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Hydrogenotrophic Denitrification of Groundwater Using a Simplified Reactor for Drinking Water: A Case Study in the Kathmandu Valley, Nepal

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Abstract: High nitrate-nitrogen (NO_3^-–N) content is a typical feature of groundwater, which is the primary water source in the Kathmandu Valley, Nepal. Considering the Kathmandu Valley's current problem of water scarcity, a user-friendly system for removing NO_3^-–N from groundwater is promptly desired. In this study, a simplified hydrogenotrophic denitrification (HD) reactor was developed for the Kathmandu Valley, and its effectiveness was evaluated by its ability to treat raw groundwater. The reactor operated for 157 days and showed stability and robustness. It had an average nitrogen removal efficiency of 80.9 ± 16.1%, and its nitrogen loading rate and nitrogen removal rate varied from 23.8 to 92.3 g–N/(m^3·d) and from 18.3 to 73.7 g–N/(m^3·d), respectively. Compared to previous HD reactors, this simplified HD reactor is a more user-friendly option for the Kathmandu Valley, as most of the materials used for the reactor were locally available and require less maintenance. The reactor is recommended for groundwater treatment at the household level. It has a current treatment capacity of 40 L/d, which can fulfill the daily requirements for drinking and cooking water in a household with 4–5 people.

Keywords: NO_3^-–N removal; user-friendly; groundwater treatment; hydrogenotrophic denitrification; drinking water; Kathmandu Valley

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1. Introduction

Drinking water is indispensable to human life, and the United Nations Sustainable Development Goal 6 (SDG) targets the securement of universal and equitable access to safe and affordable drinking water for all people [1]. The Kathmandu Valley includes Kathmandu, the capital and the largest city of Nepal, and it is facing water shortages due to the rapidly expanding population [2] and continued urbanization [3]. There is a considerable discrepancy between the water demand and the water supply in the Kathmandu Valley (the total water demand is 430 million liters per day (MLD), but the water supply averages only 103 MLD) [4]. To address the deficit in water supply, the locals have been compelled to find alternative water sources [5–10]. Among them, groundwater has been

a principal source of drinking water in the valley [11], and household dependency on groundwater is high [5]. However, nitrogenous contaminants, mainly ammonium-nitrogen (NH_4^+–N) and nitrate-nitrogen (NO_3^-–N), are often observed in the groundwater [11–15], limiting its usage. Excess NH_4^+–N can be associated with the creation of offensive odors and taste, and it can diminish the efficacy of chlorine disinfection, resulting in higher risks of pathogenic contamination [16]. Furthermore, NH_4^+–N can be converted into nitrite-nitrogen (NO_2^-–N) or NO_3^-–N by microbial activity, and a high intake of nitrate-contaminated water is known to have adverse effects on human health, leading to serious diseases such as methemoglobinemia, gastric cancer, and non-Hodgkin's lymphoma [17]. Therefore, it is necessary to eliminate contaminants from groundwater.

Although ceramic filtration has conventionally been used in households to purify water [8], this method is not effective in removing nitrogenous contaminants. The development of alternative removal processes, especially for nitrogen species, is therefore necessary. A promising reactor for NH_4^+–N removal from groundwater, which is low-cost, low-maintenance, and has excellent performance, has been developed [18–20]. However, the system is capable of removing only NH_4^+–N through oxidation of NH_4^+–N to NO_3^-–N by nitrifying bacteria, and the NO_3^-−N produced during nitrification remains of great concern [20]. Therefore, it is imperative to develop a suitable system for the removal of both preexisting and NH_4^+–N–derived NO_3^-–N in the groundwater of the Kathmandu Valley.

Myriad approaches for removing NO_3^-–N from groundwater have been studied. However, the current economic situation and the energy affairs of the Kathmandu Valley could pose a challenge for the selection of water treatment technology, and the compatibility of these systems with the current social situation must be considered [8,9]. In this context, user-friendly water treatment systems that are low-cost, energy-efficient, compact, and easy to operate and maintain are desirable to ensure their sustainability [8,21]. From this perspective, physicochemical approaches, including ion exchange, reverse osmosis, and electrodialysis may be ineligible due to high capital infrastructure costs, high energy consumption, and the costs related to the disposal of waste brine [22]. The use of a biological treatment seems to be a preferable alternative [22]. Biological denitrification processes are broadly classified into two groups: heterotrophic and autotrophic denitrification, and the former has been conventionally studied for NO_3^-–N removal. The addition of organic carbon entailed in the activation of heterotrophic denitrification raises concerns regarding the increased risk of secondary pollution associated with elevated levels of total organic carbon [21,23–25].

Recently, hydrogenotrophic denitrification (HD), which is a type of autotrophic denitrification, has drawn much attention as a promising technology for nitrate removal from groundwater [19,21,23–36] because it offers certain advantages over heterotrophic denitrification systems. First, biomass generation can be reduced, thereby reducing clogging and the cost of post-treatment [33]. Second, there is no production of toxic waste as HD is a clean process [21,24]. Third, organic carbon is not necessary, leading to lower operational costs and lower risk of secondary contamination [23]. Fourth, H_2 is economical for nitrogen removal per electron equivalent compared to other electron donors [23,33].

The HD process is generally applied using a hollow fiber membrane reactor, a gas-permeable reactor, a membrane bioreactor, or a packed bed reactor. This leads to high costs for infrastructure and operation, frequent cleaning, post-treatment requirements, and high maintenance. In contrast, simplified HD systems, for example, using an attached growth reactor, can be well suited for application in developing countries owing to their user-friendliness [19,21,23]. To date, most research on the development of HD reactors for groundwater has been conducted in the laboratory using synthetic groundwater, and little information is available on the application of HD for raw groundwater treatment and the behavior of HD systems under on-site conditions.

