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Nitrous oxide fluxes from a grain–legume crop (narrow-leafed lupin) grown in a semiarid climate

LOUISE BARTON*, KLAUS BUTTERBACH-BAHL†, RALF KIESE† and DANIEL V. MURPHY*

*School of Earth & Environment (M087), Faculty of Natural & Agricultural Sciences, The University of Western Australia, Crawley 6009, Australia, †Institute for Meteorology and Climate Research, Atmospheric Environmental Research (IMK-IFU), Karlsruhe Institute of Technology, Kreuzeckbahnstr 19, 82467 Garmisch-Partenkirchen, Germany

Abstract

Understanding nitrous oxide (N_2O) fluxes from grain–legume crops in semiarid and arid regions is necessary if we are to improve our knowledge of global terrestrial N_2O losses resulting from biological N_2 fixation. N_2O fluxes were measured from a rain-fed soil, cropped to a grain–legume in a semiarid region of southwestern Australia for 1 year on a subdaily basis. The site included plots planted to narrow-leafed lupin (*Lupinus angustifolius*; 'lupin') and plots left bare (no lupin). Fluxes were measured using soil chambers connected to a fully automated system that measured N_2O by gas chromatography. Daily N_2O fluxes were low (-0.5 to $24\,g\,N_2O\text{-}N\,ha^{-1}\,day^{-1}$) and not different between treatments, culminating in an annual loss of $127\,g\,N_2O\text{-}N\,ha^{-1}$. Greatest daily N_2O fluxes occurred from both treatments in the postharvest period, and following a series of summer and autumn rainfall events. At this time of the year, soil conditions were conducive to soil microbial N_2O production: elevated soil water contents, increased inorganic nitrogen (N) and dissolved organic carbon concentrations, and soil temperatures generally $>25\,^{\circ}C$; furthermore, there was no active plant growth to compete for mineralized N. N_2O emissions from the decomposition of legume crop residue were low, and approximately half that predicted using the currently recommended IPCC methodology. Furthermore, the contribution of the biological N_2 fixation process to N_2O emissions appeared negligible in the present study, supporting its omission as a source of N_2O from the IPCC methodology for preparing national greenhouse gas inventories.

Keywords: Australia, biological N_2 fixation, crop residue, emission factor, IPCC methodology

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Introduction

Legume crops contribute valuable nitrogen (N) inputs to farming systems throughout the world. Conservative estimates suggest 50–70 Tg N is fixed biologically in agricultural systems, despite the progressive replacement of legume rotations with synthetic N fertilizers over the past four decades (Smil, 2001; Crews & Peoples, 2004; Herridge *et al*., 2008). N fixed by legumes contributes to human food production, via fresh pods and dry grains, or as a feedstock and pasture for animals. Non-N_2 fixing crops have been shown to benefit from legume crops through a variety of mechanisms including N inputs, disease breaks, and improved soil fertility (Peoples *et al*., 2009). However, N_2 fixation by cultivated legumes is also considered to enhance anthropogenic nitrous oxide (N_2O) emissions (Stehfest & Bouwman, 2006).

Correspondence: Louise Barton, tel. + 61 8 488 2543, fax + 61 8 488 1108, e-mail: louise.barton@uwa.edu.au

N_2O fluxes from legume crops are mainly derived from decomposition of the above- and belowground legume residues, with losses from the biological N_2 fixation process *per se* considered to be negligible (Rochette & Janzen, 2005). N released from legume residues is at risk of being emitted as N_2O via a number of biological processes. For example: (i) mineralized N can be nitrified by soil microorganisms converting soil ammonium (NH_4^+) to nitrate (NO_3^-) under aerobic conditions, with N_2O emitted as a by-product of the transformation; (ii) anaerobic denitrifiers can sequentially reduce nitrogen oxides (e.g., NO_3^-) to nitric oxide (NO), N_2O, and finally N_2, with incomplete reduction resulting in N_2O emissions; and (iii) nitrifier denitrification, reduction of NO_2^- to N_2 via N_2O, may also be a significant source of soil N_2O (Wrage *et al*., 2005). These soil biological processes, and the emission of N_2O, are greatly enhanced by increased N availability, and while losses may appear low in relation to legume residue inputs (e.g., 1.25%) (IPCC, 2006), the high global warming potential of N_2O (298 times greater than CO_2 during a 100-year life time) (Forster *et al*., 2007) means accurate

estimates are required when assessing net greenhouse gas fluxes from legume crop production.

Legumes systems are estimated to emit 0.4 Mt N_2O-N annually, around 10% of total agricultural N_2O emissions; but, this value has largely been estimated from studies conducted in temperate agricultural systems (Stehfest & Bouwman, 2006). Legume crops are widely grown in semiarid and arid land regions (precipitation $<400\,mm\,yr^{-1}$, aridity index <0.5; UNESCO, 1977), which constitute one-third of the global land area (Harrison & Pearce, 2000). N_2O fluxes from legume crops grown in the absence of N inputs from synthetic or organic N are rarely reported from semiarid or arid regions (Rochette & Janzen, 2005; Stehfest & Bouwman, 2006). Dick *et al*. (2008) concluded that growing leguminous crops in a carbon-(C) and N-poor soil in a semiarid region of Mali did not increase N_2O compared with continuous cereal cropping. Furthermore, other studies investigating N_2O fluxes from rain-fed, fertilized N crops also suggest that losses from semiarid regions are likely to be lower than those from temperate regions (Galbally *et al*., 2008), and less than that estimated using international default emission factors (Barton *et al*., 2008, 2010). A better understanding of global N_2O fluxes from grain–legume crops would be gained by including more measurements, and improving the representation of agricultural systems from semiarid and arid regions (Stehfest & Bouwman, 2006). Understanding N_2O fluxes from grain–legume crops will also improve assessments comparing greenhouse gas emissions from crops produced using legume-fixed N vs. those grown using synthetic N fertilizer sources (Crews & Peoples, 2004; Hillier *et al*., 2009).

The southwestern Australian grain-growing region consists of approximately 18 million ha of semiarid land and is responsible for 40% of Australia's annual grain production, including grain–legumes (Australian Bureau of Agricultural and Resource Economics, http://www.abareconomics.com). The region has a strong seasonality characterized by cool, wet winters and hot, dry summers; and consequently, crops are only grown during the winter months as low rainfall (and availability of water for irrigation) excludes summer cropping. Grain–legumes are traditionally cropped in rotation with cereal crops, with the harvested grain removed, and the resultant aboveground material retained (but rarely incorporated into the soil). For example, lupin is commonly grown in Australia, representing 61% of global lupin production in 2008 (Food and Agricultural Organization of the United Nations, http://www.fao.org). Consequently, the aims of the following study were to (i) acquire a unique, 1 year data set of continuous subdaily N_2O fluxes from a rain-fed, grain–legume (lupin) crop; (ii) determine the contribution of a grain–legume crop to soil N_2O emissions; and (iii) improve our understanding of the soil and environment factors controlling N_2O emissions from crops grown in a semiarid region.

