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N$_2$O, NO, N$_2$, and CO$_2$ emissions from tropical savanna and grassland of Northern Australia: an incubation experiment with intact soil cores

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N$_2$O, NO, N$_2$, and CO$_2$ emissions from tropical savanna soils and grassland

C. Werner et al.

Title Page

Abstract	Introduction
Conclusions	References
Tables	Figures

◀◀ ▶▶

◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Abstract

Strong seasonal variability of hygric and thermal soil conditions are a defining environmental feature in Northern Australia. However, how such changes affect the soil–atmosphere exchange of nitrous oxide (N$_2$O), nitric oxide (NO) and dinitrogen (N$_2$) is
5 still not well explored. By incubating intact soil cores from four sites (3 savanna, 1 pasture) under controlled soil temperatures (ST) and soil moisture (SM) we investigated the release of the trace gas fluxes of N$_2$O, NO and carbon dioxide (CO$_2$). Furthermore, the release of N$_2$ due to denitrification was measured using the helium gas flow soil core technique. Under dry pre-incubation conditions NO and N$_2$O emission were
10 very low ($< 7.0 \pm 5.0\,\mu g\,NO\text{-}N\,m^{-2}\,h^{-1}$; $< 0.0 \pm 1.4\,\mu g\,N_2O\text{-}N\,m^{-2}\,h^{-1}$) or in case of N$_2$O, even a net soil uptake was observed. Substantial NO (max: $306.5\,\mu g\,N\,m^{-2}\,h^{-1}$) and relatively small N$_2$O pulse emissions (max: $5.8 \pm 5.0\,\mu g\,N\,m^{-2}\,h^{-1}$) were recorded following soil wetting, but these pulses were short-lived, lasting only up to 3 days. The total atmospheric loss of nitrogen was dominated by N$_2$ emissions (82.4–99.3 % of to-
15 tal N lost), although NO emissions contributed almost 43.2 % at 50 % SM and 30 °C ST. N$_2$O emissions were systematically higher for 3 of 12 sample locations, which indicates substantial spatial variability at site level, but on average soils acted as weak N$_2$O sources or even sinks. Emissions were controlled by SM and ST for N$_2$O and CO$_2$, ST and pH for NO, and SM and pH for N$_2$.

1 Introduction

Tropical savanna ecosystems are one of the most important biomes of the earth and cover approximately 27.6 million km^2 (Hutley and Setterfield, 2008) or 11.5 % of the global surface (Scholes and Hall, 1996). However, a clear, globally applicable, delineation is often difficult and thus all attempts on using those area estimate to scale
25 regional measurements to biome-scale will result in significant uncertainties (Davidson and Kingerlee, 1997). On a continental scale these areas can be constraint much bet-

ter. North Australian tropical savannas cover about 2 million km^2 or approximately 12 % of the global extent of the biome and are located north of 20° S in Western Australia, the northern part of the Northern Territory and northern Queensland (e.g., Lehmann et al., 2009; Williams et al., 2005). Improved pastures were introduced in Northern Australia since the 1880s to increase productivity and their area expanded by approximately 2500 km^{-2} yr^{-1} during the 1980s (Lonsdale, 1994).

Savanna systems experience pronounced intra-annual variability of rainfall (dry season conditions from 2 to 9 months) in addition to pronounced inter-annual climatic fluctuations (Frost et al., 1986). In the tropical savanna zone of Australia the wet season is controlled by the Asian monsoon and occurs from October to March. Precipitation ranges from over 2000 mm yr^{-1} at coastal areas to < 400 mm yr^{-1} inland (e.g., Russell-Smith and Yates, 2007) and declines by approximately 1 mm yr^{-1} per km inland (Cook and Heerdegen, 2001). Another key feature of savanna is a high fire frequency (∼ 20 % burnt annually, Dwyer et al., 2000). Tropical savanna of the coastal belt of Northern Australia was found to burn even more frequently (up to 35 %, Russell-Smith et al., 2007).

Climatic conditions as well as fire activity control soil conditions and nutrient turnover in these ecosystems as they affect soil temperature, soil moisture levels and thus oxygen availability in the soil matrix and availability and structure of organic matter and inorganic nutrients. Biogenic N trace gases are produced in soils through the microbial processes of nitrification and denitrification (Conrad, 1996), two key processes of N cycling in terrestrial ecosystems. The mineralization of organic material leads to the formation of ammonium (NH$_4^+$), which serves as a substrate for autotrophic and/or heterotrophic nitrification. Under aerobic conditions, autotrophic and/or heterotrophic nitrifying organisms oxidise NH$_4^+$ via hydroxylamine (NH$_2$OH) and nitrite (NO$_2$) to nitrate (NO$_3^-$). Anaerobic conditions in the soil or in micro-sites promote the denitrification process, using NO$_3^-$/NO$_2$ as alternative electron acceptors. Depending on environmental conditions, NO$_3^-$ is reduced stepwise via NO$_2$, nitric oxide (NO) and N$_2$O to inert nitrogen (N$_2$). N$_2$O as well as NO emissions from soils depend on the rates of nitrification

Full Screen / Esc

and denitrification and underlying environmental conditions. Both processes can occur simultaneously in the soil due to the heterogeneity of soils with regard to gas diffusion and metabolic activity, which can create anaerobic micro-sites even in well aerated soil (Granli and Bøckman, 1995). The primary controls of N$_2$O as well as NO production and emission are the availability of NH$_4^+$ and NO$_3^-$, the content of decomposable organic substrate and the soil aeration status, whereas soil temperature and pH are secondary controls (Granli and Bøckman, 1995). The significant variability of environmental factors (soil, vegetation and hydrological properties) across space and time (e.g., Davidson, 1991; Papen and Butterbach-Bahl, 1999) has often been demonstrated to control the magnitude and variability of N$_2$O and NO emissions from soils. Soil water status mainly regulating soil aeration conditions was found to act as the main control for N$_2$O emissions in most tropical soils (e.g., Butterbach-Bahl et al., 2004b; Davidson, 1991; Grover et al., 2012; Linn and Doran, 1984; Werner et al., 2007b), although, due to the complexity of interacting environmental controls, it is difficult to obtain relationships of predictive value (Andersson et al., 2003).

Savanna regions are characterized by climates with distinct wet and dry seasons so that biogeochemical processes and associated trace gas exchange can be expected to also exhibit strong seasonal patterns following the hygric regime (e.g., Andersson et al., 2003; Eamus and Prior, 2001). Such pronounced emission characteristics were also previously reported for seasonal tropical rainforest ecosystems (Breuer et al., 2000; Butterbach-Bahl et al., 2004b; Werner et al., 2007b, 2006). Fire is another characteristic forcing in all savanna ecosystems (Bond and Keeley, 2005). In addition to direct pyrogenic emissions (loss of trace elements to the atmosphere during combustion), fire also effects physicochemical and biological processes, and can thus have a significant direct or indirect effect on processes and controls of N cycling and N$_2$O/NO soil–atmosphere exchange (Bustamante et al., 2006; Levine et al., 1996; Rondón et al., 1993; Serça et al., 1998; Weitz et al., 1998). For the reasons cited above, most datasets of N trace gas emissions from savanna soils show a substantial variation of N$_2$O and NO emissions on a seasonal scale following the hygric conditions (e.g., Castaldi et al.,

2004, 2006; Pinto et al., 2002), whereas on a site scale emissions are mostly controlled by the variability of soil properties (Castaldi et al., 2004, 2006).

There is some literature available investigating the soil–atmosphere exchange of NO, N_2O and CO_2, however, information of N_2 losses via microbial denitrification is not available for savanna systems and also still scarce for other terrestrial ecosystems (Butterbach-Bahl et al., 2013; Dannenmann et al., 2011; Groffman et al., 2006).

In order to investigate gaseous emissions of C (CO_2) and N (N_2O, NO, N_2) compounds from tropical savanna and grassland systems in Northern Australia, we took intact soil cores from various field sites and performed a range of incubation experiments under controlled thermal and hygric conditions. The scope of the study is (a) to assess the magnitude of trace gas release under a range of environmental conditions typical for tropical savanna systems of Northern Australia, (b) to deduce environmental regulators leading to the observed emissions, and (c) to contrast the findings from natural savanna soil cores to samples derived from an improved pasture site.

2   Material and methods

2.1   Site description and soil core sampling

In this study we investigated two field sites approximately 30 km east of Darwin, Northern Territory, Australia, and extracted intact soil cores for our laboratory incubation experiments. The first site is located at 12°30′ S, 131°05′ E at 18–22 m a.s.l., Howard Springs (HS). Vegetation was *Eucalyptus tetrodonta* and *Eucalyptus minanta* dominated open-forest savanna. The second site was established on managed grassland at Beatrice Hill Farm, Middle Point, located at 12°36.5′ S, 131°18′ E, approximately 25 km south-east of the savanna site.

