LOADING OF AN ANAEROBIC PACKED BED FOR TREATMENT OF A HIGH STRENGTH GRAY WATER

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ABSTRACT

In preparation of long-duration manned space flight, biological treatment systems have been evaluated for mission specific waste streams consisting of a high-strength gray water. The purpose of the work was to determine the appropriateness of using nitrification and denitrification to treat the urine-humidity condensate waste stream and to determine the optimal loading rate for the bench scale treatment system. Biological treatment of the high strength graywater was kinetically and stoichiometrically limited. Due to the low C:N ratio of this wastewater (0.85:1), biological treatment at long HRTs is stoichiometrically limited and may be overcome by adding alkalinity to improve nitrification efficiency and organic carbon to improve denitrification efficiency.

KEYWORDS

Denitrification, nitrification, high strength gray water

INTRODUCTION

Currently, manned space missions like the International Space Station (ISS) are not self-sufficient as far as their water supply is concerned and rely upon multiple shuttle missions for resupply. Owing to the extensive use of light-weight materials in space missions, water is one of the heaviest materials used during a mission, making shuttle resupply extremely expensive propositions, as it is estimated that 1 liter of water supplied to the International Space Station (ISS) costs $ 20,000. The cost of resupply increases in direct proportion to the duration of the mission and the payload weight, which has severe monetary implications. Current conditions emphasize the need for developing more efficient environmental control and advanced life support systems for onboard conversion of wastewater to potable water, making these long-term space missions self-sufficient. This has led to the recent emphasis on the development of water-recycling systems for space missions.

Considerations in the design of water reclamation systems in space include shelf life, resupply-return logistics, crew time needed for maintenance, energy requirements to operate the system, launch weight, and stowage volume. Currently, two different processes train philosophies are under investigation for the appropriateness for long-duration space missions such as a lunar base or a trip to Mars. One approach consists of an integrated biological/physicochemical system, while the other approach consists of a purely physicochemical treatment processes.

Biological treatment has several advantages over physicochemical treatment methods (i.e., reverse osmosis, ion exchange, distillation, etc.) including: [1] less energy inputs; [2] minimal

use of expendables such as ion-exchange resins and reverse osmosis membranes, reducing payload requirements; [3] minimal wastes produced that require storage and handling until final disposal; and [4] minimal chemical additions to augment treatment efficiency, further reducing payload requirements. When used in this capacity, biological treatment systems have the advantage of significantly reducing the load on downstream physicochemical treatment processes. Physiochemical systems will be required to treat the biologically treated wastewater to potable water standards; however, the size of the physiochemical units will be significantly reduced as the contaminant concentration is reduced during biological treatment.

As NASA prepares for long-duration manned space flights, the applicability of biological wastewater treatment systems must be evaluated for the waste streams anticipated during the mission. NASA has demonstrated the appropriateness of biological treatment technologies on dilute waste streams (Campbell et al., 2003a; Campbell et al., 2003b); however, a more concentrated waste stream may present a greater treatment challenge. One such waste stream is the urine-humidity condensate waste stream, which consists of urine, humidity condensate collected in the cabin of the space craft from human and machine respiration, and dilution water created during urinal flushing and oral hygiene activities. The high strength gray water has a dissolved organic concentration (DOC) and total nitrogen (TN) concentration of approximately 1100 mg/L and 1500 mg/L, respectively. The system influent pH is approximately 9. The TN of the influent is predominately ammonia-nitrogen, which may cause free ammonia toxicity problems at the high pH values. Therefore, the purpose of the work was to assess the appropriateness of using nitrification and denitrification to treat the urine-humidity condensate waste stream and to determine the optimal loading rate for the bench scale treatment system.

MATERIALS AND METHODS

The denitrification-nitrification system consists of an anaerobic packed bed (APB) and a membrane-aerated bioreactor, respectively (Figure 1). The packed bed was approximately 46 cm long with an internal diameter of 7.62 cm. The surface area of the lava rock, the support media for biofilm attachment, was 0.2 m^2. The resulting working volume of the reactor was 1.1 L. The reactor was inoculated from a mixed heterotrophic culture from the TTU-WRS (Jackson and Morse, 2005).