Materials and methods

Site and soil

N_2O fluxes were measured on the Cunderdin Agricultural College (31°36′S, 117°13′E), in the central grain belt of Western Australia, approximately 156 km east of Perth. Cunderdin has a long-term average rainfall of 365 mm (range, 142–601 mm yr^{-1}; 1914–2008), which mainly falls during the winter months (June–August), a mean daily maximum temperature of 25.1 °C (range, 23.5–26.3 °C; 1951–2007), and a mean daily minimum temperature of 11.4 °C (10.2–12.2 °C; 1951–2007) (Commonwealth Bureau of Meteorology, http://www.bom.gov.au/climate). The experimental site is located on a flat to gently undulating land, and consists of free-draining sand overlying poorly draining clay (Natric Haploxeralf and Typic Natrixeralf; USDA, 1992). The surface soil (0–120 mm) has a pH of 6.0 (1 : 5, soil : 0.01 M $CaCl_2$ extract), electrical conductivity of 170 $\mu S\,cm^{-1}$ (1 : 5, soil : water extract), cation exchange capacity of 3.3 $cmol\,kg^{-1}$, total C concentration of 9.38 $mg\,g^{-1}$, total N concentration of 0.76 $mg\,g^{-1}$, and bulk density of 1.4 $g\,cm^{-3}$. The surface soil contains 93% sand, 4% silt, and 3% clay. For further details of the soil profile, see Barton *et al*. (2008).

The site has been planted to a variety of cereal and grain–legume crops in rotation since the 1930s, and in the 3 years before the present study, was used to measure soil N_2O fluxes from wheat (*Triticum aestivum* cv Carnamah, 2005–2006; *T. aestivum* cv Bonney Rock, 2006–2007) (Barton *et al*., 2008), and canola (*Brassica napus* cv ATR Banjo, 2007–2008) (Barton *et al*., 2010). Livestock was excluded from the site from December 2004 so as to avoid N from animal urine and dung. Crop residue [1320 kg dry matter (DM) ha^{-1}, average across all plots] from the previous canola crop was still present at the time of planting the present study.

Experimental design and approach

Plots (105 $m^2\,plot^{-1}$) were planted to narrow-leafed lupin (*Lupinus angustifolius* cv Mandelup) on 14 May 2008, with no prior cultivation. A completely randomized design with two treatments, and three replicates, was used. Plots were either planted with inoculated and fumigated lupin seed (100 kg seed ha^{-1}, 'lupin') or remained bare (no lupin). Each plot was separated by a buffer of at least 1.67 m, which was either planted to lupin or kept bare (and free of weeds). The lupin plots were topdressed with 100 kg ha^{-1} of K_2SO_4, and direct-drilled (to 30 mm) with 100 kg ha^{-1} of 'Superphosphate CuZnMo'®, at planting. An area (6.76 m^2) within each plot was designated for measuring N_2O fluxes, while the remainder of the plot was used for soil and plant sampling. Four weeks after planting, plant numbers in the chamber base areas were

adjusted, by either removing or adding seedlings, to reflect the average plant density in the field (32 plants m^{-2}), as plant germination was uneven. Lupin was harvested from the chambers on 5 November 2008 and the stubble was retained for the remainder of the study.

Measurement of N_2O fluxes

N_2O fluxes were measured for approximately 1 year, commencing on 14 May 2008 immediately after planting, and ending on 28 April 2009. Fluxes were measured in each treatment plot using soil chambers (one per plot) connected to a fully automated system. Details of the design and operation of automated gas sampling system have been described by Breuer *et al*. (2000) and Kiese *et al*. (2003). Briefly, the system consisted of a gas chromatograph (SRI 8610C, SRI Instruments, Torrance, CA, USA) fitted with an ^{63}Ni electron capture detector for N_2O analysis, an automated sampling unit for collecting and distributing gas samples, and six chambers (one per treatment plot). Chambers (500 mm × 500 mm, clear perspex) were placed on metal bases inserted into the ground (100 mm), and fitted with a top that could be automatically opened and closed. Four bases were located in each treatment plot to enable the chambers to be moved sequentially to a new position every week to minimize the effect of chambers on soil properties and plant growth. The height of the chambers (not bases) was progressively increased with clear perspex extensions to accommodate crop growth, with a maximum height of 650 mm. The detection limit for N_2O fluxes of the system was 0.6 μg N_2O-N m^{-2} h^{-1} at a chamber height of 150 mm, and the dilution via leakage considered negligible.

A full measurement cycle for the determination of N_2O fluxes commenced with chamber lid closure and finished 96 min later when the lid opened. This enabled four N_2O measurements (24 min apart) to be collected from each chamber during each cycle. The chambers remained open for at least 96 min between each measurement cycles, and the system provided up to eight (hourly) flux rates per day. The chambers were programmed to open if the air temperature in the chamber exceeded a set value (43 °C when lupin was growing in the chamber, 60 °C at other times) or if rain fell (>0.4 mm in 5 min) while the chambers were closed. This minimized the effect of the chambers on soil properties and crop growth, although it is possible that air temperature regimes were altered during the enclosure period. The system was automatically calibrated by standard calibration gases (0.5–0.6 μL L^{-1} N_2O depending upon the calibration cylinder; with a variance of ± 2%), four times during each measurement cycle. For almost 4 weeks (2 July 2008–28 July 2008), N_2O concentrations were determined by calibrating the system using the concentration of N_2O in the ambient air due to an error with the standard calibration gas.

Inorganic N, dissolved organic carbon (DOC), and soil water content

Soil inorganic N (NO_3^- and NH_4^+), total DOC, and gravimetric water content were measured to explain seasonal variations in N_2O fluxes. The inorganic N of the surface soil (0–50 and 50–100 mm) was measured the day following planting and then weekly for 2 weeks, and then generally every 2 weeks for the remainder of the study. At each sampling date, two samples were collected from each plot, with each sample containing five bulked subsamples. Every 4 weeks, soil samples were also analysed for DOC. The soil inorganic N content of the surface metre was measured before planting (8 May 2008), at 75% flowering (19 August 2008) and at the completion of the study (28 May 2009). Soil samples were collected at specified depths (0–50, 50–100, 100–200, 200–300, 300–500, 500–750, and 750–1000 mm) at two locations per plot. NO_3^-, NH_4^+, and DOC were extracted from soil samples by adding 80 mL of 1 M potassium chloride to 20 g of field-moist soil (sieved <4 mm) and shaking for 1 h. The filtered solution (Adventec MFS, Adventec 5C, Dublin, CA, USA) was frozen until analysed for NO_3^- and NH_4^+ colorimetrically using a modified hydrazine reduction method (Downes, 1978). DOC was measured by further filtering the extract through a polyethersulphone membrane (45 μm pore size, Pall Gelman Laboratory, Ann Arbour, MI, USA) and analysing for total C (TOC-5000A, Shimadzu, Kyoto, Japan). Gravimetric soil water content was determined at the same time soil samples were collected for inorganic N and DOC analysis, and after drying subsamples at 105 °C for at least 24 h.