This part of the Australian north is characterized by a strong seasonality of precipitation, with a pronounced dry season ranging from May to September. The dry season is followed by a period of intense convective thunderstorms ("build-up"), which

marks the start of the wet season (Cook and Heerdegen, 2001). The long-term annual mean of precipitation for the closest climate station Middle Point is 1411 mm (data: http://www.bom.gov.au, years 1981–2010; see Fig. 1 for a Walter & Lieth Climatic Diagram of this station). The dominant amount of annual precipitation (91 %) is received in the wet season, often as heavy afternoon thunderstorms. Due to the strong rainfall intensities, a substantial amount of this water does not penetrate the soil and is transported away via overland flow. The inter-annual variability of wet season intensity and total rainfall is quite substantially, as it is linked to large-scale circulation systems of the Southern Hemisphere. The annual average temperature is 27 °C and the seasonal temperature variability is less pronounced with mean daily temperature of 23.1 °C during the dry season and up to 29.4 °C (monthly means) during the "build-up". The daily temperature amplitude varies much stronger with a maximum daily amplitude in July/August and a minimum in February.

At site one (HS, savanna site), a transect of sampling positions was established (Transect 1, T1), stretching approximately 500 m slightly down-slope into a small topographical depression (altitudinal difference less than 4 m over the transect distance). At five positions, spaced approximately 100 m, sampling positions were established. Soil cores were extracted at positions P1, P3, P5 (positions P2 and P4 were not used for this analysis, but to ensure consistent naming, the original position identifiers are used). P1 was located at the upper-most position of the topographic gradient, P3 250 m down-slope, and P5 at the lowest position. The individual sampling positions are referenced as T1P1, T1P3, and T1P5 in the manuscript. The tree layer of this tropical savanna site is dominated by *Eucalyptus tetrodonta* and *Eucalyptus minanta*, while the dominant grasses are Shorgum spp. and *Heteropogon contortus*. Typical fire regime of this vegetation type is 2 in 3 years (Hutley et al., 2011). The site was located in a peri-urban environment where fire management reduced fire frequency (a general phenomena observed for settlements in this region, Elliott et al., 2009) and woody cover was ∼ 10 to 20 % higher at this site than the extensive surrounding savanna that is burnt more frequently. Managed fires are usually lit frequently (fire return interval ap-

proximately 2–3 years) as low-intensity fires early in the dry season reducing the risk of higher intensity late dry season fires (P3, P5). The site P1 was privately owned and was not burned for more than 6 years (D. Georges, personal communication, 2010). Tree cover was highest at P1, while the landscape was more open towards the center of the transect. Tree cover increased again at P5 due to higher water availability at the bottom of the slope (cover approx. 40–50 %).

The improved pasture at site two was established in the 1960s by clearing savanna vegetation and ploughing and seeding of Pangola grass (*Digitaria eriantha*). The area was fertilized annually or every second year with approximately 50 kg phosphorus, 100 kg superphosphate and 100 kg urea in single fertilization events. Buffalo graze the improved pasture during the wet season and for 3–4 weeks during the dry season (grazing intensity: one beast per ha), and a herd grazed the site until the soil cores were extracted. The site is referenced as T3P1 throughout the manuscript.

At each position 12 soil cores were extracted for the incubation experiments under dry season conditions in 2010 and air dried immediately. For this, plastic cylinders (height: 20 cm, diameter: 14 cm) were driven into the soil and the surrounding soil removed. The cores were then carefully lifted from the ground and the bottom of the cylinder sealed with appropriate pipe caps, packed and sent to the laboratory at IMK-IFU, Karlsruhe Institute of Technology (Garmisch-Partenkirchen, Germany) for analysis. The extracted soil cores were divided into three groups of four cores for each sample position. The three groups were used for independent analysis with different moisture levels (water filled pore space 25 %, 50 %, 75 %; see section "incubation setup" for details) and each analysis was replicated with three cores. One soil core was kept as a spare for each position.

2.2 Soil analysis

Soil organic carbon, total nitrogen, soil texture, and soil pH was analyzed by a commercial laboratory for all sampling positions (Dr. Janssen, Gillersheim, Germany). Microbial C and N was measured using the chloroform fumigation-extraction technique

(Brookes et al., 1985). We sampled and sieved soil from cores of each field position. Three sub-samples were immediately extracted with 60 mL 0.5 M K_2SO_4, while another three samples were fumigated with chloroform vapor for 24 h and extracted similarly. Total organic carbon (TOC) and total chemically bound nitrogen (TNb) was measured using a TOC analyzer (DIMATOC 2000, Dimatec Analysentechnik GmbH, Essen, Germany) and a coupled chemoluminescence detector, respectively. See Dannenmann et al. (2006) for more details.

2.3 Incubation setup and detection of N_2O, NO, N_2 and CO_2

In order to investigate the C and N trace gas exchange of the soil samples under a variety of environmental controls, we designed an incubation experiment. Soil water content of the stored cores was determined prior to the experiment gravimetrically by drying samples derived from the spare replicate core. The samples were dried at 105 °C for 24 h. Bulk density of the samples was also measured and used to calculate the pore volume and consequently the amount of water required to reach 25, 50 and 75 % water filled pore space (WFPS).

$$WFPS[\%] = \frac{W_{vol}}{\left(1 - \frac{BD}{2.65}\right)}$$

W_{vol} volumetric water content,

BD   bulk density [$g\,cm^{-3}$],

2.65   particle density [$g\,cm^{-3}$].

The cores were first exposed to 20 °C temperature for pre-incubation before the air-dried samples were wetted to the desired soil moisture levels of 25 % WFPS (M25), 50 % WFPS (M50), and 75 % WFPS (M75), respectively: at these points the weight of the watered cores was recorded. To avoid a bias of follow-up measurements, all three

soil moisture levels were analyzed with independent cores. The moisture content was adjusted bi-daily by weighing the cores and adding evaporated water at least one hour prior to the measurements. Note that we report average trace gas fluxes for the second segment of each incubation step in order to exclude transition effects from raising temperatures. Soil cores were sealed with gas-permeable foil immediately after the sampling procedure to minimize the evaporative loss of water. During the course of the incubation period, the temperature was step-wise increased from 20 to 30 and to 40 °C. In-between measurement events, all soil cores were incubated in thermostat cabinets (Lovibond ET 651-8, Tintometer GmbH, Dortmund, Germany) at the prescribed incubation temperature. The cabinets were ventilated with low-flow pumps to prevent CO$_2$ enrichment or temperature gradients.

The air-dried soil cores were measured for up to 10 days prior to the initial wetting at each incubation run to record the baseline emission and to test the detection setup. N$_2$O, NO and CO$_2$ emissions were measured approximately every second to third day. After the initial wetting, soil cores were incubated and measured until the initial "pulse-event" leveled off and flux rates approached a relatively stable emission level. Afterwards, the temperature was raised to 30 °C and measurements continued. After 2–3 weeks of incubation at 30 °C, the temperature was increased again by 10 °C to 40 °C. A schematic of the incubation and measurement schedule is given in Fig. 2.

The static chamber technique (e.g, Smith et al., 1995) was used to measure N$_2$O and CO$_2$ fluxes. The soil cores were closed with gas-tight lids equipped with a port and septum for gas sampling. The headspace of each of the 12 soil cores (T1P1, T1P3, T1P5, and T3P1; $n = 3$) was determined individually (average headspace volume 1.54 L). Four gas samples of 5 mL chamber air were drawn from the port at 0, 20, 40, and 60 min after closure using 20 mL plastic syringes fitted with three-way stopcocks and injected into vacuumized sampling veils. The sample veils and a set of veils with standard gas mixture for calibration (0.4 ppmv N$_2$O, 400 ppmv CO$_2$; Air Liquide, Düsseldorf, Germany) were placed into two auto-sampler systems (HT280T, HTA s.r.l., Brescia, Italy) attached to gas chromatographs (SRI 8610C, SRI Instruments, Torrance,

CA, USA) systems equipped with a ^{63}Ni electron detector (ECD) for N_2O analysis and an flame ionization detector and methanizer for CO_2 analysis. We used high-purity N_2 as carrier gas and installed ascarite filter to exclude any interference of CO_2 on N_2O detection. Flux rates of N_2O and CO_2 were calculated from the observed linear

5 increases of sampled chamber air using linear regression (e.g., Papen and Butterbach-Bahl, 1999).

Nitric oxide emissions were measured immediately after sampling for N_2O/CO_2 analysis using a dynamic chamber technique (e.g., Yamulki et al., 1997) and chemiluminescence detector (CLD 770 AL ppt, Ecophysics AG, Durnten, Switzerland). A sealing

10 lid with inlet and outlet fittings was attached to the soil core, and synthetic air (79 % N_2, 21 % O_2) was pumped through the headspace. The NO flux was calculated from the inlet/outlet NO concentrations in a dynamic equilibrium with constant gas flow rate. Steady-state conditions were reached approximately after 20–25 min, and the average detected flow rate over 5 min after this grace period was used to determine the

15 flux. The detector was periodically calibrated in the laboratory with standard gas and a calibration unit (for details see, Butterbach-Bahl et al., 1997).