The membrane-aerated reactor (AMR) was designed at TTU (Morse et al., 2003) to allow for bubble-less aeration of wastewater, which is necessary in microgravity environments. The reactor was 45.7 cm in length and has a diameter of 10.2 cm. The membranes were approximately three times longer than the length of the reactor and had an inner and outer diameter of 0.17 cm and 0.08 cm, respectively. The membranes were placed in a random fashion to act similarly as packing media in a packed bed reactor, resulting in membrane-membrane contact. The membrane surface area for biofilm attachment and aeration was 1.098 m^2; however, approximately 25 percent of the membrane surface area was not available due to membrane-membrane contact. Thus, the assumed available surface area for microbial growth and aeration was 0.825 m^2. The bottom air cavity of the reactor was pressurized with facility air to 6 psi to facilitate oxygen transport from the lumen side of the membranes to the wastewater. The reactor was inoculated from a mixed culture of nitrifying organisms from the TTU-WRS (Jackson and Morse, 2005).

The rest of the anaerobic packed bed and the nitrifying reactor system contained feed and effluent tanks, a peristaltic feed pump, a recycle piston pump, and Masterflex tubing. The packed bed reactor was pressurized to keep the nitrogen gas bubbles formed during denitrification in solution. A gas-liquid separator was located after the membrane-aerated reactor to allow gas bubbles in the system to escape to prevent cavitation of the piston pump.

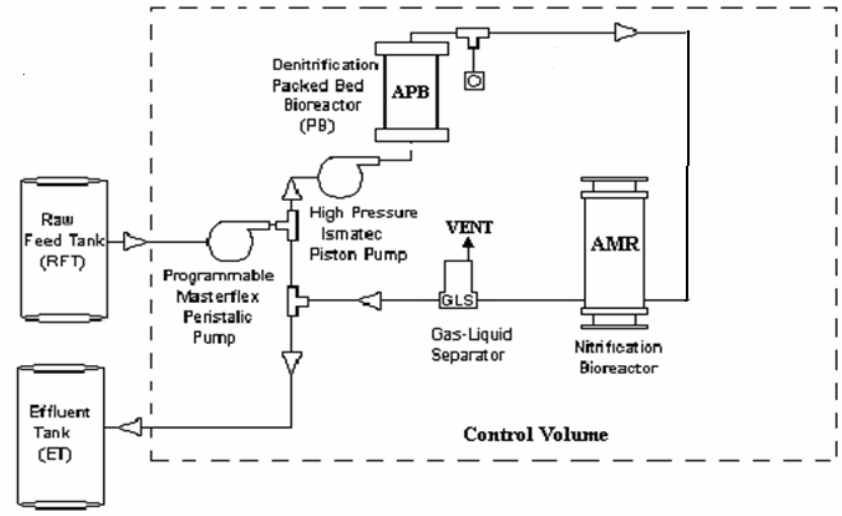

Figure 1. Schematic diagram of the anaerobic packed bed reactor and membrane-aerated reactor system (Kaparthi, 2004).

For 1 L of feed solution, 344 mL urine, 3.12 mL humidity condensate, and 522 mL of DDI as make-up water was added to the feed tank daily. The humidity condensate is mixture of chemicals designed to mimic the humidity condensate collected on the ISS (Verosko et al., 2004). The humidity condensate consists predominately of ethanol, 1, 2-propanediol, and zinc acetate dehydrate (Verosko et al., 2004). The loading study was conducted by increasing the influent flow rate and maintaining a constant recycle ratio (recycle ratio equaled 10). The detention times used in this study were 2.02, 2.31, 2.69, 3.24, 4.04, 4.62, and 5.39 d. The detention time, representing the time a drop of water remained in the system, was altered by increasing the flow rate. Due to this approach, the volume of feed solution prepared each day was modified. Additionally, 500 mL of the feed solution was maintained in the feed tank at all times to encourage urea hydrolysis and provide a volumetric buffer to minimize shock loadings to the system.