Plant N uptake

N uptake in the aboveground biomass and the grain was measured both inside and outside the chamber areas at plant maturity (5 November 2008). Samples from outside the chamber area were collected from four, 0.25 m^2 quadrants in each plot, and by cutting the shoots at the base and collecting fallen leaves. Samples were oven-dried at 60 °C for 1 week before recording the dry mass. The grain was collected, weighed, and analysed for N using a CHN analyser (Elementar Analysensysteme GmbH, Vario Macro, Hamau, Germany). Similarly, the remaining shoot was dried and ground before being analysed for total N. Inside each chamber, the aboveground biomass and grain yield were measured in a similar way to those quadrants outside the chambers, except that the shoot was cut at 150 mm above the ground to simulate harvest. Once the quadrant and chamber samples had been completed, the remaining area of the plot was harvested using an experimental harvester.

Soil environmental and climatic measurements

A weather station was installed within the experimental site to measure climatic parameters, as well as soil temperature, on an hourly basis. Rainfall was measured using a 204 mm diameter automated tipping rain gauge (TB3, Hydrological Services, Warwick Farm, NSW, Australia) with a resolution of 0.2 mm, air temperature was measured at 1.2 m height using a platinum resistance element (PT100 RTD, MTL Engineering Australia, Canning Vale, Australia), and soil temperature was measured at 100 mm using a sealed platinum resistance element (PT100 RTD, MTL Engineering Australia). All climatic

and soil temperature data were collected and stored automatically by a datalogger (DataTaker 50, Data Electronics, Scoresby, Australia). Air temperature within one chamber was recorded every minute, during chamber closure, using a temperature probe (Pt100, IMKO, Ettlingen, Germany).

Analysis of data

All data were statistically analysed using Genstat for Windows (Payne *et al*., 2009). A general linear model (using a completely randomized design) was used to determine whether lupin treatment affected measured annual N_2O fluxes. *Post hoc* pairwise comparisons of means were made using LSD (significance level of 5%). Daily N_2O emissions were related to soil and environmental properties using a correlation analysis. A linear mixed model (with REML procedure) allowing for variance heterogeneity was used to analyse the repeated measurements of soil parameters with time. Skewed data were corrected using the natural logarithm transformation before conducting analyses.

Water-filled pore space (WFPS) was calculated by dividing volumetric water content by total porosity (Linn & Doran, 1984). Volumetric water contents were calculated by multiplying gravimetric water content by bulk density. Total porosity was calculated as 1–(bulk density/particle density), and using a particle density of 2.65 g cm^{-3}. Plant N uptake in the chamber areas were calculated by multiplying aboveground dried biomass by the N concentration of the plant material. The amount of plant residue remaining as stubble, and the N content of the stubble, were estimated using data collected from outside the chamber area.

Hourly N_2O ($\mu g\, N_2O\text{-}N\, m^{-2}\, h^{-1}$) fluxes were calculated from the slope of the linear increase in N_2O concentration during the chamber lid closure period, and corrected for chamber air temperature, air pressure, and the ratio of cover volume to surface area, as described by Barton et al. (2008). Flux rates were converted to zero if the regression coefficient (r^2) was <0.80. Daily losses for each plot were calculated by averaging hourly losses for that day. Annual fluxes for each plot were calculated by integrating hourly losses with time.

Australia's national greenhouse gas inventory is prepared in accordance with both the IPCC *Revised 1996 Guidelines for National Greenhouse Gas Inventories* (IPCC, 1997) and the IPCC *Good Practice Guidance* (IPCC 2000). The amount of N_2O emitted by biological N_2 fixation was estimated by either summing fluxes during the growth phase of the current study, or using the IPCC methodology currently adopted by Australia, whereby the amount of N_2 fixed is multiplied by 1.25% (IPCC, 1997). Biological N_2 fixation was estimated by either using a national average for narrow-leafed lupin (14.2 kg N_2 fixed t^{-1} shoot DM multiplied by 1.33 to account for biological N_2 fixation associated with the nodulated roots) (Unkovich *et al*., 2010) or using the IPCC methodology currently adopted by Australia (IPCC, 1997) [Eqn (1)]. It is important to note that biological N_2 fixation is not considered a source of N_2O in the IPCC's most recent guidelines for national greenhouse gas inventories (IPCC, 2006); however, some countries, such as a Australia, still utilize the former publication.

$$M_{BNF} = P \times R \times DM \times CC \times NC, \quad (1)$$

where M_{BNF} is the mass of N fixed by grain crop (kg N), P is the annual production of crop (grain, kg DM ha^{-1}), R is the residue to crop ratio (kg crop residue kg^{-1} grain; 2.1, default value), DM is dry matter content (kg dry weight kg^{-1} crop residue; 0.8, default value), CC is the mass fraction of C in crop residue (kg C kg^{-1} crop residue; 0.4, default value), and NC is the N to C ratio in crop residue (0.05, default value).

The amount of N_2O emitted by crop residues returned to soil was estimated by either summing fluxes during the postharvest phase of the current study, using the IPCC methodology currently adopted by Australia, whereby the amount of N in the aboveground residue is multiplied by 1.25% (IPCC, 1997), or using the current recommended IPCC methodology where the amount of N in the above- and belowground residue is multiplied by 1.0% (IPCC, 2006). The amount of N in the residue returned to soil was either measured by multiplying the dried biomass of the aboveground residue by the N concentration of the plant material measured in the present study, using the IPCC methodology currently adopted by Australia where stubble is not removed by burning (IPCC, 1997) [Eqn (2)], or using the current recommended IPCC methodology that accounts for belowground residues and where stubble is not removed by burning (IPCC, 2006). [Eqn (3)],

$$MR_{1997} = P \times R \times DM \times CC \times NC \times (1 - FFOD), \quad (2)$$

where MR_{1997} is the mass of N in the crop residue (kg N ha^{-1}), FFOD is the fraction of the crop residue that is removed (kg dry weight kg^{-1} crop residue; 0.09, default value), and the remaining symbols are as for Eqn (1).

$$MR_{2006} = P \times Frac_{Renew} \times \{[R \times N \times (1 - Frac_{Remove})] + [R_{BG} \times N_{BG}]\}, \quad (3)$$

where MR_{2006} is the mass of N in the crop residue (above and belowground, kg N ha^{-1}), P is the annual production of crop (grain, kg DM ha^{-1}), $Frac_{Renew}$ is the fraction of total area under the crop that is renewed annually (1, default value for annual crops), R is the residue to crop ratio (kg crop residue kg^{-1} grain; 1.98, default value), N is the N content of aboveground crop residues (kg N kg^{-1} residue DM; 0.008, default value), $Frac_{Remove}$ is the fraction of the aboveground residues removed (kg N kg^{-1} crop N; 0, default value), R_{BG} is the ratio of belowground residue to harvest yield (kg residue DM kg^{-1} crop grain DM; 0.38, default value), and N_{BG} is the N content of belowground residue (kg residue N kg^{-1} residue DM; 0.008, default value).

