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Flooding-induced N_2O emission bursts controlled by pH and nitrate in agricultural soils

Mette Hansen [a], Tim J. Clough [b], Bo Elberling [c,*]

[a] Department of Geosciences and Natural Resource Management, University of Copenhagen, Øster Voldgade 10, DK-1350 Copenhagen K, Denmark
[b] Agriculture & Life Sciences Division, PO Box 84, Lincoln University, Canterbury, New Zealand
[c] Center for Permafrost (CENPERM), Department of Geosciences and Natural Resource Management, University of Copenhagen, Øster Voldgade 10, DK-1350 Copenhagen K, Denmark

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ABSTRACT

Agricultural soils are a major source of the greenhouse gas nitrous oxide (N_2O) to the atmosphere. Increasing frequency and severity of flooding as predicted for large intensively cropped areas may promote temporary denitrification and N_2O production but the effect of flooding events on N_2O emissions is poorly studied for agricultural systems. The overall N_2O dynamics during flooding of an agricultural soil and the effect of pH and NO_3^- concentration has been investigated based on a combination of the use of microsensors, stable isotope techniques, KCl extractions and modelling. This study shows that non-steady state peak N_2O emission events during flooding might potentially be at least in the order of reported annual mean N_2O emissions, which typically do not include flood induced N_2O emissions, and that more than one-third of the produced N_2O in the soil is not emitted but consumed within the soil. The magnitude of the emissions are, not surprisingly, positively correlated with the soil NO_3^- concentration but also negatively correlated with liming (neutral pH). The redox potential of the soil is found to influence N_2O accumulation as the production and consumption of N_2O occurs in narrow redox windows where the redox range levels are negatively correlated with the pH. This study highlights the potential importance of N_2O bursts associated with flooding and infers that annual N_2O emission estimates for tilled agricultural soils that are temporarily flooded will be underestimated. Furthermore, this study shows that subsurface N_2O reduction is a key process limiting N_2O emission and that a reduction in N_2O emissions is achievable if highly fertilized N-rich soils are limed.

1. Introduction

Future climate change will lead to changes in precipitation worldwide. A higher frequency of extreme rainfall events is predicted for temperate areas such as New Zealand and Northern Europe (IPCC, 2007; Min et al., 2011). This increases the risk of flooding for low-lying or poorly drained areas, which are the same areas receiving run off and ground water with potentially high nitrate (NO_3^-) concentrations. As a consequence it can be expected that there will be an increase in nitrous oxide (N_2O) production and emissions from these areas, particularly fertilized and nitrogen-rich agricultural fields (Knowles, 1982).

Nitrous oxide is a greenhouse gas with a global warming potential relative to CO_2 of 298 on a 100 year time horizon assuming a lifetime of 114 years in the atmosphere (IPCC, 2007). Additionally, N_2O has a negative effect on stratospheric ozone as NO and other free radical species (NO_x), generated from N_2O, deplete the ozone layer (Badr and Probert, 1993). The atmospheric concentration of N_2O has increased since pre-industrial times by 16% from 270 ppb to 319 ppb in 2005 (IPCC, 2007) and it is currently considered the dominant anthropogenic ozone depleting substance emitted (Ravishankara et al., 2009). Soils are the main source of both anthropogenically and naturally produced N_2O and changes in land use have been the primary driver for the observed increase in tropospheric N_2O concentration (IPCC, 2007). Today, agricultural fields account for 42% of the total anthropogenic contribution of N_2O to the atmosphere and N_2O is the single most important greenhouse gas when looking at agricultural soils (IPCC, 2007).

In oxygen (O_2) limited environments production of N_2O in soil occurs as microbial processes utilize nitrogenous compounds as electron acceptors (Knowles, 1982). During denitrification N_2O is an obligate intermediary product in the reduction of NO_3^- to N_2, a process performed by heterotrophic microorganisms. It is also a by-

* Corresponding author. Tel.: +45 35322520.
E-mail addresses: metha@geo.ku.dk (M. Hansen), timothy.clough@lincoln.ac.nz (T.J. Clough), be@geo.ku.dk (B. Elberling).

