Review of the technologies for nitrates removal from water intended for human consumption

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Abstract. The goal of this paper is to review the technologies used for nitrate removal from water intended for human consumption. It also reviews the effects of nitrates on human health and the various legislative provisions in different states. The treatment technologies that will be detailed are: reverse osmosis, ion exchangers, biological denitrification and electrodialysis. For every technology, the following information will be provided: advantages, disadvantages, process performance and cost. Furthermore, there will also be examples of water treatment plants across the Globe that use these types of technologies for nitrate reduction.

1. Introduction
An increase of nitrate concentrations in groundwater is observed worldwide. Although nitrogen concentrations in water sources originate from several causes, the main source of pollution with these compounds is human activity. Thus, nitrogen compounds in water can come from intensive agriculture, especially by applying nitrogen-based fertilizers, as well as discharges of insufficiently treated or untreated wastewater and lack of centralized system for collecting and treating domestic wastewater in localities where there are centralized drinking water distribution systems [1].

Intensive agriculture is considered to be the main cause of nitrogen pollution in groundwater. Because the demand for food is increasing and the principle of crop rotation is no longer sustainable, the soil is drained of resources, so farmers add nitrate-based fertilizers. Properly managed, nitrogen does not endanger human health and increases agricultural production. However, if the amount of nitrogen introduced into the soil is higher than the plants need, the excess nitrogen can seep into groundwater, contaminating them [2].

In many European countries, conducted studies have shown that in the areas where intensive agriculture is practiced, high concentrations of nitrates have been noted. Regarding the fertilizers used in agriculture, it is estimated that only 40-60% of the total amount is used by plants, the rest reaching water sources [3].

The presence of nitrates in drinking water can have negative effects on human health. In reaction with bacterial flora and saliva, nitrates can turn into nitrites, thus affecting children under three months old and inducing methemoglobinemia. Nitrates can form compounds such as nitrosamines and nitrosamides, compounds with carcinogenic potential [4].

If nitrogen or nitrate is converted to nitrite in the human body, it can cause two chemical reactions that lead to health problems: induction of methemoglobinemia, especially in infants under one year of age, and the potentially carcinogenic formation of nitroamides and nitrosamines.

Methemoglobin, normally present in 1 to 3 percent of the blood, is oxidized and cannot act as a carrier of oxygen in the blood. Certain substances, such as nitrites, are formed by the reaction of
nitrate with saliva, but in infants under one year of age, the relatively alkaline conditions in the stomach allow bacteria to form nitrites. Up to 100% of nitrates are reduced to nitrites in infants, compared to 10% in adults and children over one year of age. Also, newborns do not have the same ability as adults to convert methemoglobin back to haemoglobin. When the concentration of methemoglobin reaches values of 5-10%, symptoms may include lethargy, suffocation, and blue coloration of the skin - "blue baby syndrome". Anoxia and death can occur at high concentrations of nitrites or nitrates. However, this occurs quite rarely. According to the WHO, since 1945, there have been about 2,000 cases of illness, about 8% of which have resulted in death [4,5, 6].

In Romania, the maximum allowable limit for nitrates in drinking water, according to Law 458/2002, is 50 mg / l. The guideline level according to the World Health Organization (WHO 2011) for ammonium is 50 mg / l [6, 7, 12]. According to EPA, the maximum allowable limit for nitrates in drinking water is 10 mg N / l [10].

Over time, several methods have been developed to reduce the concentration of nitrates in water intended for human consumption.

If raw water contains nitrogen concentrations above the limit imposed by law, two alternatives can be considered: water treatment or non-treatment.

If you opt for processes that do not involve water treatment, the main methods used are mixing water with good quality water or abandoning the source and using an alternative source of water [13].

Most of the time, the use of an alternative source of water is not possible, thus making the treatment of water before it is distributed to consumers mandatory. The main treatment processes applicable to sources that exceed nitrates are:

- Biological processes;
- Reverse osmosis;
- Electrodialysis;
- Ion exchangers.

2. Main treatment processes

2.1. Biological process

The use of biological processes has been introduced relatively recently in water treatment for water with high nitrates concentration.

Compared to other existing processes, biological denitrification shows that after processing, nitrates can be transformed into nitrogen gas, which is not harmful to the environment or human health [18].