Results

Environmental conditions

A total of 299 mm rain fell during the study period (14 May 2008–28 April 2009), of which 206 mm was during the period between planting and harvesting the lupin (Fig. 1b). The 2008 annual rainfall for Cunderdin (355 mm, recorded at an official site nearby) was 97% of the 95-year average (1914–2008), while rainfall during

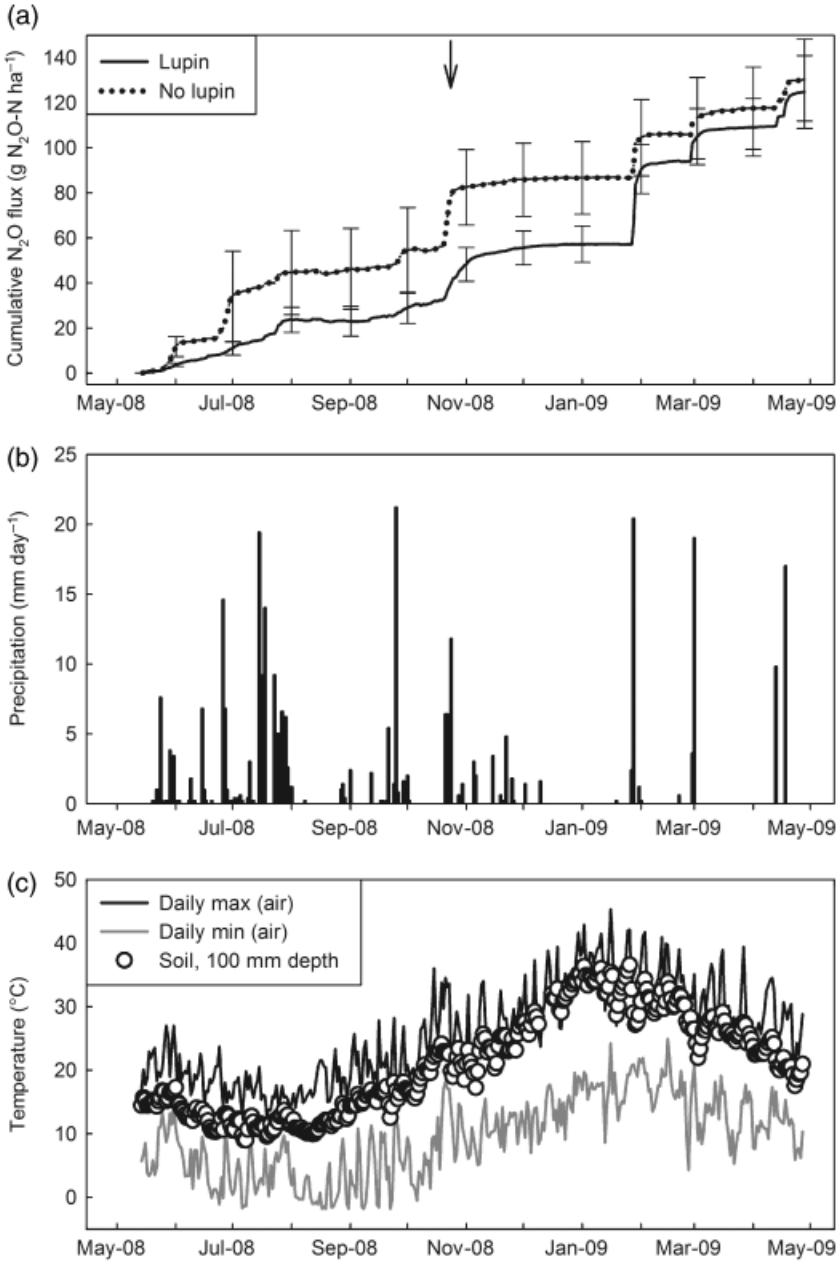

Fig. 1 Cumulative nitrous oxide (N_2O) fluxes (a), daily precipitation (b), and maximum and minimum hourly air temperature, plus average daily soil temperature (100 mm depth; c) with time at a cropped site at Cunderdin, Australia (14 May 2008–28 April 2009). The arrow indicates the timing of harvest.

the growing season (May 2007–October 2007) was 87% of the long-term average (Commonwealth Bureau of Meteorology, http://www.bom.gov.au/climate). The mean minimum daily air temperature recorded at the study site was 9.5 °C and the mean maximum daily air temperature was 25.8 °C. The lowest hourly air temperature (−1.4 °C) was recorded in June 2008, while the greatest maximum hourly temperature (45 °C) was recorded in January 2009 (Fig. 1c). Average daily soil temperatures in the surface 100 mm of the study site ranged from 9 to 36.6 °C (Fig. 1c). Soil temperatures were lowest during July 2008 (mid-winter) and highest in January 2009 (mid-summer).

Surface soil NO_3^- and NH_4^+

The amount of inorganic N ($NO_3^- + NH_4^+$) in the surface soil (0–100 mm) varied during the year, and in a

