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Biological denitrification of nitrate contaminated groundwater with moving bed biofilm reactor

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ABSTRACT/RESUME

Abstract: *In recent years, the massive and uncontrolled use of fertilizer in agriculture has increased the level of nitrate in groundwater up to make this strategic and valuable source of water useless or not convenient for drinking water purpose. Several processes have been tested to remove nitrate from groundwater and although efficient, they require high capital as well as management costs. Biological processes represent a valid and low cost alternative to remove nitrate from water. This paper, actually, presents the preliminary and encouraging results of the start-up and operating of a bench scale 1.8L Moving Bed Biofilm Reactor (MBBR) to remove biologically nitrate. Kaldnes K1were used as biofilm carrier and the reactor was fed with synthetic water simulating a typical groundwater with different concentrations of nitrate, i.e. NO_3^--N ranging from 30 to 60 mg/L. Acetate was added as carbon source.*

I. Introduction

Groundwater is globally the main source of drinking water and, in arid countries, even the only one. The quality of groundwater has been getting worse in the last decades because of high level of nitrate as consequence of the extensive use of chemical fertilizers in intensive agriculture systems as well as the discharge on soil of domestic and animal wastes. Elevated concentrations of nitrate in water can be harmful for humanhealth: nitrate can actually cause methemoglobinemia in infants (Blue-Baby) and even cancer [1].
Conventional physical-chemical methods to remove nitrate from water include ion exchange, reverse osmosis and electro-dialysis. But all of these processes are expensive and the concentrated waste brines require further treatment or disposal [2] The use of biological denitrification to convert nitrates to harmless nitrogen gas (Eq. (1)) [3] could offer an alternative treatment process for the remediation of groundwater contaminated by nitrate due to the low cost and high denitrification efficiency of the process.

$$NO_3^- \rightarrow NO_2^- \rightarrow NO \rightarrow N_2O \rightarrow N_2 \quad (1)$$

Different biological systems can be used to perform the biological removal of nitrate from water; Moving Bed Biofilm Reactors (MBBRs) is one of them and one of the most promising. The MBBRs show the advantages of both, attached and suspended growth systems and are used to treat wastewater as well as raw waters for drinking purpose. This system is based on the use of carriers where the biomass attaches and grows [4]; MBBRs are operated similarly to the activated sludge

reactors as carriers are in constantmovement in the biological tank [5].

The performance of $MBBR_S$ depends on the shape and amount of carriers used to fill the reactor: commonly the percentage of tank occupied with carriers varies from 50 and 70% in volume.Carriers are characterized by an extremely high specific surface area and this aspect allows to havea higher biomass concentration in a smaller reactor volume than in conventional suspended growth system, thus reducing the costs of the treatment. Anyway, not the whole surface area of carriers is useful for growing biomass, but at least a 70% as reported in the literature [6].

The performance of MBBR is the result of attachment, growth and detachment of biofilm, and all these processes are influenced by the environmental conditions: shape of carriers, thickness of biofilm, mixing intensity, pH, nutrient levels, ionic strength and temperature of water.

Several studies have been conducted on biological removal of nitrate from groundwater [7- 8- 9] using attached growth systems, but really few with MBBR, the aim of this study actually, has been to test the performance of this system in removing nitrate from synthetic water with characteristics similar to a real groundwater. Nitrate concentration was varied from 30 to 60 mg/L of NO_3^--N and acetate was used as external source of carbon for the denitrifying heterotrophic bacteria.

II. Materials and methods

II.1. MBBR Configuration

The MBBR was set up in an air-tightly closed 2L plastic cylinder (fig.1.)The working volume was set equal to1.8 L. The bioreactor was filled with kaldnes K1 as carriers with a filling percentage of 50 %. Carries are made of high density polyethylene (HDPE) (fig.2.).

A magnetic stirrer system was placed on the bottom of the reactor to perform the mixing of the bulk, thus avoiding the settlement of carriers and promoting the contact between biomass and substrates.