Inert nitrogen (dinitrogen, N_2), the final product of the microbial transformation process "denitrification", is abundant in the atmosphere and thus, measurements of N_2 production of soil have to be carried out with great sophistication and care. They are

20 commonly only available from laboratory measurements under controlled atmosphere, as the amounts of emitted N_2 collected in the sampling headspace are multiple magnitudes lower than concentrations in ambient air. N_2 emission was measured after the end of the incubation experiments using the helium gas flow soil core method (Butterbach-Bahl et al., 2002).

25 Here, we used the custom-build system developed for soil core incubation described in detail in Dannenmann et al. (2011). The system contains two vessels for N_2 and N_2O emissions measurements. Every vessel contains seven soil cores of 4 cm height and 100 cm^3 volume each, which are incubated together in order to account for soil spatial heterogeneity. Prior to flux measurements, the soil and headspace atmosphere

N_2O, NO, N_2, and CO_2 emissions from tropical savanna soils and grassland

C. Werner et al.

Title Page

Abstract	Introduction
Conclusions	References
Tables	Figures

|◄ | ►|

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

in the vessels is replaced by an artificial gas mixture (20 ppm N_2, 400 ppb N_2 and 20 % O_2 in helium) by two days of gas flushing from bottom to top through the soil cores. The required extreme gas tightness of the vessels against intrusion of atmospheric N_2 is ensured by double sealings purged with helium, and by placing the incubation vessels with its tubing connections under water (Dannenmann et al., 2011). After the soil and headspace flushing period, N_2 and N_2O concentrations are measured hourly in the headspace over a period of 8 h in order to allow for calculation of N_2 and N_2O gas fluxes (Butterbach-Bahl et al., 2002). Similar to previous use of the system (Dannenmann et al., 2011), regular measurements with empty vessels showed no increase of N_2 in the system over the measurement period. In order to measure N_2 emissions immediately after the C and N gas incubation experiments, the three replicated soil cores of one incubation measurement were destructively subsampled to gain seven soil cores for the helium soil core incubation vessel. Subsequent N gas flux measurements were conducted for the defined incubation moisture level and a series of soil temperature settings (20 °C, 30 °C, and 40 °C).

2.4 Data analysis and presentation

All analysis and plotting was carried out using R 2.15 (R Development Core Team, 2011). Differences between measured gas fluxes at the various incubation settings and tests for comparable air-dried soil conditions were investigated using a multiple comparison of means procedure (Herberich et al., 2010) as group sizes and distributions varied substantially in the comparison. Relative importance of environmental factors controlling trace gas emissions was assessed using multiple linear regression analysis and a tool for variance decomposition (proportional marginal variance decomposition, R package "relaimpo", see Grömping, 2007). Statistical significance is given at the 95 % difference level ($p \leq 0.05$), and error bars denote standard deviation if not specified otherwise.

3 Results

3.1 Soil analysis

Physico-chemical properties of the sampled soil cores are given in Table 1. All cores have a very sandy texture composition (T1 > 80 %, T3 = 68 %), but the pasture site T3 has a notably higher fraction of clay (15 %). Bulk density was highest at position T1P5 (1.7 g cm^{-3}) and in a range of 1.4 to 1.5 g cm^{-3} for the other soil cores. Soil pH was ranging from 4.4 to 5.1. With 0.8 and 0.7 %, total organic carbon was very low for sites T1P3 and T1P5, respectively, higher at the grassland site T3P1 (1.5 %) and highest for site T1P1 (2.8 %). Total nitrogen concentrations were also low and ranged between 0.04 % and 0.1 %. The C/N ratios were also widest for site T1P1 (24.1) and most narrow for site T1P5 (15.5). With 222.1 µg C g^{-1} soil dry weight (sdw) microbial carbon and 35.7 µg N g^{-1} sdw microbial nitrogen, the samples from the improved pasture site were approximately 2.3–3.6 times (carbon) and 4.7–7.8 times (nitrogen) richer, respectively, in microbial nitrogen than the samples from natural savanna (T1). Ammonium and nitrate concentrations were low for all sample sites and ranged from 1.5–2.8 and 0.04–0.1 µg N g^{-1} sdw, respectively (both nutrient concentrations were highest at locations T1P1 and T3).

3.2 Gas fluxes

3.2.1 CO_2 emissions

The CO_2 baseline emission during pre-incubation (unwatered, 20 °C) was 0.1 ± 2.7 mg C m^{-2} h^{-1} (T1) and 1.0 ± 3.8 mg C m^{-2} h^{-1} (T3) and not significantly different between transect positions or transects. Immediately after watering the soil cores, CO_2 emissions rose for all three watering levels (M25, M50, and M75) and all replicates (see Fig. 3). Peak emissions generally occurred immediately after watering (day 0 of incubation section: D0), and emissions at the following day (D+1) dropped to 70 % (M25),

38 % (M50), and 37 % (M75) of the initial pulse emission response recorded at D0. After two days (D+2), CO_2 emissions declined further to levels of 26–34 % before stabilizing at those levels and eventually increasing again (see Fig. 3). During the pulse emission event (D0−D+2), the average CO_2 emission was $62.8 \pm 43.7\,\mathrm{mg\,C\,m^{-2}\,h^{-1}}$ (min: $7.7\,\mathrm{mg\,C\,m^{-2}\,h^{-1}}$, max: $195.1\,\mathrm{mg\,C\,m^{-2}\,h^{-1}}$). Pulse intensity was found to differ significantly between natural savanna soil cores of transect T1 ($55.5 \pm 39.8\,\mathrm{mg\,C\,m^{-2}\,h^{-1}}$) and improved pasture at transect T3 ($75.1 \pm 46.4\,\mathrm{mg\,C\,m^{-2}\,h^{-1}}$), but not between individual transect positions. After the initial pulse event, CO_2 emissions remained relatively stable for all three moisture levels at 20 °C soil temperature (Fig. 3).

Average CO_2 emission at 20 °C (after the pulse event) were $19.2 \pm 6.4\,\mathrm{mg\,C\,m^{-2}\,h^{-1}}$ (M25), $23.1 \pm 6.9\,\mathrm{mg\,C\,m^{-2}\,h^{-1}}$ (M50), and $36.2 \pm 18.5\,\mathrm{mg\,C\,m^{-2}\,h^{-1}}$ (M75), indicating a direct link between soil moisture and CO_2 emission strength. Fluxes recorded at M75 were significantly higher than those obtained at M25 and M50 ($p < 0.001$). In contrast to the pulse emission, CO_2 emissions from T3 ($30.5 \pm 16.2\,\mathrm{mg\,C\,m^{-2}\,h^{-1}}$) were not significantly higher than those from T1 ($25.2 \pm 13.9\,\mathrm{mg\,C\,m^{-2}\,h^{-1}}$). No significant difference between individual transect positions was detected. It is noteworthy that CO_2 emissions for some individual cores (T1P1R1, T1P3R1) were substantially higher for M75 conditions than those from the other two replicates, resulting in high standard deviations. At 30 °C soil temperature, average CO_2 emissions were again not found to differ significantly between T1 ($36.0 \pm 17.2\,\mathrm{mg\,C\,m^{-2}\,h^{-1}}$) and T3 ($43.3 \pm 20.8\,\mathrm{mg\,C\,m^{-2}\,h^{-1}}$, $n = 45$), but moisture levels were again significantly correlated with higher emissions (M25: $34.1 \pm 14.5\,\mathrm{mg\,C\,m^{-2}\,h^{-1}}$, M50: $35.6 \pm 20.4\,\mathrm{mg\,C\,m^{-2}\,h^{-1}}$, M75: $44.0 \pm 18.6\,\mathrm{mg\,C\,m^{-2}\,h^{-1}}$). No significant differences were recorded for CO_2 fluxes from cores originating from different transect positions, indicating that physico-chemical soil properties did not have a major influence on CO_2 emissions. Since no data is available from the 40 °C incubation temperature run with M25 cores, we do not report results for transect positions or transects, as the higher moisture levels (M50 and M75 only) lead to biased mean fluxes due to the strong response of CO_2 fluxes to moisture levels. See the discussion section for an in-depth analysis of temperature effects on emissions.