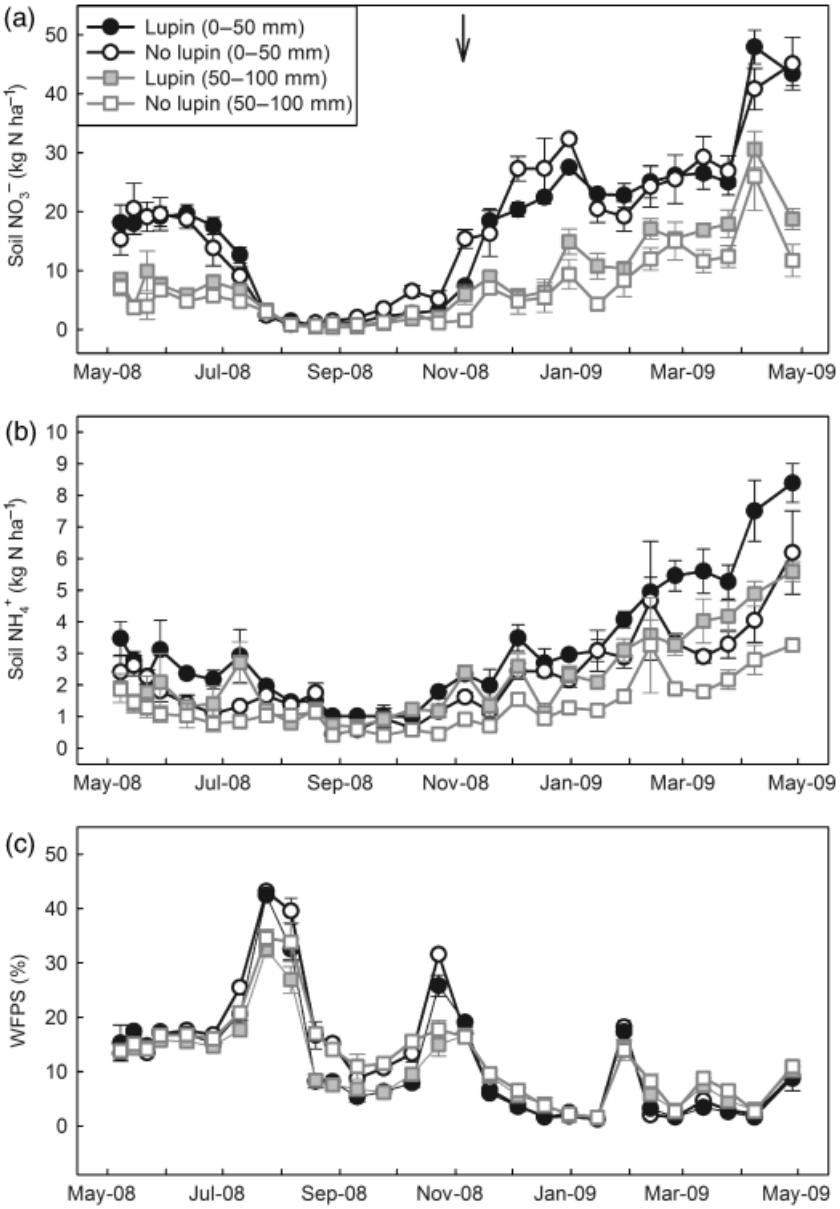

Fig. 2 Soil nitrate (NO_3^-; a), ammonium (NH_4^+; b), and water-filled pore space (WFPS; c) with time at a cropped site at Cunderdin, Australia (14 May 2008–28 April 2009). Values represent means (± standard errors) of three replicates. The arrow indicates the timing of harvest.

similar way for both treatments (Fig. 2a and b) ($P<0.05$). The surface 50 mm tended to contain more inorganic N than the 50–100 mm depth for the first 2 months following planting (May–July 2008), with both depths declining to <5 kg N ha^{-1} from late July (winter) through to late October 2008 (spring, Fig. 2a and b). Soil inorganic N in both lupin treatments (and at both soil depths) progressively increased following summer rainfall, especially soil NO_3^- in the surface 50 mm, and continued to rise following further, periodic summer and autumn rainfall. The amount of surface soil NO_3^- measured at the end of study was greater than values observed at the commencement of the study (Fig. 2a). A large proportion of inorganic N in the surface soil was in the NO_3^- form, rather than NH_4^+ (Fig. 2a and b). For example, soil NO_3^- in both treatment ranged from <1 to 48 kg N ha^{-1} in the surface 50 mm, and <1 to 31 kg N ha^{-1} in the 50–100 mm depth; while the soil NH_4^+ ranged from <1 to 8.4 kg N ha^{-1} in the surface 50 mm and <1 to 5.6 kg N ha^{-1} in the 50–100 mm depth (Fig. 2a).

The concentration of inorganic N in soil solution did not differ between the lupin and no lupin treatments

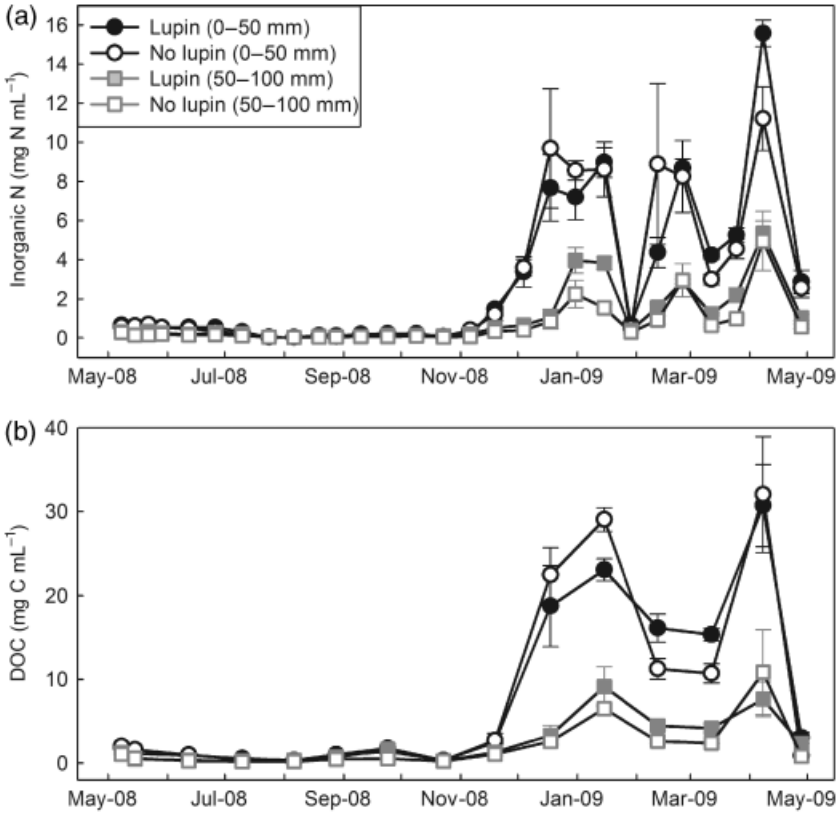

Fig. 3 Inorganic nitrogen (N) (NO_3^- + NH_4^+; a) and dissolved organic carbon (DOC; b) concentrations with time at a cropped site at Cunderdin, Australia (14 May 2008–28 April 2009). Values represent means (± standard errors) of three replicates.

($P < 0.05$), but appeared to be greater in the surface 50 mm than the 50–100 mm depth (Fig. 3a). For example, soil inorganic N concentrations in both treatment ranged from < 0.5 to 15 mg N mL^{-1} in the surface 50 mm and < 0.5 to 5 mg N L^{-1} in the 50–100 mm depth. Inorganic N concentrations were greatest during the summer when the soil was dry, but decreased following summer rainfall events.

Soil inorganic N with depth

The total amount of inorganic N contained in the surface metre of soil did not vary between the lupin and no lupin treatment, but did vary depending on the sampling dates ($P < 0.05$) (Fig. 4). The amount of inorganic N in the surface metre was larger ($P < 0.05$) at the end of study (132 kg N ha^{-1}) than at the start (49 kg N ha^{-1}) and at the flowering stage (40 kg N ha^{-1}). A large proportion (> 48%) of the inorganic N in the surface metre at the start of the study, and again at the end of the study, was in the surface 100 mm. At flowering, inorganic N declined in the surface 200 mm by 28 kg N ha^{-1}, but increased in the 200–1000 mm depth by 18 kg N ha^{-1}, and mainly due to increases in the 500–1000 mm soil depths ($P < 0.05$). Between flowering and the end of the study, there was an increase in the amount of inorganic N in each of the depth increments to 500 mm, with no changes below this depth ($P < 0.05$). The increased inorganic N content in the surface

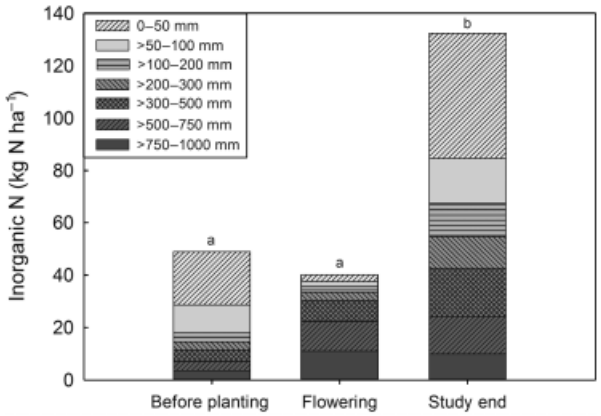

Fig. 4 Soil inorganic nitrogen (N) content (NO_3^- + NH_4^+) with soil depth before planting (8 May 2008), at crop flowering (19 August 2008), and at the end of the study (28 April 2009). Values represent means of six replicates (lupin and no lupin combined), and bars with the same letters are not significantly different ($P > 0.05$).

