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# Nitrogen Removal from a Secondary Effluent by Sandy Soil Percolation


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## Abstract

Nitrogen is a required nutrient by all living beings. However, nitrate, the most soluble form of nitrogen, may pollute groundwater used for human consumption; in high concentrations, it is responsible for causing child methemoglobinemia. Nitrogen in groundwater may result from sewage disposal systems, livestock facilities and fertilized cropland. Other possible sources are effluents from sewage secondary treatment plants which are used for green area irrigation. There are several physical-chemical nitrogen-removal techniques, but biological treatment process is used most commonly. With this method, organic nitrogen and ammonia are converted into nitrate in an aerobic environment by means of biological nitrification. Posterior removal of nitrate is reached by anaerobic denitrification, converting it to various gaseous forms of nitrogen.

Chihuahua City is located in the Mexican Republic, in an arid region with very scarce hydrological resources. The drinking water supply is covered by groundwater coming from several overexploited aquifers. To alleviate the water scarcity problem in the city, wastewater is being treated in conventional activated sludge systems, and part of reclaimed water is reused in green area irrigation. In the treatment, organic matter and suspended solids are efficiently removed; however, dissolved solids, including nitrogen in different forms, remain in the effluent. Given the possibility that nitrogen present in the treated water used in gardens could represent a risk of groundwater contamination, a pilot study in experimental systems packed with sand (ESPS) was done, reproducing the conditions of grass irrigation with reclaimed water in Chihuahua's gardens and studying its behavior during infiltration. Removal of ammonia nitrogen, nitrite, nitrate and remnant organic matter was evident in the ESPS, presumably through volatilization of ammonia and combined biological nitrification and denitrification. These results showed that irrigation with the secondary effluent on sandy soils of Chihuahua's gardens doesn't represent a risk of underlying aquifer contamination.

**Keywords**: groundwater, nitrogen contamination, reclaimed water irrigation, biological treatment.

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## Introduction

Nitrogen is widely spread in the environment. It may be found in a variety of chemical forms and different media: as a gas in the atmosphere, in soils, or forming solutions in surface water and groundwater. Nitrogen is a required nutrient by all living beings as a primary constituent in amino acids, which are the building blocks of proteins. It is transported through the food chain from primary producers (algae and plants) to different types of consumers, including humans. Otherwise, excess of nitrogen is undesirable in surface waters, where it can cause eutrophication. In oxidized form, nitrogen may cause pollution in groundwater, so there is a limit to nitrates in water used for human consumption. Potential pollutant sources of nitrogen in groundwater include fertilizers, raw wastewaters and the effluents from secondary treatment, which are discharged into rivers or lakes, or are used for the irrigation of green areas or agricultural crops. Elimination of nitrogen in secondary effluents can be done through various processes involving different levels of technology. An experimental study carried out with water from a municipal wastewater treatment plant showed that nitrogen present in a secondary effluent can be removed by percolation through sandy soils, so the possible contamination risk on underlying aquifer is discarded.

*Presence of Nitrogen in the Environment*

In gas form ($N_2$), nitrogen is the principal constituent of the atmosphere (up to 78% by volume). In this form, nitrogen is relatively inert or unreactive; however, at high temperatures, it may react with other elements and form oxides (FEO, 2013). Some of them are nitrous oxide ($N_2O$), known as laughing gas, nitric oxide (NO), nitrogen dioxide ($NO_2$), and other forms such as $N_2O_3$ and $N_2O_5$. Some of these oxides, often known as $NO_x$, are important constituents of urban air pollution (FEO, 2013). Gaseous nitrogen can also be present in soil pore spaces, where it occurs in both organic and inorganic forms. The predominant inorganic forms of nitrogen in soils are ammonium ($NH_4^+$) and nitrate ($NO_3^-$). Nitrite ($NO_2^-$) and nitrous oxide are present in smaller concentrations (PSSL, 2013). Nitrates enter the soil from rain and fertilizer applied to crops, as ammonium does. Furthermore, nitrogen gas is converted into nitrogen compounds by nitrogen-fixing bacteria, which are present in nodules on the roots of certain plants, acting as a natural fertilizer (WHH, 2007). In this way, the formation of proteins in plants (primary producers) begins to pass them through the food chain to consumers including man.

