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## DEVELOPMENT AND OPTIMIZATION OF BIOLOGICAL TREATMENT OF QUARRY WATERS FROM MINERAL NITROGEN IN THE SUBARCTIC

ABSTRACT. The new concept of bioremediation of anthropogenic water bodies and guarry wastewaters treatment by phytoextraction and phytotransformation in the Subarctic conditions is presented. This technology is based on transformating the mancaused water reservoirs into nature-like marsh ecosystems. At the first stage, a new patented method for advanced waste treatment using floating bioplate was developed and implemented. After implementing the bioplate, the concentration of ammonium ions in water decreased by 53-90%, nitrate nitrogen reduced by 15-20%. At the second stage, the floating bioplate technology was modified into the highly efficient purificating marsh ecosystem, which allowed to cover the waterbody territory to the greatest possible extent. The technology is based on the creation of phytomats enabling in the accelerated mode to form plant blocks of three different types. They are aimed both at local grassing down, and at swamping deep and shallow areas of sediment ponds. In forming phytomats, two soil substitutional substrates (thermovermiculite and wood sawdust) and regionally-optimized assortment of 24 plant species are used. The proposed technology does not require energy, chemicals and soil components which are scarce in the region. The predominance of natural ecosystem processes in the formed phytocenoses allows to achieve maximum efficiency, and the use of available materials contributes to minimizing the costs of creating and maintaining the system. The introduction of this technology and formation of the artificial phytocenosis with the area of about 30% of the man-caused reservoirs territory made it possible to increase the efficiency of wastewater treating from mineral nitrogen compounds by 22%.

**KEY WORDS:** bioremediation, sewage quarry, sediment pond, mineral nitrogen compounds, phytocenosis, phytomats

**CITATION:** Maria V. Korneykova, Vladimir A. Myazin, Lyubov A. Ivanova, Nadezhda V. Fokina, Vera V. Redkina (2019) Development and optimization of biological treatment of quarry waters from mineral nitrogen in the Subarctic. Geography, Environment, Sustainability, Vol.12, No 2, p. 97-105 DOI-10.24057/2071-9388-2019-5

#### INTRODUCTION

The arctic regions attract attention with their enormous resource potential. This leads to increased environmental problems related to the vulnerability of nature and purification of industrial wastewater from mining enterprises. Ammonium nitrates and additives, such as nitromethane and sodium nitrate, are the main components of the explosives used in drilling and blasting operations at mines. Use of explosives is accompanied by contamination of mine and quarry waters with compounds of the nitrogen group.

So far, alongside physico-chemical and microbiological methods, the approaches based on the use of natural processes occurring in landscape and aquatic ecosystems are thought to be the most promising for cleaning quarry waters of nitrogen compounds (Yakovlev et al. 1985; Vurdova and Fomichev 2001; Birman and Vurdova 2002; Jin et al. 2002; Mattila et al. 2007; Savichev 2008; Ksenofontov 2010; Nefedyeva et al. 2017). To implement these purification methods, constructed wetlands with higher vegetation, substrates, and associated microbial communities are created. They are reliable and effective, do not require energy and chemicals, and do not have an additional negative impact on the environment. Such systems are widely used for post-treatment of sewage water from various pollutants, including mineral nitrogen compounds after primary purification of sewage by mechanical and physicochemical methods (Ran et al. 2004; Stewart et al. 2008; Miranda et al. 2014; Vymazal 2014; Zhang et al. 2014).

The experience of applying this method in Sweden, Finland, Norway, Canada, and Russia shows that constructed wetlands as post-treatment facilities are effective even at low temperatures (Jenssen et al. 1993; Maehlum et al. 1995; Nyquist and Greger 2009). However, the creation and operation of artificial phyto-cleaning systems and plant communities in the northern areas (short vegetation period, prolonged low temperatures, strong winds, and lack of soil resources) are sure to be difficult and requirean individual approach. The aim of the given research is the development and optimization of biotechnology for post-treatment of quarry waters from nitrogen compounds using phytoremediation in the natural-climatic environment of the Murmansk region, Russia.

#### MATERIALS AND METHODS

The research was carried out at the settling pond of quarry for iron ore mining in the Murmansk region of Russia. The settling pond was an excavated pond, consisting of two sections, separated by a bulk dam of sandy-gravelly-rocky soil.