3.2.2 N₂O emissions

Nitrous oxide emissions under pre-incubation conditions were low and did not differ significantly between transect positions or transects (T1: $0.0 \pm 1.4\,\mu\text{g}\,\text{N}\,\text{m}^{-2}\,\text{h}^{-1}$, T3: $-0.2 \pm 1.9\,\mu\text{g}\,\text{N}\,\text{m}^{-2}\,\text{h}^{-1}$). We measured low N₂O uptake or zero N₂O emissions for 8 out of 12 cores under these conditions (min: $-6.2\,\mu\text{g}\,\text{N}\,\text{m}^{-2}\,\text{h}^{-1}$), but variance ranged from 0.3 to $3.6\,\mu\text{g}\,\text{N}\,\text{m}^{-2}\,\text{h}^{-1}$. Increased N₂O emissions were detected immediately after watering the cores (D0), but average emissions during the defined peak event (D0 – D+2) were relatively modest for 33 out of 36 cores sampled at T1 (avg: $3.2 \pm 3.1\,\mu\text{g}\,\text{N}\,\text{m}^{-2}\,\text{h}^{-1}$, max: $13.7\,\mu\text{g}\,\text{N}\,\text{m}^{-2}\,\text{h}^{-1}$) and T3 (avg: $4.9 \pm 6.2\,\mu\text{g}\,\text{N}\,\text{m}^{-2}\,\text{h}^{-1}$, max: $24.2\,\mu\text{g}\,\text{N}\,\text{m}^{-2}\,\text{h}^{-1}$), but the results for T1 and T3 were not significantly different. Very strong N₂O emissions were recorded for three individual cores at M75 (T1P1R1: $85.8\,\mu\text{g}\,\text{N}\,\text{m}^{-2}\,\text{h}^{-1}$, T1P1R2: $106.2\,\mu\text{g}\,\text{N}\,\text{m}^{-2}\,\text{h}^{-1}$, T1P3R1: $180.2\,\mu\text{g}\,\text{N}\,\text{m}^{-2}\,\text{h}^{-1}$) after a delay of two or six days and remained systematically higher throughout the incubation experiment (see Fig. 4). Due to their distinctly different emission patterns, those three cores are excluded from further analysis here and discussed in detail at the end of this section. Intensity of pulse emissions correlated positively with saturation levels (M25: $2.7 \pm 3.2\,\mu\text{g}\,\text{N}\,\text{m}^{-2}\,\text{h}^{-1}$, M50: $3.2 \pm 4.1\,\mu\text{g}\,\text{N}\,\text{m}^{-2}\,\text{h}^{-1}$, M75: $5.8 \pm 5.0\,\mu\text{g}\,\text{N}\,\text{m}^{-2}\,\text{h}^{-1}$). After the initial pulse, N₂O emissions at 20 °C were $1.2 \pm 3.8\,\mu\text{g}\,\text{N}\,\text{m}^{-2}\,\text{h}^{-1}$ (T1) and $0.6 \pm 1.0\,\mu\text{g}\,\text{N}\,\text{m}^{-2}\,\text{h}^{-1}$ (T3), and no significant differences were detected for transect positions or transects. At 30 °C, N₂O emissions increased slightly to $1.4 \pm 1.8\,\mu\text{g}\,\text{N}\,\text{m}^{-2}\,\text{h}^{-1}$ (T1) and $2.6 \pm 5.1\,\mu\text{g}\,\text{N}\,\text{m}^{-2}\,\text{h}^{-1}$ (T3). At 40 °C, N₂O emission substantially increased over the results obtained at 30 °C, but due to missing results for the M25 incubation run, we do not report site averages here.

3.2.3 NO emissions

Pre-incubation emissions were $6.7 \pm 4.3\,\mu\text{g}\,\text{N}\,\text{m}^{-2}\,\text{h}^{-1}$ (T1) and $7.0 \pm 5.0\,\mu\text{g}\,\text{N}\,\text{m}^{-2}\,\text{h}^{-1}$ (T3), and no significant differences were detected between the transect positions or

transects. Large NO emission pulses of 27.7–$306.5\,\mu g\,N\,m^{-2}\,h^{-1}$ were recorded after the addition of water at D0 (see Fig. 5). Furthermore, NO emission strength was negatively correlated with water saturation (M25: $141.2 \pm 70.9\,\mu g\,N\,m^{-2}\,h^{-1}$, M50: $48.4 \pm 26.6\,\mu g\,N\,m^{-2}\,h^{-1}$, M75: $22.1 \pm 15.2\,\mu g\,N\,m^{-2}\,h^{-1}$), and pulse strength did not significantly differ between transects. NO emissions at the following day (D+1) dropped sharply to 16 % (M25), 40 % (M50), and 28 % (M75) of the initial pulse emission response at D0. After two days (D+2), NO emissions further decreased to 10–26 % of the emission of D0. After the initial pulse, average NO emissions at 20 °C were below pre-incubation levels and ranged from 3.2 ± 1.8 to $5.8 \pm 2.9\,\mu g\,N\,m^{-2}\,h^{-1}$ for the four transect positions being $4.9 \pm 2.7\,\mu g\,N\,m^{-2}\,h^{-1}$ at T1 and $3.2 \pm 1.8\,\mu g\,N\,m^{-2}\,h^{-1}$ at T3 (differences not significant). NO emissions increased at 30 °C incubation temperature to $6.7 \pm 4.3\,\mu g\,N\,m^{-2}\,h^{-1}$ (T1) and $6.5 \pm 6.9\,\mu g\,N\,m^{-2}\,h^{-1}$ (T3), respectively. At 40 °C, a marked increase of NO emission was observed for the majority of soil cores watered to 50 % WFPS, while NO emissions at 75 % WFPS only increased slightly or remained constant (see Fig. 5). Again, as no M25 measurements are available, we do not give site averages for 40 °C, but the significant increase of NO emissions can be highlighted when M50 and M75 NO emissions obtained at 40 °C are compared to the fluxes measured under 30 °C. NO emissions under medium moisture levels (M50) increased from $6.7 \pm 5.1\,\mu g\,N\,m^{-2}\,h^{-1}$ at 30 °C to $52 \pm 87.5\,\mu g\,N\,m^{-2}\,h^{-1}$, while high moisture levels (M75) only led to an increase from $5.0 \pm 4.9\,\mu g\,N\,m^{-2}\,h^{-1}$ to $11.5 \pm 13.7\,\mu g\,N\,m^{-2}\,h^{-1}$. A negative correlation between moisture levels and NO emission strength was observed for all episodes of the incubation experiment. We did not detect significant differences between both land-use types for temperatures up to 30 °C, but at 40 °C, NO emission was significantly higher at the savanna transect than at the improved pasture.

3.2.4 N$_2$ emissions

An incubation setup with the same number of replications as performed for N$_2$O, NO and CO$_2$ measurements was not feasible due to the time required to perform

a single N_2 measurement (see methods' section). We thus report results as transect averages (T1, $n = 3$) and single measurement incubation results (T3). However, in each measurement cycle, soil from all replicate cores was pooled in the sampling cuvette and thus, the results are based on multi-core soil conditions (in case of T3 without true measurement replication). Dinitrogen emissions varied between 16.7 and 2350.4 $\mu g\, N\, m^{-2}\, h^{-1}$. As shown in Table 2, N_2 emissions were strongly influenced by water saturation. Average N_2 emissions did increase from M50 to M75 by a factor of 11 to 16 for savanna soil (T1), whereas emission strength did not differ between M25 and M50. Although no true replication is available for T3 results, a similar pattern can be observed, but we do not contrast results from the two transects due to differences in sample numbers.

In general, N_2 emissions dominate the gaseous nitrogen losses at T1 (see Table 3). While N_2O emissions account only for 0.3–3.6 % of gaseous N losses, N_2 emissions represent up to 99.3 %. Note that inclusion of N_2O outlier emissions reduced the share of N_2 losses only by one percent (Table 3). NO emissions contributed to 5.6–16.5 % at low to medium soil saturations and 20 to 30 °C soil temperatures. Only at 40 °C and medium soil saturation (M50), NO emissions did account for a substantial fraction of total N losses to the atmosphere (43.2 %). At high water saturation, NO emissions were insignificant and contributed only 0.3–1.1 %.

4 Discussion

4.1 CO_2 emissions

Decomposition of organic matter and root respiration lead to the emission of CO_2 from the soil, but since the soil cores were removed from the field and any existing fine roots were cut, the observed fluxes represent only soil respiration CO_2 efflux. Even though our major focus was on the emission of nitrogen gases, CO_2 emissions can

also be useful as a proxy to identify the relative availability/production of substrate for the microbial processes responsible for N_2O, NO and N_2 losses.

Carbon dioxide emissions increased with rising temperature and high moisture levels (Table 2), and soil temperature was identified to have the highest relative importance, followed by soil moisture variations, in explaining the observed CO_2 emissions (Fig. 6). The strong temperature dependency of decomposition and associated CO_2 emissions, given a certain moisture level, is well known in the literature and was reported for many ecosystems (Raich and Schlesinger, 1992). In general, CO_2 emissions from tropical savanna in Northern Australia were found to increase through the seasons (dry, wet-dry transition, wet), but due to relatively minor seasonal variations in temperature, changes in soil moisture are considered to be the dominant control in these ecosystems (Grover et al., 2012). This was also reported for African savannas (Brümmer et al., 2009a) and the wet-dry tropics of the Amazon Basin (Davidson et al., 2000). Strong break-of-season (pulse) emissions ranging between 100–250 mg C m^{-2} h^{-1} were previously reported for natural savanna in Africa (Brümmer et al., 2009a) and are in-line with our observations. We found no significant differences in average CO_2 emissions between pasture cores and cores from the natural savanna site.