*Consequences of Nitrogen in Water*

In addition to the benefits provided by nitrogen in the formation of proteins, which are essential constituents of living being cells, nitrogen can generate serious problems in water bodies. In surface waters, such as rivers and lakes, high levels of nitrogen compounds cause eutrophication; it consists of excessive growth of algae and cyanobacteria, which produce toxins and consumption of oxygen, killing water life (WHH, 2007). Moreover, nitrogen can be a potential contaminant for groundwater used for human consumption. The most oxidized nitrogen form, nitrate, is responsible for causing cyanosis or methemoglobinemia (blue baby syndrome) in children under six months. This illness consists of the accumulation of methemoglobin (MeHb) in the blood, which is a form of hemoglobin (Hb) containing oxidized iron that can no longer bind oxygen; and therefore, reduces the oxygen-carrying capacity to tissues of the body (BCDPIC, 2011). According to Denshaw-Burke (2013) "symptoms are proportional to the fraction of methemoglobin level, and include skin and blood color changes at levels up to 15%. As levels rise above 15%, neurologic and cardiac symptoms arise as a consequence of hypoxia; levels higher than 70% are usually fatal". The methemoglobinemia hazard from drinking water with nitrate occurs when bacteria in the digestive system transform nitrate ($NO_3^-$) to nitrite ($NO_2^-$), and the last oxidizes iron in hemoglobin of red blood cells to form methemoglobin. Because infants under six months of age have a higher concentration of the digestive system bacteria known to transform nitrate to nitrite, and a lower than normal concentration of the enzyme known to reduce methemoglobin back to hemoglobin, they are at higher risk for methemoglobinemia. Consuming water from a source containing 10 or less mg/L of nitrate-nitrogen provides assurance that methemoglobinemia should not result from drinking water (Skipton & Hay, 1998).

According to Skipton *et al.* (2008), the EPA Maximum Contaminant Level (MCL) for nitrate-nitrogen in a public water supply (10 mg/L) is based on acute health effects, specifically the risk of methemoglobinemia; acute health effects are those that result from ingestion of a contaminant over a short period of time. Moreover, some studies have shown a correlation between long-term ingestion of elevated nitrate and increased incidence of certain cancers and increased birth defects.

The World Human Organization (WHO) guideline for nitrite is 3 mg/L. This stricter guideline on nitrite relative to nitrate is due to its ability to lead to methemoglobinemia at low concentrations, to which infants are susceptible (Wu *et al.*, 2013).

*Origin of Nitrogen in Water*

Nitrate in groundwater may result from point sources such as sewage disposal systems and livestock facilities, from nonpoint sources such as fertilized cropland, parks, golf courses, lawns, and gardens, or from naturally occurring sources of nitrogen (Skipton *et al*., 2008). Wastewater without treatment coming from sewers, septic tanks or direct discharges in watercourses may contaminate the aquatic environment too.

Nitrate is very soluble in water, so it is the nitrogen form most susceptible to leaching. In irrigated soils, nitrate below the root zone (four to six feet) will leach downward until it reaches the saturated zone of underlying aquifer. The rate of nitrate movement downward depends on a variety of factors, including soil texture, precipitation and irrigation amounts, and crop uptake of water and nitrate (PSSL, 2013).

Other possible sources of nitrogen in groundwater are the effluents from sewage secondary treatment plants. In these systems, biodegradable organic compounds are reduced to a minimum, but nutrients such as nitrogen and phosphorus, as well as other mineral constituents, persist. Effluents from secondary wastewater treatment plants are especially used in arid regions for agricultural and landscaping watering, irrigation of pasture, crops, orchards, vineyards, plantation forests, golf courses, racecourses and other recreation grounds. Irrigation with secondary effluents implies the risk that underlying groundwater may be downgraded. The risk is greatest when effluents have high quantities of nutrients, salts, pathogens or other contaminants (DEC, 2004). Nitrogen is the most important nutrient in effluents from municipal sewage treatment plants. Total nitrogen concentrations are generally between 5 and 50 mg/L and can be present in organic and mineral forms. The mineral compounds include ammonia ($NH_3$), ammonium ($NH_4^+$), nitrate ($NO_3^-$), nitrite ($NO_2^-$) and urea ($NH_2CONH_2$). The relative amount of each of these forms depends on the original constitution of the wastewater and the treatment processes used. When effluents are employed for irrigation, interactions between organic nitrogen, plant growth and the environment are much slower than for most mineral forms (DEC, 2004). Organic nitrogen is converted to ammonium and nitrate through mineralization and nitrification processes, respectively. On the other hand, nitrate can be converted to nitrogen gas through the process of denitrification. In soils, the rate of this process is dependent on a number of complex factors including the availability of water and carbon sources, concentration of oxygen and temperature (DEC, 2004). Nitrogen losses can also occur in soils by volatilization of ammonia.