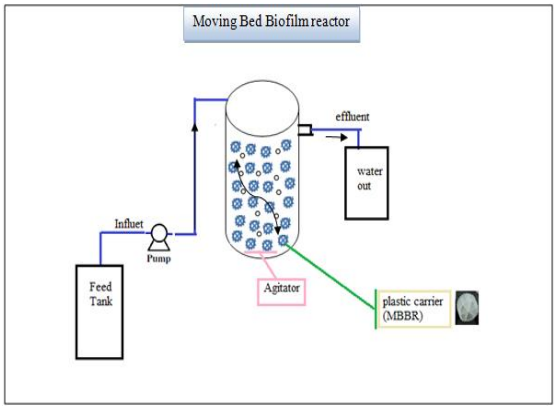


Figure 1.*Schematic diagram of the experimental apparatus.*

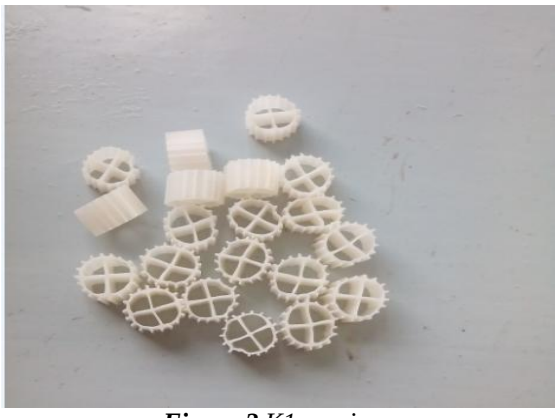


Figure 2.*K1 carriers*

II.2. MBBR Operation

The MBBR was inoculated with activated sludge from a municipal wastewater treatment plant located in Nola, Italy. The biomass concentration in sludge was 13.43 g/L as total solids (TS).The bioreactor during the start-up was operated for 40 days with a hydraulic retention time (HRT) of 24 h. The inlet flowrate and initial nitrate concentration were 1.25 ml/min and 30 NO_3^--N, respectively. When the bioreactor reached stationary condition, the content of nitrate in the influent was increased gradually up to the final concentration of 60 NO_3^--N. In table 1 the operating conditions of the MBBR are listed.

Table 1. *Operating conditions of the anoxic MBBR*

Days of operation	COD/ NO_3^--N	HRT(h)	NO_3^-N(mg/L)
0-40	*3*	*24*	*30*
41-52	3	*24*	*40*
53-67	*3*	*24*	*50*
68-77	*3*	*24*	*60*

The bioreactor was fed with a peristaltic pump (WATSON MARLOW 520 Du) from a 10 L influent storage tank. Sludge was not recirculated.

II.3. Synthetic Water Composition

The synthetic water was composed of NO_3^-as electron acceptor and sodium acetate as electron donor. A COD/ NO_3^- -N ratio equal to 3 was set according to a previous study [10]. This value is lower than stoichiometric (i.e. 3.74 [11]) to take into account the occurrence of other biological reactions that consume COD. The most commonly carbon sources used in heterotrophic denitrification are methanol, ethanol and acetate. In this study was tested acetic acid because is more readily metabolized than methanoland more safety for human health than methanol and ethanol [12-13].

In detail, the synthetic water was prepared by adding various amounts of KNO_3 and $C_2H_3NaO_2$ (sodium acetate) to a demineralized water containing 150 mg/L ofKH_2PO_4, 325 mg/L of $NaHCO_3$ and 1% (v/v) of a solution composed of $FeSO_4.7H_2O$ (0.20 mg/L), titriplex (0.565 mg/L), 0.1% (v/v) of a trace nutrient solution containing $ZnSO_4.7H_2O$ (0.1g/L), $MnCl_2.4H_2O$(0.03g/L),H_3BO_3 (0.3 g/L), $CoCl_2.6H_2O$ (0.2g/L), $CuCl_2.2H_2O$ (0.01g/L), $NiCl_2.6H_2O$ (0.02g/L), and $NaMoO_4.2H_2O$ (0.03g/L) [14].

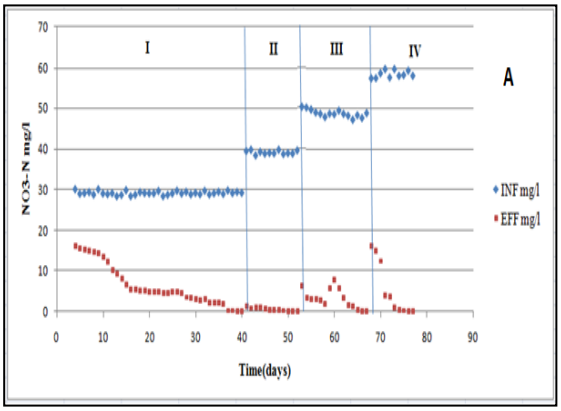
II.4. Analytical Methods

Samples were collected from the influent and effluent stream once a day and filtered through 0.45 µm membranes. All the analyses were conducted according to the standard analytical methods for water and wastewater [15]. Nitrate concentrations were measured with 761 compact IC (Metrohm), COD through titration, T and pH with digital probes.

III. Results and discussion

As reported in figure 3A and 3B, the start-up of the reactor lasted 40 days (phase I). Subsequently the NO_3^--N concentration in the influent was increased by 10 mg/L, from 30 mg NO_3^--N/L to 40 mg NO_3^--N/L.MBBR showed a high resilience since a negligible reduction in the efficiency was observed only in the first day after the increase of the nitrate load. Then the efficiency rapidly reached again percentage next to 100% (phaseII), proving that the reactor was supplied with an amount of biomass higher than that strictly necessary to degrade the input substrate during the start-up phase.

