

CONTRIBUTION OF ION EXCHANGE PROCESSES IN THE ELIMINATION OF NITRATES CONTAINED IN THE WATERS OF THE ABIDJAN AQUIFER (SOUTH COTE D'IVOIRE)

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ABSTRACT

Nitrogen compounds, including nitrates, are threatening the quality of water in the Continental Terminal aquifer in the Abidjan district. Consumption of nitrate-rich water (over 50 mg/l) can have harmful effects on human health. This study was prompted by the observation that nitrate levels were high in the water from certain boreholes in the Adjamé Nord catchment area, with the aim of reducing nitrate levels in drinking water. To achieve this objective, the water from a borehole was filtered on a weakly basic anionic resin pilot plant to reduce the nitrate content in the drinking water. The variation in the flow rate of raw water passing through the resins was studied to see the effect of the flow rate on nitrate removal. Three flow rates were tested, taking into account the treatment capacity of the pilot plant $(200 \text{ l/h}; 300 \text{ l/h}$ and 800 l/h). The results of the elimination tests show that the resin is effective in denitrating the water. The duration of the cycle varies according to the flow rate (78h for 200l/h; 46h for 300l/h and 14h for 800l/h). At the start of each cycle, the raw water nitrate removal rate reaches 95% and decreases over time. Whatever the flow rate of the raw water used, nitrate removal was satisfactory (95.68% for the 200l/h cycle, 90.81% for the 300l/h cycle and 91.08% for the 800l/h cycle).

Keywords: Denitration, Anionic resin, Ion exchange, Groundwater, Abidjan.

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INTRODUCTION

Groundwater remains the most important source of drinking water in Africa (Ketchemen-Tandia et al. 2017; Jean-Eudes et al., 2022; Remini, 2021). It is currently under threat from nitrate pollution (Aboubakar et al., 2017; Poromna et al., 2022; Savadogo et al., 2023; Laghzal et al., 2018). High nitrate levels have been observed in groundwater in some African countries (Lamribah et al., 2013; Ohou-Yao et al., 2020). In Togo, nitrate levels in groundwater from the Continental Terminal aquifer in the town of Aneho range from 25.84 to 165.35 mg/L (Poromna et al., 2022). In Niger, studies by Rabilou et al. (2018) show that nitrate levels in groundwater in the Zinder region range from 62.04 to 146.96 mg/L. Koussa and Berhail, (2021) show that nitrate levels vary from 4.20 to 160 mg/l in groundwater in the Djelfa region (Algeria). In Côte d'Ivoire, and more specifically in the Abidjan district, the groundwater exploited in the Continental Terminal aquifer to supply drinking water is also threatened by nitrates (Oga, 1998; Jourda et al.*,*2006; Oga et al., 2007; Ahoussi, 2008; Soro et al.*,* 2010; Savadogo et al., 2023). Recent studies by Savadogo et al (2023) show that nitrate levels vary between 1 and 198 mg/L in groundwater from the Abidjan Continental Terminal. This groundwater is contaminated by domestic wastewater during the infiltration of rainwater during recharge (Oga, 1998; Kheliel et al., 2017; Faye et al., 2020).

In the rural municipality of Antanifotsy (Malaysia), the underground water (Wells) was characterized by high seasonal shifts. Spatial variation of Nitrite concentration was observed. Almost all of the measured variables were very high during the rainy season, but lower in the dry season. Underground water contamination by nitrate was found during the rainy season (P5 = 60.644 mg/L and P6 = 53.68 mg/L); this is a main threat to the local people's health (Heriarivony et al., 2016).

In the Biskra region (Algeria), the analysis of the physicochemical water quality included 12 samples from boreholes in different aquifers exploited in the area, used for human consumption. The results showed that the water of the limestone aquifer (Maestrichian) is better than other aquifers (phreatic, Miopliocene, Eocene). This affects more particularly the pH, conductivity (mineralization), total hardness and the concentration of the major elements. The study of the pollution parameters showed that there was an excess in the concentration of nitrate, manganese, and ammonia. This may be due to the proximity of agricultural land as well as industrial areas in the case of some boreholes (Bouchemal and Achour, 2015).

