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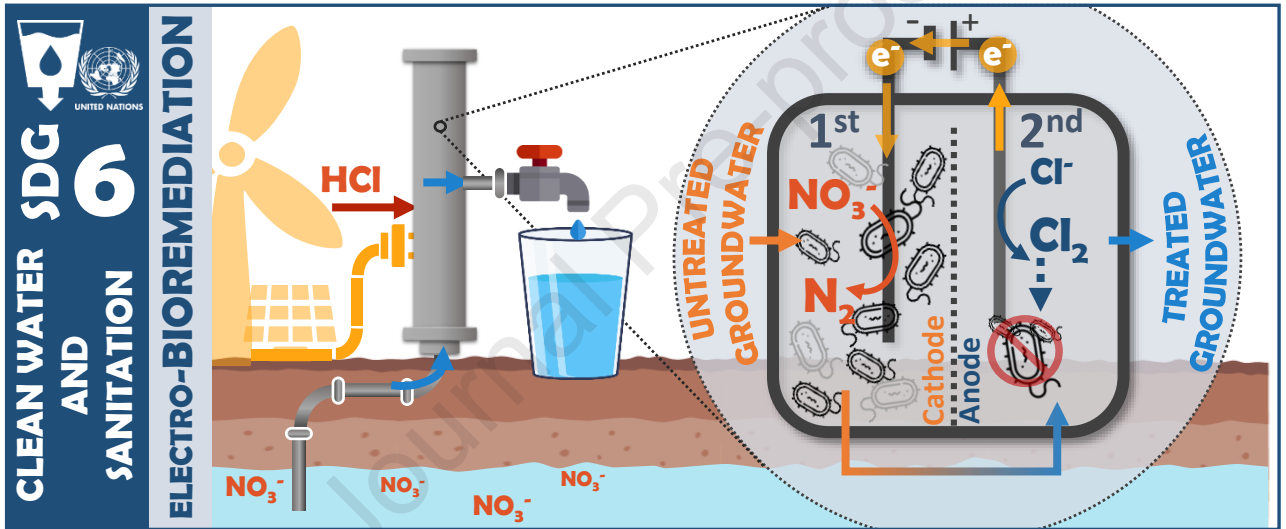
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# **Nitrate electro-bioremediation and water disinfection for rural areas**

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## Abstract:

Nitrate-contaminated groundwater is a pressing issue in rural areas, where up to 40% of the population lacks access to safely managed drinking water services. The high costs and complexity of centralised treatment in these regions exacerbate this problem. To address this challenge, the present study proposes electro-bioremediation as a more accessible decentralised alternative. Specifically, the main focus of this study is developing and evaluating a compact reactor designed to accomplish simultaneous nitrate removal and groundwater disinfection. Significantly, this study has established a new benchmark for nitrate reduction rate within bioelectrochemical reactors, achieving the maximum reported rate of  $5.0 \pm 0.3 \text{ kg NO}_3^- \text{ m}^{-3}_{\text{NCC}} \text{ d}^{-1}$  at an  $\text{HRT}_{\text{cat}}$  of 0.7 h. Furthermore, the on-site generation of free chlorine was effective for water disinfection, resulting in a residual concentration of up to  $4.4 \pm 1.1 \text{ mg Cl}_2 \text{ L}^{-1}$  in the effluent at the same  $\text{HRT}_{\text{cat}}$  of 0.7 h. These achievements enabled the treated water to meet the drinking water standards for nitrogen compounds (nitrate, nitrite, and nitrous oxide) as well as pathogens content (*T. coliforms*, *E. coli*, and *Enterococcus*). In conclusion, this study demonstrates the potential of the electro-bioremediation of nitrate-contaminated groundwater as a decentralised water treatment system in rural areas with a competitive operational cost of  $1.05 \pm 0.16 \text{ € m}^{-3}$ .

## Keywords:

Bioelectrochemical system; Contaminated groundwater; Free chlorine production; Microbial electrochemical technology; Nitrate reduction; Water treatment.

## Abbreviation:

HRT	Hydraulic retention time
$\text{HRT}_{\text{cat}}$	Cathodic hydraulic retention time
NCC	Net liquid cathode compartment
NAC	Net liquid anode compartment
Ti-MMO	Titanium covered with mixed metal oxide
WE	Working electrode
CE	Counter electrode

## 1. INTRODUCTION

38

39 The United Nations has established the objective of universal access to safe drinking water through  
40 the adoption of the 2030 Agenda (SDG 6, A/RES/70/1). Unfortunately, by 2020, around two billion  
41 people will still lack access to safely managed drinking water services. This challenge is particularly  
42 prominent in rural areas, where only 60% of the population has access to safely managed services,  
43 compared to 86% in urban areas (WHO and UNICEF, 2021). This highlights the urgent need for  
44 significant efforts to expand access to safe drinking water in rural areas. Therefore, developing and  
45 implementing novel treatments and technologies are pivotal in bridging this gap and ensuring  
46 universal access to safe drinking water.

47 Intensive agricultural and livestock production practices in rural areas are a major concern, leading to  
48 nitrate contamination of groundwater (Suthar et al., 2009; Yu et al., 2020). Such contamination  
49 threatens freshwater quality and safety, making it inappropriate for human consumption. The  
50 European Directive 2020/2184 has established a nitrate concentration threshold of  $50 \text{ mg NO}_3^- \text{ L}^{-1}$  to  
51 ensure the safety of the drinking water. Furthermore, water is a passive carrier for many pathogens,  
52 including viruses, bacteria, protozoa and larvae (Ashbolt, 2004; Gerba, 2015). This risk arises in rural  
53 areas due to localised contamination, such as faecal and manure leaching, and during water  
54 transportation from the source to the point of use due to unhygienic practices (Chique et al., 2021;  
55 Peter-Varbanets et al., 2009). The same directive (EU 2020/2184) sets *Escherichia coli* and intestinal  
56 *Enterococcus* guideline values of 0 UFC  $\text{mL}^{-1}$  for drinking water.