*Methods to Remove Nitrogen from Water*

There are three basic physical-chemical nitrogen-removal techniques available for application today: ammonia stripping, selective ion exchange and breakpoint chlorination.

Ammonia stripping is carried out through several steps: raising the pH to values close to 11, formation of drops of water on a stripping tower and circulation of large quantities of air through the tower to cause the air-water contact. Ammonia is discharged to the atmosphere as a stable gas that is not converted into nitrogen oxides (EPA, 2013).

The second method is the selective ion exchange process, which involves the use of a zeolite that is selective for ammonia, called clinoptilite. After wastewater is passed through a bed of this resin at a rate of about 10 bed volume per hour, an average ammonia removal of 96% is obtained (EPA, 2013).

The third method, breakpoint chlorination, consists in the addition of chlorine to wastewater containing ammonia nitrogen; ammonia reacts with the hypochlorous acid formed to produce chloramines. Further addition of chlorine to the break point converts the chloramines to nitrogen gas, which is released into the atmosphere (EPA, 2013).

The three methods mentioned above for nitrogen removal have advantages and disadvantages. However, the biological treatment process is used most commonly. With this method, organic nitrogen and ammonia are converted into nitrate in an aerobic environment by means of biological nitrification. Microbial activity is responsible for the two steps of nitrification. First, bacteria *Nitrosomonas* convert ammonium to nitrite. The second step of nitrification occurs through *Nitrobacter* species, which convert nitrite to nitrate. This step rapidly follows ammonium conversion to nitrite, and consequently nitrite concentrations are normally low in water and soils (PSSL, 2013).

Removal of nitrate is reached by biological denitrification; it consists of convert nitrate to various gaseous forms of nitrogen (nitric oxide, nitrous oxide, dinitrogen), which can be lost into the atmosphere. Denitrification occurs under oxygen-limiting conditions when anaerobic bacteria use nitrate in respiration in the presence of carbon sources such as organic matter (PSSL, 2013). Denitrification is considered an effective method since nitrate is converted in nitrogen gas, which is dispersed into the atmosphere without causing secondary pollution.

A technique that combines several physical-chemical and biological processes is the method known as “soil aquifer treatment”, consisting of the application of partially treated sewage effluent to infiltrate into the soil and move down to the groundwater. The unsaturated or "vadose" zone then acts as a natural filter and can remove essentially all suspended solids, biodegradable materials, bacteria, viruses, and other microorganisms. Significant reductions in nitrogen, phosphorus, and heavy metal concentrations can also be achieved (Pescod, 1992). Soil aquifer treatment systems tend to promote nitrification of influent wastewater and transform a large fraction of influent nitrogen to nitrate, as long as aerobic conditions are predominant (Güngör & Ünlü, 2005). The second part of the process consists on flooding and drying periods with a succession of aerobic and anaerobic conditions in the upper part of the soil profile, which stimulates nitrification and denitrification. As mentioned before, in this case, anaerobic bacteria reduces nitrate to free nitrogen gas and oxides of nitrogen that return to the atmosphere. According to Pescod (1992), with this process, about 75% of the nitrogen in sewage can been removed. Denitrification requires the presence of nitrate and organic carbon (an energy source for denitrifying bacteria) under anaerobic conditions. About 1 mg/L of organic carbon is required for each mg of nitrate nitrogen to be denitrified.