Therefore, the objective of this research is to develop a simplified, user-friendly HD reactor for people in the Kathmandu Valley, Nepal, and use it for raw groundwater treatment to evaluate its effectiveness, understand its behavior under on-site conditions, and

fill the gap in knowledge between laboratory and practical applications. In this research, groundwater from the Jwagal area located in the Kathmandu Valley, Nepal, where the groundwater is highly contaminated by NH_4^+–N, at levels up to 60 mg–N/L [11], was selected for a case study. The simplified HD reactor was developed using as many locally available materials as possible and installed after a nitrification system, in order to eliminate the NO_3^-–N generated during the nitrification process. This research was designed to support future efforts for deeper insight into the simplified HD reactor, which could potentially have important implications for the application of this technology in developing countries.

2. Materials and Methods

2.1. Description of the Study Area

The Kathmandu Valley is situated between the latitudes 27°32′13″ and 27°49′10″ N, and the longitudes 85°11′31″ and 85°31′38″ E. The study area, Jwagal, is located in the middle of the Kathmandu Valley (Figure 1). In Jwagal, Kathmandu Upatyaka Khanepani Limited (KUKL) operates a water treatment unit to extract deep groundwater for intermittent distribution. The extracted groundwater is treated by aeration, coagulation and flocculation, sedimentation, sand filtration, and chlorination, as shown in Figure 2, before distribution to households.

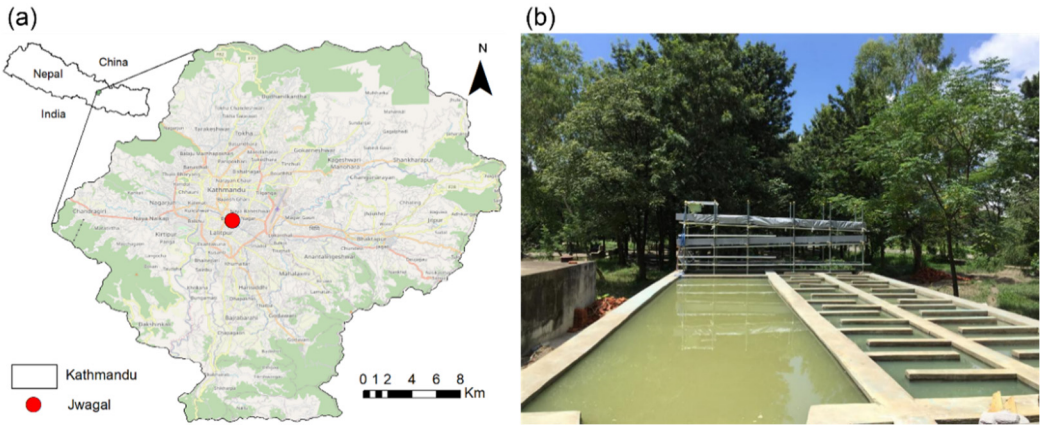

Figure 1. (**a**) Location and (**b**) picture of the experiment site in Jwagal in the Kathmandu Valley.

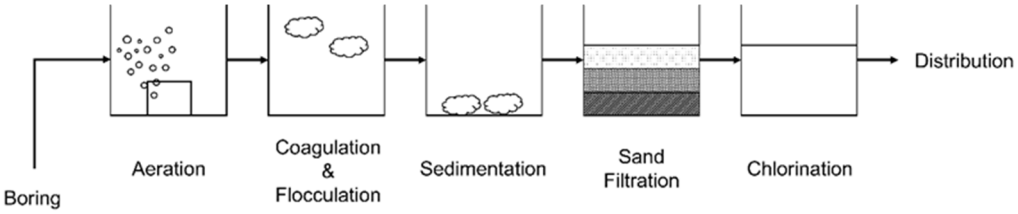

Figure 2. Schematic of the existing water treatment system operated by KUKL in Jwagal.

2.2. Characteristics of Groundwater at Jwagal

Table 1 summarizes the groundwater quality of Jwagal, with data partially adapted from previous reports [11,37]. As can be seen in Figure 1b, the groundwater is turbid. The groundwater is heavily contaminated by NH_4^+–N at concentrations ranging from 41.2 to 57.3 mg–N/L, which exceeds the drinking water standard (1.2 mg–N/L) set by the World Health Organization (WHO) [16], and which cannot be removed by the current treatment process [9]. Thus, a nitrification system described by the Japan International Cooperation Agency (JICA) in 2019 [38] was supplementarily installed after the sedimentation system to remove NH_4^+–N from the groundwater.

Table 1. Composition of the raw groundwater in Jwagal in the Kathmandu Valley.

Parameter	Concentrations (mg/L)	Reference
NH_4^+–N	41.2–57.3	[11]
NO_3^-–N	0.4–3.4	[11]
Na^+	61	[37]
K^+	12	[37]
Ca^{2+}	68.8	[37]
Mg^{2+}	14	[37]
Cl^-	2.1	[37]
SO_4^{2-}	Not detected	[37]
HCO_3^-	579.5	[37]
Fe^{2+}	9.9–10.9	This study
DOC (dissolved organic carbon)	10.6–14.0	This study
pH	6.29–6.79	This study