57 To increase access to treated water in rural areas and drive technological transition in the water  
58 sector, compact decentralised water treatment systems have become crucial. These decentralised  
59 solutions offer effective and sustainable methods, characterised by low operating costs, sustainability,  
60 minimal maintenance, and independence from utilities such as energy sources. Within this context,  
61 electro-bioremediation is one of the emerging decentralised treatments for sustainable groundwater  
62 remediation. Electro-bioremediation involves the utilisation of electroactive microorganisms to carry  
63 out specific oxidation and reduction reactions using solid electron conductors (Wang et al., 2020).  
64 This approach addresses the constraints associated with electron donor/acceptor availability in

65 groundwater. It facilitates the efficient removal of various pollutants, including inorganic substances  
66 (e.g. metals and nutrients) and organic compounds (e.g. hydrocarbons) (Pous et al., 2018).

67 Electro-bioremediation of nitrate has emerged as a promising approach. When nitrate is the target  
68 contaminant, autotrophic denitrification is performed using the cathode as an electron donor and  
69 inorganic carbon as a carbon source. This overcomes the lack of electron donors in groundwater,  
70 avoiding chemical dosing. In particular, nitrate electro-bioremediation offers competitive advantages  
71 over conventional treatments by minimising environmental impacts such as brine formation and the  
72 accumulation of undesirable by-products such as nitrite. It also has a competitive energy consumption  
73 ( $0.25 \text{ kWh m}^{-3}$ , Cecconet et al., 2018) compared to methods such as reverse osmosis, which typically  
74 consumes  $0.9 - 2.2 \text{ kWh m}^{-3}$  (Twomey et al., 2010). Nevertheless, conventional treatments still have  
75 higher treatment capacities with shorter hydraulic retention times (HRT) in the range of seconds to  
76 minutes (Xu et al., 2018). In contrast, studies on electro-bioremediation typically reported higher HRTs  
77 of some hours (e.g., 15.6 h Cecconet et al., 2018, 2.4 h Puggioni et al., 2022 or 3.3 h Wang et al.,  
78 2021). Only one study in this field reported a minimum HRT in the cathode compartment of 0.5 h,  
79 although without reaching the nitrate threshold of  $50 \text{ mg NO}_3^- \text{ L}^{-1}$  in the effluent (Pous et al., 2017).  
80 Therefore, reducing HRT, which would significantly increase the nitrate reduction rate and reduce the  
81 number of reactor units, is necessary to achieve a more competitive treatment.

82 Simultaneously, electro-bioremediation is a versatile treatment option, offering potential water  
83 disinfection through diverse anodic evolution reactions, including chlorine, hydrogen peroxide, ozone,  
84 or radical formations (Bergmann, 2021). Recent research has explored the fusion of electro-  
85 bioremediation with anodic disinfection through chloride oxidation to chlorine to address various  
86 challenges, such as wastewater treatment in secondary settlers (Botti et al., 2023) and nitrate-  
87 contaminated saline groundwater (Puggioni et al., 2021). Hence, integrating water disinfection and  
88 nitrate reduction via electro-bioremediation simplifies the treatment process. This approach can  
89 significantly improve the cost-effectiveness of nitrate-contaminated groundwater potabilisation  
90 through a single-step treatment.

91 This study presents an innovative electro-bioremediation system that combines water disinfection with  
92 nitrate reduction to nitrogen gas in a compact reactor to treat nitrate-contaminated groundwater. The  
93 nitrate reduction rate was significantly enhanced by controlling the cathodic pH to  $6.8 \pm 0.2$ . This  
94 cathodic pH is decisive for the performance due to its strong influence on the denitrification process  
95 in terms of both rate and selectivity to nitrogen gas, as mentioned by other authors (Clauwaert et al.,  
96 2009; Puggioni et al., 2021; Zhao et al., 2022). In addition, the hydrochloric acid used for pH control  
97 was recovered by oxidising chloride to chlorine in the anodic compartment, serving as an effective *in-*  
98 *situ* water disinfectant. In parallel, there is an increasing need to assess future implementation. This  
99 study evaluated treatment performance in terms of nitrate reduction and disinfection capacity. For the  
100 first time, this study prioritised achieving standard drinking water quality using electro-bioremediation,  
101 focusing on addressing nitrate risks and pathogen presence. Finally, the techno-economic  
102 implications were critically evaluated in terms of its benefits and associated operating costs.

## 104 2. Materials and methods

### 105 2.1 Reactor setup

106 A compact tubular bioelectrochemical fixed-bed reactor (Fig. 1) was built with PVC (55 mm diameter  
107 and 350 mm length). The cathode and anode compartments were separated with a tubular cation-  
108 exchange membrane (40 mm diameter, 1 mm thickness and 300 mm length; CEM, CMI-7000,  
109 Membranes Int., USA). The cathode (inner compartment) was filled with granular graphite (average  
110 diameter of 3.25 mm, enViro-cell, Germany) with a bed porosity of 50%, resulting in an estimated  
111 electrode surface area of  $0.4 \text{ m}^2$  and a net cathode volume (NCC) of 0.22 L. The cathode was initially  
112 inoculated with a denitrifying community mainly composed of *Sideroxydans* sp. from another running  
113 denitrifying bioelectrochemical reactor, which was characterised previous studies (Ceballos-Escalera  
114 et al., 2024, 2021). The anode (outer compartment) was a cylindrical titanium mesh covered with  
115 mixed metals oxide (Ti-MMO, 45 mm diameter, 0.5 mm thickness and 200 mm length, Special Metals  
116 and Products, SL, Spain), which is a stable material to promote chlorine formation. The anode surface

117 was 0.2 m<sup>2</sup> with a net anode volume (NAC) of 0.43 L. A potentiostat (VSP, BioLogic, France) was  
118 used to control the reactor electrically in a potentiostatic mode, with the cathode (working electrode,  
119 WE) potential fixed at -0.32 V vs. Ag/AgCl to facilitate complete nitrate reduction to nitrogen gas (Pous  
120 et al., 2015). Under potentiostatic conditions the working electrode (WE, the cathode in this work) is  
121 controlled at a specific value, while the counter electrode (CE, the anode in this work) varies in order  
122 to meet the cathode current requirements. Along the operational study, the potentiostat recorded the  
123 voltage difference between the anode and the cathode (i.e., cell voltage), which was used to calculate  
124 the power requirements of the system.