Several studies on land treatment of effluents have been conducted. Sharma *et al.* (2011) showed that advanced primary effluent with coagulation followed by SAT could be an attractive option for wastewater treatment and reuse in developing countries. Essandoh *et al.* (2013) performed laboratory studies simulating soil aquifer treatment in 1 m columns containing silica sand under saturated and unsaturated soil conditions; they showed that dissolved organic carbon removal and nitrification did enhance when the wastewater travelled a longer length through the unsaturated zone. Motz *et al.* (2012)

studied the efficacy of nutrient treatment in the vadose zone below a distribution system receiving secondary treated municipal wastewater in cold winter conditions. Results showed that ammonium-N from the applied effluent was effectively nitrified in the top 30 cm of the soil profile. There was also evidence for subsequent nitrate removal by denitrification in this same zone, but it was limited to the warm summer months. Zhang *et al.* (2005) operated a pilot subsurface wastewater infiltration system filled with a mixed soil of red clay and cinder to treat rural sewage. An intermittent operation was adopted to improve nitrogen removal over 90%. Nitrogen balance calculation suggested that nitrification–denitrification was the main mechanism of nitrogen removal.

## Nitrogen Removal Study

*Wastewater Treatment and Reuse in Chihuahua*

Chihuahua City, in Northern Mexico, is located in an arid region where the surface hydrological resources are very scarce. Actually, the drinking water supply of the city is covered by groundwater coming from several regional aquifers, which suffer from overexploitation due to the high demand of the resource. One contribution strategy to alleviate the water scarcity problem in the city has been the treatment of wastewater in two facilities: Planta Norte (Figure 1) and Planta Sur. Reclaimed water is reused for activities where drinking water is not required, as public and private green area irrigation. Both wastewater treatment plants operate with biologic or secondary treatment processes consisting of conventional activated sludge systems. The processes consist of the typical elements related to sieving, sand clearing, grease elimination, primary clarification, aeration and secondary sedimentation with sludge recycling to biological reactor, and finally, water disinfection. Residual sludge is yielded to thickening, anaerobic digestion and dehydration with filter press before its final disposition. According to the existing norms for the biosolid advantages in Mexico (NOM-004), this sludge is classified into excellent quality.

Table 1 presents the characteristics of influent and effluent of Planta Norte corresponding to April 2010. For this period (prior to the completion of this study) the performance in that plant showed efficiencies near or higher than 90% in organic matter (COD and BOD), surfactants, as well as in suspended solids. Grease removal is about 50%; however, nitrogen in different forms is only partially removed, so significant quantities of this element remain in the effluent.