2.3. Simplified HD Reactor

Figure 3 shows a schematic of the on-site HD reactors. To ensure the sustainability of the water treatment unit and its compatibility with local conditions, reactors were made using as many locally available materials as possible. A cylindrical plastic water jar that is commonly used by households in Nepal [5,39] was selected for two attached growth reactors with a working volume of approximately 20 L. Polyolefin sponges (1 cm × 1 cm × 1 cm; Sekisui Aqua Systems Co., Ltd., Japan) were selected as the durable carrier material for better bacterial attachment and long-term applicability, and 1000 pieces were introduced into the inside of each reactor. The seed sludge for the on-site HD reactor was obtained from an HD reactor in operation [38,40]. A fiber carrier with 1.5 ± 0.4 g of volatile suspended solid (VSS) was used for acclimatizing the reactor. The two reactors were continuously operated in parallel from May 2017 to November 2017 with H_2 (hereafter D1) and without H_2 (hereafter D2) gas supply. The H_2 flow rate, flow rate of the influent, and hydraulic retention time (HRT) were maintained at 120 mL/min, 28 mL/min, and 12 h, respectively, during the operation. H_2 gas was supplied from a water electrolytic H_2 generator (HG-26; GL Science, Tokyo, Japan) through a commercially available aeration diffuser designed for aquariums (WP-1680, Sobo, China). Two reactors were installed after a nitrification system that treated the pretreated groundwater. The nitrification system comprises a dropping nitrification system, where the groundwater drips down from the top to the bottom by gravity. Nitrifying bacteria on the hanging materials consume oxygen in the air to convert NH_4^+–N to NO_3^-–N [20,40]. The nitrified groundwater was then supplied to the bottom of the HD reactors and passed through the carriers towards the outlet. The reactors were installed inside a room (dark) to prevent the algal growth. D1 and D2 were continuously operated for 157 and 113 d, respectively, without any internal cleaning. Samples of the influent and the effluents of D1 and D2 were taken randomly (at least once in every 10 days) to monitor the nitrogen concentrations, pH, dissolved oxygen (DO), water temperature, and turbidity during the operation period. In total, 21 samples from D1 and 18 samples from D2 were collected.

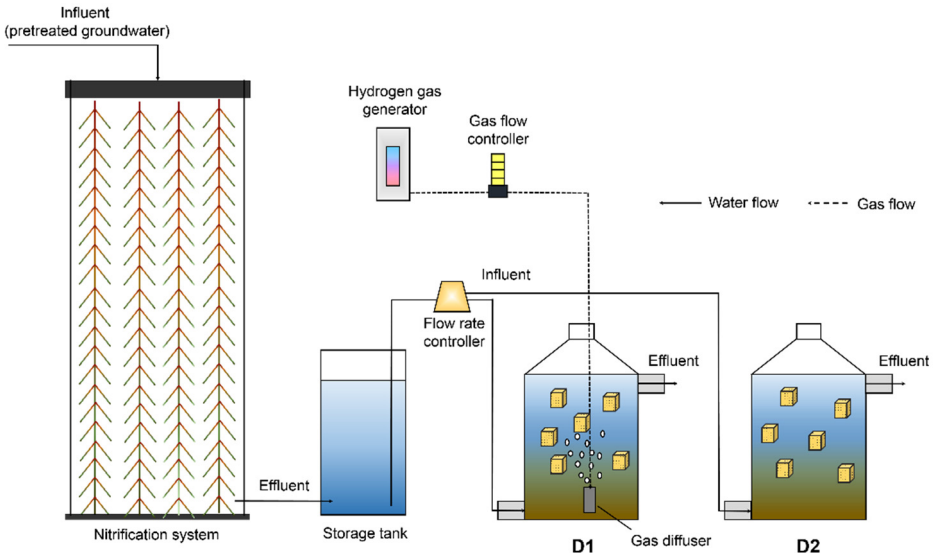

Figure 3. Schematic of the simplified HD reactor.

2.4. Batch Test for the Determination of NO_3^-−N Removal and Denitrification Rate

Batch tests were conducted immediately before the start of the long-term operation and on Day 112 to determine the NO_3^-–N and denitrification (NO_3^-–N and NO_2^-–N) rates in the same manner as previously described [21]. The rate was obtained from linear regressions based on the results of the test. The H_2 flow rate was maintained at 120 mL/min during the batch test. The samples were collected from the reactor at regular intervals.

2.5. Analytical Methods

Collected samples were immediately filtered using a 0.45 µm pore-size membrane filter and stored in a freezer (−18 °C) until water quality analysis was performed. NO_3^-–N, NO_2^-–N, NH_4^+–N, and bicarbonate (HCO_3^-) concentrations were analyzed by ultraviolet spectrophotometric screening, colorimetric, colorimetric-phenate, and titration methods, respectively, in accordance with standard methods [41]. The pH, DO, water temperature, and turbidity were measured using a pH meter (Horiba-B712, Kyoto, Japan), a portable DO meter (Multi 3410; WTW, Weilheim, Germany), a digital thermometer (WT-6, China), and a digital turbidity meter (TU-2016, Sato Shouji Inc., Saitama, Japan), respectively. The average temperature data of Kathmandu from May 2017 to November 2017 were obtained from the Historical Weather and Climate Data [42].