125  
126

## 127 *2.2. Synthetic groundwater*

128 Synthetic nitrate-contaminated groundwater was used in this study. It mimicked the groundwater of  
129 the village of Navata (Spain). The synthetic groundwater was prepared with distillate water and  
130 contained 203.9 mg L<sup>-1</sup> NaNO<sub>3</sub>, 420.0 mg L<sup>-1</sup> NaHCO<sub>3</sub> as inorganic carbon source, 7.5 mg L<sup>-1</sup>  
131 KH<sub>2</sub>PO<sub>4</sub>, 1.9 mg L<sup>-1</sup> Na<sub>2</sub>HPO<sub>4</sub>, 100.0 mg L<sup>-1</sup> NaCl, 75.2 mg L<sup>-1</sup> MgSO<sub>4</sub> × 7H<sub>2</sub>O, 10.0 mg L<sup>-1</sup> NH<sub>4</sub>Cl  
132 and 0.1 mL L<sup>-1</sup> of a trace minerals solution (Balch et al., 1979). In addition, the influent contained 10%  
133 of effluent from a parent denitrifying bioelectrochemical reactor to simulate the presence of  
134 microorganisms in the groundwater (Ceballos-Escalera et al., 2021). The inorganic medium only  
135 incorporated nitrate as a contaminant (169 ± 5 mg NO<sub>3</sub><sup>-</sup> L<sup>-1</sup>) and bicarbonate as a carbon source. The  
136 resulting influent had an electric conductivity of 1.3 ± 0.1 mS cm<sup>-1</sup> and a pH of 8.0 ± 0.3. The analysis  
137 of pathogens revealed that *Enterococcus* was present in the effluent of the parent denitrifying  
138 bioelectrochemical reactor. The concentration was 1.3 ± 0.9 ufc per 100mL<sup>-1</sup> (Table S1,  
139 Supplementary data).

## 140 *2.3. Reactor continuous operation*

141 The reactor was operated in continuous flow mode. Synthetic groundwater was fed at various  
142 hydraulic retention times (HRT) ranging from 7.0 h to 2.1 h. The HRT was reduced by approximately  
143 25% within one week or until a steady state was achieved. To focus specifically on the cathodic



144 process of nitrate reduction, and make the results comparable to literature, the HRT was also  
145 expressed in terms of cathodic hydraulic retention times ( $HRT_{cat}$ ) ranging from 2.4 h to 0.7 h.

146 Synthetic groundwater was pumped directly through the bottom of the cathode compartment and spilt  
147 over the top into the anode compartment towards the bottom where the outlet was located (Fig. 1).  
148 The outlet of the cathode compartment was recirculated to the influent at a flow rate of  $85 \text{ L d}^{-1}$  to  
149 improve fluid distribution and enhance mass transfer (Vilà-Rovira et al., 2015). Specifically,  
150 recirculation is highly recommended when the electrical conductivity of the water is low, such as in  
151 groundwater (Ceballos-Escalera et al., 2021). A pH probe was installed in the cathodic recirculation  
152 to control the cathodic pH at  $6.8 \pm 0.2$ . Hydrochloric acid ( $0.2 \text{ M HCl}$ ) was used for this control as the  
153 supplied chloride ions could subsequently be oxidised to chlorine.

#### 154 *2.4. Analyses methods and calculations*

155 Liquid samples were collected and analysed following the standard water measurement methods  
156 specified by the American Public Health Association (APHA, 2005). The ion concentration was  
157 determined using an ionic chromatography system (ICS 5000, Dionex, USA) with a detection limit of  
158  $0.01 \text{ mg L}^{-1}$ . Nitrous oxide ( $\text{N}_2\text{O}$ ) was monitored by a liquid-phase microsensor (Unisense, Denmark)  
159 located at the cathodic recirculation. Free chlorine was measured immediately after sampling with a  
160 specific kit (Free Chlorine DPD Reagent Powder Pillows, HACH Company, Loveland, CO, USA). Total  
161 coliforms, *E. Coli* and *Enterococcus* concentrations were analysed externally (Cat-Gairín Laboratory,  
162 Girona). The pH and electrical conductivity of the samples were measured with a pH meter (pH meter  
163 basic 20+, Crison, Spain) and a conductivity meter (EC-meter basic 30+, Crison, Spain), respectively.

164 The performance of electro-bioremediation was assessed based on various factors, including nitrate  
165 removal efficiency, energy consumption, free chloride concentration, and operational costs. The  
166 nitrate reduction rate was determined (Eq. S1, Supplementary data) and normalised by the net  
167 cathode volume ( $\text{kg NO}_3^- \text{ m}^{-3} \text{ d}^{-1}$ ). Energy consumption was calculated (Eq. S5, Supplementary data)  
168 and expressed relative to the amount of nitrate removed ( $\text{kWh kg NO}_3^-$ ) or the volume of water treated  
169 ( $\text{kWh m}^{-3}$ ). The calculation of the cathodic coulombic efficiency considered the presence of potential  
170 intermediates such as nitrite and nitrous oxide (Eq. S6, Supplementary data) (Pous et al., 2017).