4.2 N_2O emissions

Measurements of gaseous nitrogen losses from tropical savanna are still scarce in the literature (e.g., Castaldi et al., 2006; Dalal and Allen, 2008; Stehfest and Bouwman, 2006; Steinkamp and Lawrence, 2011) and to our knowledge no previous study investigated the emission of N_2O, NO and N_2 simultaneously. Annual N_2O emission from tropical savanna systems were reported to range from 0.06 to 1.46 kg N yr^{-1} (mean: 0.6 kg N yr^{-1}) and to be lower for Australian savannas (0.06–1.08 kg N yr^{-1}, median: 0.19 kg N yr^{-1}) due to drier conditions and smaller amounts of N deposited from the atmosphere (Dalal and Allen, 2008). From previous in-situ measurements at an area close to transect T1, it was calculated that soils only acted as minor sinks or sources for N_2O annually (-0.015 kg N ha^{-1} yr^{-1}, Livesley et al., 2011), a result also found to be

true for savanna soils of the Daly River south of our site, which receives substantially less annual precipitation ($0.02\,\mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$, Grover et al., 2012).

For total site exchange, it has to be noted that termite mounds have been identified as hot-spots of N_2O emissions in savannas (e.g., Brümmer et al., 2009b) and are abundant in savannas of tropical Australia (Dawes-Gromadzki, 2008), but the total contribution to the soil–atmosphere exchange is highly variable in space. It was also reported that below-ground termite activity can (often only temporarily) add to on-site variability of measured in-situ fluxes (Livesley et al., 2011). Very low N_2O emission or even soil N_2O uptake was also previously reported for tropical savanna soils under dry season conditions (Andersson et al., 2003; Castaldi et al., 2006; Donoso et al., 1993; Livesley et al., 2011; Sanhueza et al., 1990), and low mineral N content was considered to be a major controlling factor (Rosenkranz et al., 2006), although many other controls and processes are discussed (Chapuis-Lardy et al., 2007).

We also observed very low N_2O emissions and occasional N_2O uptake by the soil cores with a maximum soil uptake rate of $-6.2\,\mathrm{\mu g\,N\,m^{-2}\,h^{-1}}$ during pre-incubation measurements of our air-dried soil samples at $20\,^{\circ}\mathrm{C}$. At low (M25) and medium (M50) moisture conditions, average N_2O emissions ranged from 0.3 to $5.4\,\mathrm{\mu g\,N\,m^{-2}\,h^{-1}}$ and were inline with the low N_2O emissions previously observed from nearby sites (Grover et al., 2012; Livesley et al., 2011).

The N_2O emissions observed for the improved pasture site under dry to medium moisture levels, were also low and did not differ significantly to the natural savanna site, which was to be expected as the analysis of physicochemical properties did not reveal significant differences between the two plots either (Table 1). This similarity is somewhat remarkable as the improved pasture site did receive substantial amounts of P and N over 40 years. Similar results were reported for young and old pastures under dry season conditions in the Daly region, Northern Territory, Australia, but emissions from old pasture under wet conditions were higher ($> 25\,\mathrm{\mu g\,N\,m^{-2}\,h^{-1}}$). A direct comparison is difficult, however, as soil water content reported (dry: $\sim 5\%$ WFPS, medium: $\sim 37\%$ WFPS, wet: $\sim 53\%$) does not align with our incubation setup. However, the

general range of reported in-situ flux measurements from savanna and pasture shows good agreement with our laboratory results (Grover et al., 2012).

As stated before, we excluded the results for three of the nine savanna soil cores measured with 75 % WFPS from the previous discussion as they clearly did exhibit different emission patterns. In contrast to the other cores, N$_2$O emission increases of those cores were delayed by 2 to 3 days, and strong N$_2$O emissions (T1P1R1: $33.7 \pm 29.8\,\mu g\,N\,m^{-2}\,h^{-1}$, T1P1R2: $26.3 \pm 17.8\,\mu g\,N\,m^{-2}\,h^{-1}$, T1P3R1: $52.3 \pm 37.0\,\mu g\,N\,m^{-2}\,h^{-1}$, $n = 8$) were measured throughout the incubation run. If those outlier results are included (see Table 2), average N$_2$O emissions are 1.4 to 3.8 times higher and thus substantially higher than those reported for savanna soil and on par with reported pasture fluxes under wet conditions (see, Grover et al., 2012). Although the cause for these substantially different emission patterns could not be determined, we hypothesize that small-scale mineralization hotspots (e.g., due to availability of organic material from dead soil fauna or plant residuals) account for this observation. These findings underline the importance of taking spatial heterogeneity into account when designing field samplings, and it is a strong indication that more spatial replicates would help to fully assess site scale soil–atmosphere. However, the nature of incubation experiments (i.e., the need for timely replicate sampling, laboratory constrains and climate controlled storage) severely limits the number of samples, though the gas pooling technique may offer ways to overcome spatial heterogeneity (Arias-Navarro et al., 2013; Morris et al., 2013).

Nitrous oxide emissions were positively correlated with temperature and soil moisture (see Table 2), and their variations were found to explain 58.6 and 41.3 %, respectively, of the variance of N$_2$O emissions if a simple simple linear two-parameter regression model was used (pmvd method, total variance: 46.0 %). Inclusion of soil properties did not further improve the linear model (see decomposition plot, Fig. 6). Previous results also show a positive correlation between soil temperature and N$_2$O emissions (Castaldi et al., 2010), but other authors found no clear linear correlation (Brümmer et al., 2008; Scholes et al., 1997), although observing clear diurnal patterns (Brümmer et al., 2008).

Previous studies from tropical savanna and seasonally dry tropical forest sites identified changes in WFPS to dominate the N_2O emission patterns (Brümmer et al., 2008; Castaldi et al., 2010; Davidson et al., 1993; Grover et al., 2012; Rees et al., 2006), but in general, N_2O emissions from undisturbed savanna soil ranked lowest for sea-
5 sonally dry ecosystems due to low nutrient availability and soil textures favoring quick drainage (Castaldi et al., 2006). In accordance, significant positive correlations were reported for clay content and soil volumetric water content (Castaldi et al., 2004), factors which strongly determine the pore space and water saturation and thus soil aeration and anaerobiosis. It is well established that oxygen saturation in the soil air controls
10 the level of aerobic nitrification and anaerobic denitrification microbial activity (Davidson et al., 1993) and thus nitrogen pathways and process rates. Compared to tropical biomes of seasonally dry or humid tropical forests (e.g., Butterbach-Bahl et al., 2004b; Gharahi Ghehi et al., 2012; Kiese et al., 2003; Werner et al., 2007b), the annual N_2O emissions from savanna soils seem to be substantially lower, which is hypothesized
15 to originate from N depletion via pyro-denitrificatory losses (bush fires), shorter wet periods with prevailing denitrificatory conditions in combination with well-drained soils and lower amounts of total nitrogen cycling within the system (Castaldi et al., 2006; Davidson et al., 2001). Other studies have not found significant effects of soil moisture on N_2O emissions under in-situ conditions (Livesley et al., 2011), and is assumed that
20 a lack of seasonality and low nitrogen levels provide the rationale for this observation.

4.3 NO emissions

Nitric oxide emissions measured at low moisture conditions (M25) and 20 to 30 °C soil temperature were 6.4 ± 2.4 and $9.1 \pm 4.8 \, \mu g \, N \, m^{-2} \, h^{-1}$, respectively, and therefore higher than the fluxes reported for African sites during the dry season ($1.6–2.2 \, \mu g \, m^{-2} \, h^{-1}$,
25 Meixner et al., 1997; Serça et al., 1998) but comparable to dry season conditions observed in Venezuela (Cárdenas et al., 1993). A link between the magnitude of NO emissions from savanna soil and soil water content and nutrient availability has been postulated by various authors (Cárdenas et al., 1993; Feig et al., 2008; Parsons et al.,

1996; Poth et al., 1995; Rondón et al., 1993). It was reported that soil moisture is the dominant factor whenever it is not limiting the NO flux, but emission strength is modulated by daily variation of soil temperature (Meixner et al., 1997). A significant correlation of NO emissions with soil pH was also reported (Poth et al., 1995), and it is in agreement with the outcome of our relative importance analysis where pH scored second after soil temperature in explaining observed NO emissions post pulse (Fig. 6). The low score of soil moisture in this analysis can be explained by the non-linear response of NO to changing soil moisture levels and is also reflected in the low total explanatory power of 19.4 %. High soil temperatures (40 °C) in combination with medium moisture levels (M50) resulted in strongest NO emissions ($65.0 \pm 97.8 \, \mu g \, N \, m^{-2} \, h^{-1}$), with high intra-core variability. This indicates a substantial importance of aerobic ammonia oxidation based pathways (Butterbach-Bahl et al., 2013) for NO emission. Previous studies from African savannas on sandy soils suggest that maximum NO emissions occur at a narrow WFPS range between 10 to 25 % WFPS (Aranibar et al., 2004; Feig et al., 2008; Otter et al., 1999) and that a strong linear increase of NO emissions occurs with increasing temperatures starting at 25 °C to 30 °C (Aranibar et al., 2004). It was also reported that NO emission decreases again at a soil temperature exceeding 40 °C (Passianoto et al., 2004), but such extreme soil temperatures were not included in our experimental setup. In accordance with this, the analysis of T1 site cores (savanna) showed strongest emissions for pulsing after the initial rewetting and the following incubation steps for the lowest soil moisture treatment (M25). It is noteworthy that pulse emissions again were slightly stronger at M25 conditions for pasture cores, the following measurements revealed no difference in emission strength between the three moisture levels at the first temperature interval. At 30 °C soil temperature, emissions for the driest cores remained low but M50 cores exhibited higher NO emissions (see Fig. 5).