Nitrate can lead to pathologies such as methaemoglobinaemia when present in large quantities in the human body (Oluyomi et al., 2008). This disease, also known as "blue disease" or "cyanosis", is common in infants. Ingestion of large quantities of nitrate in humans can lead to the formation of nitrosamine, a compound presumed to be carcinogenic (Moinet, 1990; Bockman, 1990). To avoid this health risk, some boreholes affected by the high presence of nitrate have been abandoned in the District of Abidjan. This is the case for some boreholes in the communes of Adjamé and Plateau (Oga, 1998; Ahoussi, 2008), and also in Benin (Hountondji et al., 2020). These abandoned boreholes have favored a reduction of drinking water production volume and disruption to the water

supply. In the context of rapid population growth, increasing drinking water requirements and growing nitrate contamination of boreholes, it is important to find a way of removing nitrates from groundwater. Several processes have been developed to remove nitrates from water. These include biological processes (Bouteraa, 2023) and physico-chemical processes such as ion exchange processes on resins (Monzie, 2003; Maamar; 2016), adsorption processes on clays and activated carbons (Kheliel et al., 2015; Kheliel, 2018; Fambi et al., 2021; Song et al., 2023) and reverse osmosis (Schoeman and Steyn, 2003). According to Korngold (1992), the process of eliminating nitrates by ion exchange is the most effective and rapid method of water treatment because the other physico-chemical processes are not specific for eliminating nitrates. They are therefore only used if demineralisation of the water is sought at the same time. Numerous studies have been carried out on nitrate removal using the resin ion exchange method, and many studies have been carried out on synthetic waters (Samatya et al., 2006; Nur et al., 2012; Kalaruban et al., 2016). Others have been carried out on real waters such as groundwater (De Heredia et al.,2006; Keranen et al., 2015; Maamar, 2016) and wastewater (Leakovic et al., 2000). The objective of this study is to contribute to the reduction of nitrate levels in groundwater using the ion exchange method on weakly basic anionic polystyrene resins. This study will enable the continued operation of the offending boreholes in the District of Abidjan.

MATERIAL AND METHODS

Presentation of the study area

The District of Abidjan is located in the south of Ivory Coast. It is located between latitudes $5^{\circ}10$ and $5^{\circ}38$ North and longitudes $3^{\circ}45$ and $4^{\circ}21$ West (Fig.1). It is composed of thirteen (13) municipalities: Abobo, Adjamé, Attécoubé, Cocody, Koumassi, Marcory, Plateau, Port-Bouët, Treichville, Yopougon, Bingerville, Songon and Anyama. The District covers an area of $1,160 \text{ km}^2$, or 0.6% of the national territory. It is bordered to the north by the department d'Agboville, to the south by the atlantic ocean, to the southeast by the department of Grand-Bassam, to the south –west by the department of Jacqueville, to the east by the department of Alépé and to the west by the department of Dabou. The climate is equatorial, with four alternating seasons (Kouamé, 2018). A long rainy season (May - July), a short dry season (August - September), a short rainy season (October-November) and a long dry season (December-April). The District has a very extensive hydrographic network, consisting of the Ebrié, Aghien and Potou lagoons, as well as the Ebrié, Aghien and Potou rivers, Aghien, Potou and the Anguédédou, Gbangbo and Banco rivers. The variation in temperature from 1981 to 2020 shows that the average monthly temperature varies between 24. 28°C and 27.59°C, with an average value of 26.32°C. January, February, March and April are the hottest months, with temperatures above 27°C. Average monthly rainfall varies between 18.65 mm and 258.16 mm, with an average of 109.4 mm. January records the lowest rainfall (18.65 mm) and June the highest (258.16 mm).

Figure 1: Map of the District of Abidjan (Savadogo et al., 2023)

Fig. 2 shows the geological map of the various aquifers encountered in the Abidjan District (Jourda, 1987). The study area is located in the Ivorian sedimentary basin of Cretaceous-Quaternary age. The geological formations are sand, clay and marl. The geological formations and the favourable climatic conditions mean that the district of Abidjan has groundwater resources at its disposal. This groundwater is present in three types of aquifer.

The Quaternary aquifer is highly threatened by anthropogenic pollution because of its depth near the surface (Savané et al., 2006; Ahoussi, 2008). The Continental Terminal aquifer or Abidjan aquifer is used to supply drinking water in the Abidjan district. It is the water from this aquifer that is studied in this survey. The Maestrichtian aquifer (upper Cretaceous) is the deepest aquifer in the Ivorian sedimentary basin, and can reach a depth of 200 m.