Also, compared to other existing processes (like reverse osmosis, ion exchangers, or electrodialysis), biological denitrification is more efficient and cheaper. [18]

Biological denitrification is a method of reducing nitrates to nitrogen gas with the help of denitrifying bacteria. This process is performed in steps and nitrates are successively reduced to compounds of nitrogen as shown in the following figure:

\[ \text{NO}_3^- \rightarrow \text{NO}_2^- \rightarrow \text{NO} \rightarrow \text{N}_2\text{O} \rightarrow \text{N}_2 \]  \hspace{1cm} (1)

Based on utilized food and energy sources, bacteria are classified into two main categories, named „heterotrophic” and „autotrophic” [14].

Heterotrophic denitrifying bacteria require a source of organic carbon for respiration. If sufficient amounts of organic carbon are not dissolved in water, added substances such as methanol, ethanol, etc. may be used [7, 16].

In order to study the effectiveness of denitrification of groundwater using ethanol as a carbon source, an experiment was conducted at the Haifa Institute of Technology. The results confirmed the theoretical values of the amounts of ethanol used for nitrogen denitrification, namely that the ratio between the amount of ethanol used by bacteria and the amount of reduced nitrogen is equal to 0.5 [8].
Autotrophic denitrifying bacteria are bacteria that do not require carbon sources for respiration and therefore produce denitrification by themselves. They use hydrogen or reduced sulphur species as a substrate and carbon dioxide or bicarbonate is used as the carbon source. [3]

The denitrification process is influenced by certain inhibitory factors such as temperature, dissolved oxygen concentration, pH and alkalinity, inhibitors and the ratio between electron donor and nitrogen [7, 18].

**Advantages**
The main advantages of using biological processes are:

- Low costs, compared to other technologies used (reversed osmosis, ion exchangers, etc.);
- Good efficiency, regardless of the amount of nitrates from the raw water;
- The process does not need chemical substances that make the processes used after this step easier.

**Disadvantages**
The main disadvantages of the biological denitrification technique are:

- The amount of time required for the commissioning of the biological process is larger than all the other available technologies;
- If the water parameters are variable, so are the treated water parameters;
- Additional processes are needed to eliminate substances resulting from microbial activity [16];
- For a proper operation, careful maintenance and monitoring of biomass and its composition are required.

2.2. **Reverse osmosis**
Reverse osmosis is a physical process that uses semipermeable membranes. Water is forced through a semi-permeable membrane under pressure, allowing water molecules to pass through while retaining most of the dissolved materials. [10]. Reverse osmosis can be used to remove multiple contaminants simultaneously (ionic, particles, and organic constituents).

Using a high-pressure system can achieve water efficiency of 85% percent but requires high energy consumption. Reverse osmosis is one of the most expensive forms of centralized water treatment and is cost-effective only if water demand is very low or multiple contaminants require removal.

Reverse osmosis requires a careful review of raw water characteristics and pre-treatment to prevent membranes from degrading. Pre-treatment consists of removal of suspended solids from water. This usually involves passing the water through a series of filters before the reverse osmosis stage.

After the reverse osmosis, all the ions from the water will be removed. As a result, the pH of the treated water will fluctuate unless controlled. Every treatment plant that uses this system, should provide a post-treatment pH and alkalinity adjustments for water stabilization [9].

**Monitoring and maintenance of the installation**
Reverse osmosis systems are usually highly automated, and can be used almost anywhere, regardless of the operator's training.

The frequency of membrane replacement and cleaning is given by the characteristics of the raw water. Periodic cleaning of the membranes used in the reverse osmosis process is required. Acidic substances are used to clean the membranes, which have the role of removing the pollutants stored on the membranes. After cleaning, treated water is used to clean them and they are put back into operation.

Over time, membrane degradation will lead to a gradual decrease in efficiency and the membrane will have to be replaced with a new one. Membrane life varies from 5 to 20 years [13].

As a result of the process, a fairly large volume of concentrated stream (15-50% of the starting volume of water) has to be disposed. Because it has a very high concentration of dissolved substances, especially salts, treatment is required before it can be discharged into the sewer system (if possible). Other possibilities for disposal of the concentrate are represented by discharging the concentrate in salt
waters (seas, oceans), injecting it in deep drillings, in infiltration basins, or the use of drying beds. The management of the residue resulting from the reverse osmosis process is conditioned by the substances dissolved in the concentrate. The characteristics of the raw water influence the quality of the concentrate and thus strongly influence the costs necessary for the safe removal of both the environment and the human health of the concentrate resulting from reverse osmosis.

