deposition, groundwater movement, and velocity of movement, precipitation and evaporation.

Ammonification represents the first step in so called mineralization of organic nitrogen and it is defined as biological conversion of organic into ammonia nitrogen. In anaerobic conditions, the accumulation of ammonia nitrogen appears due to prevention of nitrification. Ammonia volatilization is a physico-chemical process where ammonia nitrogen is in equilibrium with a gas phase and hydroxyl ions. This reaction is pH dependable, alkaline values of pH are convenient for the existence of the aquatic form of  $\text{NH}_3$  in solution, while acid or neutral pH values favour the existence of the ion shape of ammonia nitrogen. Nitrification is defined as biological oxidation of ammonia nitrogen to nitrate. Nitrification occurs in two levels, as a result of chemoautrophic aerobic bacteria of the genera *Nitrosomonas* and *Nitrobacter*. These organisms require oxygen for the oxidation of ammonia nitrogen to nitrite and the oxidation of nitrite to nitrate. Factors affecting the level of nitrification are pH, water alkalinity, temperature, inorganic carbon, microorganisms, and the concentration of ammonia nitrogen. Denitrification is defined as a process of biological reduction to the final nitrogen product—gaseous nitrogen  $\text{N}_2$  or  $\text{N}_2\text{O}$ . In anaerobic conditions and in the presence of an available organic substrate denitrified organisms can use nitrates as electron acceptors during respiration. Genera included in processes of denitrification are *Pseudomonas*, *Thibacillus*, *Acinetobacter*. In anaerobic conditions some microorganisms oxidize carbohydrates to carbon dioxide and water using oxygen from nitrates as an electron acceptor and transforming nitrates to gas nitrogen (Canter L.W., 1997).

Nitrogen can be brought into soil in organic and inorganic shape, depending on the source. Organic nitrogen originates from compounds such as amino acids, amines and proteins. Inorganic nitrogen occurs as ammonia, ammonia ion, nitrites and nitrates. Nitrogen from untreated and partially treated industrial and communal waste waters and manure can appear in organic and inorganic shape, while in chemical fertilizers, it appears mostly in the shape of ammonia and nitrate ions. Ammonia nitrogen submits readily to ion exchange in soil, while nitrates remain soluble, and are carried easily by groundwater. If ion exchange zones become saturated, as it is the case with sandy soils, ammonia nitrogen breaks through to groundwater before it is nitrified.

The presence of nitrates in groundwater, especially drinking water affects human health badly and also has negative impact on animals and plants. With human beings, nitrate toxicity is the consequence of processes of nitrate reduction in nitrite. The reaction begins in spittle in all age groups as well as in a gastrotestinal tract of babies during first three months of life. In high doses, nitrite toxicity is demonstrated by cardiovascular disorders, namely methemoglobinemia at lower doses. Methemoglobinemia refers to processes in which hemoglobin oxidises to methemoglobin. When the level of methemoglobin in blood rises, the level of oxygen falls. These processes result in disappearance of oxygen, and the disease is called cyanosis. Effects of methemoglobinemia are fast reversible and do not have cumulative effects. Babies to three months of age are the most sensitive population in relation to nitrates. This is the consequence of the fact that about 10% of taken nitrates is transformed to nitrites. In that case babies fall ill with cell anoxia and clinic cyanosis ( they get blue colour, so called blue baby syndrome). Accompanying effects include low activity of enzymes reducing methemoglobin, increasing methemoglobin sensitivity to oxidation and increase pH in a stomach, which accelerates bacterial reduction of nitrates in nitrites.

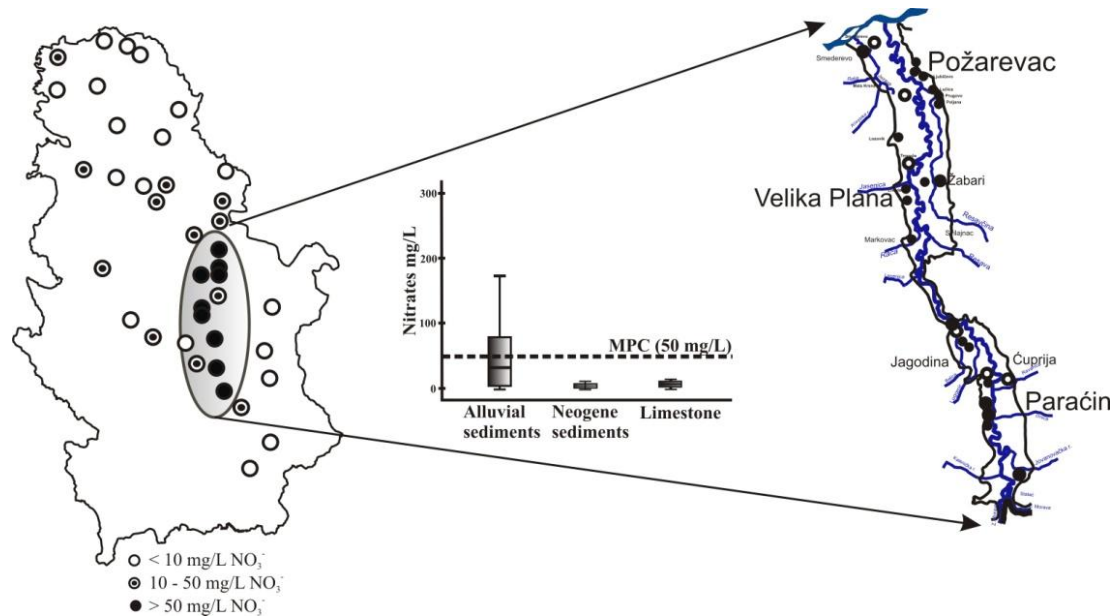
Another consequences of drinking water contaminated by nitrates should be emphasised, among which the most significant one is their cancerous impact on many organs in a body. Mutagenic activity of nitrates and nitrites is related to the basis separation and the cleavage of chromosomes. The remaining toxic effects of nitrate and nitrite competitively inhibit taking iodine from a thyroid gland, modify the composition of phospholipid of various organs (liver, brain), cause pseudoallergies and reduce the concentration of the vitamin B group.