Unfortunately, no measurements were conducted for low moisture conditions (M25) and high soil temperatures (40 °C) during the first incubation sequence of our work and thus, we can only speculate that NO emissions at low moisture levels (M25) would

have even exceeded our maximum NO emissions recorded due to nitrification activity peaking at moisture levels below 50 % WFPS (see literature citations above).

4.4 N_2 emissions and total gaseous nitrogen losses

Dinitrogen emissions varied greatly between the different incubation setups (17–2350 µg N m^{-2} h^{-1}), and this range is comparable to results reported for temperate forests (Butterbach-Bahl et al., 2013; Dannenmann et al., 2008). The N_2 emission rates were clearly dominated by soil saturation (see Table 2 and Fig. 6). Average N_2 emissions for savanna soil below 50 % WFPS and under any implemented temperature setting was ranging from 45 to 108 µg N m^{-2} h^{-1}, while average N_2 emissions with highest water saturation (M75) were ranging from 627 to 1201 µg N m^{-2} h^{-1}. It is thus not surprising that the analysis of relative importance revealed that approximately 75 % of the variance of N_2 emissions was explained by changes in WFPS. Variations in pH accounted for another 23 % of variance (Fig. 6), while the other factors did not contribute to explain the observed N_2 emissions. The dominance of WFPS is in line with common knowledge about the functioning of denitrification, given that N_2 production is catalyzed by the enzyme N_2O-reductase, which in-turn is up-regulated by low oxygen concentrations (Spiro, 2012). From calculating fractions it is clear that N_2 is the dominant compound of gaseous losses, often exceeding N_2O and NO emissions by several magnitudes (Table 3). N_2 accounted for 82–99 % of all gaseous N losses from savanna soils for the investigated incubation intervals, except for the 50 % WFPS/40 °C setup, where strong NO emissions were accounting for 43 % of all N emissions.

The N_2-dominated N gas product ratios across all soil moisture levels might appear surprising at first, since strong N_2 soil emission has been associated with strictly anaerobic soil conditions and the investigated sandy soil textures may facilitate good aeration and thus impair formation of anaerobic microsites. It is however increasingly recognized that our previous knowledge on rather low N_2 loss from ecosystems is falsified by systematic but irreproducible underestimation of N_2 emission estimates due to the failure of the widely applied acetylene inhibition technique to indirectly estimate N_2

loss (Butterbach-Bahl et al., 2013). In contrast, the few available reliable direct measurements of N_2 emission from upland soil obtained with state-of-the-art methods such as the helium soil core incubation technique generally revealed large ecosystem N_2 losses (Butterbach-Bahl et al., 2013). This is in line with recent evidence about unexpectedly large abundance and diversity of the N_2O reducing microbial community in soil, not restricted to denitrifiers (Jones et al., 2013; Sanford et al., 2012). The latter findings could provide a microbiological explanation for high importance of N_2 loss in upland soil in general and probably also for the savanna soils investigated in this study.

4.5 Pulse emissions

Significant N_2O pulse emissions following the first rains after a dry season, often with a small temporal delay, have been reported for different seasonally dry (sub-)tropical ecosystems (Brümmer et al., 2008; Davidson et al., 1993; Scholes et al., 1997; Werner et al., 2007a) and are often preceded by significant CO_2 emissions immediately after the soil is re-moistured. We also measured significant CO_2 pulse emissions for all three watering levels immediately after water addition, but emissions dropped significantly in the following days. It has been stated that this pattern is caused by water-induced activation of soil microbes after pronounced dry season periods leading to immediate mineralization of accumulated organic material and subsequent consumption of freshly produced NH_4^+/NH_3^- trough nitrification (Davidson et al., 1993).

The initial watering lead to N_2O pulse emissions with peak emissions immediately after water addition, but they were, with three days duration, only short-lived (a pattern that was previously reported for African savanna soil, Brümmer et al., 2008; Scholes et al., 1997). The occurrence of N_2O pulsing emissions was previously reported by Grover et al. (2012) for in-situ measurements at a nearby savanna site, but emissions were, in accordance with our laboratory results, several magnitudes below reported break-of-seasons pulse emissions reported for tropical rainforests (Butterbach-Bahl et al., 2004b; Werner et al., 2007b). We did observe a positively correlated increase of N_2O pulse emissions with watering intensity, but with an average pulse emission of

$5.8 \pm 5.0\,\mu g\,N\,m^{-2}\,h^{-1}$ for our highest watering intensity (M75) the amount of N released to the atmosphere was still minor. It has to be noted that, due to experimental design restrictions (consecutive incubation variations of temperature), we only measured initial pulse emissions under rather low temperature settings, and given the observed positive correlation of N_2O emissions with temperature, pulse emissions at higher temperatures would have been even more pronounced.

Strong NO pulse emissions were reported for many sites from seasonally-dry tropics (Brümmer et al., 2008; Levine et al., 1996; Scholes et al., 1997) and rainforests with seasonality (Butterbach-Bahl et al., 2004b) and are now an essential part in models simulating global NO emissions (Steinkamp and Lawrence, 2011; Yan et al., 2005; Yienger and Levy, 1995). The immediate emission of NO after watering was also observed by Pinto et al. (2002), who measured up to 100-fold increased NO emissions above pre-watering levels to $105\,\mu g\,N\,m^{-2}\,h^{-1}$, but noted that results varied substantially between collars. These results are inline with our observed peak pulse emissions, which also varied between 27.7 and $306.5\,\mu g\,N\,m^{-2}\,h^{-1}$. In contrast to N_2O peak emission, the response was negatively correlated with moisture saturation. For soils of seasonally dry tropical rainforests in Mexico and Australia, high rates of NO emissions and quick temporal responses to the first rainfall events were observed (Butterbach-Bahl et al., 2004b; Davidson et al., 1993, 1991). The authors also state that rainfall events during the wet season triggered only minor emission events, providing pointers that the duration and severity of antecedent dry periods ultimately determines the magnitude of response of NO emissions to wetting events (Butterbach-Bahl et al., 2004a; Davidson et al., 1993). Thus, these pronounced pulse emissions events are thought to contribute substantially to the annual total of NO emissions (Butterbach-Bahl et al., 2004b; Ludwig et al., 2001).

Due to incubation design and the required destructive sampling of soil cores for N_2 analysis, pulsing effects on N_2 emission were not investigated (N_2 analysis for the various incubation conditions was carried out after the N_2O, NO and CO_2 incubation runs were completed).

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

5 Conclusion

We reported the N_2O, NO, N_2 and CO_2 emission response of intact soil cores from tropical savanna and pasture to a range of climatic conditions. The laboratory analysis of intact soil cores allows for an analysis under controlled soil moisture and temperature conditions while largely preserving the structural components of the soil column, although influence of flora and macro-fauna is lost compared to in-situ measurements. Key findings of our analysis are:

1. Soil–atmosphere exchange of NO and CO_2 under late dry season conditions was very low and for N_2O often even negative.

2. Total atmospheric loss of nitrogen was dominated by N_2 emissions under all but one tested soil climatic conditions (82.4–99.3 % of total gaseous N loss), but NO emissions contributed almost 43.2 % to the total gaseous loss at 50 % WFPS and 30 °C soil temperature.

3. N_2O emissions were substantially higher for 3 out of 12 soil cores investigated and indicate substantial spatial variability at site level (maximum N_2O emission rate: 180 µg N m^{-2} h^{-1}). This has implications for site-scale estimates and extrapolation of soil–atmosphere exchange rates.

4. Pulse emissions of varying magnitudes (dependent on the amount of moisture added) were observed for N_2O, NO and CO_2, but they were short-lived (24–72 h). Again, three of the cores displayed a different emission pattern with no initial response, but prolonged elevated N_2O after the first days.

5. We generally found no significant differences in emissions from savanna and pasture sites despite long-term fertilization of the pasture.

6. Major controls on emission levels were soil moisture and temperature for N_2O and CO_2 emissions, soil temperature and pH for NO (soil moisture effect was not

detected due to non-linear response in the analysis), and soil moisture and pH for N_2.