Ground and thawed waters, as well as precipitation enter the collection unit at the bottom of the pit. There, they are piped through the pipe system to the first section of the settling pond. Using sedimentation, the sewage is mechanically treated. Later, passing through the dam, the water is cleaned by filtration, adsorption, precipitation, and oxidation-reduction reactions carried out during the life of microorganisms in the filtering stratum. The latest stage for post-treatment of quarry waters from mineral compounds of nitrogen by phytoremediation takes place in the second section of the settling pond. Its depth is not greater than 2 m and is characterized by a frequently changing level of water. From it, the purified water enters a natural watercourse through the collector.

The growth of the coastal strip with local plant species around the pond is very slow; the vegetation occupies an extremely small part of the surface, and the forming groups of plants are thin. The cover is usually in the range from 1–2 to 0.5%. Often, only lone individuals are found.

In the settling pond, vegetation is mainly represented by the coastal-water communities of the helophytes with an insignificant development of the attached and free-floating mono-species of hygrophytic communities. The submerged-water macrophytes are absent.

The development of higher hygrophytic vegetation is impossible in the I-st settling

pond section because of rather muddy water conditions. The bottom cannot be seen from the depth of 0.5 m. Community formation occurs only due to coastal overgrowth by helophytes and temporarily flooded hydrophilic species. In the II-nd settling pond section, the water is less muddy, and the bottom is viewed from the depth of 1.0–1.2 m. However, the bottom of the reservoir is mostly large-stony and strongly cluttered with flooded wood. Aquatic vegetation is found only at the silted and flattened bottom of the coastal shallow. It is represented by mono-species horsetail communities and abundant sedge-cotton grass communities, passing into small coastal sedge marshes.

Due to the use of explosives during iron ore extraction, 5000–6000 kg of nitrates, 30–50 kg of nitrites, and 60–80 kg of ammonium nitrogen are supplied monthly with quarry water to the settling pond. At the beginning of the study, the average content of nitrogen mineral forms in water was:  $134.24\pm10.10$  mg/l of nitrates,  $0.75\pm0.18$  mg/l of nitrites, and  $1.26\pm0.30$  mg/l of ammonium ion. The content of chlorides, sulfates, and iron did not exceed the MPC (maximal permissible concentration). The water pH varied from 7.6 in the I-st section, to 6.8 in the II-nd section, and to 6.6 in the natural water flow.

The analytical work was carried out in the specialized accredited laboratories of INEP

KSC RAS (Apatity) and the JSC "Olkon" (Olenegorsk). The water quality indicators were analyzed in accordance with the current standard technical documentation. The content of nitrates was determined by the ionometric method with electrochemical laboratory fluid analyzer "Multitest IPL-102" and measuring electrode Elite-021 (NO3-). The content of nitrites in water was determined by the photometric method with Griess reagent. Determination of ammonium ions was carried out by photometric method with Nessler's reagent.

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## **RESULTS AND DISCUSSION**

During the period of 2012 to 2017, we conducted laboratory and field pilot industrial experiments on the phytosystem to inspect wastewater post-treatment for mineral nitrogen compounds. The research was fulfilled in two stages.

The first stage. At this stage, a new patented method for post-treatment of waste quarry waters using the floating bioplate was developed and introduced (Evdokimova et al. 2015). It is represented by the floating clusters of connected frames with biological loading (Fig. 1). It was experimentally established that the bioplate area should be at least 40% of the total area of the treated reservoir for more rapid and effective treatment of the reservoir from nitrogen compounds (Evdokimova et al. 2016).