Figure 2: Geological map of the different aquifers in the Abidjan District (Jourda, 1987)

Raw water or water to be treated

The experiments were carried out on raw water from a borehole used to supply drinking water in the District of Abidjan, more specifically in the zone of Adjamé. This water is drawn directly from the Continental Terminal aquifer, also known as the Abidjan aquifer. The geographical coordinates of the borehole are X=385637.00 and Y=591857.00. Raw water samples from this borehole were taken and analysed from 2019 to 2022 in the dry and wet seasons. The pH of the groundwater was measured in situ using a WTW-type pH meter. Before to pH measurements, the pH meter was calibrated in the laboratory using the following buffer solutions pH 4, pH 7 and pH 10. Turbidity was measured using a turbidimeter of the type HACH 2100Q. Conductivity and temperature were measured using a WTW multiparameter. In the laboratory, the concentrations of chemical elements such as nitrate (NO₃⁻), sulphate (SO₄²⁻), phosphate (PO₄³⁻) and chloride (Cl⁻) were determined spectrophotometrically using a HACH DR 6000 spectrophotometer. Table 1 shows the methods used to analyse the physico-chemical parameters.

Paramètres	Unit	Chemical analysis methods
Temperature	$\rm ^{\circ}C$	ISO27888
Conductivity	μ S/Cm	ISO27888
pH		ISO10523
Turbidity	NTU	EPA1801
Nitrate	mg/L	HACH 8139
Chloride	mg/L	HACH 8113
Sulphate	mg/L	HACH 8051
Phosphate	mg/L	HACH 8048

Table 1: Methods for analysing physico-chemical parameters

Resins

The weak basic anionic resins have been used for the denitration of raw water. The exchange capacity of weakly basic resins is significant for solutions with a pH between 3 and 8. These resins are characterised by the presence of an amine group, responsible for their basicity. The choice of the weak basic resins is linked to the acidic nature of the raw water studied. The weak basic resins are easy to exchange ions with when the water to be treated is acidic (Monzie, 2003). These resins were supplied by CHEMDOC Water Technologies (France).

Water denitration method

Ion exchange on anionic resins is the denitration method used in our pilot study. Ion exchange on anionic resins is the denitration method used in our pilot study. Ion exchange resins are macromolecules that are insoluble in water, carrying ionisable functions and having the potential property of exchanging the mobile ions they contain with other ions of the same sign from the water to be treated (Monzie, 2003). Before treatment, the resins were regenerated with a sodium chloride (NaCl) solution to charge them with chloride ions. The choice of regenerating solution was based on the low concentration of chloride ions in the raw water. During the ion exchange process, the nitrate ions existing in the raw water are replaced by chloride ions from the resins. In practice, before treatment, a solution of sodium chloride (NaCl) is passed through a column of ten (10) litres of the weak basic anionic resins. Next, fifteen cubic metres (15 m^3) of borehole water go through the resin column, which have previously been charged with chloride ions. Finally, samples of raw and treated water were collected for chemical analysis. It is remembered that the raw water comes directly from the offending borehole via a bypass. Taking into account the treatment capacity of the pilot plant, three water flow rates through the resin column were tested: 200 l/h; 300 l/h and 800 l/h to study the effect of water flow rate on nitrate removal. Fig. 3 shows the water denitration pilot unit. The water denitration system is connected directly to a borehole. It comprises a bottle containing 10 litres of resin and a salt tank. The commissioning process involves connecting the device to a power source and opening the raw water supply valve. This allows water to flow through the resin column and into the brine tank. A quantity of salt (NaCl) is added to the brine tank until it is completely dissolved. This will allow the resins to be regenerated and the device to be rinsed. Once the regeneration process is complete, the water is directly denitrated.