171 Two main costs were considered in the estimation of the operational cost for the treatment: (i) the  
172 cost of hydrochloric acid and (ii) the energy consumption from the power supply to sustain the  
173 electrochemical reactions. The price of hydrochloric acid was determined by its commercial  
174 concentrate cost (1.40 € L<sup>-1</sup>, 35% HCl, Ref. 13235T-00/B02, Vadequimica, Spain). The energy cost  
175 in this study was estimated using the electricity price for industrial consumers from the second period  
176 of 2022 in Europe (Eurostat statistics, 0.20 € kWh<sup>-1</sup>).

### 177 3. Results and discussion

#### 178 3.1. Quality of treated groundwater: compliance with drinking water standards

179 For the first time, the overall characteristics of the treated water in the electro-bioremediation process  
180 were evaluated in accordance with the European Directive 2020/2184 (Table 1). This directive  
181 establishes both chemical and microbiological standards to ensure drinking water quality.

182 The nitrate concentration in the treated water remained below the safe limit of 50 mg NO<sub>3</sub><sup>-</sup> L<sup>-1</sup>, with a  
183 concentration range of 4.5 ± 0.6 to 15.1 ± 7.7 mg NO<sub>3</sub><sup>-</sup> L<sup>-1</sup> between the HRT<sub>cat</sub> from 2.4 to 0.7 h.  
184 Besides, the treatment exhibited high selectivity to nitrogen gas (<99%). Harmful denitrifying by-  
185 products, neither nitrate nor ammonium, were detected in the effluent, and the concentrations  
186 remained below the prescribed limits of 0.5 mg NO<sub>2</sub><sup>-</sup> L<sup>-1</sup> and 0.5 mg NH<sub>4</sub><sup>+</sup> L<sup>-1</sup>. Furthermore, the  
187 absence of nitrous oxide in the liquid phases, a greenhouse gas, reinforces the environmental  
188 sustainability of the treatment. Hence, the treatment effectively eliminated nitrate without producing  
189 any harmful by-products in the treated groundwater.

190 In parallel, the free chlorine concentration in the effluent increased from 0.3 ± 0.1 to 4.4 ± 1.4 mg Cl<sub>2</sub>  
191 L<sup>-1</sup> as the HRT<sub>cat</sub> decreased from 2.4 h to 0.7 h. In this scenario, chlorine was considered suitable as  
192 a disinfectant due to the absence of organic matter in the groundwater, preventing the formation of  
193 toxic by-products (Mazhar et al., 2020). The typical residual chlorine concentration in conventional  
194 potable water plants ranges from 0.2 to 2.0 mg L<sup>-1</sup>, with a possible increase in dosage during extreme  
195 contamination scenarios (Brandt et al., 2017). Thus, it was assumed that the chlorine production  
196 attained in this study was satisfactory for the *in-situ* disinfection, ensuring the microbiological quality

197 of the treated water up to the point of use. The disinfection capacity was also evaluated during the  
198 HRT<sub>cat</sub> test of 1.3 h, with a free chlorine concentration of  $1.7 \pm 0.8$  mg Cl<sub>2</sub> L<sup>-1</sup>. This analysis revealed  
199 the absence of Total coliforms, *E. coli*, and *Enterococcus*, as the European Directive 2020/2184  
200 required.

201 The European Directive 2020/2184 indicates other less stringent parameters for monitoring and  
202 ensuring human health protection. Some of these quantitative and qualitative parameters are pH,  
203 electrical conductivity, other ion content, colour, taste and odour. The pH was maintained at neutral  
204 levels, as recommended by the same guideline ( $6.5 < \text{pH} < 9.5$ ), for HRT<sub>cat</sub> higher than 1.3 h (Table  
205 1 and Fig. 2). Only when the HRT<sub>s\_cat</sub> were lower than 1.3 h the pH was dropped below 6.5. At these  
206 conditions, the higher current densities exacerbated the pH difference between the anode-cathode,  
207 implying: (i) higher anode potential from  $1.49 \pm 0.02$  (HRT<sub>cat</sub> of 2.4 h) to  $2.05 \pm 0.12$  V vs. Ag/AgCl  
208 (HRT<sub>cat</sub> of 0.7 h) and (ii) higher requirement for acid dosage (Table 1). The electrical conductivity  
209 gradually increased due to the addition of acid, reaching  $2.0 \pm 0.1$  mS cm<sup>-1</sup> at the lower HRT<sub>cat</sub> of 0.7  
210 h. However, it never surpassed the recommended value of 2.5 mS cm<sup>-1</sup>. Meanwhile, chloride  
211 concentration overcame the suggested value of 250 mg Cl<sup>-</sup> L<sup>-1</sup>, increasing from  $92 \pm 1$  mg Cl<sup>-</sup> L<sup>-1</sup> in  
212 the influent to  $387 \pm 24$  mg Cl<sup>-</sup> L<sup>-1</sup> in the effluent. Finally, this chloride concentration should not have  
213 any health risks. Finally, the colour appears unchanged, while chlorine accumulation would only  
214 slightly influence the taste and odour.

215 In conclusion, electro-bioremediation is a powerful treatment to meet the mandatory drinking water  
216 requirements. Specifically, the more sustainable HRT<sub>cat</sub> to meet the neutral pH was 1.3 h (Table 1).  
217 Nevertheless, to sustain operation at lower HRT<sub>s\_cat</sub> and achieve a higher treatment rate, a viable  
218 approach is to blend the treated groundwater with a fraction of untreated groundwater. This method,  
219 commonly used in drinking water services, would balance the pH in lower HRT<sub>s\_cat</sub> and decrease  
220 chloride levels while maintaining safe nitrate levels.

### 221 *3.2 Techno-economical implications for decentralised water treatment*

222 Electro-bioremediation is a promising option for sustainable decentralised water treatment, even  
223 though its real applicability is currently being evaluated. The success of this transition relies on the

224 treatment's effectiveness, competitiveness, and feasibility. Therefore, after verifying the satisfactory  
225 quality of the treated water, the present study carefully evaluated key factors such as reaction rates,  
226 efficiency and costs to assess the competitiveness of the treatment (Fig. 2).