Samples were collected from the influent and effluent of the system as well as at intermediate points in the system, the filtered with a 0.45 μm filter, and analyzed for pH, TN, DOC, ammonia, nitrate (NO_3^--N), and nitrite (NO_2^--N). Ammonia samples were preserved with sulfuric acid and the concentration was measured using an ammonia probe (Thermo Orion Corporation, Model number 951201) in conjunction with an Orion meter (Model 250A). Ammonia measurements were converted to ammonium-nitrogen values (NH_4^+-N). Ion chromatography (AS40, DIONEX) was used to measure the concentrations of NO_2^--N and NO_3^--N, while TN was measured using a TN-TOC analyzer (TNM-1, Shimadzu). DOC was measured using a combustion method

(Shimadzu Corporation, Model number TOC-V_{CSH}). TN and DOC samples were preserved with hydrochloric acid.

As real urine was used in the loading study, nitrification was estimated by determining the amount of NO_3^--N and NO_2^--N produced in the system rather than the amount of NH_4^+-N removed as the input of NH_4^+-N to the system was never at steady-state. Urea hydrolysis, which is the breakdown of urea in to ammonium, has been documented to take up to 60 hours in the feed tanks of these systems (McLamore, 2004). Due to urea hydrolysis occurring in the feed tanks as well as in the treatment system, NH_4^+-N concentrations were highly variable. Additionally, ammonia is produced during denitrification (Metcalf and Eddy, 2003), compounding the flux of NH_4^+-N in the system. Therefore, NO_x-N production rather than NH_4^+-N removal was the preferred methodology to estimate nitrification efficiency. The NO_x-N production across the system was estimated using the following formula. The equation below accounts for the NO_x-N produced in the membrane reactor that is converted to N_2 gas in the packed bed reactor.

$$NO_x\text{-N produced (mg/L)} = (NO_x\text{-}N_{eff} - NO_x\text{-}N_{inf}) + (TN_{inf} - TN_{eff})$$

In the equation above, NO_x-N_{eff} is the NO_x-N in the system effluent (mg/L), NO_x-N_{inf} is the NO_x-N in the system influent (mg/L), TN_{inf} is the total nitrogen in the system influent (mg/L), and TN_{eff} is total nitrogen in the system effluent (mg/L). The equation presented above assumes all of the TN entering the system is ammonia, which has been confirmed during research of the system (McLamore, 2004).

RESULTS

Due to the dependence of denitrification on nitrification for nitrate (i.e., the terminal electron acceptors in denitrification) in a combined system, the performance of the nitrifying reactor will be presented. Specifically, the production of nitrite (NO_2^--N) and nitrate (NO_3^--N) in the nitrifying reactor will be presented.

Nitrifying Membrane Reactor

The nitrogen loading rates and the NO_x-N (NO_2^--N + NO_3^-–N) mass production rates in the membrane-aerated reactor for each HRT were averaged, and a graph for the variation in these values with HRT was obtained. Figure 2 shows a roughly linear relationship between the average TN loading rate and the average (and standard deviations) NO_x-N mass production rate (mg/m^2-d) for the different HRTs. The NO_x-N production is an important indicator of the membrane-aerated reactor performance as it estimates the amount of ammonia removed during nitrification. The reason for the large standard deviations observed during the experiments is due to urine. Urine contains many nitrogen compounds, vitamins, hormones, organic acids, amino acids and various organic compounds (Kaparthi, 2004). The presence and concentration of these compounds is a function of the donor, the donor's diet, and the donor's exercise regime.