2.6. Performance of the Simplified HD Reactor

The nitrogen loading rate (NLR), nitrogen removal rate (NRR), and nitrogen removal efficiency were calculated based on Equations (1)–(3).

$$\mathrm{NLR}\left[\mathrm{g\text{–}N/(m^3\cdot d)}\right] = \frac{(\mathrm{NO_3^-\text{–}N_{In}} + \mathrm{NO_2^-\text{–}N_{In}})\left[\frac{\mathrm{g\text{–}N}}{\mathrm{L}}\right] \times \text{Flow rate}\left[\frac{\mathrm{L}}{\mathrm{d}}\right]}{\text{Reactor volume}\left[\mathrm{m^3}\right]} \quad (1)$$

$$\mathrm{NRR}\left[\mathrm{g\text{–}N/(m^3\cdot d)}\right] = \frac{\{(\mathrm{NO_3^-\text{–}N + NO_2^-\text{–}N})_{In} - (\mathrm{NO_3^-\text{–}N + NO_2^-\text{–}N})_{Eff}\}\left[\frac{\mathrm{g\text{–}N}}{\mathrm{L}}\right] \times \text{Flow rate}\left[\frac{\mathrm{L}}{\mathrm{d}}\right]}{\text{Reactor volume}\left[\mathrm{m^3}\right]} \quad (2)$$

$$\text{Nitrogen removal efficiency}\ (\%) = \frac{\mathrm{NRR}\left[\mathrm{g\text{–}N/(m^3\cdot d)}\right]}{\mathrm{NLR}\left[\mathrm{g\text{–}N/(m^3\cdot d)}\right]} \times 100 \quad (3)$$

where NO_3^-–N_{In}, NO_2^-–N_{In}, NO_3^-–N_{Eff}, and NO_2^-–N_{Eff} represent the influent NO_3^-–N, influent NO_2^-–N, effluent NO_3^-–N, and effluent NO_2^-–N concentrations, respectively.

In this study, NH_4^+–N concentration was not considered for NLR, NRR, or nitrogen removal efficiency, as HD is not meant to remove NH_4^+–N, and the difference in NH_4^+–N concentrations between the influent and effluent were not significant. Furthermore, the NH_4^+–N observed in the influent is attributed to the degraded performance of the nitrification system (NH_4^+–N concentrations should be less than 1.2 mg–N/L for drinking water).

2.7. Statistical Analysis

Data were first tested for normality using the Shapiro-Wilks test, and it was found that the data did not meet the normality assumption. Thus, to compare statistical differences among the three groups (influent, D1 effluent, D2 effluent) the non-parametric Kruskal-Wallis test and Dann-Bonferroni test were used. In all data, a p-value of less than 0.05 was considered statistically significant. The data were processed using the statistical analysis software package SPSS v.22 (IBM Corp., Armonk, NY, USA).

3. Results and Discussion

3.1. Variations in Nitrogen Concentrations

Figure 4 presents the variation in N-species concentrations. Clear NO_3^-–N removal was observed in D1 as soon as the operation was initiated, and the NO_3^-–N concentrations remained low with an average value of 2.7 ± 2.9 mg–N/L, even though the NO_3^-–N concentrations in the influent fluctuated from 8.3 to 39.5 mg-N/L (Figure 4a). This demonstrates the rapid start-up ability of the HD and its stable performance even with the pretreated groundwater. The NO_3^-–N threshold for drinking water, 11.3 mg–N/L [16], was met for the entire test period. D2, however, showed a slight increase in NO_3^-–N concentration and a decrease in NH_4^+–N. The average NO_3^-–N concentration of the D1 effluent was significantly lower ($p < 0.001$) than that of the influent and D2 effluent, which clearly shows the effectiveness of the HD process for NO_3^-–N removal from raw groundwater.

The NO_2^-–N concentration should be less than 0.9 mg-N/L for drinking water [16]. Although NO_2^-–N concentrations in the D1 effluent were found to be close to 0 mg-N/L (Figure 4b), the average was 2.1 ± 2.9 mg-N/L and the NO_2^-–N concentrations exceeded the limit in 11 out of the 21 samples taken. To avoid undesirable NO_2^-–N accumulation in the system and enhance the complete denitrification, the abundant addition of HCO_3^- (more than 3000 mg/L) could be a potential option [25].

The fluctuation of NH_4^+–N concentrations in the influent is attributed to the performance of the nitrification system (Figure 4c), which should produce concentrations lower than 1.2 mg–N/L for drinking water in practical application. The simplified HD reactor did not significantly affect the changes in NH_4^+–N concentrations, which is consistent with previous research [43,44].

The nitrogen removal efficiencies of D1 gradually increased throughout the experiment and reached 100% on Day 61, with an average of 80.9 ± 16.1% (Figure 4d), whereas nitrogen removal was not found in D2. Additionally, NRR varied from 18.3 to 73.7 g–N/(m^3·d), while NLR fluctuated from 23.8 to 92.3 g–N/(m^3·d). Furthermore, the batch experiments revealed that the NO_3^-–N removal and denitrification rate drastically increased compared to before the operation and Day 112 (Table 2). These results confirmed that the simplified HD reactor has rapid start-up ability, stability, and robustness for processing pretreated groundwater.