Maximal permissible nitrate concentration in drinking water is 50 mg/L. The World Health Organization (WHO) recommends concentrations to 10 mgN/L for nitrates and 1mgN/L for nitrites.

### Example

In order to define the quality of groundwater for public supply on the territory of Serbia from the aspect of nitrate presence, groundwater samples have been analyzed from 100 sources of regional and local significance. Most analyzed samples are taken from alluvial and karst aquifer and Neogen

basins. The quality of the groundwater of the alluvial aquifer is under the direct influence of rainfall, surface water and human activities. The water bearing sediments in Neogene basins are mainly formed of sand which alternate with low permeable clays. The water of karst aquifers are characterized by good quality, low mineralization on the one hand and the very unfavorable conditions for protection against pollution, on the other. Based on analyzed data hydrochemical map was prepared to show nitrate concentrations in groundwater in order to understand water quality in terms of this parameter (Figure 1).



**Fig. 1** – Hydrochemical map of nitrate concentration in groundwater sources of Serbia and concentrations of nitrates in groundwater by aquifer type (minimum and maximum values of the median)

The mean value of nitrate concentration, on the basis of chemical analysis of groundwater from the groundwater sources, is 29.81 mg/L, with a range of concentrations ranging from 0 to over 253 mg/L, and 77% of samples do not exceed MPC in drinking water. Analyzing the results by litological composition, it was concluded that water from the Neogene sediments are not loaded with nitrates (Figure 1). These groundwaters from deeper aquifer are not affected by the penetration of pollutants from the shallower parts of the terrain, and are protected by impermeable clay layers. Water from the karst aquifer have negligible concentrations of nitrate, while waters most loaded with nitrates are from the alluvial aquifer, where as the most polluted groundwater sources are those formed in alluvion of Velika Morava river. The quality of groundwater in the area of Velika Morava alluvion is affected by the application of fertilizers in agriculture, industrial, comunal and animal wastewater. One of the biggest problem in this area is the lack of sewerage system in rural areas and wastewater discharged into the primitively made septic tank, or abandoned wells, thus establishing a direct connection with the contaminated aquifer waters.

In order to determine the prevailing source of contamination and the nitrate impact on the quality of groundwater of one part of the Velika Morava River alluvion (the source zone for the water supply of the town of Požarevac), the exploration has been carried out based on the application of the isotope method. The application of dual isotope method (proportion of  $^{15}\text{N}/^{14}\text{N}$  and oxygen ( $^{18}\text{O}/^{16}\text{O}$ )), has identified two main sources of contamination: organic soil nitrogen on one side and sewage on the other. In spite of agricultural activities, the presence of nitrates from this source is irrelevant (Miljevic N. et. al 2011).

This can be explained by the fact that the source is recharged from the Velika Morava River direction and from the direction of the settled hinterland lacking a regulated sewage system. The average annual value of nitrate concentration in groundwater of this source wells reached the value of 90 mg/L  $\text{NO}_3$  in 2011.

## Conclusions

Nitrates occur naturally as part of the nitrogen cycle and their presence in groundwater, especially that used for drinking has extremely harmful consequences on human health. Maximal allowed nitrate concentration in drinking water is 50 mg/L. In order to define the quality of groundwater for public supply on the territory of Serbia from the aspect of nitrate presence groundwater samples were taken from 100 sources of regional and local significance. Most samples were processed from compact and karst aquifers. Sources formed within the compact aquifer are mostly related to two environments: alluvion and Neogene basins. The results have shown that 77% samples meet prescribed MPC of 50 mg/L. Among the listed lithological environments, the most contaminated are waters from alluvial aquifers where, from the regional point of view, the most endangered are waters of the Velika Morava River alluvion.

By isotope methods, it has been determined that the source of nitrates in groundwater in part of this alluvion by its origin from soil organic nitrogen and waste water of rural population situated in the recharge zone of the alluvial aquifer where there is not a regulated sewage system.

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