The straight line in Figure 2 has a slope of 0.49 and a Y-intercept of 340.6 mg/m^2-d. This trend can be justified as the low HRTs are expected to have high TN loading rates (mg/m^2-d). At both

high and low HRTs, the system exhibited poor nitrification performance, which is a function of low loading rates (HRT equals 4.69 d and 5.39 d) and insufficient contact time (HRT equals 2.02 d and 2.69 d), generating low NO_x-N production. Any point with a high TN loading rate because of the high influent flow rate (which means a low HRT) will have a less than proportional increase in NO_x-N production over the preceding point on the graph. This causes the graph to have a slope significantly less than 1.

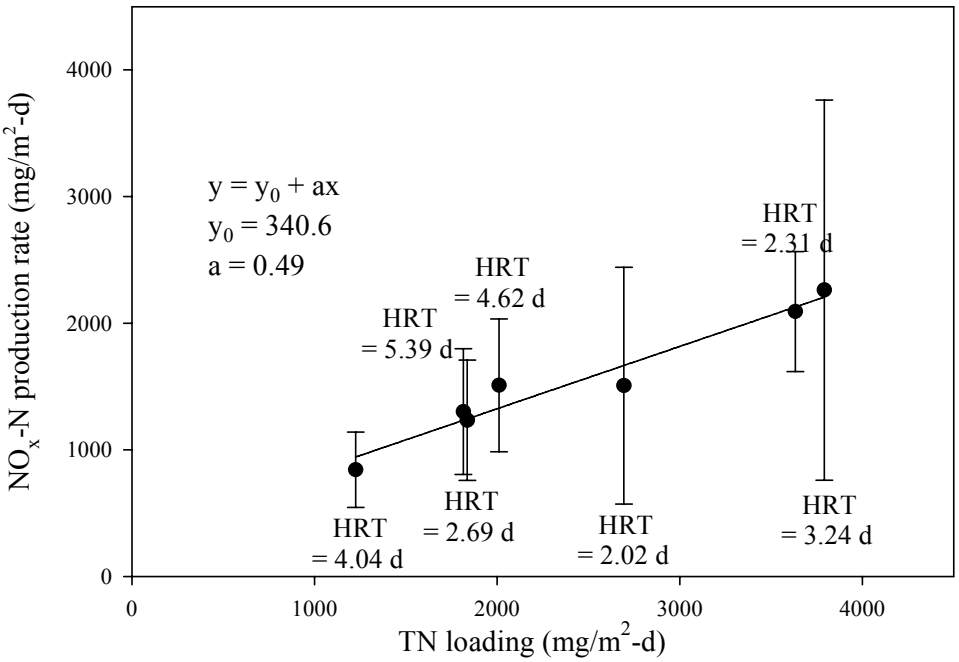

Figure 2. Variation in average NO_x-N mass production per unit membrane surface area with average nitrogen loading rate for different HRTs.

Figure 3 presents the variation in average NO_x-N production per unit membrane surface area for different loading rates. At HRT values of 4.04 d, 4.69 d and 5.39 d, the effluent pH was approximately 6.6, suggesting that alkalinity was limiting. Therefore, at high HRTs (low flow rates), stoichiometric limitations controlled reactor performance. However, at low HRTs (i.e., high flow rates) the reactor performance was kinetically limited. Effluent pH values for HRT values of 3.24 d, 2.69 d, 2.31 d, and 2.02 d ranged between 7.6 and 8.6, suggesting that alkalinity was not limiting. Despite kinetic or stoichiometric limitations, effluent NH_4^+-N concentrations ranged from 361 mg/L to 590 mg/L, generating an NH_4^+-N removal efficiency of 4 to 33 percent.