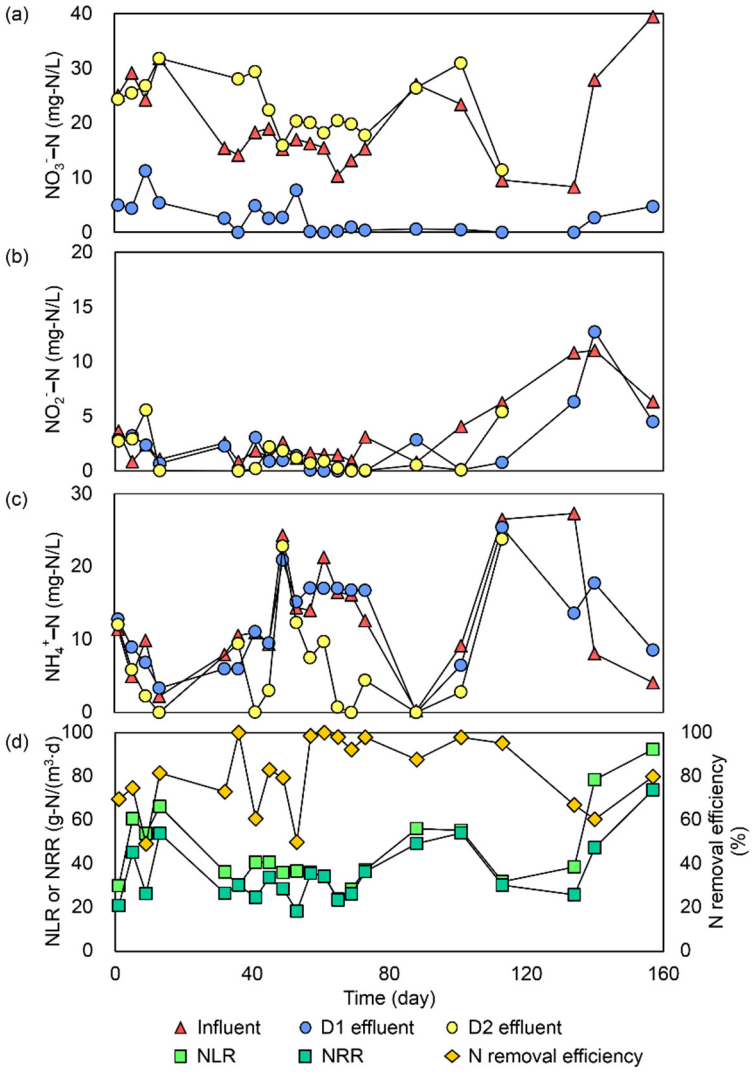

Figure 4. Profiles of (**a**) NO_3^-–Nconcentrations, (**b**) NO_2^-–N concentrations, (**c**) NH_4^+–N concentrations, and (**d**) the nitrogen loading rate (NLR), nitrogen removal rate (NRR), and nitrogen (NO_3^-–N + NO_2^-–N) removal efficiencies of D1.

Table 2. Changes in nitrogen removal rate resulting from the batch experiments.

Day	NO_3^-–N Removal Rate (mg-N/L/d)	Denitrification Rate (mg-N/L/d)
Before Operation	7.5	4.2
Day 112	20.3	26.7

3.2. Ambient Condition and Changes in Operational Parameters

Figure 5a shows the variation of the average ambient temperature and the water temperature inside the reactors. The ambient temperature varied in the range of 14.4–26.5 °C, while the water temperature ranged from 22.5 to 28.1 °C. It is known that denitrification can occur over a wide range of temperatures, from 2 to 50 °C [45], and that HD occurs at temperatures between 15 and 50 °C [46]. Although a higher nitrate removal rate was obtained at higher temperatures [46], the HD efficiencies were maintained at a certain level (>70%) with a water temperature range of 15–30 °C [31]. In the present study, the water temperature inside the reactor stayed within this range for HD performance without temperature control.