Figure 3: Water denitration pilot unit

Raw and treated water samples were taken every (02) two hours. The water sampling protocol consisted of rinsing the sampling bottles with the water to be tested and then collecting the water samples in the bottles of 1litre capacity without leaving any air bubble in them. Chemical analyses were carried out in the laboratory on the taken water samples. Nitrate levels in the raw and treated water were determined spectrophotometrically using a HACH DR 6000 spectrophotometer. The nitrate content of the raw and treated water was used to calculate the percentage of nitrate elimination from the water according to equation 1:

$$
TA = (Ci - Cf) * \frac{100}{ci}
$$
 (1)

- TA: Percentage of nitrate removal from water
- Ci: Nitrate levels in raw water
- Cf: Nitrate levels in treated water

The average of the percentage of nitrate removal from the water was calculated as a function of the flow rates (200l/h ;300l/h and 800l/h) to see if the nitrate removal performance differed from one flow rate to another. The Kruskall Wallis ANOVA test was used to compare these means. The temperature, conductivity, turbidity and hydrogen potential of the raw and treated water were measured. The Student's t test was used to compare the data from the raw and treated waters. There are significant differences between the means of the removal rate if the p-value is less than 0.05. Otherwise, the difference is said to be insignificant. The content of chloride ions was measured in two samples of raw and treated water to see the evolution of chloride ions in the treated water. Chloride ions were measured spectrophotometrically.

RESULTS

Physico-chemical analyses of raw water from the borehole

Tables 2 and 3 show that all the hydrogen potential (pH) values are below the WHO limit value (6.5-8.5). This shows the acidic nature of the borehole water. With the exception of nitrate, the chemical elements analysed comply with WHO guidelines for water intended for human consumption. Chloride, sulphate and phosphate levels are low in the borehole water. Nitrate levels sometimes exceed the WHO limit value (50 mg/L). This water can not be used directly for human consumption. The nitrate content must therefore be reduced before distribution.

Parameters	2019	2020	2021	2022	WHO limit
Temperature $(^{\circ}C)$	25.00	27.60	28.00	27.60	25
Conductivity $(\mu S/Cm)$	190.20	178.60	204.80	204.80	250
Hydrogen potential	4.76	4.77	4.61	4.61	$6.5 - 8.5$
Turbidity (NTU)	0.42	0.42	0.27	0.40	5
Nitrate (mg/L)	39.60	54.60	58.30	61.00	50
Chloride (mg/L)	20.60	20.40	20.00	18.00	200
Sulphate (mg/L)	4.00	3.00	4.00	3.87	250
Phosphate (mg/L)	0.29	0.05	0.12	0.18	0.5

Table 3: Results of physico-chemical parameters of borehole water during the rainy season

Results of the water treatment process

Hydrogen potential of raw and treated water

Table 4 shows that the pH values of the raw water vary between 4.67 and 5.03, with an average of 4.83 ± 0.02 . Those of treated water vary between 4.73 and 5.31 with an average of 4.88 ± 0.11 . All the raw and treated waters were acidic. The p-value of 0.001 shows that there is a significant difference between the pH values of the raw and treated waters.

Raw and treated water temperature values

Table 5 shows the temperature values for the raw and treated water. The p-value is equal to 0.39, showing a non-significant difference between the temperature values of the raw and treated water.

Table 5: Raw and treated water temperature values

Temperature (°C)	Number of samples	Minimum	Maximum	Average	p-value
Raw water	70	23.20	27.70	26.15 ± 1.03	0.39
Treated water	70	23.10	27.80	26.29 ± 0.93	

Conductivity values for raw and treated water

Observation of the table 6 shows that the mean conductivity value of the raw water is 228.38 ± 19.74 µS/Cm and that of the treated water is 224.8714 ± 18.01 µS/Cm. The pvalue (0.28) shows a non-significant difference between the conductivity values of the raw and treated waters.

Turbidity values for raw and treated water

The table 7 shows that the mean turbidity value of the raw water is 0.37 ± 0.14 NTU and that of the treated water is 224.87 ± 18.01 NTU. The p-value (0.48) shows a significant difference between the turbidity values of the raw and treated waters.

Table 7: Turbidity values for raw and treated water

Turbidity (NTU)	Number of samples	Minimum	Maximum	Average	p-value
Raw water	70	0.10	0.74	0.37 ± 0.14	0.48
Treated water	70	0.16	0.75	0.38 ± 0.12	

Results of nitrate removal from water at a flow rate of 200 L/H

Fig. 4 shows the results of nitrate removal from raw water at a flow rate of 200l/h. Analysis of this figure shows that the 200l/h cycle takes 78 hours. During this cycle, the nitrate content of the raw water exceeds the value recommended by the WHO for water intended for human consumption. Up to 30 hours of treatment, the nitrate removal rate varies from 95.61% to 74.05%. This shows very good nitrate elimination at the start of the process. After 30 hours and up to 56 hours of treatment, the nitrate elimination rate gradually decreases from 62% to 2%. After 56 hours, the concentration of the nitrate in the treated water progressively exceeds that of the raw water, in other words, the treated water becomes loaded with nitrate.