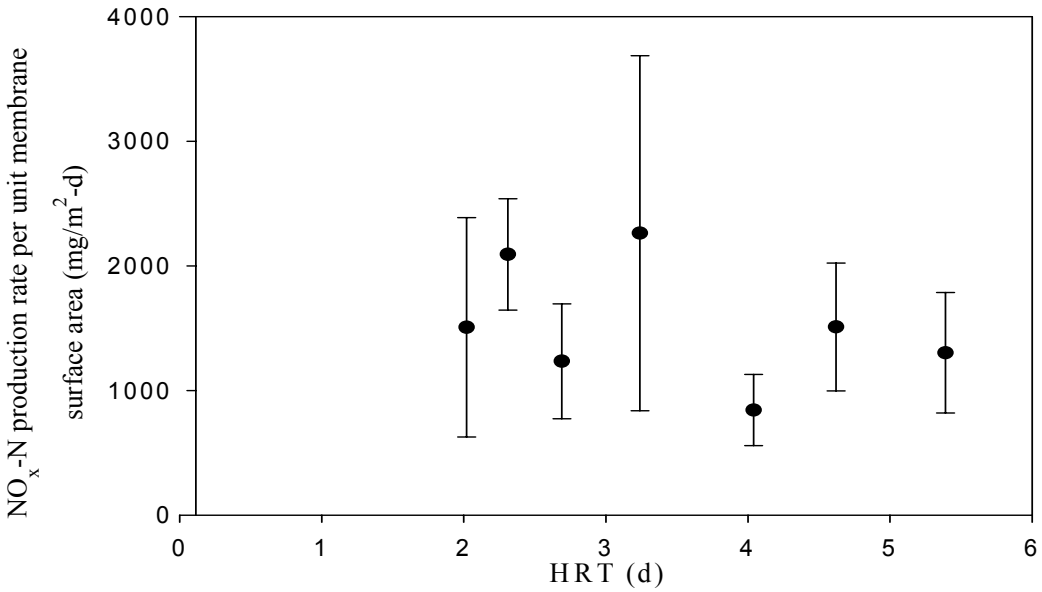

Figure 3. Variation in average NO_x-N production per unit membrane surface area for different loading rates.

Anaerobic Packed Bed Reactor

The TN loading rates and TN removal rates of the packed bed reactor for each HRT were averaged, and a graph for the variation in these values with HRT was obtained (Figure 4). The same was done for the DOC loading rates and the DOC removal rates (Figure 5). A roughly linear relationship is obtained between the values for removal of both TN and DOC. The line through the plot for TN removal rate (Figure 4) has a slope of 0.5, and a negative Y-intercept. The negative Y-intercept can be attributed to the fact that DOC was limiting in the waste stream. Therefore, stoichiometric limitations controlled reactor performance rather than kinetic limitations.

Figure 4 shows average TN removal per unit anaerobic packed bed surface area for different loading rates. Again, the data suggests at long HRTs, the performance of the reactor is stoichiometrically limited while at higher HRTs, the reactor is kinetically limited. The relationship is expected as the performance of the nitrifying reactor will affect the performance of the packed bed reactor as denitrification will be limited by the amount of NO_3^--N present in the system.