Fig. 1. Floating clusters of bioplate

As the result of using the floating bioplate with higher vegetation in the course of 3 years, its viability and possibility for absorbing nitrogen compounds, mostly ammonium and nitrite forms, and in a lesser degree nitrate forms, are shown (Table 1).

| Table 1. The content of mineral nitrogen compounds in the settling pond water in |
|--|
| 2013-2016, mg/l  |

| Date       | NH <sub>4</sub> + |           | NC         | ) <sub>3</sub> - | NO <sub>2</sub> - |           |
|------------|-------------------|-----------|------------|------------------|-------------------|-----------|
| Date       | l section         | Collector | l section  | Collector        | l section         | Collector |
| 27.06.2013 | 1.29±0.07         | 1.10±0.08 | 155.3±11.2 | 153.1±10.8       | 0.73±0.06         | 0.17±0.02 |
| 10.07.2013 | 0.26±0.02         | 0.07±0.01 | 133.0±9.5  | 137.2±9.6        | 1.06±0.07         | 0.55±0.05 |
| 09.10.2013 | 3.30±0.19         | 2.04±0.15 | 107.9±7.6  | 95.0±6.9         | 0.91±0.07         | 0.80±0.07 |
| 09.07.2014 | 1.51±0.09         | 0.38±0.08 | 138.5±9.8  | 127.3±9.2        | 0.63±0.05         | 0.57±0.04 |
| 16.09.2014 | 3.62±0.18         | 0.52±0.07 | 178.4±12.5 | 180.1±12.6       | 0.41±0.05         | 0.37±0.03 |
| 02.10.2014 | 1.50±0.08         | 0.35±0.05 | 128.6±9.1  | 109.7±7.9        | 0.36±0.04         | 0.14±0.01 |
| 01.07.2015 | 4.20±0.21         | 0.45±0.06 | 168.2±12.5 | 157.4±11.2       | 0.67±0.05         | 0.55±0.06 |
| 29.07.2015 | 1.47±0.11         | 0.42±0.08 | 157.8±11.4 | 150.5±10.5       | 0.58±0.06         | 0.41±0.02 |
| 09.10.2015 | 2.81±0.14         | 0.63±0.12 | 241.1±17.1 | 230.1±17.2       | 0.71±0.05         | 0.40±0.03 |
| 06.06.2016 | 3.30±0.15         | 0.46±0.05 | 131.4±9.2  | 150.5±9.7        | 0.68±0.05         | 0.35±0.03 |
| 26.07.2016 | 8.92±0.48         | 6.45±0.51 | 203.9±14.3 | 206.9±14.6       | 1.23±0.09         | 1.40±0.12 |
| 19.09.2016 | 9.30±0.67         | 6.90±0.45 | 214.6±12.6 | 165.4±11.7       | 1.00±0.08         | 1.10±0.11 |

\*Note: "-" - was not defined

Prior to starting research, ammonium ions decreased by 15% in the second section from the initial content in first section of settling pond. After the bioplate introduction, their content decreased by 53–90%. The nitrate nitrogen concentration in the water decreased by 15–20% after the bioplate construction.

Further observations revealed that the floating bioplate allowed the post-treatment to be carried out exclusively in the deep-water areas of the settling pond. In these circumstances, the coastal strip, dam, shoal, and backwaters were not involved into the process. The swamping of these areas would help to increase the pond cover area with plants significantly and accelerate the restoration succession at the site.

*The second stage.* The goal of this stage was to modify the floating bioplate into a

more efficient phytoremediation ecosystem, allowing maximum coverage of the territory. The research was carried out in 2017. The basis of the development was the technology of creating phytomats, which allowed forming plant communities of different types.

A phytomate is a plastic mesh bag with the dimensions of 0.4 x 0.7 m. Of the mixture, 12.5 dm<sup>3</sup> consisted of sawdust and thermovermiculite. It was taken in the ratio of 4:1 (by volume), and 50 g of the grass mixture was placed into the bag. This technology allowed to form of phytomats up to 10 cm in height and an area of about 0.3 m<sup>2</sup> each. (Fig. 2). The composition of grass mixture included seeds of the grasses, which intensively grow in the Murmansk region, on sandy-stony soils: *Agropyron intermedium* (Host.) Beauv., *Elytrigia repens* (L.) Desv. ex Nevski, *Festuca rubra* L., *Phleum pratense* 

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L., *Leymus arenarius* (L) Hochst., *Polygonum weyrichii* Fr. Schmidt, and *Bistorta vivipara* (L.) Delarbre. The manufactured goods can be stored in a dry state for a long period of time and transported to any distance.