### 227 3.2.1 Improving Nitrate Reduction Performance

228 Nitrate removal in the presented treatment was achieved through the denitrifying biocathode, utilising  
229 the cathode as the sole electron donor. Previously, a cathode potential of -0.32 V vs. Ag/AgCl was  
230 identified as the optimal potential for achieving the highest nitrate reduction rate in similar  
231 bioelectrochemical reactors (Pous et al., 2015). Specifically, the biocathode used to inoculate the  
232 reactor in this study exhibited a robust electroactive response at a cathode potential of -0.32 V vs.  
233 Ag/AgCl in the presence of nitrate in the media. Further electrochemical characterisation by cyclic  
234 voltammetry revealed a formal potential of approximately -0.20 V vs. Ag/AgCl associated with nitrate  
235 reduction (Figure S1, Supplementary Data) (Ceballos-Escalera et al., 2021). Meanwhile, testing  
236 similar denitrifying biocathodes with cyclic voltammetry indicated a clear electrochemical response in  
237 the presence of nitrate, with a wide range of formal redox potentials between -0.200 V and -0.70 V  
238 vs. Ag/AgCl (pH 7.0-8.0) (Ceballos-Escalera et al., 2024; Korth et al., 2022; Pous et al., 2016, 2014).  
239 Moreover, the electric current of the reactor was enhanced by reducing the  $HRT_{cat}$ , which resulted in  
240 higher nitrate reduction rates (Table 1). At the same time, the reactor demonstrated a remarkable  
241 coulombic efficiency in nitrate reduction, assuming the cathode as the sole electron donor, with an  
242 average of  $101 \pm 6\%$  observed in all tests (data not shown). This highlights the strong selectivity of  
243 nitrate removal using the electrode as the electron source.

244 The nitrate removal efficiency remained consistently above 90% in all tests. Complete reduction to  
245 nitrogen gas was achieved without accumulating intermediates such as nitrite or nitrous oxide. As a  
246 result, by decreasing the  $HRT_{cat}$  from 2.4 h to 0.7 h, the nitrate removal rate increased from  $1.7 \pm 0.0$   
247 to  $5.0 \pm 0.3 \text{ kg NO}_3^- \text{ m}^{-3}_{NCC} \text{ d}^{-1}$  (Table 1, Fig. 2). Although the highest nitrate reduction rate, while  
248 maintaining the recommended neutral pH according to the European Directive 2020/2184, was  
249 achieved at an  $HRT_{cat}$  of 1.3 h with a rate of  $2.9 \pm 0. \text{ kg NO}_3^- \text{ m}^{-3}_{NCC} \text{ d}^{-1}$ . As far as the author knows,  
250 this study has achieved the highest reported nitrate reduction rate in a bioelectrochemical system,

251 reaching a maximum rate of  $5.0 \pm 0.3 \text{ kg NO}_3^- \text{ m}^{-3}_{\text{NCC}} \text{ d}^{-1}$ . Previous studies have reported a maximum  
252 nitrate reduction of up to  $3.7 \text{ kg NO}_3^- \text{ m}^{-3}_{\text{NCC}} \text{ d}^{-1}$  at  $\text{HRT}_{\text{cat}}$  of 0.5 h (Pous et al., 2017).

253 The intensification of the process in the cathode compartment can be mainly attributed to two key  
254 operating procedures: (i) applying pH control at the cathode and (ii) implementing internal  
255 recirculation. pH plays a critical role in the denitrifying bioelectrochemical reactor, with neutrality being  
256 identified as the optimal pH (Rogińska et al., 2023). In particular, a more fundamental study of  
257 denitrifying biocathodes has shown a suitable pH in the range of 6 to 8 (Korth et al., 2022).  
258 Furthermore, nitrate reduction is a pH-dependent process that consumes protons. This consumption  
259 leads to an increase in pH within the cathodic compartment. This is particularly challenging when  
260 dealing with groundwater due to the low electrical conductivity, which also limits proton transport from  
261 the anode to the cathode. Additionally, internal recirculation enhances reactor hydrodynamics,  
262 overcoming mass transfer limitations inherent in systems with low electrical conductivity (Ceballos-  
263 Escalera et al., 2021). This improvement enhances reactor homogeneity, reducing pH and substrate  
264 (i.e., nitrate) gradients along the biocathode.

### 265 3.2.2 In-situ chloride recovery for disinfection

266 After the reduction of nitrate in the cathode compartment, the treated groundwater flowed into the  
267 anode compartment, where two potential abiotic reactions could occur due to the presence of the  
268 anode material (Ti-MMO) and the operational anode potential ( $> +1.49 \pm 0.02 \text{ V vs. Ag/AgCl}$ ): (i) water  
269 oxidation to oxygen ( $E^{\circ}_{\text{H}_2\text{O}/\text{O}_2} = +1.03 \text{ V vs. Ag/AgCl}$ ) and (ii) chloride oxidation to chlorine ( $E^{\circ}_{\text{Cl}^-/\text{Cl}_2} =$   
270  $+1.16 \text{ V vs. Ag/AgCl}$ ). While oxygen has a low economic interest, chlorine is widely used as a  
271 disinfectant in drinking water systems (Bereiter et al., 2021). Specifically, in the context of groundwater  
272 with low organic matter content, chlorination is a sustainable disinfection method due to its minimal  
273 risk of toxic by-product formation (Mazhar et al., 2020). Additionally, the present system took  
274 advantage of the addition of hydrochloric acid in the cathodic compartment, which increased the  
275 chloride concentration ( $385 \pm 25 \text{ mg Cl}^- \text{ L}^{-1}$ , Table 1). The rise in chloride concentration promoted the  
276 *in-situ* electrochemical production of chlorine.