At the day 53 the NO_3^--N concentration in the effluent was further increased setting its value equal to 50 mg NO_3^--N/L (phase III). In this phase, although the increase of NO_3^--N concentration was, as previously done, by 10 mg/L, the MBBR required a pretty longer, but anyway reasonably short, time to recover its efficiency. This longer time was necessary for the microorganisms to increase their number up to have a value adequate to remove all the NO_3^--N contained in the influent. Phase III was also characterized by a drop of MBBR efficiency due to an intentionally caused failure in the air-tight sealing of the reactor with the aim of testing the influence of the oxygen on the anoxic MBBR. The occurrence of oxygen concentration higher than decimals actually inhibited the denitrifying bacteria and consequently reduced the efficiency of process. Once the concentration of oxygen was back to a negligible value the efficiency showed again values close to 100%. At the day 68 the NO_3^--N concentration in the influent was further increased by 10 mg/L up to reach the final value of 60 mg NO_3^--N/L (phase IV). In this phase the time required by the reactor to recover the efficiency was longer than in phase III showing the tendency that recovery times are proportional to the NO_3^--N content in the influent



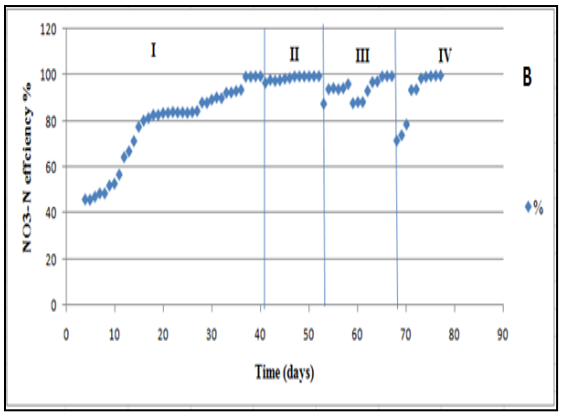
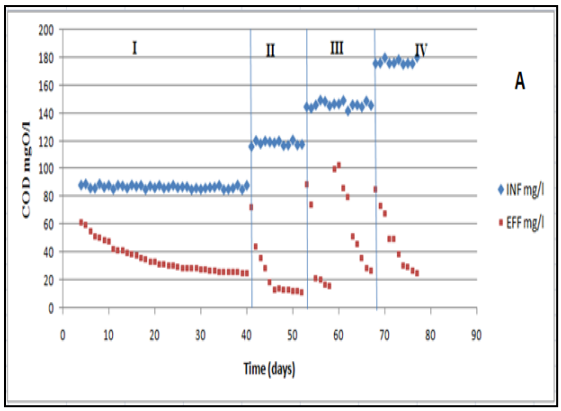


Figure 3 .*Effect of NO_3^--N /Lconcentration in the effluent on the denitrification process: (A) numeric values; (B) percentage values. I = 30 mgNO_3^--N/L II = 40 mgNO_3^--N /L; III = 50 mgNO_3^--N /L; IV = 60 mgNO_3^--N /L*

In figure 4A the concentrations of COD in the influent as well as in the effluent are reported during the 4 phases through which the anoxic MBBR was operated, whereas in figure 4B the COD removal efficiency is shown. As it can be easily noticed, comparing figure 3 with figure 4, a perfect correspondence between nitrate removal and COD consumption is found as proof of the occurrence of the denitrification process. Although the nitrate removal efficiency reached values of about 100%, COD removal efficiency was around 72% and this result ensured that the reactor was not carbon limited. The slight increase of residual COD in the system when the NO_3^--N concentration was increased in the influent proves that the COD/ NO_3^- -N ratio set equal to 3 is not the optimal value. It has to be searched among values lower than 3, because other reactions rather than denitrification involving heterotrophic microorganisms take place promoted by the low concentration of O_2 present in the influent [16-17].



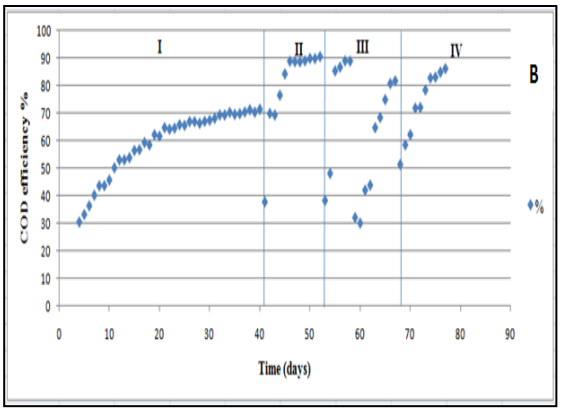


Figure 4.*COD removal efficiency: (A) numeric values; (B) percentage values. I = 30 mgNO_3^--N /L II = 40 mg NO_3^--N /L; III = 50 mg NO_3^--N /L; IV = 60 mg NO_3^--N /L*

IV. Conclusion

This study proves the high efficiency as well as resilience of anoxic MBBR system used to biologically remove nitrate from groundwater. Kaldnes K1 showed to be really effective to grow the biomass, but other types of carriers are expected to be as performing as K1. The study was limited to 60 mg NO_3^--N /L as nitrate load and 24 hours as HRT. Moreover, the short time required to MBBR to fully recover its efficiency when the operating conditions were changed and intensified leads to think that this system has wide margins to successfully treatment waters with a higher load of nitrate in a smaller volume and a shorter time. Furthermore, the residual COD concentration that represents the main drawback of the system can be reasonably reduced up to value close to zero by decreasing the COD/ NO_3^- -N ratio and/or supplying the system with a activated carbon filter phase.

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