**Advantages**
The main advantage in using reverse osmosis is that it produces high-quality water, regardless of the raw water quality. Other advantages include multiple contaminant removal, high automation process, easy to use nature, and the system’s suitability for very small stations.

**Disadvantages**
The main disadvantages of reverse osmosis include:
- High energy costs (the water has to be pumped through the membrane at high pressure),
- Water needs pre- and post-treatment;
- A potentially large waste volume that has to be disposed of;
- Changes in water pH, potentially requiring pH adjustment.

**Brighton water treatment plant**
The City of Brighton, Colorado has a population of 40,000 inhabitants. It uses six groundwater wells with a capacity of 160 m³/h. The source is impacted by nitrate with concentrations ranging from 49 to 89 mg/l of nitrate.

The treatment consists of reverse osmosis with blending and it was commissioned in 1993. Since then, the system has never produced water exceeding the maximum allowance limit for nitrates. In the water mixing process, 40% of the total raw water entering the treatment plant passes the reverse osmosis plant and 60% of it enters the pre-treatment stage. After the pre-treatment stage, the water passes through the reverse osmosis installation, the concentrate is removed from the system and the permeate is remineralized and then mixed with raw water, resulting in water with low nitrate content. The concentrate is discharged into the South Plate River. A biological process is used for the treatment of the concentrate. It has the role of reducing the amount of nitrates from the brine.

The benefits of using this technology are represented by the high performance of the treatment and the simple operation. At the same time, the decision was made in the locality to use reverse osmosis to the detriment of ion exchange technology to reduce the salt loading of the South Platte River. Before this (1990), residents used ion exchange technology to treat water at home.

The main disadvantages of using this technology are generated by the amount of concentrate resulting from the process and the large amount of energy required for the operation of the system [1, 13, 15].

| Table 1. Treatment costs [5]. |
|-----------------------------|
| Capital cost                | 8,253,000  |
| Operating and monitoring equipment | 100,000 |
| Annual O&M costs            | 210,000   |
| COMPLETE Cost (including treatment, distribution, everything): | 0.836 $/m³ |

2.3. **Electrodialysis**
Electrodialysis is an efficient and flexible process in terms of nitrate reduction. This process can be compared to reverse osmosis due to the rate of nitrate reduction. Due to the high costs involved,
Electrodialysis is a process that is not widely used. It is suitable for low water volumes, not for high water volumes and thus is not widely used in water treatment.

In the water treatment process, electrodes are inserted into the volume of water and a direct or pulsating current is applied between the electrodes, leading to water electrodialysis [6, 13].

In the process of electrodialysis, ions migrate through selective semipermeable membranes as a result of the electrically charged surfaces of the membrane. A positive electrode (cathode) and a negative electrode (anode) are used to charge the membrane. Nitrate ions move through the membrane to the anode. Continuing to the anode, the nitrate ion is rejected by the ion exchange impermeable membrane and is trapped in the recirculated concentrate stream. The following figure shows how the electrodialysis works.

![Diagram of electrodialysis membrane stack](https://example.com/diagram.png)

**Figure 1.** Illustration of electrodialysis membrane stack [13].

**Monitoring and maintenance of the installation**

The electrodialysis process requires the pre-treatment of raw water. In the case of groundwater, it must be filtered before electrodialysis. If the raw water is loaded with iron and manganese, the pre-treatment must ensure values lower than 0.3 mg/l in the case of manganese and 0.1 mg/l in the case of iron. Hydrogen sulphide can be tolerated up to 0.3 mg/L and turbidity up to 2 NTU.

Membranes used in electrodialysis can be cleaned using dilute acid solutions. Membrane cleaning is recommended to be performed at least once a week, depending on the quality of water influencing the process. [13]

Monitoring is required to assure that the electrodialysis treatment is effective in reducing nitrate concentration. Monitoring includes a daily grab sample or continuous nitrate monitoring of the electrodialysis treated water, or continuous monitoring of a surrogate such as conductivity [9].