277 In all  $\text{HRT}_{\text{cat}}$  tested, the effluent consistently kept an adequate free chlorine concentration to ensure  
278 effective *in-situ* disinfection. This concentration progressively increased from  $0.3 \pm 0.1$  to  $4.4 \pm 1.4$  mg  
279  $\text{Cl}_2 \text{ L}^{-1}$  by decreasing the  $\text{HRT}_{\text{cat}}$  (Fig. 2). Lower  $\text{HRT}_{\text{cat}}$  induced higher nitrate reduction rates  
280 associated with higher current densities. Under such conditions, the anode potential increased from  
281  $1.49 \pm 0.02$  ( $\text{HRT}_{\text{cat}}$  of 2.4 h) to  $2.05 \pm 0.12$  V vs. Ag/AgCl ( $\text{HRT}_{\text{cat}}$  of 0.7 h) to sustain this current,  
282 promoting chlorine accumulation on the effluent. Besides, the low pH under lower  $\text{HRT}_{\text{cat}}$  (pH of  $3.0$   
283  $\pm 0.1$  at 0.7 h) forces a displacement of the chemical equilibrium (hypochlorite) to chlorine.

### 284 3.2.3 Operational costs

285 The sustainability of electro-bioremediation relies on minimising reagent usage and substituting them  
286 with electrochemical reactions to reduce treatment costs. This reduction in reagent dependency also  
287 enables the implementation of the treatment in remote areas. The operational costs of the presented  
288 treatment were attributed to the hydrochloric acid for pH control and the electrical power required to  
289 maintain the bio- and electrochemical reactions. It is important to note that previous studies have  
290 identified the power supply as the primary energy consumer in bioelectrochemical reactors (Ceconet  
291 et al., 2018; Zou and He, 2018). The operational cost estimation does not include additional costs  
292 associated with external pumping systems or personnel costs. Additionally, expenses can vary based  
293 on the specific settings used in each scenario, as well as variations in reagent and electricity costs in  
294 different regions. However, the price structure is expected to remain the same.

295 The estimated operational cost was  $1.05 \pm 0.16$  €  $\text{m}^{-3}$  based on the outcome achieved at an  $\text{HRT}_{\text{cat}}$   
296 of 1.3 h, which kept a neutral effluent pH while achieving the highest nitrate reduction rate. The  
297 breakdown of costs reveals that power supply accounted for about 12% of the total cost, equivalent  
298 to  $0.13 \pm 0.01$  €  $\text{m}^{-3}$ . Additionally, the low energy demand ( $0.63 \pm 0.07$  kWh  $\text{m}^{-3}$ ) and the resilience  
299 of the bioelectrochemical systems to power fluctuation support the feasibility of using renewable  
300 energy sources (Rovira-Alsina et al., 2021). This opens up the possibility of utilising solar panels to  
301 achieve self-sufficiency and reduce the operational costs associated with energy consumption. On  
302 the other hand, the cost of hydrochloric acid accounted for 88% of the operating costs. Considering  
303 the hydrochloric acid market price, the estimated cost was approximately  $0.92 \pm 0.15$  €  $\text{m}^{-3}$ .

304 It is worth noting that electro-bioremediation presents a competitive cost compared to other commonly  
305 used nitrate removal treatments such as reverse osmosis (0.04-2.67 € m<sup>3</sup>) and ion-exchange resin  
306 (0.07-2.85 € m<sup>3</sup>) (Jensen et al., 2012). Moreover, electro-bioremediation offers a lower environmental  
307 impact by removing nitrate instead of concentrating it in brines (Fig. 2). Furthermore, this process  
308 combines the reduction of nitrates with disinfection, removing the requirement for further treatment to  
309 achieve potable water. Decentralised water treatment systems often require separate chlorination-  
310 based disinfection devices, resulting in a cost between 0.01 - 0.93 € m<sup>-3</sup> (Dossegger et al., 2021). The  
311 main operating cost associated with acid consumption (88%) was recovered by utilising in-situ  
312 chlorine production. Besides, the need for transporting and handling hazardous disinfectant chemicals  
313 was eliminated. Thus, this approach may reduce costs and enhance the overall sustainability and  
314 safety of the treatment.

## 315 4. Conclusions

316 This study optimised the cost-effectiveness of treating nitrate-contaminated groundwater through  
317 electro-bioremediation, effectively merging nitrate reduction and chlorine production in a single unit.  
318 The nitrate reduction rate was enhanced by implementing cathodic pH control at  $6.8 \pm 0.2$ , reaching  
319 the maximum reported rate in the literature so far ( $5.0 \pm 0.3 \text{ kg NO}_3^- \text{ m}^{-3} \text{NCC d}^{-1}$  at an  $\text{HRT}_{\text{cat}}$  of 0.7 h).  
320 Subsequently, the hydrochloric acid utilised to control the cathodic pH was recovered to produce  
321 chlorine *in-situ* with a final concentration ranging from  $0.3 \pm 0.1$  to  $4.4 \pm 1.4 \text{ mg Cl}_2 \text{ L}^{-1}$ . Chlorine  
322 evolution allowed a correct disinfection of effluent water. For the first time in electro-bioremediation,  
323 the overall quality of treated water has been assessed taking into account both the chemical and  
324 biological requirements for drinking water. The results demonstrated nitrate and nitrite concentrations  
325 below the specified limits and the absence of pathogens such as T. coliforms, *E. coli*, and  
326 *Enterococcus*. Finally, the competitiveness of electro-bioremediation compared with conventional  
327 treatments was demonstrated by the estimated operating cost of  $1.07 \pm 0.17 \text{ € m}^{-3}$  and the lower  
328 environmental impact. In addition, the technology is attractive for meeting drinking water standards in  
329 rural areas due to its minimal chemical dependency and complete absence of residue formation. In  
330 conclusion, these results strongly encourage further research into electro-bioremediation of nitrate-

331 contaminated groundwater and open the door to real implementation of this technology in the rural  
332 areas.