**Figure 1**. *Planta Norte Diagram*

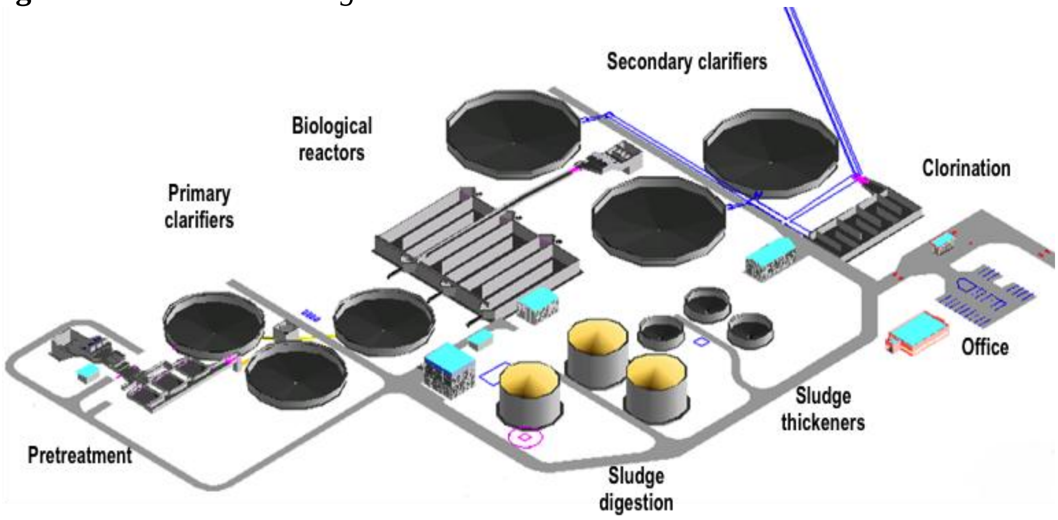



**Table 1**. *Influent and Effluent Characteristics of Planta Norte**

| Parameter | Influent | Effluent |
|---|---|---|
| | Mean value (Range) | Mean value (Range) |
| pH | 7.5 (7.3 – 7.9) | 7.1 (7.0 – 7.3) |
| Total suspended solids (mg TSS/L) | 206 (162 – 359) | 6 (4 – 11) |
| Chemical oxygen demand (mg COD/L) | 439 (341 – 579) | 45 (33-59) |
| Biochemical oxygen demand (mg BOD/L) | 164 (135 – 177) | 17 (9 – 23) |
| Grease and oil (mg/L) | 20 (14 – 30) | 9 (7 – 11) |
| Surfactants (mg MBAS/L) | 24 (18 – 27) | 2.5 (2.1 – 2.9) |
| Fecal Coliform bacteria (MPN/100 mL) | - | 9.1 (<2.2 – 16) |
| Ammonia (mg $NH_3$/L) | 40.7 (39 – 44) | 35 (29 – 37) |
| Nitrate (mg $NO_3$/L) | - | 12.8 (11-15) |
| Nitrite (mg $NO_2$/L) | - | 0.3 (0.1 – 0.5) |

*Data from April 2010 (JCAS)

The qualitative characteristics from effluent regarded to Planta Norte show certain variations throughout the year, depending on the quality of the influent. However, they usually remain in compliance with Mexican Official Norm

(NOM-003), which establishes the required quality limits for the use of treated water in services that involve direct contact, as green area irrigation is. Such limits are 20 mg/L for BOD and TSS concentrations, 15 mg/L related to oil and grease and 240 MPN/100 mL for Fecal Coliform bacteria. Based in such compliance, since 2000 Chihuahua has had a conduction system of secondary effluent from Planta Norte to different city points. In this way, 150 liters per second are distributed for watering public parks, gardens, boulevards, public and private educative institutions, sports and recreational centers, golf and equestrian fields, etc.

*Experimental Study for Nitrogen Removal*

Given the possibility that nitrogen present in treated water that is used in Chihuahua for garden irrigation could represent a risk of groundwater contamination by percolation through the vadose until the saturated zone of the aquifer, a pilot study in an experimental system was done. The objective was to reproduce the actual conditions of irrigation in a small-scale system to study the behavior of the constituents present in the applied water (ammonia, nitrite, nitrate and organic matter) during their infiltration through the soil to the underlying aquifer.

Percolation Unit Description

Behavior of nitrogen and organic matter in a pilot irrigation system was studied from September to December 2010, using the effluent of the activated sludge wastewater treatment facility from the city of Chihuahua (Planta Norte). The application of treated water was done superficially on a percolation experimental unit (PEU) one depth meter packed with sand, in which grass crops were developed to simulate the irrigation that is currently done on the main green areas of the city (Figure 2). The bulk density of the sand was 1.62 $g/cm^3$, with 1.17% of organic matter. The experimental prototype was fitted with structures to collect the percolated water in order to determine both its volume and its physicochemical quality and compare them with the applied water by surface irrigation.

Results of Experimental Tests

The experimental results are shown in Figures 3, 4 and 5.

Figure 3 allows us to appreciate that influent ammonia to the PEU (mean concentration: 20 mg $NH_3$/L) was removed almost at all, while nitrate was diminished from 11 to 5 mg/L, as average values (Figure 4). Nitrite (not shown in plot) was also removed from 3 to less than 1 mg/L. Also, organic matter in terms of chemical oxygen demand (COD) decreased in 25 %, as it is possible to observe in Figure 5.

**Figure 2**. *Experimental Unity for Percolation (PEU)*

Removal of ammonia, nitrite, nitrate and organic matter was evident in the PEU with sand. The processes to reduce nitrogen may have combined nitrification of ammonia followed by denitrification of both nitrate and nitrite to gaseous nitrogen forms. In the first step, occurring in aerobic conditions in upper layers of the PEU, similar to the unsaturated zone of groundwater, ammonia has been oxidized to nitrite and nitrate by autotrophic nitrifying bacteria (*Nitrosomonas* and *Nitrobacter* species) using $CO_2$ as carbon source. In the second step, carried out in absence of oxygen when packed sand was flooded, nitrite and nitrate has been reduced to nitrogen gas by the action of denitrifying heterotrophic bacteria. The organic energy source of carbon for denitrifying bacteria under anaerobic conditions was the remnant organic mater (COD) present in the effluent coming from the secondary treatment.

Other contributions to reduce the nitrogen forms in the PEU presumably were removal of harvestable plant matter from the system and volatilization of ammonia.