**Advantages**
The main advantages of using electrodialysis for nitrates reduction are:

- It requires lower pressure than reverse osmosis;
- The membranes have a fairly long lifespan;
- It can suit any size system.

**Disadvantages**
The main disadvantages of using electrodialysis for nitrates reduction are:
- It requires pre-treatment if high levels of Fe, Mn, H₂S contamination are recorded in raw water;
- The electrodialysis stage effluent requires pH regulation;
- The concentrate resulting from washing the membranes may require special disposal [9].

**Gandia water treatment plant**

Gandia is a city located in the eastern part of Spain, 65 km away from Valencia. The water distributed to the consumers comes from the underground source. Over time, a degradation of the wheels has been observed by increasing the amount of nitrates in the raw water. Because in Spain the maximum limit of nitrates in drinking water is 50 mg/l and the boreholes were worth 80 mg/l, it was decided to implement a new water treatment technology.

Reverse osmosis and electrodialysis solutions were considered the viable solutions for water treatment, the last one being selected due to lower electricity costs and treatment stage with a capacity of 740 m³/h was implemented, consisting of 4 modules with 5 lines for each module. [13]

The following table shows the quality of raw water influencing the treatment stage and the treated water.

**Table 2. Efficiency of the electrodialysis treatment stage, Gandia treatment plant [13].**

| Ion   | Raw water | Treated water | Percent Removal |
|-------|-----------|---------------|-----------------|
| Ca    | 82        | 24.9          | 70%             |
| Mg    | 24        | 8.3           | 65%             |
| Na    | 23        | 10            | 57%             |
| K     | 1         | 0.3           | 70%             |
| HCO₃⁻ | 250       | 99.1          | 60%             |
| SO₄²⁻ | 58        | 12.7          | 78%             |
| Cl    | 29        | 7.5           | 74%             |
| NO₃⁻  | 60        | 16.6          | 72%             |

The main advantages observed in the use of this technology are:
- Relatively low costs due to the long life of the membranes, estimated at 15 years;
- A high rate of water recovery;
- A small amount of concentrate.

2.4. **Ion exchangers**

Ion exchange is defined as a substitution of an ion bound to an inert matrix with another ion by dissolving an ionic bond and forming a new bond, without triggering significant structural changes. Depending on the nature of the exchanged ions, the ion exchange process can be of two types: anion exchange and cation exchange.

The anion exchanger is called the anionite and the cation exchanger is called the cationite.

Ion exchangers are granular, insoluble substances whose molecular structure contains basic radicals or acids that can exchange negative or positive ions with the liquid they are put in contact with. In the process of ion exchange, the total number of ions in the liquid remains unchanged.

The use of ion exchangers is an efficient and economical method for reducing nitrates from water intended for human consumption. The method provides process control, is easy to automate and is not affected by temperature in the usual operating range. The ion exchange method is specially used for small and medium capacity water treatment plants [7, 15]. The water volume is treated in its entirety or only part of it, depending on the concentration of nitrate in the raw water and the concentration required in the distributed water.

The main types of ion exchangers are:
- Organic;
- Anorganic.
Ion exchangers can retain a well-defined amount of ions - ion exchange capacity. After reaching the maximum amount of retained ions, it is necessary to regenerate them. Regeneration involves removing retained ions and replacing them with hydroxide ions (for anionites) or protons (for cations). Depending on the flow direction of the regeneration solution, there are two methods:

- Cocurrent regeneration - the regenerating substance flows in the same direction as depletion;
- Countercurrent regeneration - the regeneration solution flows in the opposite direction to the depletion of the resin. [7, 10, 15]

Countercurrent regeneration is the most commonly used solution. It has two important advantages:

- Higher efficiency and consequently reduced reagent requirements;
- Better quality of the treated water, given the fact that the bottom layers are regenerated with a larger amount of re-agent [10].

**Monitoring and maintenance of the installation**

Nitrate is easily reduced from water effluent by ion exchange resin, but it is difficult to remove from the resin during regeneration; more chlorine ions are needed to remove a single nitrate ion.

The amount of solution resulting from the regeneration may represent about 1% of the treated water. It is loaded with chloride, nitrate, sulphate, bicarbonate and usually, it is disposed of at the wastewater treatment plant.

Also, the dispose of used regenerant contributes to increased treatment costs of using ion exchangers [7].