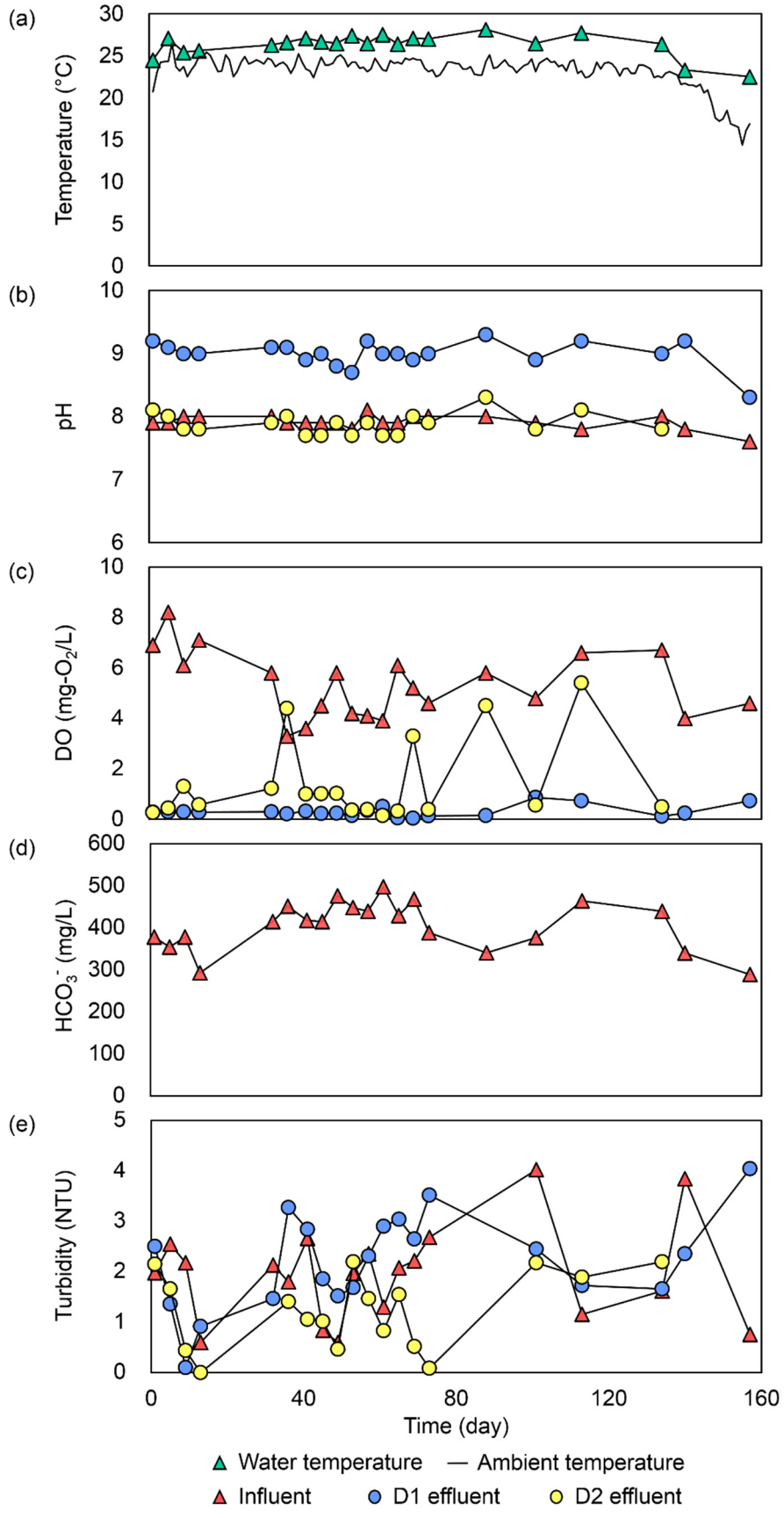

Figure 5. Variations of operating conditions: (**a**) temperature, (**b**) pH, (**c**), dissolved oxygen (DO) concentrations, (**d**) HCO_3^- concentrations, (**e**) turbidity during the operation.

The pH during HD usually increases because the process produces OH^- ions as a byproduct [25]. The average pH of the influent and the effluents of D1 and D2 were 7.9 ± 0.1, 9.0 ± 0.2, and 7.9 ± 0.2, respectively. The pH of the D1 effluent was significantly higher ($p < 0.001$) than that of the influent and D2 effluent (Figure 5b). These results indicate the occurrence of microbial activity in the D1 reactor.

The DO concentration in D1 was kept low, ranging from 0.05 to 0.87 (Figure 5c), with an average value of 0.3 ± 0.2 mg–O_2/L, which could be the result of H_2 gas supply and denitrification. Meanwhile, the DO concentrations in the influent and D2 ranged from 3.2 to 8.2 and 0.15 to 5.40 mg–O_2/L, respectively. The effluent of the nitrification system contains high DO as previously reported [20], which is the same as the influent of the simplified HD reactor. However, the HD system can be inhibited by high DO concentrations [47,48], and Singhopon et al. has suggested that the DO value should be between 0.5 and 0.8 mg–O_2/L to improve NRR [48]. In the present study, supplying H_2 gas into the reactor was instrumental in maintaining the preferred range of DO concentrations for HD.

Figure 5d depicts the variations in HCO_3^- concentration during the operation. A total of 0.171 moles of HCO_3^- was consumed to remove 1 mole of NO_3^- based on the stoichiometry of the HD [49]. The maximum NO_3^-–N concentration found in the influent during the operation was 39.5 mg–N/L; thus, approximately 30 mg/L of HCO_3^- would be required for the complete removal of NO_3^-–N. In previous research, inorganic carbon sources were added to HD systems to improve stability [50], but this increases their operational costs. The average HCO_3^- concentration in the influent was 404.5 ± 56.8 mg/L. This shows that the groundwater in Jwagal naturally contains enough bicarbonate for the HD to proceed without additional inorganic carbon input.

Figure 5e profiles turbidity in the simplified HD reactor. Turbidity during the HD process has not been widely reported to date. The turbidity can influence not only the aesthetics or acceptability of drinking water but also disinfection treatment [51]; thus, it is important to know the impact of the simplified HD reactor on turbidity. In the case of household water treatment, the WHO suggests that turbidity should be at least less than 5 NTU, and ideally turbidity should be less than 1 NTU as regards aesthetics and disinfection efficiency [51]. In this study, turbidity was higher than 1 NTU, with averages of 2.0 ± 0.9, 2.2 ± 0.9, and 1.2 ± 0.7 NTU for the influent, D1 effluent, and D2 effluent, respectively. Thus, a post-treatment after the simplified HD reactor is recommended.

3.3. Comparison with Various HD Reactors

Most research on HD technologies developed for groundwater treatment has been conducted in laboratories using synthetic groundwater. To compare the reactor performance between synthetic and raw groundwater treatment, various studies on HD reactors are summarized in Table 3. In the table, experiments using different types of reactor, substrates, bacteria inoculums, temperatures, HRTs, H_2 availability, influent NO_3^-–N concentrations, carrier materials, reactor volumes, NLR, NRR, and nitrogen removal efficiencies are compared. The simplified HD reactor developed in this research exhibited nitrogen removal efficiencies as high as those of other reactors in Table 3 (~100%), and showed 73 g–N/(m^3·d) of NRR, which is higher than other techniques shown in Table 3, such as bio-ceramite reactors [31], heterotrophic denitrification coupled with electro-autotrophic denitrifying packed bed reactors [32], and HD with electrolytic reactors [36]. Although hollow fiber reactors [26], submerged membrane reactors [28], and membrane biofilm reactors [30] possessed higher NRR (110–770 g–N/(m^3·d)) than attached growth reactors, clogging often occurs using these techniques, necessitating frequent cleaning and cost for the replacement of the fiber or membrane [45]. Fluidized bed reactors can also achieve a high NRR (2160 g–N/(m^3·d)) [29]; however, the system itself is far more complicated and hence is difficult to control [45]. The unsaturated flow pressurized reactor also exhibited a high NRR (2100 g–N/(m^3·d)) and has simplicity, a small size, and is safe, although frequent cleaning of these reactors might be needed [33].

Table 3. Comparison of reactor performance and operating conditions.