These results indicate that garden irrigation with a secondary effluent does not represent a risk to groundwater contamination because nitrogen present can be removed by percolation through sandy soils.

**Figure 3.** *Behavior of Ammonia in Experimental Unit*

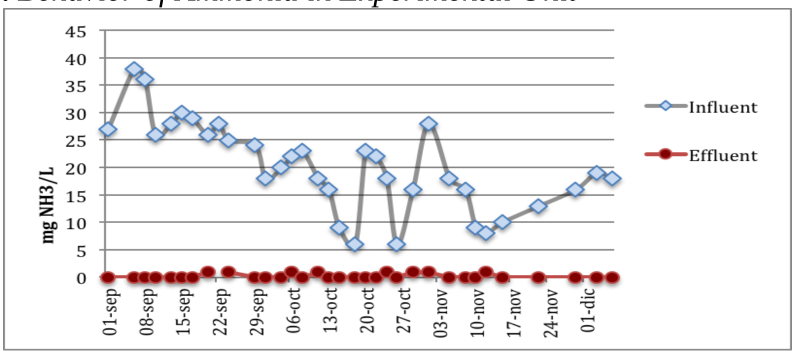


**Figure 4.** *Behavior of Nitrate in Experimental Unit*

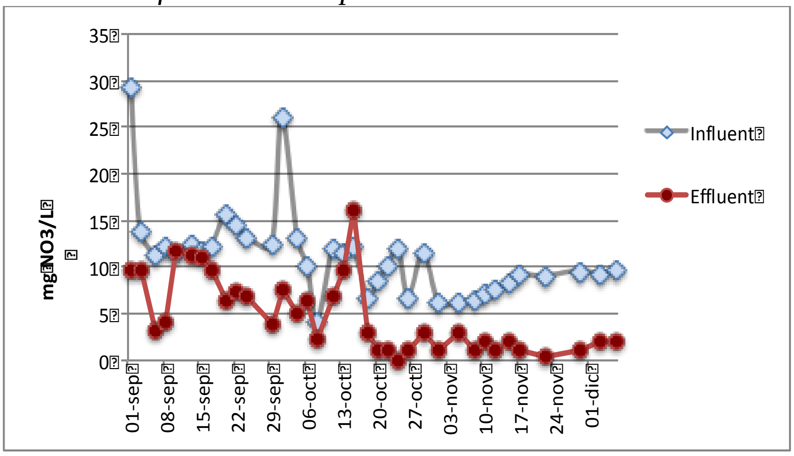
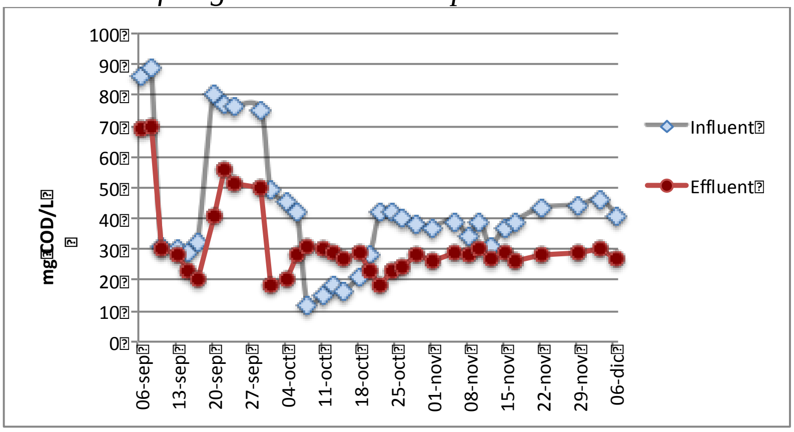



**Figure 5.** *Behavior of Organic Matter in Experimental Unit*




## Conclusions

Just as nitrogen is necessary for all forms of life, it can represent a risk of groundwater contamination. The effluent from Planta Norte (PN) in the city of Chihuahua is a potential source of nitrogen pollution of water used for human consumption in the area.

This experiment showed that infiltration of secondary effluent from PN on sandy soil favors biological removal of nitrogen. So, in green areas of the city of Chihuahua, which are characterized mainly by sandy soils, irrigation with reclaimed water does not represent a risk of nitrogen pollution in the underlying aquifer.

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