Figure 4: Removal of nitrates from water at a flow rate of 200 l/h

Removal of nitrates from water at a flow rate of 300 l/h

The results for nitrate removal percentages at a flow rate of 300l/h are shown in Fig.5. Good nitrate removal is observed, which gradually decreases as function of time.

- From 0 to 22 hours of treatment, the nitrate elimination rate is greater than 70% and can reach 95% at time.
- From 24 to 28 hours of treatment, the rate of nitrate elimination rapidly decreases until it reaches zero, with a rise in nitrate concentration in the treated water that continues up to 32 hours.

• After 34 hours, the nitrate content of the treated water exceeds that of the raw water.

Figure 5: Removal of nitrates from water at a flow rate of 300 l/h

Removal of nitrates from water at a flow rate of 800 l/h

The results of nitrate removal from water at a flow rate of 800 l/h are shown in Fig. 6. This treatment cycle lasts 15 hours. Throughout the cycle, the nitrate content in the raw water remains above the value recommended by the WHO for drinking water. Despite the high speed in which the raw water passes through the resins, very good nitrate elimination is observed, which gradually decreases as a function of time.

- From 0 to 6 hours of treatment, the nitrate elimination rate varies from 90 to 80%.
- From 8 to 14 hours of treatment, the nitrate elimination rate falls from 50 to 20%.

Figure 6: Removal of nitrates from water at a flow rate of 800 l/h

Comparison of water nitrate removal performance as a function of flow rate

Table 8 shows the statistical comparison of nitrate removal percentages as a function of flow rate. The p-value obtained is 0.227. This value is greater than 0.05, showing that the flow rates used give almost similar results. In other words, there is no significant difference between the percentages of nitrate removal from the water, regardless of the flow rates used.

Results of chloride ion concentrations in raw and treated water

Table 9 shows the concentration of chloride ions released in the treated water during the ion exchange process on the resins. Treated water is richer in chloride ions than raw water. But the concentration of chloride ions in treated water remains below 200 mg/l, the WHO guideline for water intended for human consumption.

DISCUSSION

The raw water used to carry out the nitrate removal tests is acidic. This acidity of groundwater in the Abidjan district has been observed by several authors (Jourda et al., 2006; Oga et al., 2007; Ahoussi, 2008; Savadogo et al., 2023). It is linked to the partial pressure of carbon dioxide (Oga, 1998; Oga et al., 2007; Adiaffi et al., 2009). Rainwater becomes acidic as it carries carbon dioxide dissolved in the soil to the water table. The high nitrate content of the water to be treated indicates anthropogenic pollution. This borehole is located in a highly urbanised area of Abidjan where agriculture is rarely practised. The main source of nitrate contamination of groundwater is the infiltration of domestic wastewater into the aquifer. Studies using nitrogen-15 as a tracer indicate that the nitrates present in the groundwater of the city of Abidjan come mainly from urban wastewater (Oga, 1998). The removal of nitrates from groundwater using weakly basic anionic resins gives satisfactory results. Progressively, the cycle time decreases as the flow rate increases. It is longer at low flow rates (200l/h) than at high flow rates (800l/h). At the start of treatment in each cycle, the nitrate removal rate is high, reaching 90% regardless of the flow rate of water through the resin column. This is due to the high availability of resin binding sites, which diminish over time. This phenomenon of rapid nitrate retention at the start of treatment has been observed by certain authors. According to the work of Maamar (2016), using 50 ml of highly basic resins (Purolite A 520 E) showed a good nitrate retention capacity of up to 99% as soon as it was put into service. This phenomenon was also observed by Dziubek and Mackiewicz (2009), who obtained a 99% nitrate removal rate using highly basic anionic resins (IONACSR-7). Nur and colleagues (2012) report nitrate removal rates up to 85%with purolite A 520 and 65%with purolite A 500PS from a solution containing 20mg/l nitrate and 10mg/l phosphate for a resin dose of 1.5g/l. Studies by Nujić and colleagues (2017) indicate a nitrate elimination rate of over 85% with 6 g of highly basic resins (relite). They also report a nitrate removal rate of over 60% obtained using 6 g of highly basic resin (duolite). Studies by Samatya and colleagues (2006) found similar results. The quantity of resins used in the process of eliminating pollutants from water is an important parameter because it determines the saturation time of the resins. Work by Xu and colleagues (2010) on the adsorption of phosphate ions on ion exchangers has shown that availability of pollutant binding sites