200 mm between flowering and the end of the study was presumably due to the mineralization of organic N following summer rainfall (compare Figs 2 and 4). Although there was generally no effect of lupin on the inorganic N content of each soil depth, at the end of the study the amount of inorganic N in the 50–100 mm soil depth was greater in the lupin treatment than in the no lupin treatment (lupin, 22 kg N ha^{-1}; no lupin 12 kg N ha^{-1}).

WFPS

Soil WFPS of the surface soil (0–100 mm) varied seasonally in response to rainfall (compare Figs 1 and 2c) ($P < 0.05$). Soil WFPS ranged from <2% to 43% in the surface 50 mm, and <2% to 35% in the 50–100 mm depth (Fig. 2c). For both treatments and sampling depths, soil WFPS was greater during the growing season when a large proportion of the annual rainfall occurred. However, soil water contents periodically increased following summer and autumn rainfall (Fig. 2c).

DOC

The concentration of DOC in soil water varied seasonally at both soil depths ($P < 0.05$), and ranged from 0.17 to 32 mg C mL^{-1} (Fig. 3b). Soil DOC was often greater in the surface 50 mm than the 50–100 mm depth, but generally did not differ between the lupin and no lupin treatments. Dissolved organic C concentrations were particularly high preceding summer (January–February) and autumn (April) rainfall, after which concentrations declined.

Plant yield, N uptake, and biological N_2 fixation

Grain yield from the lupin treatment was 1.2 t ha^{-1}, and the same as the average yield recorded for the region in 2008 (Planfarm Pty Ltd., pers. comm.). The amount of residue remaining following harvest was 2.3 t dry weight ha^{-1}, representing 66% of the total aboveground biomass. Lupin N_2 fixation during the growing season was estimated to be 67 kg N ha^{-1} (standard error, 9 kg N ha^{-1}) based on average biological N_2 fixation rates for lupin grown in Australia (Unkovich *et al.*, 2010), compared with 61 kg N ha^{-1} (standard error, 17 kg N ha^{-1}) estimated using the IPCC methodology adopted by Australia (IPCC, 1997). The amount of N actually measured in the aboveground biomass (shoot + seed) was 91 kg N ha^{-1} (standard error, 14 kg N ha^{-1}). The mass of N contained in the aboveground crop residue was 26 kg N ha^{-1} (standard error, 3.4 kg N ha^{-1}), compared with 55 kg N ha^{-1} (standard error, 15 kg N ha^{-1}) estimated using the IPCC methodology adopted by Australia (IPCC, 1997), and 22 kg N ha^{-1} (standard error, 3.3 kg N ha^{-1}) as estimated using the current IPCC methodology (IPCC, 2006).

N_2O fluxes

Daily N_2O fluxes ranged from −0.5 (October 2008) to 24 g N_2O-N ha^{-1} day^{-1} (January 2009) in the lupin treatment, and −0.7 (August 2008) to 10 g N_2O-N ha^{-1} day^{-1} (January 2009) in the no lupin treatment (Fig. 1a). Sudden increases in cumulative N_2O-N emissions occurred during summer and autumn and were generally associated with rainfall >5 mm day^{-1} (Fig. 1a). On a few occasions, emissions were less than zero indicating N_2O uptake, which is consistent with findings from other soils containing low amounts of mineral N (Rosenkranz *et al.*, 2006). Overall, daily N_2O emissions were positively correlated with WFPS ($P < 0.05$; Table 1). Hourly N_2O fluxes following summer and autumn rainfall events tended to peak on the day of rain, or the day following with greater losses from the lupin treatment than the no lupin treatment (Fig. 5). For example, hourly N_2O fluxes following summer and autumn rainfall were as high as 164 μg N_2O-N m^{-2} h^{-1} for the lupin treatment, and 95 μg N_2O-N m^{-2} h^{-1} for the no lupin treatment on 29 January 2009. Greatest hourly N_2O fluxes occurred following the first summer rainfall, and were not exceeded by subsequent rainfall of similar magnitude (Fig. 5).

The total amount of N emitted as N_2O after almost 1 year (350 days) did not differ between the lupin and no lupin treatment, with an average of 127 g N_2O-N ha^{-1} (standard error, 11 g N_2O-N ha^{-1}; $P < 0.05$) (Fig. 1a). For the lupin treatment, a large proportion (58%) of the loss occurred postharvest, whereas for the no lupin treatment only 37% of the annual loss occurred postharvest (Table 2). However, postharvest cumulative N_2O losses did not vary between treatments ($P < 0.05$). The methodology currently used by Australia for calculating N_2O emissions predicted 760 g N_2O-N ha^{-1} would be

Table 1 Correlation between daily N_2O fluxes and measured soil/environmental parameters in the surface 50 mm over the study period (14 May 2008–28 April 2009)

Parameter	*n*	*R*
Soil nitrate (NO_3^-), kg N ha^{-1}	54	−0.32
Soil ammonium (NH_4^+), kg N ha^{-1}	54	−0.26
Inorganic nitrogen, kg N ha^{-1}	54	−0.32
Water-filled pore space (WFPS), %	54	0.44**
Soil temperature (50 mm), °C	686	−0.05