To create phytomats, two types of substrates were used. The first substrate is small-fractioned thermovermiculite from the Kovdor deposit obtained by the electric roasting method (Ivanova and Kotelnikov 2006; Ivanova 2010). The second substrate is sawdust. All the used substrates were not the source of secondary pollution that was established by the laboratory studies.

Thermovermiculite and sawdust are known to have high air, moisture capacity, and sorption. They provide optimal conditions (humidity, aeration, and temperature) for the assured, rapid, and harmonious germination of seeds included in phytomate composition. They also are conducive to intensive growth and development of plants in subsequent stages of ontogeny. They enhance the purifying capacity of the phytosystem not only due to sorption properties, but also as the result of consuming mineral nitrogen compounds by microorganisms, which transforms plant materials.

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The main purpose of phytomates is creating high-quality artificial phytocenoses (plant blocks) of 3 types:

- Plant blocks of type I are for sodding sandy-gravel coastal strips, slopes of filtering dams, and other land areas lacking natural fertile soils. These blocks transform these territories into phyto-barriers (Fig. 3A). After the phytomates have been spread out, they are moistened (5 liters of water/1 phytomate) to start the process of seed germination. The initial germination of seeds in the phytomats occurs on the 5th–7th day, and the mass germination takes place on the 10th–13th day. Throughout the whole period of phytomate functioning, the organic-mineral-vegetable base continues to work as a filter and nutrient layer.

- *Plant blocks of type II* are for creating plant communities in the backwater or shallows. The nutrient organic-mineral substrate



Fig. 2. The plastic mesh bags consist of sawdust, thermovermiculite, and seeds mixture

becomes the basis for strengthening and growing plants at the bottom (Fig. 3b). The phytomats are spread out in groups and submerged partially into water. If the air temperature is favorable, the germination of seeds will be observed on the 7th day. At this point, different types of hygrophytes and hydrophytes in the form of prepared seedlings, cuttings, or adult plants can be introduced in to phytomates. In such conditions, plants take root within 3-5 days and guickly grow at the bottom of the pond.

- Plant blocks of type III are for originating plant communities in the deep-water sites of the ponds (more than 0.5 m). Phytomats are placed on the floating constructions, which ensures their retention and partial submerging into water (Fig. 3c). The floating bioplate, which was used in our earlier work, can serve as an example of such a construction.

The efficiency of guarry water treatment depends to a large extent on the adequate choice of the plant species. There were 24 types of plants used as the main plants of cenosis: Typha latifolia L., Carex sp., Eryophorum angustifolium Honck = E. polystachion L., Eriophorum vaginatum L., Menyanthes trifoliate L., Comarum palustre L., Calla palustris L., Caltha palustris L., Equisetum fluviatile L., Equisetum palustre L., Salix phylicifolia L., Salix caprea L., Sphagnum sp., Lemna minor L., Hippuris sp., Ranunculus repens L., Agropyron intermedium (Host.) Beauv., Elytrigia repens (L.) Desv. ex Nevski, Festuca rubra L., Phleum pratense L., Leymus arenarius (L) Hochst., Polygonum weyrichii Fr. Schmidt, Tussilago farfara L., and Bistorta vivipara (L.) Delarbre

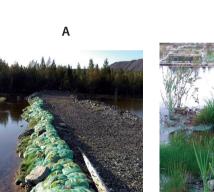
Efficiency assessment of the suggested technology. In the course of monitoring the content of mineral forms of nitrogen in the settling pond water after introducing the proposed technology, the following results were obtained (Table 2).



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Fig. 3. Groups of phytomate, forming various types of phytocenoses: A – plant blocks of type I; B – plant blocks of type II; C – plant blocks of type III Table 2. Nitrate content in waste water in 2017, mg/l

| Place of       | 01.06.           | 20.06.     | 06.07.     | 14.07.     | 04.08.     | 30.08.     | 26.09.     |  |
|----------------|------------------|------------|------------|------------|------------|------------|------------|--|
| sampling       | NO <sup>3-</sup> |            |            |            |            |            |            |  |
| l section      | 90.5±9.4         | 112.5±11.9 | 100.3±12.0 | 100.7±11.3 | 124.1±9.1  | 112.1±9.8  | 125.5±12.3 |  |
| II section     | 11.6±0.8         | 95.8±6.7   | 93.1±6.8   | 90.2±8.2   | 120.0±10.2 | 103.2±8.5  | 111.5±10.1 |  |
| Collector      | 12.2±0.9         | 84.0±5.9   | 85.3±5.9   | 80.6±6.4   | 116.0±9.4  | 106.9±10.5 | 108.5±9.2  |  |
| Natural stream | 16.5±1.2         | 31.5±2.2   | 42.6±4.5   | 45.5±5.1   | 68.2±8.5   | 59.3±5.8   | 70.6±7.5   |  |