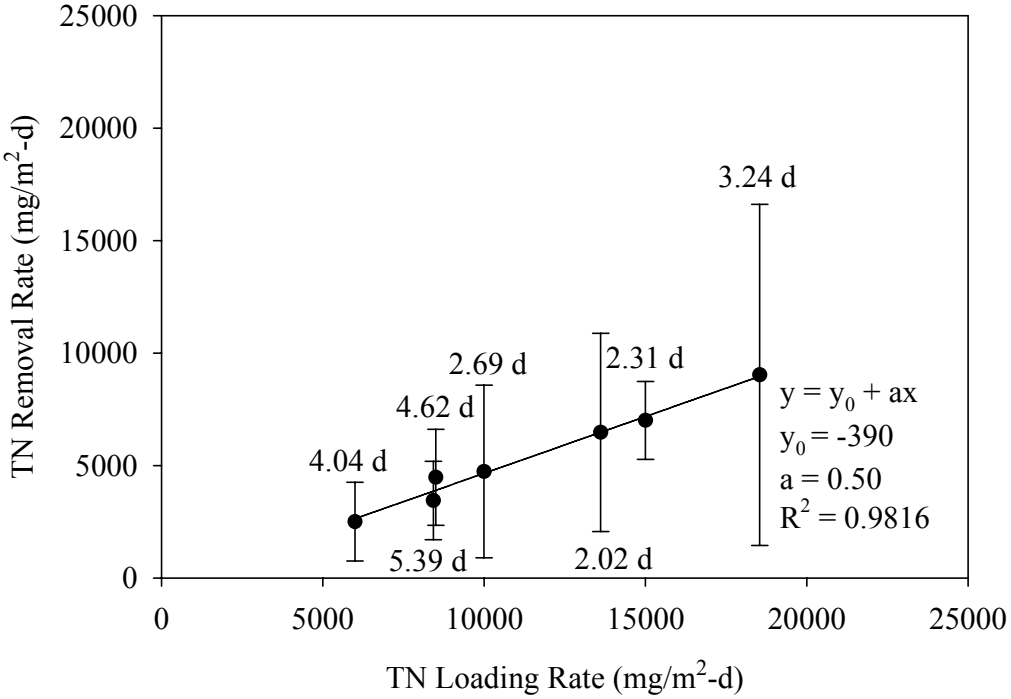

Figure 4. Variation in TN removal rate across the system with TN loading rate for different HRTs.

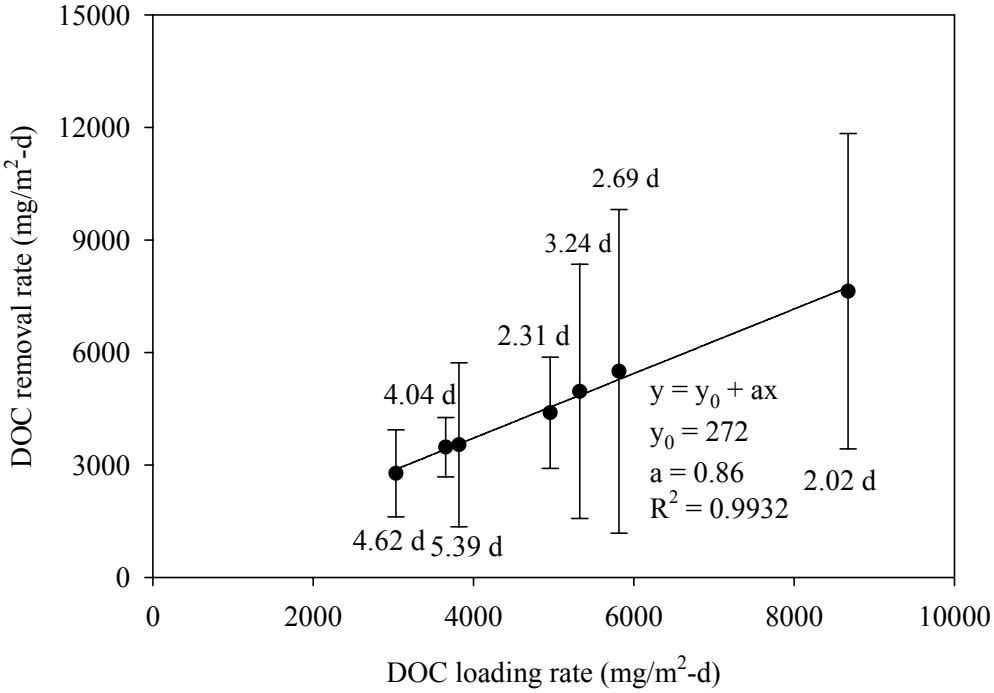

Figure 5. Variation in DOC removal rate across the system with DOC loading rate for different HRTs.

In Figure 5, the line through the plot for DOC removal has a slope of 0.86 and a Y-intercept of 272 mg/m^2-d. From this figure, the points with poor nitrogen removal performance (i.e., HRTs 2.02 d, 2.31 d, and 3.24 d) (Figure 4) occur in the higher range of the DOC loading rates experienced during the system operation. Since these points have poor denitrification performance, DOC removal was also affected owing to the stoichiometric relationship between TN and DOC removal, leading to a slope lower than 1.0.