**Values are significantly different from zero at the 0.05 and probability levels.

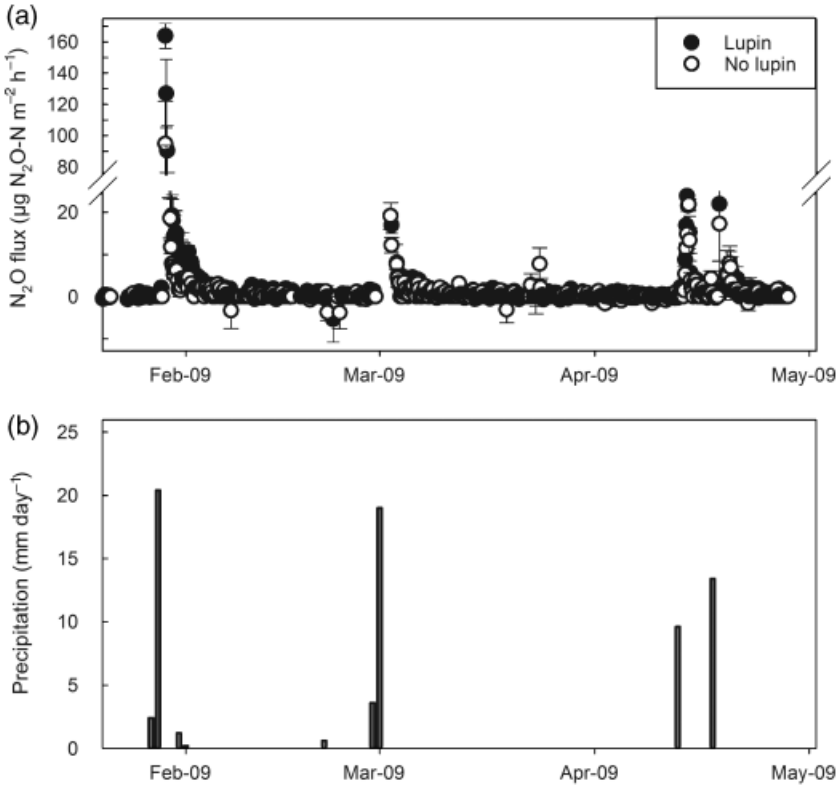

Fig. 5 Hourly nitrous oxide (N_2O) fluxes (a) following a series of summer and autumn rainfall events (b) at a cropped site at Cunderdin, Australia (20 January 2009–28 April 2009). Values represent means (± standard errors) of three replicates.

Table 2 Measured and IPCC predicted soil N_2O (g N_2O-N ha^{-1}) emissions during the growing season and postharvest period of a soil either cropped to lupin or left bare

	Growing season* (g N_2O-N ha^{-1})		Postharvest† (g N_2O-N ha^{-1})		
Treatment	IPCC (1997)‡	Measured	IPCC (1997)‡	IPCC (2006)§	Measured
Lupin	760 (211)	52 (7.3)[a]	692 (192)	225 (33)	73 (11)[a]
No lupin (bare)	na	83 (17)[a]	na	na	47 (9.1)[a]

Values represent means (standard errors) of three replicates. For each column, values followed by the same letter are not significantly different ($P < 0.05$).
*Growing season, 14 May 2008 (planting) to 5 November 2008 (harvest).
†Postharvest, 5 November 2008 (harvest) to 28 April 2009 (completion of study).
‡Calculated using IPCC method published in 1997 (IPCC 1997), which is the methodology currently utilised by Australia.
§Calculated using IPCC method published in 2006 (IPCC 2006), which is the most recently published methodology and omits N_2O emissions from biological N_2 fixation.
na, not applicable.

emitted from biological N_2 fixation during the growing season, and 692 g N_2O-N ha^{-1} would be emitted from the decomposition of residue, giving a predicted annual emission of 1452 g N_2O-N ha^{-1} from lupin production in the present study (Table 2). Whereas the current IPCC methodology predicted a total of 225 g N_2O-N ha^{-1} would be emitted from lupin production as a result of residue decomposition (Table 2).

Discussion

N_2O fluxes from grain–legume crops

Annual N_2O fluxes calculated using subdaily measurements from a rain-fed, grain–legume crop grown in a semiarid region have not been reported previously (Stehfest & Bouwman, 2006; Galbally *et al*., 2008).

Although annual N_2O emissions have been reported from haricot beans grown in a semiarid environment dominated by summer rainfall, these losses were based on only 15 measurements and were extensively grazed during the dry season (Dick *et al.*, 2008). N_2O fluxes from grain–legumes have also been measured in Canadian semiarid environments, but the data are largely unpublished (Rochette & Janzen, 2005). Investigations of N_2O fluxes from grain–legume crops have mainly been confined to soybean in temperate climates (Rochette & Janzen, 2005; Parkin & Kaspar, 2006; Stehfest & Bouwman, 2006), with most measurements conducted for <12 months.

N_2O fluxes from a rain-fed, lupin crop in a semiarid climate appear to be low, and less than grain–legume crops grown in other climates. In the present study, daily fluxes ranged from -0.5 to 24 g N_2O-N ha^{-1} day^{-1} (median, 0.1 g N_2O-N ha^{-1} day^{-1}) for the lupin treatment, culminating in an annual loss (125 g N_2O-N ha^{-1}) not different from the no lupin treatment (130 g N_2O-N ha^{-1}). Daily losses reported in the present study are comparable with those reported for a grain–legume crop grown without N fertilizer in a semiarid region of Mali, where daily fluxes ranged from -0.05 to 5 g N_2O-N ha^{-1} day^{-1} (Dick *et al.*, 2008). In other climates, daily N_2O fluxes (or equivalent) reported for unfertilized grain–legume crops have ranged from 0.3 to 25 g N_2O-N ha^{-1} day^{-1} for field peas grown in a temperate region of Belgium (Goossens *et al.*, 2001), 0 to 43 g N_2O-N ha^{-1} day^{-1} for soybean grown in a temperate regions of the United States (Bremner *et al.*, 1980; Jacinthe & Dick, 1997), with reports of >100 g N_2O-N day^{-1} for soybean grown in humid continental climates in north America (MacKenzie *et al.*, 1998; Rochette *et al.*, 2004; Parkin & Kaspar, 2006). Only a limited number of studies have had sufficient data to calculate annual N_2O fluxes from grain–legume crops, with losses ranging from <50 to ca. 8000 g N ha^{-1} yr^{-1} (Bremner *et al.*, 1980; Rochette & Janzen, 2005; Parkin & Kaspar, 2006; Dick *et al.*, 2008), with the greatest annual flux reported for soybean grown in a fine-loamy soil in central Iowa, USA (Parkin & Kaspar, 2006). The annual N_2O flux from the aforementioned study conducted in Mali was greater (635 g N_2O-N ha^{-1}) than the present study, despite the low daily losses, but presumably because the annual flux was calculated using the arithmetic mean flux of 15 daily measurements (Dick *et al.*, 2008). A review of N_2O flux data from legumes concluded that the mean annual emission from annual legume crops was 1.0 kg N ha^{-1} (Rochette & Janzen, 2005).

N_2O fluxes from grain–legume crops grown in semiarid regions are similar to losses from rain-fed, N fertilized crops grown in the same environment. For example, the annual N_2O flux from a legume crop in the current study (125 g N_2O-N ha^{-1} yr^{-1}) was similar to those rates reported from the same site when cropped previously to N fertilized wheat and canola (110 to 128 g N_2O-N ha^{-1} yr^{-1}; 75–100 kg N ha^{-1} yr^{-1} applied as urea depending on the year) (Barton *et al.*, 2008, 2010). Similarly, recent papers and reviews of N_2O fluxes from various agro-ecosystems have concluded that there is a tendency for legume crops to emit similar, if not less, N_2O than fertilized nonlegume crops (MacKenzie *et al.*, 1998; Rochette *et al.*, 2004; Helgason *et al.*, 2005; Rochette & Janzen, 2005; Parkin & Kaspar, 2006; Dick *et al.*, 2008). Our findings support the general observation that under similar climatic and management regimes, N_2O fluxes from legume cropping systems will not necessarily be greater than emissions from N fertilized nonlegume crops.