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product during dissimilatory NO_3^- reduction (DNRA) to ammonium (NH_4^+) as NO_3^- is reduced to NH_4^+ via nitrite (NO_2^-) by fermentative microorganisms (Tiedje et al., 1982). Denitrification rates increase with organic C and NO_3^- availability, soil water content, pH and temperature (Knowles, 1982; Šimek and Cooper, 2002). The N_2O:N_2 ratio, describing the end product of denitrification, shifts in favour of N_2O as soil NO_3^- concentrations and acidity increase (Knowles, 1982; Weier et al., 1993). Not all N_2O produced in a soil will be emitted as it can be consumed during denitrification to N_2 a process controlled by the presence of N_2O reductase (NOS) (Knowles, 1982). Highly anoxic conditions, caused by high soil water content and high availability of easily degradable organic matter, favour the consumption of N_2O (Wrage et al., 2001) as NOS is strongly inhibited by the presence of O_2 (Knowles, 1982). Thus the balance between N_2O consumption and production rates controls N_2O emissions as well as the transport properties of N_2O in the soil (Clough et al., 2005). The primary mode of transport for N_2O in the soil is diffusion, which is controlled by concentration gradients according to Fick's law of diffusion.

The environmental factors for production of N_2O are optimal when fertilized fields are flooded. Non-steady state draining experiments have established the relationship between water-filled pore space and N_2O emissions (Castellano et al., 2010), however, to the authors knowledge, no studies on agricultural soils and only a few studies on natural soils have examined the effect of soil flooding on N_2O dynamics: the temporal and spatial trends of subsurface N_2O concentrations and net surface emissions. Jørgensen and Elberling (2012) found a distinct pulse pattern in N_2O concentrations and emissions when flooding an un-managed wetland peat soil. An increase in N_2O concentrations was observed within the first 24 h followed by a rapid decline in concentration, until the N_2O concentration was below detection after 40 h. It was concluded that for these wetland peat soils the increase in N_2O production would not affect the annual N_2O emission budget, even if flooding event frequency increases in the future (Jørgensen and Elberling, 2012). This may not be the case for agricultural fields, where tillage events can increase the availability of NO_3^-–N (Eriksen and Jensen, 2001; Silgram and Shepherd, 1999) and thereby the potential for N_2O production via denitrification.

The aim of this study was to investigate the overall N_2O dynamics during a flooding event of a New Zealand agricultural soil as affected by soil pH and NO_3^- concentration. Specific aims of the study were to determine the balance between produced, consumed and emitted N_2O from the soil and to determine the depth- and time-specific production and consumption of N_2O. Two methods were used in combination to achieve the aims: depth-specific profiling of the soil N_2O concentration and the redox potential using microsensors as well as 2 M KCl extractions of 3 soil layers per soil core after ^{15}N labelled NO_3^- addition. The study was designed based on the hypothesis that it is possible to mitigate N_2O emissions by changes in agricultural practises (with a focus on changes in soil pH and N-input) and that annual N_2O inventories made to date have potentially been underestimated because the impact of flooding has not been included in annual budgets.

2. Materials and methods

A Templeton silt loam soil (Udic Ustochrept) was collected from a field, with a management history of perennial pasture, from the top layer (0–10 cm deep) during cultivation for pasture renovation, Lincoln, Canterbury (43° 38.720S; 172° 26.753E Lat/Lon). The Canterbury region is temperate with mean annual precipitation of 600–700 mm and a daily mean air temperature range of 1–10 °C in the coldest months and 12–22 °C in the warmest (Cappelen and Jensen, 2001). The Templeton soil and similar inceptisols represents app. 25% of the Canterbury Plains (Molloy, 1988). Inceptisols in temperate areas are soils with high inputs of fertilizer N (Potter et al., 2010) with crop types typically consisting of cereals such as barley and wheat (Leff et al., 2004).