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Fig. 1: Illustration of the reactor (right) and perpendicular section scheme (left).

1.5 -column fitting image

Fig. 2: Main results at the different  $HRT_{cat}$  tested (A) and economic assessment of the presented treatment (electro-bioremediation) compared with the range of the operational cost of other conventional treatments for nitrate removal at very small scale (10 to 190  $m^3 d^{-1}$ ) (Jensen et al., 2012) and chlorination-based disinfection devices (1 to 3  $m^3 d^{-1}$ ) (Dossegger et al., 2021) (B). Cost values were converted from dollars to euros using the exchange rate of the year of publication.

2 -column fitting image

Table 1: Treatment performances and water characteristics according to the different  $HRT_{cat}$  tested ( $n \geq 2$ ). The water characteristics are represented with a colour-coded according to the fulfilment of drinking water standards (Directive EU 2020/2184). Neither nitrite, ammonium, nor nitrous oxide were accumulated. NaN: Not a Number.

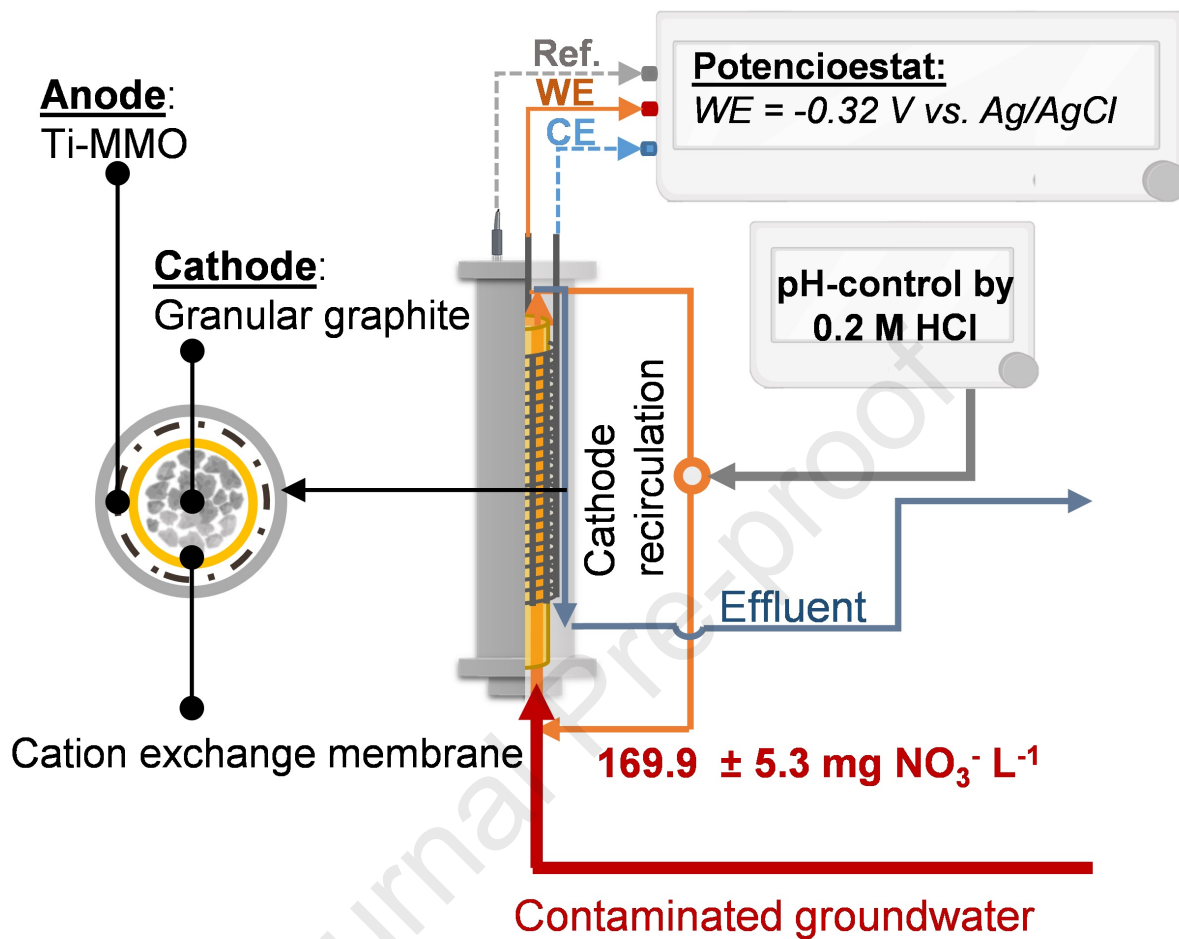
Operation performance			Treatment performance				
$HRT_{cat}$	Current	Cell voltage	Nitrate reduction rate	Nitrate reduction efficiency	Energy consumption	HCl consumption <sup>a</sup>	
[h]	[mA]	[V]	[kg $NO_3^-$ m <sup>-3</sup> <sub>NCC</sub> d <sup>-1</sup> ]	[%]	[KWh m <sup>-3</sup> <sub>water</sub> ]	[L m <sup>-3</sup> <sub>water</sub> ]	
2.4	35 ± 1	1.81 ± 0.02	1.7 ± 0.0	97 ± 1	0.69 ± 0.03	0.76 ± 0.17	
1.6	49 ± 1	2.10 ± 0.12	2.6 ± 0.1	98 ± 0	0.73 ± 0.03	0.66 ± 0.07	
1.3	53 ± 5	2.08 ± 0.12	2.9 ± 0.1	95 ± 2	0.63 ± 0.07	0.66 ± 0.10	
0.9	92 ± 5	2.43 ± 0.08	4.3 ± 0.0	98 ± 1	0.95 ± 0.02	0.89 ± 0.10	
0.7	104 ± 5	2.37 ± 0.12	5.0 ± 0.3	90 ± 4	0.83 ± 0.06	0.99 ± 0.12	