The results indicate that the gaseous loss of nitrogen in savannas from northern Australia is largely controlled by NO emissions at high soil temperatures and low moisture conditions and N_2 losses, whereas the soil–atmosphere exchange of N_2O is generally low or even negative after pronounced dry periods. We did encounter strong N_2O emissions for few individual cores, but no explanation for this emission pattern could be detected from the soil physico-chemical properties of these cores. This indicates that a higher replication or larger sample areas are required to determine the N_2O exchange of these systems more reliably.

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Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

◄◄ | ►►

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

|◄ | ►|

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Table 1. Physico-chemical properties of the sampled soil and mean microbial biomass (sampling depth 0–15 cm, * µg C/N g [dry weight] soil^{-1}).

Land use	Pos.	Soil texture (%)			Density	Org. C	Total N	pH	Microbial biomass (*)		NH$_4^+$	NO$_3^-$
		Sand	Silt	Clay	(g cm^{-3})	(%)	(%)		C	N	(*)	(*)
Natural	T1 P1	81	12	7	1.4	2.8	0.1	5.0	80.9	12.2	2.8	0.1
savanna	T1 P3	87	8	5	1.5	0.8	0.04	4.4	87.1	8.2	0.8	0.04
	T1 P5	87	10	3	1.7	0.7	0.05	5.1	87.4	10.1	0.7	0.05
Pasture	T3 P1	68	17	15	1.5	1.5	0.1	4.6	67.7	16.7	1.5	0.1

Table 2. Average CO$_2$, N$_2$O, NO, and N$_2$ emission at different moisture and temperature incubation settings (fluxes reported post-pulse, [a] value excl. outliers/brackets incl. outliers, [b] N$_2$: T1 $n = 3$, T3: no replicates).

Temperature [°C]		WFPS [%] 25	50	75	Transect
		carbon dioxide (mg CO$_2$-C m^{-2} h^{-1})			
20		19.7 ± 6.8	21.5 ± 5.6	33.3 ± 18.7	T1
		17.6 ± 5.0	28.1 ± 8.7	45.1 ± 15.9	T3
30		32.0 ± 15.9	25.4 ± 15.5	66.1 ± 20.2	T1
		38.8 ± 14.4	18.6 ± 8.7	31.3 ± 18.3	T3
40			53.6 ± 26.0	75.8 ± 30.3	T1
			31.6 ± 10.8	38.4 ± 12.4	T3
		nitrous oxide (µg N$_2$O-N m^{-2} h^{-1})[a]			
20		0.3 ± 0.4	0.5 ± 0.6	3.6 ± 7.2 (13.7 ± 29.0)	T1
		0.3 ± 0.3	0.5 ± 0.7	1.1 ± 1.4	T3
30		0.6 ± 0.4	1.5 ± 2.4	3.1 ± 1.2 (9.6 ± 12.4)	T1
		0.4 ± 0.2	1.9 ± 2.3	8.2 ± 10.2	T3
40			5.4 ± 4.8	13.7 ± 9.4 (19.6 ± 15.3)	T1
			4.1 ± 4.4	17.1 ± 14.5	T3
		nitric oxide (µg NO-N m^{-2} h^{-1})			
20		6.4 ± 2.4	4.9 ± 2.1	3.4 ± 2.6	T1
		3.6 ± 1.7	4.2 ± 1.7	1.7 ± 1.1	T3
30		9.1 ± 4.8	6.1 ± 3.1	3.5 ± 2.4	T1
		3.2 ± 1.5	8.4 ± 9.0	9.5 ± 8.2	T3
40			65.0 ± 97.8	13.2 ± 15	T1
			12.9 ± 11.0	6.1 ± 6.7	T3
		dinitogen (µg N$_2$-N m^{-2} h^{-1})[b]			
20		107.6 ± 82.8	65.3 ± 26.5	1041.1 ± 255.8	T1
		400.6	110.1	1281.2	T3
30		45.4 ± 33.2	56 ± 13	627.4 ± 789.1	T1
		311.3	118.9	348.3	T3
40			79.9 ± 11.5	1201 ± 1625	T1
			207.0	614.4	T3

Table 3. Fractional contribution of N_2O, NO and N_2 to total gaseous N losses of savanna soil at T1 (units: % of total gaseous N loss; $N_2O : NO : N_2$; [a] value excl. outliers/brackets incl. outliers).

		WFPS [%]		
		25	50	75 [a]
Temp. [°C]	20	0.3 : 5.6 : 94.1	0.7 : 6.9 : 92.4	0.3 : 0.3 : 99.3 (1.7 : 0.3 : 98)
	30	1.1 : 16.5 : 82.4	2.4 : 9.6 : 88.1	0.5 : 0.6 : 99.0 (1.6 : 0.5 : 97.9)
	40		3.6 : 43.2 : 53.2	1.1 : 1.1 : 97.8 (1.7 : 1.1 : 97.3)
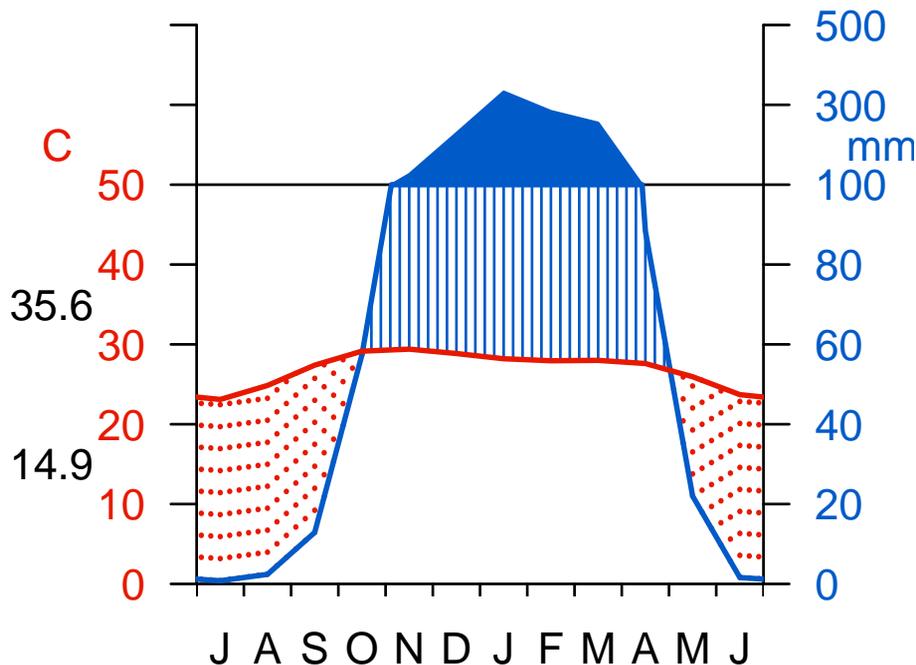
Figure 1. Climatic conditions at sampling sites (data: Bureau of Meteorology, Australia, http://www.bom.gov.au, years 1981–2010).

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

|◄ | ►|

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion
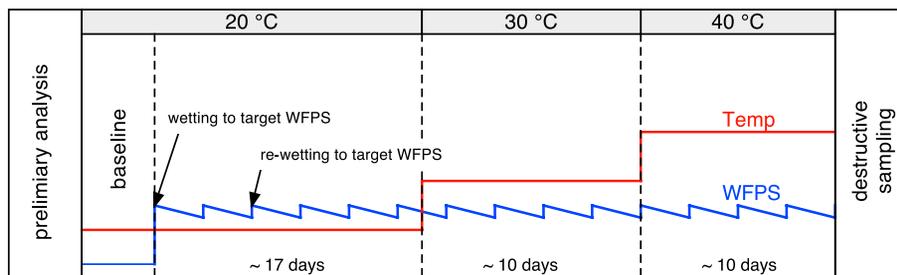
Figure 2. Schematic of the incubation setup. The soil moisture level of the cores was kept constant be reading water every second day, the temperature was raised stepwise from 20 °C, to 30 °C, and finally 40 °C. The incubation outlined in this figure was performed three times for separat soil cores (once for each of the three soil moisture levels; WFPS: water filled pore space).
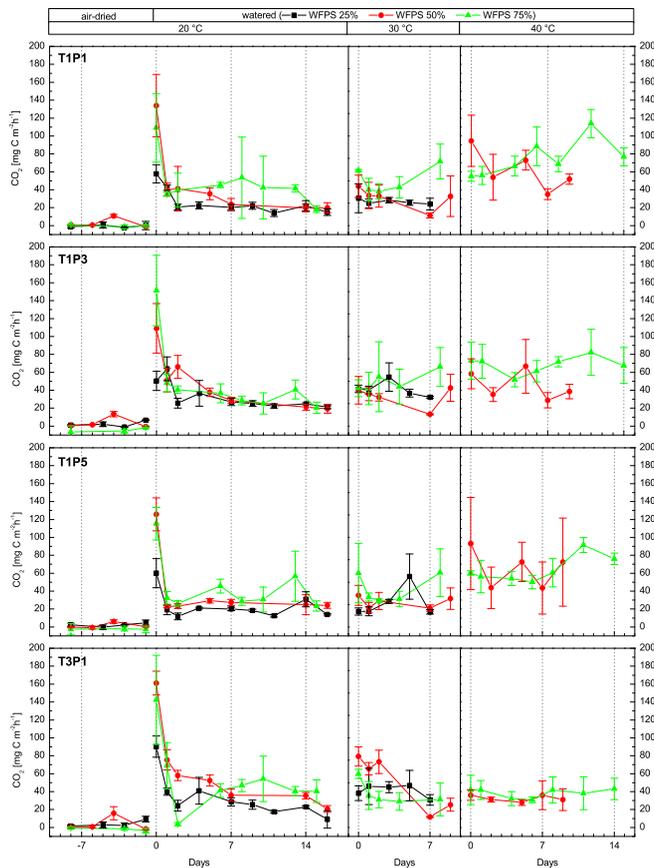
Figure 3. Average CO_2 emission per site (savanna: T1P1, T1P3, T1P5, grassland: T3P1) for three soil moisture (black: 25 % water filled pore space (WFPS), red: 50 % WFPS, green 75 % WFPS) and three temperature levels (x-axis: days since incubation change/negative values indicate pre-incubation period).