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ions was not significant and amounted to

10–25% of the initial level. In the natural

watercourse, the nitrate concentration de-

creased to values not exceeding or equal to MPC (45 mg/l) in June–July; in August–Sep-

tember, it exceeded the MPC by 30–50%. On average, during the observation period,

the nitrate concentration in the II section

water was 84.8±14.9 mg/l, which exceeded the established value of MPC by 2 times.

The ammonium and nitrite ions concentra-

tions in the II section water of the settling

In the course of the work, we noted that the increase of the delay of water in the

settling tank increased, so the duration of

contact with water and coastal vegetation resulted in a marked decrease in the con-

The study of the nitrate ions content in plants revealed their predominant accu-

pond were below MPC values.

centration of nitrate ions.

As the result of dilution during intensive snowmelt, research indicated the nitrate concentration decreased. Subsequently, the decreased concentration of nitrate mulation in the roots of plants growing on the bioplato, compared with the green biomass (Table 3).

Generally, throughout the study period, the efficiency of clearing quarry waters increased by 22% (Table 4).

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#### CONCLUSIONS

This multi-year research-based, low-cost technology has been developed for the transformation of man-caused water bodies into a nature-like wetland ecosystem for the after-treatment of waste quarry waters from mineral nitrogen compounds. The technology is based on the use of phytomats, allowing an accelerated creation of different plant block combinations for landscapes with different moisturizing (coastal areas, backwater or shallows).

The proposed purification system is based on natural mechanisms using aborigine species of higher plants and substrates-soil substitutes (thermovermiculite and wood sawdust); it does not require the use of the

Nitrate content in plant rootsPlant speciesPlants from natural<br/>wetlandsPlants from constructed<br/>wetlandsCómarum palústre L.67.05195.52Eryophorum angustifolium Honck79.78154.12Salix phylicifolia L.77.9695.25

## Table 3. Nitrate content in plant roots, mg/kg

#### Table 4. Efficiency of waste water treatment in the settling pond

| Table in Enterency of Master Mater deather in the setting point |              |              |              |              |             |  |  |
|---|--------------|--------------|--------------|--------------|-------------|--|--|
|   | 2013         | 2014         | 2015         | 2016         | 2017        |  |  |
| Constructed<br>wetlands area,<br>m <sup>2</sup> (%)             | 360 (7.5)    | 660 (13.75)  | 960 (20)     | 1260 (26.25) | 1560 (32.5) |  |  |
| Concentration<br>of nitrates*,<br>mg/l                          | 135.12±10.78 | 169.74±34.16 | 175.82±14.14 | 171.00±12.37 | 84.8±14.9   |  |  |
| Efficiency of<br>wastewater<br>treatment, %                     | 0            | 3.4          | 5.3          | 8.6          | 22.5        |  |  |

Note: \* - the average value for the investigated vegetation period in the collector of the settling pond

energy, chemicals, and scarce soil components in the region. Predominance of natural ecosystem processes in the formed phytocenoses allows for maximum efficiency, and the use of cheap materials allows for the minimization of costs for creation and maintenance. Over the entire period of research, the efficiency of quarry waters treatment averaged 22% with a 30% coverage of the man-caused reservoir with phytomats.

This research is significant because it proposes an integrated approach to forming nature-like wetland ecosystems for wastewater treatment. This has been the first time it was implemented in the practice of existing mining enterprises in the extreme conditions of the subarctic region.

#### ACKNOWLEDGEMENTS

This work was carried out with the financial and technical support of the JSC "Olkon" (the Olenegorsk Mining and Processing Plant).

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Received on January 15<sup>th</sup>, 2019

Accepted on May 17<sup>th</sup>, 2019