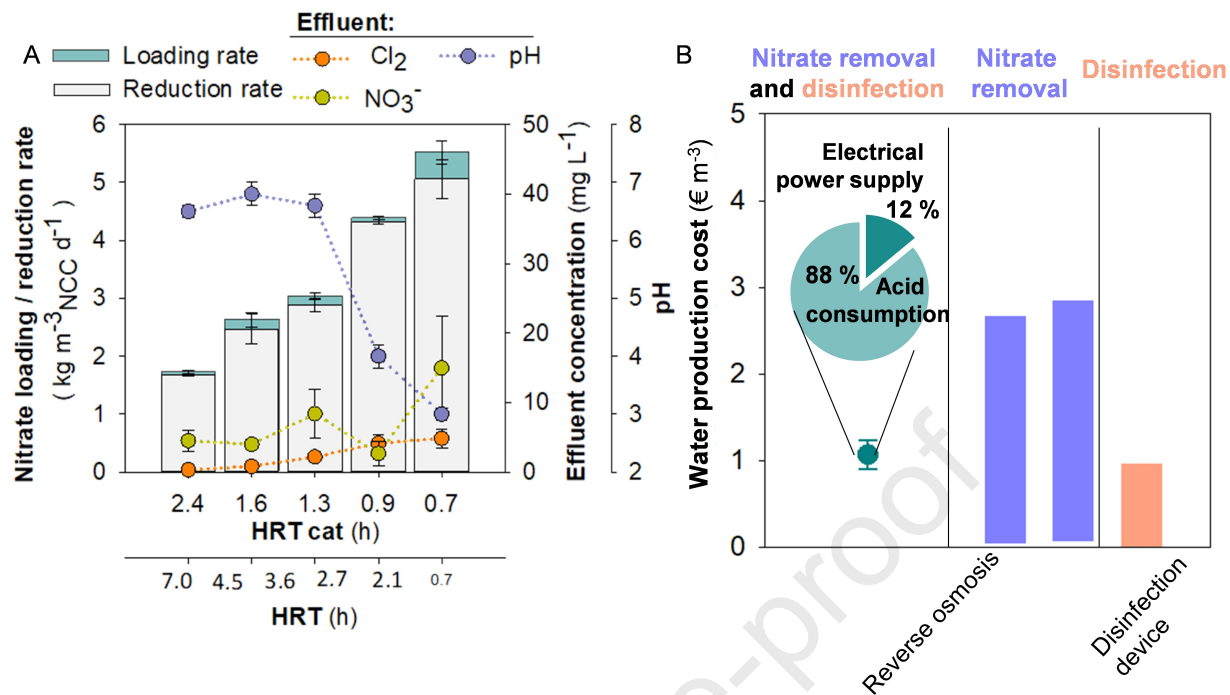
**PARAMETRIC VALUE**  
  
 Drinking water

Water characteristics								
$HRT_{cat}$	pH	Electrical conductivity	Nitrate	Chloride	Free chlorine	T. coliforms	<i>E. coli</i>	<i>Enterococcus</i>
[h]		[mS cm <sup>-1</sup> ]	[mg $NO_3^-$ L <sup>-1</sup> ]	[mg Cl <sup>-</sup> L <sup>-1</sup> ]	[mg Cl <sub>2</sub> L <sup>-1</sup> ]	[ufc 100 mL <sup>-1</sup> ]	[ufc 100 mL <sup>-1</sup> ]	[ufc 100 mL <sup>-1</sup> ]
<b>Untreated groundwater</b>								
-	8.0 ± 0.3	1.3 ± 0.1	169.9 ± 5.3	93 ± 1	0.0 ± 0.0	0 ± 0	0 ± 0	1.3 ± 0.9
<b>Treated groundwater</b>								
2.4	6.5 ± 0.1 <sup>b</sup>	1.4 ± 0.1	4.5 ± 0.6	385 ± 1	0.3 ± 0.1	NaN	NaN	NaN
1.6	6.8 ± 0.2 <sup>b</sup>	1.5 ± 0.2	3.8 ± 0.6	380 ± 7	0.8 ± 0.1	NaN	NaN	NaN
1.3	6.6 ± 0.2 <sup>b</sup>	1.7 ± 0.1	8.2 ± 3.5	395 ± 6	1.7 ± 0.8	0 ± 0	0 ± 0	0 ± 0
0.9	4.0 ± 0.2 <sup>b</sup>	1.9 ± 0.3	2.7 ± 1.8	361 ± 33	4.1 ± 1.2	NaN	NaN	NaN
0.7	3.0 ± 0.1 <sup>b</sup>	2.0 ± 0.1	15.1 ± 7.7	399 ± 29	4.4 ± 1.4	NaN	NaN	NaN

<sup>a</sup> The acid consumption was recalculated to represent the consumption of concentrated acid (HCl, 35%) instead of the diluted acid (HCl, 0.2 M) used in the laboratory-scale experiment. This adjustment allows for a more accurate estimation of acid usage in a real treatment plant scenario. <sup>b</sup> The pH value of the effluent discharged from the anode compartment.







## Highlights:

- Treated groundwater meets standards for nitrogen compounds and pathogens
- Highest reported nitrate reduction rate of  $5.0 \text{ kg NO}_3^- \text{ m}^{-3} \text{ d}^{-1}$  at  $\text{HRT}_{\text{cat}}$  of 0.7 h.
- Water disinfection ensured through in-situ electrochemical chlorine evolution
- Cost-effective treatment with an estimated competitive operational cost of  $1.05 \text{ € m}^{-3}$

**Declaration of interests**

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

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Journal Pre-proof