

Long Term Nitrogen Management in Bioreactor Landfills

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Introduction and Objectives

Landfill leachate typically contains high concentrations of ammonia-nitrogen long after it is stable with respect to both organic and metal concentrations. Thus, the removal of ammonia from landfill leachate will be a factor in determining when an overall landfill can be considered stable. One scenario for ammonia removal is the aerobic treatment of leachate outside of the landfill to convert ammonia to NO_3 , followed by use of the landfill as an anaerobic bioreactor for the conversion of NO_3 to N_2 . The objective of this research was to develop an understanding of the nitrogen cycle in landfills sufficient to develop a long-term strategy for the management of nitrogen in landfill leachate. Specific questions include (1) How much nitrate-nitrogen ($\text{NO}_3\text{-N}$) can be added to refuse in various states of decomposition without adversely affecting the ability of the refuse to resume methanogenesis after NO_3 depletion?, (2) Do denitrification reactions result in an inhibitory pH increase?, (3) Are there conditions under which NO_3 addition to landfills would lead to the production of ammonium (NH_4^+) in place of N_2 by dissimilatory NO_3 reduction to ammonium (DNRA)?, and (4) What types of organic carbon and what concentrations are required to support denitrification in landfills?

Experimental Design

Nine reactors (1-9) were filled with a mixture of fresh and old refuse to evaluate the extent of NO_3 consumption and the effect of NO_3 addition on CH_4 production, leachate pH and N_2O production. The old refuse served as a seed to initiate CH_4 production. The reactors were operated until they reached an approximate peak in CH_4 production at which time NO_3 was added to eight of nine reactors (1-6, 8-9) while the ninth reactor (7) served as the control. This first set of NO_3 additions will be referred to as Phase 1 NO_3 additions. In these reactors, sufficient NO_3 was added every other day to increase the leachate concentration by 400 mg $\text{NO}_3\text{-N/L}$ in each reactor.

In the Phase 2 NO_3 addition regime, NO_3 was added to 3 of 9 reactors (1, 6, 8) to measure the denitrification potential in well-decomposed refuse as defined by a relatively low CH_4 production rate. Initially, NO_3 was added to the decomposed refuse. Next, high concentrations of degradable organic carbon in the form of acetate were added to selected reactors along with low concentrations of NO_3 to evaluate whether ammonia was produced by DNRA. Next, a humic acid solution was added along with NO_3 to evaluate whether it can serve as an electron donor to support denitrification. Finally, the top 25 % of the refuse in reactor 1 was replaced with fresh refuse, which simulated the placement of a layer of fresh refuse over well decomposed refuse. Nitrate was then added 3 times to evaluate whether the organic carbon from the fresh refuse could support denitrification.

Results: Phase 1 - Denitrification During Active Methane Production

The refuse ecosystem exhibited strong capacity to consume NO_3 when added at 400 mg-N/L to actively decomposing refuse. As expected, CH_4 production was inhibited when NO_3 was added and the CH_4 recovery time after cessation of NO_3 addition was generally proportional

to the duration of the NO_3 addition, which varied from 19 to 59 days. The CH_4 production recovery times suggest that methanogens were inhibited but not killed by the high NO_3 additions. The Phase 1 CH_4 yields were inversely proportional to the mass of NO_3 added, which is consistent with the diversion of the carbon substrate from the reduction of CO_2 to CH_4 to the reduction of NO_3 to N_2 . Towards the end of the NO_3 addition regime, when refuse decomposition was advanced, NO_3 consumption rates decreased such that not all NO_3 was consumed in 48 hr. Reduced NO_3 depletion rates corresponded with the lowest measured COD concentration in a reactor, suggesting that the decrease in available organic was the reason why NO_3 depletion slowed down in these reactors. Total N recovery ranged from 95 to 106% of the added $\text{NO}_3\text{-N}$. While most of the added NO_3 was recovered as N_2 (89.5 to 101.2%), 2.3 – 11.7% was recovered as N_2O . This amount is expected to be above the upper limit of what would occur in a full-scale landfill where there would be longer retention times, allowing for more complete reduction of N_2O to N_2 .

Results: Phase 2 - Denitrification in Well Decomposed Refuse

In decomposed refuse with relatively low CH_4 production rates, the depletion of NO_3 required 5 – 15 days and this period of slow NO_3 depletion corresponded with lower BOD and COD concentrations relative to Phase 1. The addition of excess acetate stimulated NO_3 depletion and resulted in a significant increase in BOD. Even after acetate additions ceased, NO_3 was rapidly consumed until the excess acetate had been depleted, which corresponded with a decrease in BOD. Nitrate depletion was also stimulated by removing the top 25% of the well-decomposed refuse and replacing it with fresh refuse. This simulated the addition of fresh refuse to the top layers of a landfill. Nitrate depletion was not stimulated by humic acid addition. Thus, the results of the acetate, humic acid and fresh refuse addition are consistent in showing that degradable organic carbon limits the rate of NO_3 depletion in decomposed refuse.

The addition of a high ratio of degradable organic carbon to NO_3 did not appear to lead to the production of ammonia by DNRA which is good as ammonia production would be counterproductive in landfills. A possible explanation is that a significant portion of the added carbon was consumed by methanogens. Methane production was inhibited but did not cease when NO_3 was added. Therefore, it is possible that the NO_3 reducing bacteria were exposed to a lower electron donor/electron acceptor ratio than that which was added to the reactors.

Summary

Methane production rates were inhibited while the refuse was exposed to high concentrations of NO_3 , but rates recovered after NO_3 addition was terminated. Thus, it would appear that the use of a landfill as a bioreactor in which NO_3 -rich leachate is recirculated for conversion to N_2 gas is viable, and denitrification will not adversely upset the landfill ecosystem in a manner from which it cannot recover. From a field-scale perspective, the results suggest that NO_3 addition can be expected to most completely reduce CH_4 production closest to the point of injection. Given the capacity of refuse to rapidly consume NO_3 , it is unlikely that CH_4 production in the deeper sections of the landfill will be affected. This has implications for the emissions of uncollected CH_4 , which likely originate near the landfill surface.