The low slope value in Figure 4 is due to limited nitrification at the HRT values of 2.02 d, 2.31 d, and 3.24 d. The points at HRT values of 2.02 d and 2.31 d experience high pH values (approximately 8.0) due to high influent NH_4^+-N loading (approximately 1600 mg/L), thus affecting nitrification performance. Nitrification was also adversely affected by the detention time, (i.e., as HRT decreased, NO_x-N production decreased). Alkalinity is consumed during nitrification, decreasing pH. This is significant because at the short HRTs (high flow rates), the pH drop due to nitrification was insignificant compared to the initial pH of the wastewater (approximately 9.0). The pH range considered optimal for nitrification is between 7.5 and 8.0 (Metcalf and Eddy, 2003) and at pH values above 8, the ammonia gas dominates in the system resulting in high concentrations of free ammonia. Ultimately, the high pHs resulting from poor nitrification generated a compounding feed back loop that shut down nitrification. Due to the high pHs, free ammonia, which is toxic to the nitrifying organisms, accumulated in the system, nitrification ceased and pH remained high. When nitrification ceased, denitrification ceased due to the lack of a terminal electron acceptor (NO_3^--N or NO_2^--N).

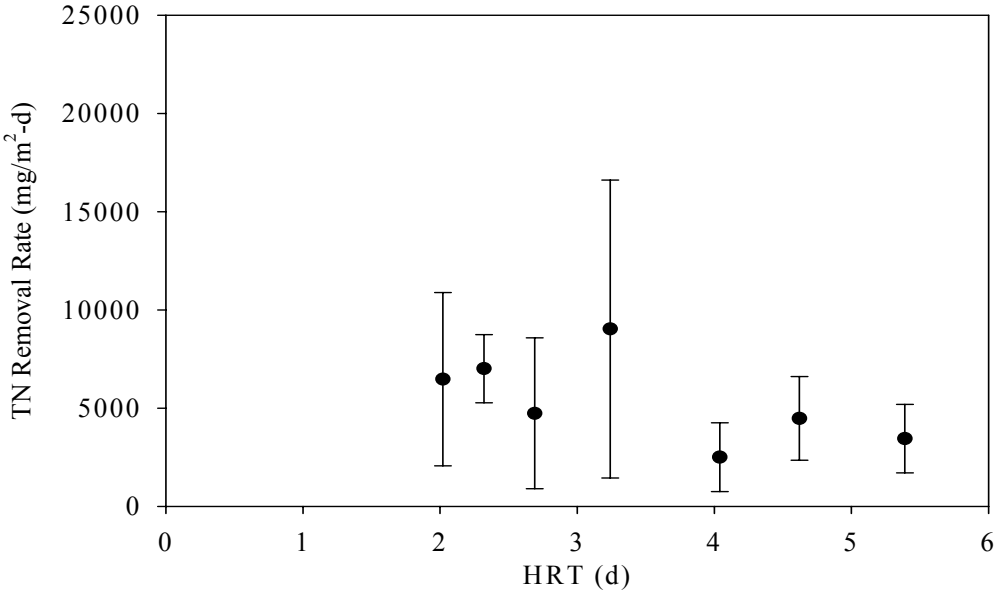

Figure 6. Variation in average TN removal per unit anaerobic packed bed surface area for different loading rates.

DISCUSSION

From this analysis, the performance of the anaerobic packed bed reactor is limited by nitrifying membrane-aerated reactor performance, which is a function of stoichiometric and kinetic limitations. Stoichiometric limitations are inherent due to the urine-humidity condensate waste stream composition, whereas kinetic limitations were a function of system operation. The ratio of C:N in the influent waste stream is approximately 0.85:1, which is significantly less than recommended range of 4:1 (Metcalf and Eddy, 2003). Additionally, not all of the influent DOC is usable. Urea will contribute to the DOC measurement and subsequent loading of the system. During the combustion method, two moles of CO_2 are released during the combustion of urea, which inflates the amount of organic carbon in the system. As denitrification requires organic carbon not inorganic carbon, all influent DOC measurements must be corrected to remove the amount of DOC contributed by urea. From the correction, the actual C:N ratio is 0.5:1 for the urine-humidity condensate waste stream.