increases with the quantity of resins. The greater the quantity of resin, the greater the availability of pollutant binding sites. The nitrate content of the water to be treated plays an important role in the nitrate removal process, as it influences the saturation time of the resin (Yaragal et al., 2022). A high concentration of nitrate in the water to be treated leads to a short saturation time for the resins.

The principle of ion exchange is based on the electrostatic attraction between the ions dissolved in the water and the functional groups present in the resins (sun et al., 2019). The presence of anions such as phosphate $(PO₄³⁻)$, sulphate $(SO₄²⁻)$ and chloride $(Cl₁)$ in groundwater can also influence nitrate removal (Sun et al., 2019). During the ion exchange process, certain ions are trapped first depending on the affinity of the resin. The affinity of a resin expresses its tendency to bind certain ions more than others. Ions with a high valency will be trapped first. This is a purely electrostatic phenomenon that does not take into account other types of interaction (swelling, specific interactions). The resin prefers counter ions of higher valency. The order of selectivity for the most common ions in water is shown below (Kawamura, 2000).

 $CrO_4^2 > SO_4^2 > SO_3^2 > HPO_4^2 > CNS > NO_3 > NO_2 > Br \cdot > Cl \cdot CN \cdot H > CO_3 >$ $HSIO_3$ ⁻ OH^- > F ⁻

Phosphate and sulphate ions, which have a high valency compared to nitrate, are preferentially adsorbed by anionic resins first, which can lead to a reduction in the percentage of nitrate removed from water. It should therefore be remembered that sulphate and phosphate levels are low in raw water, which does not have a major impact on the elimination of nitrate ions.

After a few hours of treatment (56h for the 200l/h flow rate; 26h for the 300l/h flow rate and 14h for the 800l/h flow rate), the nitrate content of the treated water exceeds that of the raw water. This occurs when all the nitrate binding sites on the resins are saturated. The nitrates, thus trapped by the resins are released into the water to be treated, increasing its nitrate concentration. At this stage, the resins need to be regenerated to restore their original exchange capacity. This phenomenon of nitrate release into the water to be treated has been observed by several authors (Kney and Zhao, 2004; Dziubek and Mackiewicz, 2009; Yaragal et al., 2022). The effect of the flow rate of water through the resin column on nitrate removal is also the subject of this study. However, it should be noted that the nitrate content in the water to be treated varied significantly during the trials, which makes comparison difficult. However, the data collected shows that there is no significant difference in nitrate removal performance from one flow rate to another. Regardless of the flow rate used, nitrate elimination was observed at the start of the process and decreased over time. The effect of flow rate on nitrate removal does not correctly appear in this study. In addition, work by Maamar and colleagues (2016) and Sanou and colleagues (2020) has shown that the water flow rate has an impact on the elimination of pollutants. Efficiency decreases as the water flow rate increases.

CONCLUSION

This study was carried out in a context where groundwater is heavily contaminated with nitrates. Water treatment by nitrate removal is based on the use of weakly basic anionic resins with different water flow rates through the resin column. The results obtained show the effectiveness of this technique. At the start of treatment, the nitrate removal rate reached 90% and decreased over time, regardless of the flow rate. A study of the variation in the flow rate of raw water through the resin column (200 l/h, 300 l/h and 800 l/h) shows good nitrate elimination by the pilot plant for all the flow rates chosen with different execution times.

We recommend that the authorities use ion exchange technology on resins in treatment plants in order to produce water of sufficient quality and quantity.

Looking ahead, it will be interesting to study the influence of sulphate and phosphate ions in the process of eliminating nitrates from water.

Declaration of competing interest

The authors declare that they have no know competing interests or personal relationships that could have appeared to influence the work reported in this paper.

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