Reactor	Substrate	Bacteria Inoculum	Temperature (°C)	HRT (d or h) Flow Rate (mL/min)	H_2 Flow Rate (mL/min) H_2 Pressure (MPa or atm) DH (mg/L) Applied Current (A)	NO_3^-–N (mg-N/L)	Carrier Materials	Reactor Volume (L)	NLR (g-N/(m^3·d))	NRR (g-N/(m^3·d))	Max or Average Efficiency (%)	Reference
Hollow fiber	Well water and synthetic groundwater	Biomass from an anoxic rotating biological reactor in a wastewater treatment plant		4.1 h	0.3–0.6 atm	145	Polypropylene hollow fiber	1.2	770	770	100	[26]
Suspended growth membrane	Synthetic groundwater	Enriched autotrophic denitrifying biomass	16.0 ± 1.1	12 h	DH 1.6	48		7	37.7	37.7	100	[27]
Submerged membrane	Synthetic groundwater	HD bacteria	25–28	3 h	DH 1.6	25	Hollow fiber membrane	5.6	110	110	100	[28]
Fluidized-bed biofilm using solid-polymer electrolyte membrane	Synthetic groundwater		30	1 h	4.0 A	20–90	Polyvinyl alcohol	2.2	2160	2160	100	[29]
Membrane biofilm	Synthetic drinking water	Anaerobic activated sludge		0.5 h	0.05 MPa	10	Hollow fiber	0.024	480	384	80	[30]

Table 3. *Cont.*

Reactor	Substrate	Bacteria Inoculum	Temperature (°C)	HRT (d or h) Flow Rate (mL/min)	H_2 Flow Rate (mL/min) H_2 Pressure (MPa or atm) DH (mg/L) Applied Current (A)	NO_3^-–N (mg-N/L)	Carrier Materials	Reactor Volume (L)	NLR (g-N/(m^3·d))	NRR (g-N/(m^3·d))	Max or Average Efficiency (%)	Reference
Attached growth	Synthetic groundwater	Activated sludge	30	6.7 h	70 mL/min	20	Fiber	3	71.7	69.1	96.4	[23]
Attached growth	Synthetic groundwater	Activated sludge		2.7 h	70 mL/min	20	Fiber carrier	3	176	167	90	[19]
Bio-ceramite	Synthetic wastewater	Anaerobic activated sludge	30	24 h	0.01 MPa	30	Ceramite	2.3	30	28.9	96.2	[31]
Suspended growth	Synthetic groundwater	Enriched HD bacteria in lab	32 ± 0.5	12 h	15 mL/min	40		2	80	77.2	96.5	[24]
Heterotrophic denitrification coupled with electro-autotrophic denitrifying packed bed	Synthetic groundwater	Activated sludge	Room temperature	24 h	0.1 A	50	Haycite Pine sawdust		27.5	27.2	99	[32]
Unsaturated-flow pressurized	Synthetic groundwater	HD bacteria in lab	25.5 ± 1	430 mL/min	DH1.5	25	Plastic biofilm		2100	2100	100	[33]
Suspended growth	Synthetic groundwater	Enriched HD bacteria in lab	32 ± 0.5	12 h	1 mL/min	40		2	80	78.4	98	[34]

Table 3. *Cont.*

Reactor	Substrate	Bacteria Inoculum	Temperature (°C)	HRT (d or h) Flow Rate (mL/min)	H_2 Flow Rate (mL/min) H_2 Pressure (MPa or atm) DH (mg/L) Applied Current (A)	NO_3^-–N (mg-N/L)	Carrier Materials	Reactor Volume (L)	NLR (g-N/(m^3·d))	NRR (g-N/(m^3·d))	Max or Average Efficiency (%)	Reference
Attached growth	Synthetic groundwater	Enriched HD bacteria in lab	32 ± 1	4 h	40 mL/min	40	Polyolefin sponge	2	210	209.9	98	[21]
Attached growth	Raw groundwater treated by on-site nitrification reactor in Chyasal, Nepal	Bacteria from on-site nitrification		13.3 h	70 mL/min	10	Fiber carrier	3	133	107	>80	19
HD with two stage injection of electrolytic H_2	Raw groundwater in Saitama, Japan	Bacteria from their lab	29.0 ± 3.1	4.2 d	2 A	8.1 ± 0.6	Sand gravel	1290	1.9	1.6	81.6 ± 4.4	36
Attached growth	Raw groundwater treated by on-site dropping nitrification in Jwagal, Nepal	Bacteria from on-site HD reactor	20.7–28.1	12 h	120 mL/min	8.3–45.9	Polyolefin sponge	20	73.7	73.7	100	This study

The NRR of the simplified HD reactor (73 g–N/(m^3·d)) was similar to that of suspended or attached growth reactors (e.g., 69.1 [23], 77.2 [24], and 78.4 g–N/(m^3·d) [34]). Higher NRRs of 167 and 107 g–N/(m^3·d) were obtained using attached growth reactors [19] for synthetic and raw groundwater, respectively. However, the supplied H_2 gas for these two reactors was 23 mL/(min·L), whereas 6 mL/(min·L) was supplied for the simplified HD reactor developed in this study.

To ensure sustainability and the compatibility of the water treatment unit with local conditions, the simplified HD reactor was made out of an inexpensive jar that is available all over Nepal, resulting in a low initial cost for the installation of the reactor. It should be noted that the reactor was continuously operated for 157 days without temperature control, cleaning, or complex maintenance. This demonstrates the ease and simplicity of the system, which facilitates local engagement with the technology. Therefore, the simplified HD reactor is a more user-friendly option for NO_3^-–N removal from groundwater in the Kathmandu Valley, Nepal.