The sampled soil was air-dried, sieved (<2 mm) and kept dry and cold (4 °C). The soil pH was determined (10 g air-dried soil:25 mL water). Half of the soil was treated with 2.08 g $Ca(OH)_2$ kg^{-1} dry soil (quicklime) in powder form to increase the pH by one unit. Lime treatment and the resulting pH increase were made consistently with previous experiments (Clough et al., 2003). Inorganic-N and dissolved organic carbon (DOC) were determined for both the un-treated and the limed soil. Inorganic-N was determined in a 2 M KCl extraction (4 g soil:70 mL KCl, shaken on an end-over-end shaker for 60 min and filtered through Whatman 42 filter paper. Filtered samples were analysed using an Alpkem FS3000 twin channel flow injection analyser (FIA) with Alpkem Winflow 4.03 software). The DOC was analysed by a DI water extraction (1:10 soil:water ratio), shaken on an end-over-end shaker for 30 min followed by centrifuging at 3500 rev/min for 20 min and filtered through a Whatman 42 filter paper into a 30 mL sample vial (Ghani et al., 2003). The DOC was determined based on the difference between the total organic carbon (TOC) and the inorganic carbon (IC) analysed using a Shimadzu Total Organic Carbon Analyser (TOC-5000A) fitted with a Shimadzu ASI-5000A autosampler.

2.1. Core preparations

Soil was packed into either stainless steel metal cores (D = 7.4 cm) for microsensor measurements or PVC plastic cores (D = 7.5 cm) for KCl extractions (see below). Soil core bases were covered with a 1 mm nylon mesh and packed to a depth of 3.5 cm. The soil was packed in layers to ensure an even bulk density of 1 g cm^{-3} throughout the profile. Four treatments were made: control (soil with no additions, **TC**), limed soil (soil plus lime, **TL**), soil with N added (soil plus nitrate-^{15}N, **TN**), and soil with N and lime added (soil plus nitrate-^{15}N and lime, **TLN**). For treatment TN and TLN a known volume of ^{15}N enriched (50 atom%) KNO_3 solution (0.0154 M) was sprayed onto a designated mass of dry soil prior to packing the soil cores, supplying 100 μg NO_3–N g^{-1} soil. Since NO_3^- is evenly distributed in the surface of a cultivated soil, the ^{15}N–NO_3^- was applied to the entire depth of soil in the packed soil core. Soil cores were packed and adjusted with KNO_3 immediately before flooding. The soil cores were then flooded from below immediately prior to commencement of microsensor measurements, to mimic the rise of a high groundwater table, by placing them in a water-filled box. This method of soil flooding also minimised soil drainage during wetting. In total 108 cores were made, of which 12 were used for microsensor measurements and 96 for KCl extractions. KCl extractions were performed on three replicates at eight time steps for each treatment. The timing of the KCl extraction was distributed throughout the pulse of N_2O production (see supporting information (SI) (Fig. S1)). Due to measurement constraints of the microsensor, replication was done in time by sequentially measuring one soil core from one treatment at any given time. For each treatment a total of 3 replicates were measured at t_1, t_2 and t_3. In practice, two replicates of each treatment (at t_1 and t_2) were measured after each other. All t_1 and t_2 measurements for all four treatments were finished within 50 days. All t_3 measurements were subsequently measured after this period (see SI Fig. S2 for exact specifications of the timing of t_1, t_2 and t_3 for all treatments). Each soil core was followed until no more N_2O evolved (up till 7 days) before the next soil core was measured, hence only one soil core for microsensor measurements was flooded at a time. Soil remained sieved but unpacked at 4 °C and unamended with ^{15}N, with these procedures performed prior to microsensor measurements starting.