The performance of the anaerobic packed bed reactor-nitrifying membrane reactor system as a whole largely depends on the nitrifying reactor performance. The reduction in the nitrifying membrane reactor performance adversely affected the performance of the anaerobic packed bed reactor. As less NO_x-N was produced, less nitrogen was removed by conversion of this NO_x-N to N_2 gas. The point of HRT equal to 3.24 d was a start-up point, which also had high pHs prevailing in the system for long periods, affecting nitrification performance, and the subsequent nitrogen removal and denitrification performance. At the start-up point (HRT 3.24 d), the reactor contained less biomass hence both the anaerobic packed bed reactor and the nitrifying membrane reactor had less ability to reduce substrates in the waste stream. The three points with highest HRTs (4.04 d, 4.69 d, 5.39 d) (i.e., lowest flow rates) had the lowest nitrogen removal, whereas the four points with highest TN loading rates (i.e., lowest HRTs) had higher TN removals. Therefore, a trend line could not be fit to the complete data set in Figure 6 and two operating conditions are evident. It is apparent that TN removal expressed as denitrification was organic carbon limited for HRTs between 4.04 d and 5.39 d and denitrification was kinetically limited for HRTs between 2.02 d and 3.24 d. The data in Figures 3 and 6 suggests that the optimal operational point is 2.31 d. At this HRT, NO_x-N production is optimized, while maintaining high TN and DOC removal.

To improve the performance of the system, operational parameters (i.e., reactor sizing, influent flow rates, etc.) should be adjusted to optimize the performance of the nitrifying membrane-aerated reactor, which will optimize the performance of the whole system. Increasing the hydraulic residence time of the nitrifying membrane-aerated reactor may improve nitrification, improving the anaerobic packed bed reactor performance. Additionally, the increased volume of the reactor may minimize nitrogen shock loadings to the system, which is illustrated by the large standard deviations in TN removal observed in Figures 4 and 6.

Ideally, the C:N ratio of the gray water should be altered so that the C:N ratio is closer to 4:1. However, the composition of the urine-humidity condensate gray water is unlikely to change as the wastewater composition is dictated by the personal hygiene equipment aboard the space craft.

Therefore, biological treatment of the high strength urine-humidity condensate waste stream may not be a viable approach due to the long hydraulic retention times necessary for treatment. Modifications must be made to the gray water to decrease initial pH values to less than 9.0 and to decrease carbon limitations. By increasing the organic carbon content of the influent gray water, nitrification may be improved as denitrification produces alkalinity. Approximately 3.57 grams of alkalinity as $CaCO_3$ is produced per gram of nitrate nitrogen consumed. To quantify the true benefit of increasing the organic carbon on alkalinity production during denitrification, the stoichiometric relationship should be developed for the wastewater of interest.

As indicated by the performed work, biological treatment of terrestrial gray water streams with similar C:N ratios may not be feasible. For biological treatment to be successful, the C:N ratio must be altered to a value closer than the recommended 4:1. Another alternative is to add alkalinity to the gray water to improve nitrification efficiency. Lastly, terrestrial systems should have a long hydraulic residence time to insure treatment is not kinetically limited.

CONCLUSION

Biological treatment of the high strength gray water known as the urine-humidity condensate gray water is both kinetically and stoichiometrically limited. Due to the low C:N ratio of this wastewater (total 0.85:1; usable 0.5:1), biological treatment at long HRTs is stoichiometrically limited and may be overcome by adding alkalinity to improve nitrification efficiency and organic carbon to improve denitrification efficiency. Although biological treatment, without modification of the wastewater, will not meet typical permit BOD and TN discharge values (30 and 10 mg/L, respectively), biological treatment may serve as an acceptable pre-treatment method for physiochemical removal techniques.

ACKNOWLEDGEMENTS

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