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

|◀ | ▶|

◀ | ▶

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion
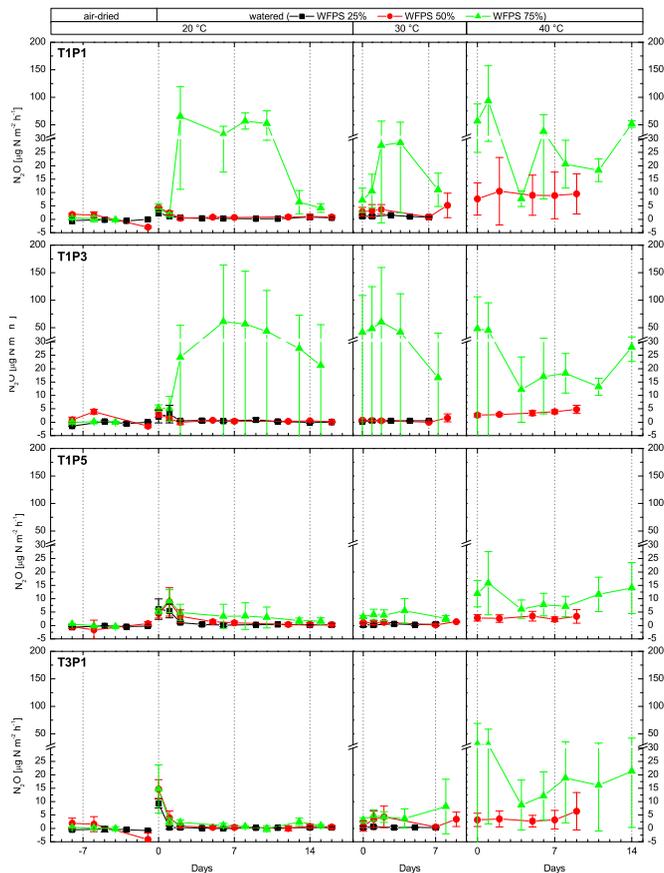
Figure 4. Average N_2O emission per site (savanna: T1P1, T1P3, T1P5, grassland: T3P1) for three soil moisture (black: 25 % water filled pore space (WFPS), red: 50 % WFPS, green 75 % WFPS) and three temperature levels (x-axis: days since incubation change/negative values indicate pre-incubation period).

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

|◄ | ►|

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion
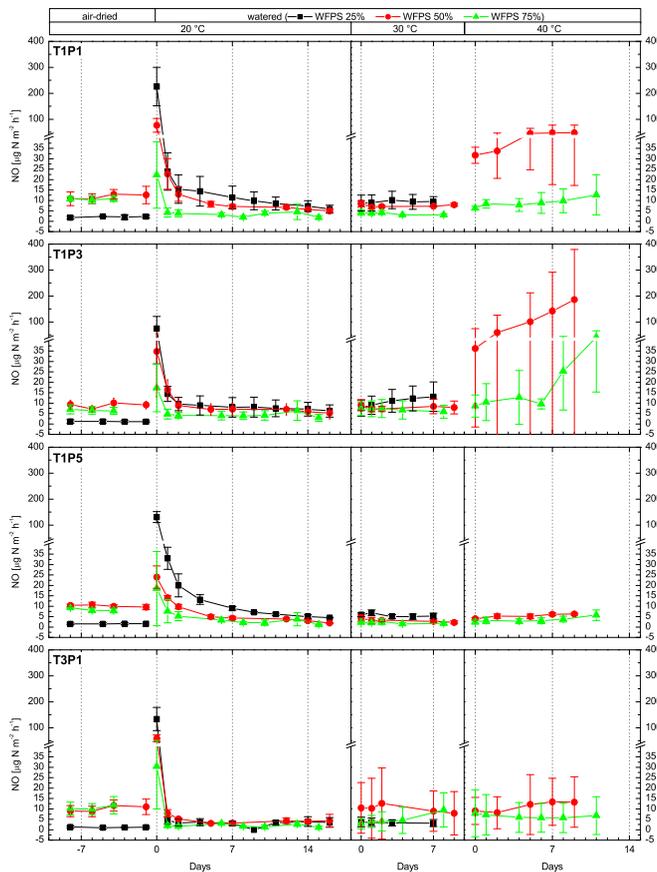
Figure 5. Average NO emission per site (savanna: T1P1, T1P3, T1P5, grassland: T3P1) for three soil moisture (black: 25 % water filled pore space (WFPS), red: 50 % WFPS, green 75 % WFPS) and three temperature levels (x-axis: days since incubation change/negative values indicate pre-incubation period).

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

◄ | ►►

◄ | ►

Back | Close

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Interactive Discussion
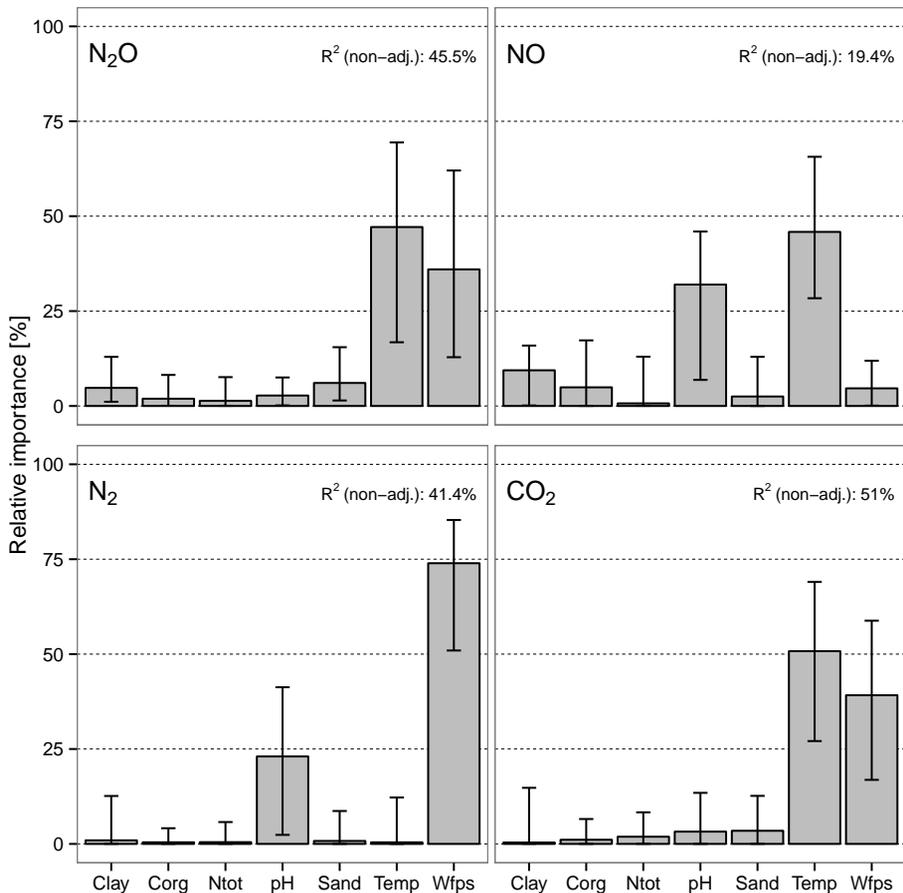
Figure 6. Relative importance of environmental factors clay, organic carbon (Corg), total nitrogen (Ntot), sand, temperature and water filled pore space (Wfps) on N_2O, NO, N_2 and CO_2 emissions (based on proportional marginal variance decomposition, R^2 values give total predictability of each model).