**Advantages**

Technologies based on ion exchangers ensure process control, are easy to automate, the process can be started in minutes and ensure stable operation, regardless of temperature. Depending on the amount of water to be treated and the concentration of nitrate in it, the whole raw water flow or only a part of it can be treated, and then mixed with the rest of the water flow, which makes it a suitable solution for small and medium capacity treatment plants.

**Disadvantages**

The main disadvantages of this technology are the management of the solution resulting from the regeneration of the resin, the increase of the corrosivity and the aggressiveness of the water due to the replacement of the bicarbonate and sulphate ions in the water. If the technology is used to treat the entire influential flow in the treatment plant, the process can become expensive.
Ion exchange technology used for nitrate reduction was first used in France in 1985. The best-known systems used for denitrification using ion exchangers are the Azurion system and the Ecodenit system [7].

**Ecodenit system**
This system was developed by OTV (Omnium de Traitement et Valorisation) and it is used in Binic treatment plant. Binic is a commune situated in the west part of France.

The treatment plant has a capacity of 160 m³/h and it is used to treat water from a surface source. The concentration of nitrate varies from 45 to 165 mg NO₃⁻/l in the raw water. After the process, 75% of the nitrates are reduced, along with 90% of sulphate ions and 30% of bicarbonates [7].

Regeneration is done using NaCl in cocurrent. The solution resulted from regeneration (55 m³/day) is transported to a wastewater treatment plant.

The same system is used in Pleven where the raw water has a concentration of 70 mg/l of nitrate. Only a part of the whole volume of raw water is treated by ion exchangers and, the treated water is blended with raw water so that the resulted nitrate concentration is below 25 mg/l.

**Azurion system**
This system was developed by Degremont and it is used in two treatment plants from France, Ormes-sur-Voulzie, near Paris and Plouenan, Brittany. Both were commissioned in 1987.

The Ormes-sur-Voulzie treatment plant has a capacity of 27 m³/h, the raw water has a concentration of 60 mg/l and the concentration of nitrate in treated water is below 25 mg/l.

The Plouenan treatment plant has a capacity of 600 m³/h. The raw water is extracted from a surface source and has a medium concentration of 80 mg/l nitrates. During the treatment process, only a part of the volume is treated by ion exchange. After this process, the treated water is mixed with untreated water for obtaining a concentration below 25 mg/l nitrates in distributed water. The solution resulted from regeneration is discharged in the sea.

3. **Conclusions**
As stated, an increase of nitrate concentrations in groundwater is observed worldwide. The main treatment processes applicable to sources that exceed nitrates are:

- Biological processes;
- Reverse osmosis;
- Electrodialysis;
- Ion exchangers.

Biological processes are based on the development of specific biomass for denitrification. To maintain the biomass, a food source must be added for the bacteria, methanol being used in most cases, which means an increase in the concentration of total organic carbon and implicitly the risk of the formation of THM, which are susceptible of being carcinogenic. The process is unstable, must be controlled very rigorously, is very sensitive to inhibitory substances, the initiation of the process with biomass recovery takes about a month and if the biomass is lost, the process must be stopped during its regeneration [11].

Reverse osmosis is a physical process that involves the passage of water through a semipermeable membrane that can retain almost the entire amount of salts in the water. The process takes place at pressures higher than the osmotic pressure. The process results in permeate (ultrapure water) - between 65% and 80% and concentrate between 20% -35%, regardless of the quality of raw water. In general, reverse osmosis is applied only at partial flow, so that by mixing the permeate with raw water that does not pass through the osmosis installation, water that results is within the limits imposed by the legislation in force on nitrogen and doesn’t need remineralization. The fundamental problem is the formation of concentrate, which, like the brine resulting from ion exchange, must be treated, and managed properly. The process has a very high degree of automation, which makes it suitable in rural areas, without the need for a permanent operator with a high degree of qualification [5, 7, 10].
Electrodialysis is a process that is not widely used. It is suitable for low flows. In the water treatment process, electrodes are inserted into the volume of water and a direct or alternating current is applied between the electrodes, thus leading to water electrodialysis [6, 13].

