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Moisture effects on gas-phase biofilter ammonia removal efficiency, nitrous oxide generation, and microbial communities

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HIGHLIGHTS

- Increase moisture content from 35 to 55% greatly improved NH_3 removal.
- Increase moisture content from 55 to 63% triggered N_2O generation.
- Ammonia oxidizers (*amoA* communities) resisted to moisture disturbance.
- Denitrifiers (*nosZ* communities) were resilient to moisture disturbance.
- Decrease of *nosZ* abundance caused N_2O generation at high MC.

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ABSTRACT

We established a four-biofilter setup to examine the effects of moisture content (MC) on biofilter performance, including NH_3 removal and N_2O generation. We hypothesized that MC increase can improve NH_3 removal, stimulate N_2O generation and alter the composition and function of microbial communities. We found that NH_3 removal efficiency was greatly improved when MC increased from 35 to 55%, but further increasing MC to 63% did not help much; while N_2O concentration was low at 35–55% MC, but dramatically increased at 63% MC. Decreasing MC from 63 to 55% restored N_2O concentration. Examination of *amoA* communities using T-RFLP and real-time qPCR showed that the composition and abundance of ammonia oxidizers were not significantly changed in a "moisture disturbance-disturbance relief" process in which MC was increased from 55 to 63% and then reduced to 55%. This observation supported the changes of NH_3 removal efficiency. The composition of *nosZ* community was altered at 63% MC and then was recovered at 55% MC, which indicates resilience to moisture disturbance. The abundance of *nosZ* community was negatively correlated with moisture content in this process, and the decreased *nosZ* abundance at 63% MC explained the observation of increased N_2O concentration at that condition.

1. Introduction

Over 70% ammonia emissions originate from livestock operations [1], and through reactions with sulfuric and nitric acid aerosols, these emissions generate 5–11% of the total $PM_{2.5}$ (particulate matter with aerodynamic equivalent diameter of 2.5 μl or less) in the United States [2,3]. Biofilters are commonly used to mitigate livestock emissions [4,5]. These engineered systems are bioreactors filled with organic packing materials such as wet woodchips or composts that remove ammonia from contaminated air streams through absorption, and then oxidize it into nitrite and nitrate using microorganisms growing on the surface of packing media [6]. Most studies have focused on the value and importance of improving biofilter ammonia capture ability [7–9], however, recent reports about the generation of nitrous oxide from biofilters have prompted designers to consider the consequences of greenhouse gas (GHG) emissions [10–12]. Nitrous oxide can be produced by both ammonia oxidizers and denitrifiers within a biofilter [13]. A very small group of ammonia oxidizers are able to directly reduce NO_2^- under microaerophilic conditions through the activities of denitrifying enzymes [14,15], but the majority of nitrous oxide is believed to come from denitrification [5,16]. Within a large scale biofilter, given the compaction of small

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Nomenclature

ANOSIM	Analysis of similarity
ANOVA	Analysis of variance
ARISA	Automated ribosomal intergenic spacer analysis
BF	Biofilter
CTAB	Cetyl trimethyl ammonium bromide
GHG	Greenhouse gas
ID	Inner diameter
LSD	Least significant difference
MC	Moisture content
NMDS	Non-metric multidimensional scaling
OD	Outer diameter
PCR	Polymerase chain reaction
PM	Particulate matter
PerMANOVA	Permutational multivariate analysis of variance
qPCR	Quantitative polymerase chain reaction
RE	Removal efficiencies
RH	Relative humidity
T-RFLP	Terminal restriction fragment length polymorphism
VOC	Volatile organic compound

media [17], there exist anaerobic zones which favor denitrification [18].

Moisture content is a major determinant of biofilter performance [19]. Too little moisture slows absorption of gaseous ammonia and microbial activities [20]; while too much water fills biofilter media pores and retards the transport of NH_4^+, O_2, and nutrients across the water film coating the microbes. Despite the explicit role of moisture for determining ammonia removal [21], the effects of moisture on nitrous oxide generation are rarely specifically considered or tested. A primary reason for this gap is the lack of awareness about the extent of localized oxygen limitation in media due to high moisture contents [11], because a biofilter is generally considered to be an aerobic environment since air continuously goes through it. Moreover, the adsorption of nitrous oxide to media surfaces complicates observations as several studies reported mitigation of nitrous oxide through biofilters [22,23]. However, water competes with nitrous oxide for adsorption spots, especially when the media is decorated with hydrophilic functional groups such as —OH or —COOH [24,25]. Therefore, at high moisture content, less nitrous oxide adsorption and higher nitrous oxide generation rate are expected [26,27]. Maia et al. [10] reported that 0.6–2.0 ppm nitrous oxide was produced in a start-up biofilter with about 60% media moisture content.