3.4. Application and Implementation of the Simplified HD Reactor

Groundwater in the Kathmandu Valley is contaminated by NH_4^+–N and NO_3^-–N, as previously reported [11–15]. NH_4^+–N removal can be conducted by combining the previously developed nitrification system [18–20] and the simplified HD reactor developed in this study (Figure 6a), whereas NO_3^-–N removal can be performed using the simplified HD reactor alone (Figure 6b). However, the simplified HD reactor sporadically emitted an odor during operation, which might affect the acceptability of the treated water. Therefore, filtration with activated carbon, followed by disinfection, is recommended for installation after the simplified HD reactor. The filtration with activated carbon is to remove the odor, turbidity, and dissolved organic compounds (if any) [40]. The activated carbon is recommended to be replaced on a yearly basis [40]. Although chlorination is more common in Nepal for water disinfection, ultra-violet (UV) disinfection could be a better alternative for household scale.

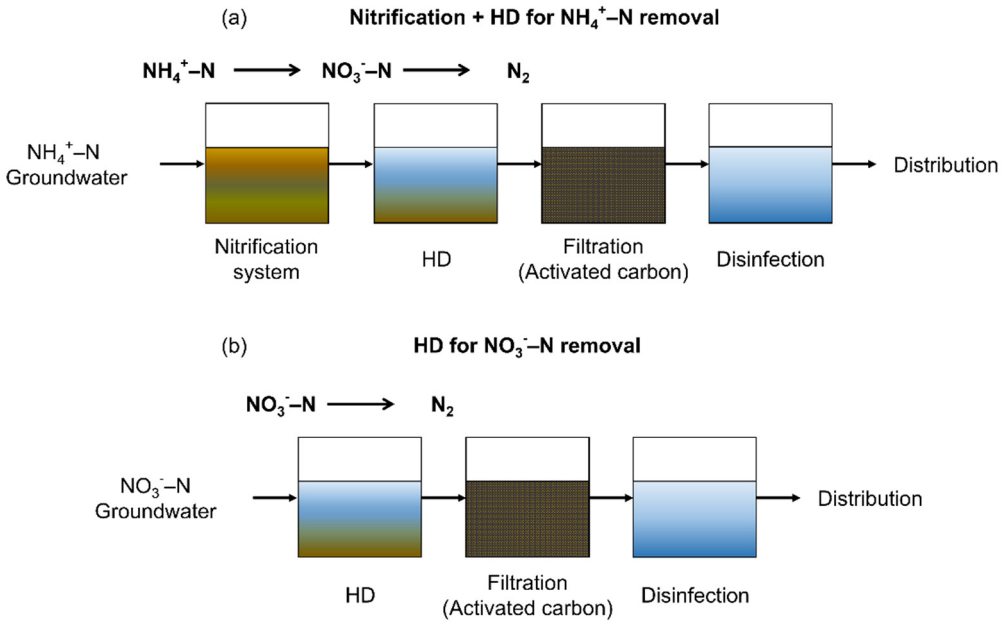

Figure 6. Proposed layout ((**a**) NH_4^+–N removal and (**b**) NO_3^-–N removal) of groundwater treatment in the Kathmandu Valley.

According to the results obtained in this study, the simplified HD reactor can currently produce 40 L of drinking water per day, operating with a working volume of 20 L and an HRT of 12 h. Basic water requirements comprise four basic human needs: drinking water for survival, water for human hygiene, water for sanitation services, and modest household

needs for preparing food [52], and these needs are estimated to require 50 L per capita per day (LPCD) [52]. Although the current simplified HD reactor is not capable of meeting all of these basic water requirements, it could provide the drinking water and water for cooking for a household. The WHO estimates that daily drinking water consumption per capita is 2 L for adults, and it is reported that the average size of a family in Nepal is 4.6 persons [53]. Thus, it can be assumed that a household requires approximately 9–10 L of drinking water per day, which is consistent with the value of 9.4 L from previously reported questionnaire survey results [8]. Furthermore, water consumption for cooking was reported to be 3–6.5 LPCD, depending on household income [54], which is equivalent to 12–33 L for a household per day. Therefore, the simplified HD reactor can cover the water requirements for drinking and cooking purposes at the household level.

In the present study, H_2 gas was supplied from a water electrolytic H_2 generator, resulting in high installation costs and frequent maintenance, such as the need to add water and replace the generator's drying agent [40]. Frequent power cuts in Nepal [55], which interrupt the operation of H_2 generators, should also be considered. In the long run, the generator should be substituted with easier and cheaper alternatives, and H_2 should be efficiently used to reduce maintenance and cost. Furthermore, the sponge materials used in this study were not locally available; thus, a simplified HD reactor with locally available materials should be tested to investigate treatment capacities using possible substitutes for the current sponge material.

4. Conclusions

A case study was conducted to develop a simplified HD reactor using as many locally available materials as possible, and the reactor was installed in the Kathmandu Valley, Nepal to evaluate its effectiveness, understand its behavior under on-site conditions, and fill the gap in knowledge between laboratory and practical applications. NO_3^-–N removal by the reactor was found to be effective, and the reactor showed rapid start-up ability, stability, and robustness against pretreated groundwater. The reactor had an average nitrogen removal efficiency of 80.9 ± 16.1%, while NLR and NRR varied from 23.8 to 92.3 g–N/(m^3·d) and from 18.3 to 73.7 g−N/(m^3·d), respectively. The simplified HD reactor can be operated under on-site conditions without temperature control, supplementary DO concentration control, an additional inorganic carbon source, or internal cleaning. Compared to the results of previous research, the simplified HD reactor was considered a more user-friendly option for people in the Kathmandu Valley, Nepal. Additionally, the reactor is recommended for groundwater treatment at the household level, as it can currently produce 40 L/d, fulfilling the daily requirements for drinking and cooking water in a household with 4–5 people. The findings of this research would be helpful for practical applications of HD in developing countries.

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