Ion exchangers are masses of ion exchangers especially dedicated to nitrate retention. However, the process has several limitations due to the concentration of chlorides in the water because the nitrogen ion in the water is exchanged with the chloride ion grafted onto the anion exchanger. There may be situations in which concentrations of nitrates are reduced but exceedances of chlorides may occur. Also, although anionites have been designed to be selective, there is still a competitive action given by sulphate and bicarbonate ions. Groundwater generally has high concentrations of bicarbonate that, when competing with the nitrogen ion, lead to premature depletion of the anionite's exchange capacity.

On the other hand, when the exchange capacity is exhausted, the anionite mass must be regenerated and has significant consumption of regeneration brine and the regeneration results in significant quantities of brine with high nitrogen content, quantities which represent a difficult waste to be managed.

The process is relatively stable, but one of the drawbacks is that it can only work fully automated because the concentration of nitrates in the effluent is not constant over a cycle, but increases progressively with the reduction of anionite exchange capacity. The process is only applicable to low exceedances of nitrogen concentrations and requires permanently qualified operating personnel.

Regarding the efficiency of the processes in reducing the amount of nitrates in the water, it differs depending on the process and is in the range of 30-96%.

The following table shows the efficiency of the different technologies used.

| Technology                  | Efficiency (%) |
|-----------------------------|----------------|
| Biological Denitrification  | 70-95          |
| Ion exchangers              | 80-99          |
| Reverse osmosis             | 50-96          |
| Electrodialysis             | 30-80          |

In terms of costs, they vary depending on the concentration of nitrogen and other pollutants in the raw water. An estimate of the costs involved in using the technologies described is presented in the following table.

| Technology                  | Investment cost ($/m³) | Operation cost ($/m³) | Waste storage ($/m³) | Total ($/m³) |
|-----------------------------|------------------------|-----------------------|----------------------|--------------|
| Reverse osmosis             | 0.12-0.23              | 0.29-0.79             | 0.11-0.69            | 0.41-1.71    |
| Ion exchangers              | 0.06-0.31              | 0.12-0.2              | 0.01-0.08            | 0.18-0.57    |
| Biological denitrification  | 0.11-0.24              | 0.13-0.21             | 0.003-0.005          | 0.24-0.45    |

It can be concluded that the use of biological denitrification or ion exchangers is cheaper than the use of reverse osmosis. However, reverse osmosis has the advantage of removing more pollutants without the need for additional steps.

A summary of the main technologies used to reduce the amount of nitrates from the water intended for human consumption, as well as their main advantages and disadvantages is presented in the following table.
Table 5. Technologies used for nitrate reduction.[9, 13,15].

| Quality parameters of raw water that can influence the process | Ions exchangers | Reverse osmosis | Electrodialysis | Biological denitrification |
|---------------------------------------------------------------|-----------------|-----------------|-----------------|---------------------------|
| Sulphates, Iron, Manganese, Bicarbonates, Heavy metals, Organic matter | Turbidity, Iron, Manganese, Total hardness, Heavy metals, Organic matter | Turbidity, Iron, Manganese, Total hardness, Heavy metals, Organic matter | Temperature, pH |
| Pre-treatment | Filtration | Filtration | Filtration | pH adjustment |
| Post-treatment | pH adjustment | pH adjustment, remineralization | pH adjustment, remineralization | Disinfection |
| Process start time | Minutes | Minutes | Minutes | Days/months |
| Quantity of treated water | 97% | 85% | 95% | Almost 100% |
| Advantages | High efficiency; Easy to operate; | High efficiency; Easy to operate; | High efficiency; | Low costs No consumption of chemical reagents |
| Disadvantages | Pre-treatment needed; Automation required; High costs in the case of waters with high mineralization; The solution from regeneration is difficult to manage; | Pre-treatment needed; Automation required; High costs if all the water flow passes through the installation; Remineralization required; The concentrate is difficult to manage; | High energy consumption; The concentrate is difficult to manage; | The process is dependent on the quality of the raw water; Possible formation of nitrites; Heavy process initiation; Additional carbon source required |

Determining the most appropriate treatment technology is not an easy task due to many factors that need to be considered. The most important factors are represented by the characteristics of the influent and the desired nitrate concentration to be obtained in the effluent.

The most appropriate treatment scheme can be determined only after studies and tests are performed for each type of technology. Therefore, pre-treatment and/or post-treatment must be determined and residue storage should also be considered.

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