Besides the influence of moisture content on the physicochemical interactions of ammonia and nitrous oxide with biofilter media, the response of biofilter microorganisms to moisture changes is another key issue, particularly given the lack of effective moisture managements in many agricultural biofilters. The relationship between community dynamics and system functional stability has been debated, and different phenomena were observed from case to case [28–32]. The concepts of resistance, resilience and redundancy have been applied to describe the relationships between microbial community structure and biofilter performance [33]. Long-term stable function has been sustained in a biofilter treating ammonia and VOCs emissions, although the microbial communities were highly dynamic [34]. Li et al. also observed functional redundancy when phosphorus was added to nitrate removal bioreactors [35]. Another study carried out by Gentile et al. [36] demonstrated resilience of microbial communities to environmental disturbances in a denitrifying reactor; and studies in soil science showed that resilience depends on both soil structure and soil microbial community composition [37,38]. Resistance of disturbances was observed in several natural systems [39,40]. Given these observations in other systems, it seems the design of reliable biofiltration systems will require better understanding of the relationship among microbial community structure, moisture fluctuations (disturbance), and biofilter performance in an engineered, well-controlled system.

Unlike contaminant gradients [34] or toxic chemical stress [41,42], moisture variation not only influences the microbial communities which eventually determine biofilter performance, but also directly affects absorption of ammonia into biofilters [43]. In other words, moisture may affect biofilter performance in both biological and chemical ways. In this study, moisture content was manipulated for a wide range of 35–63% (wet basis) to discover its roles. Particular microbial communities such as ammonia oxidizers and denitrifiers are responsible for ammonia oxidation and nitrous oxide production. Thus, their communities were examined in this study.

In this investigation, four bench-scale biofilters were constructed to evaluate the effects of moisture on biofilter performance and microbial community structure. The objectives were: (i) to test the influence of moisture content on biofilter ammonia removal and nitrous oxide generation, and to determine the appropriate moisture range for desirable performance; (ii) to explore the response of bacterial communities, ammonia oxidizers and denitrifiers to moisture variance, including community structure shifting and abundance change; and (iii) to link the microbial communities to biofilter function stability.

2. Materials and methods

2.1. Reactor design and operation

The system includes a gas preparation unit, a gas analysis unit, a control unit, and four biofilters (Fig. 1). Anhydrous ammonia (99.99%, S. J. Smith Co., Urbana, IL) regulated by a mass flow controller was mixed with pre-humidified air to provide 70 liter per minute (lpm), 40 ppm ammonia gas for each biofilter (BF). The loading rate was 5.24 $g\text{-}NH_3\ h^{-1}\ m^{-3}$. Air from the blower was 2–3 °C higher than normal room conditions. Therefore, a cooling coil was used to bring the gas closer to room temperature. Four cylindrical biofilters (column ID = 0.45 m, column H = 0.50 m) were made of transparent plastics. A layer of 0.25 m media, 1:1 volume mixture of compost and woodchip, was supported 0.10 m above the bottom of the biofilter tank by a perforated plate. For each biofilter, there were four sampling ports (4 × 90°) located in both upper (0.05 m bellow top surface) and lower (0.05 m above bottom surface) layer. Water was pumped through a coiled micro soaker hose (OD = 0.0064 m) onto the top of the media. Inlet gas was treated by biofilters and then the purified gas left via an outlet port. Empty bed retention time was 34 s. The retention time is high comparing to some other studies, mainly to reduce pressure drop. A control and data acquisition system (National Instruments Co., Austin, TX) was used to record data and control the whole system including the water pump (170DM5, Stenner Pumps & Parts, Indianapolis, IN) and solenoid valves. No extra nutrients or inoculum cultures were added, and a 10-day start-up step was allowed.

The test was composed of four steps. Each step took 22–35 days, depending on operation conditions. To increase moisture content between steps, a certain amount of water was added based on calculation and moisture content measurement; to reduce moisture content between steps 3 and 4, the biofilters were air-dried for several days, during which air stream (no ammonia input) remained unchanged but water addition was stopped; and to maintain moisture content during each step, water was added regularly based