Sources of N_2O fluxes

N_2O emissions from semiarid soils cropped to legumes are most likely to be derived from plant residue decomposition rather than biological N_2 fixation *per se*. During the growing season in the present study, N_2O fluxes from the no lupin soil were similar to fluxes from the soil cropped to a grain–legume. This suggests that a large proportion of N_2O emitted during crop growth was from the turnover of soil organic matter and previous crop residues, rather than biological N_2 fixation. Numerous studies and reviews have also concluded that there is limited evidence that biological N_2 fixation is a significant source of N_2O emissions from cropped soils (Helgason *et al.*, 2005; Rochette & Janzen, 2005; Peoples *et al.*, 2009).

N_2O emissions from the decomposition of legume residues appear to be low from semiarid environments and not as significant as those reported from agricultural systems with higher rainfall (Rochette *et al.*, 2004; Helgason *et al.*, 2005; Rochette & Janzen, 2005). Daily N_2O fluxes following lupin harvest ranged from -0.5 to 24 g N ha^{-1} from the lupin plots in the present study, and were rarely greater than the no lupin treatment with cumulative postharvest losses not different between the lupin treatments. Low daily N_2O losses following harvest in the present study could be attributed to low postharvest rainfall, not incorporating the residue into the bulk soil following harvest, and low soil organic carbon content. Residues are not typically incorporated into the poorly structured soils of Western Australia following harvest to minimize soil structure disturbance and erosion. The influence of legume crop residues on N_2O fluxes was only investigated for approximately months following harvest in the present study, and there is a possibility that N_2O emissions from the lupin treatment may have been greater than the no

lupin treatment the following winter. Yet, others have shown including grain–legume in cropping rotation does not necessarily increase cumulative N_2O emissions from the subsequent crop (Parkin & Kaspar, 2006; Jantalia *et al*., 2008; Guo *et al*., 2009). Studies that have reported elevated N_2O emissions following the harvest of a leguminous crop (no N applied) have either incorporated or surface mulched the residue into relatively fertile surface soils, and then allowed the soil to remain fallow at times of the year when N_2O emissions are likely to be greatest (e.g., following spring thaw or summer in a temperate climate) (Wagner-Riddle *et al*., 1997; Wagner-Riddle & Thurtell, 1998; Baggs *et al*., 2003). Consequently, we do not expect that the decomposition of unincorporated legumes residues will greatly enhance N_2O emissions from low organic matter, cropped soils in semiarid climates. Further investigation of the effect of legume residue, and residue management, on N_2O fluxes from semiarid soils is recommended given the complexity of the interactions between residue management and tillage over time and space, and the paucity of data from these regions.

The particularly low N_2O fluxes reported in this paper, and in the previous studies conducted at the same site (Barton *et al*., 2008, 2010), may be due to a high production of NO relative to N_2O. Wetting of dry soils after an extended hot and dry period has shown to rapidly increase NO relative to N_2O fluxes (Scholes *et al*., 1997; Butterbach-Bahl *et al*., 2004; Galbally *et al*., 2008; Hall *et al*., 2008) with nitrification considered to be the likely source in well-aerated soils (Garrido *et al*., 2002; Galbally *et al*., 2008). Soil water content is considered to be a critical factor determining the rate of NO flux in semiarid regions, and is also strongly influenced by N availability and soil temperature in other climates (Stehfest & Bouwman, 2006; Galbally *et al*., 2008). The proportion of NO : N_2O tends to vary with soil type and soil water content; thus, using an empirical relationship to predict NO fluxes from the observed N_2O or CO_2 emissions is difficult (Garrido *et al*., 2002; Butterbach-Bahl *et al*., 2004). Galbally *et al*. (2008) suggest NO fluxes from semiarid and arid regions may represent as much as 20% of the global loss. Increasing the number of NO measurements from semiarid agricultural soils is required to decrease the uncertainty of global NO estimates (Stehfest & Bouwman, 2006).

Predicting N_2O emissions using IPCC methodology

The proportion of biologically fixed N_2 by the lupin that was finally emitted as N_2O was low, and 12-fold less than that currently predicted by Australia using the *Revised 1996 Guidelines for National Greenhouse Gas Inventories* (IPCC, 1997). The discrepancy between the IPCC predicted value and that measured in the present study comes from overestimating N_2O emissions from both the biological N_2 fixation process and the decomposition of crop residues (Table 2). As discussed previously, numerous studies have concluded that there is limited evidence to suggest that biological N_2 fixation is a source of N_2O emissions *per se*, and consequently the latest guidelines published by the IPCC no longer includes a provision for N_2O emissions from biological N_2 fixation (IPCC, 2006). Instead, it is currently recommended that N_2O emissions induced by the growth of legume crops are estimated solely from the decomposition of crop residues (IPCC, 2006). Findings from the present paper support removing biological N_2 fixation as a source of N_2O emissions from national greenhouse gas inventories.

N_2O emissions from crop residue decomposition are overestimated by both the IPCC methodology currently adopted by Australia (IPCC, 1997), and to a lesser extent by the current methodology recommended by the IPCC (2006) in the present study (Table 2). The method used by Australia overestimates emissions for a number of reasons. Firstly, the predicted N contained in the residue is more than double the measured amount due to discrepancies in the residue to crop ratio (*R*), the N to C ratio in crop residue (NC), and the fraction of crop removal (FFOD; Table 3). Secondly, the emission factor (1.25%) for predicting the proportion crop residue N emitted as N_2O also overestimates fluxes. For example, multiplying the measured N content of the residue ($26\,kg\,N\,ha^{-1}$) by 1.25% predicts that $325\,g\,N_2O\text{-}N\,ha^{-1}$ will be emitted from residue decomposition, which is more than four times the measured postharvest emissions ($73\,g\,N\,ha^{-1}\,yr^{-1}$). The recommended IPCC method (IPCC, 2006) for predicting N_2O emissions from crop residue overestimates losses because the emission factor is too high for semiarid environments (1.0%). For example, if the current Australian emission factor for N_2O emissions from synthetic fertilizer (0.3%) was also used to calculate N_2O emissions from the crop residues in the present study ($26\,kg\,N\,ha^{-1}$), then the predicted N_2O emissions ($78\,g\,N\,ha^{-1}\,yr^{-1}$) would be similar to the measured postharvest emissions ($68\,g\,N\,ha^{-1}\,yr^{-1}$). Further research is needed to confirm that the current IPCC methodology accurately predicts the contribution of different types of legume residues to N_2O emissions in semiarid environments, especially when the residue is not incorporated into the soil.

Factors regulating N_2O fluxes and management implications

N_2O fluxes from a rain-fed legume crop in a semiarid region are regulated by the same factors as nonlegume

Table 3 A comparison between the IPCC default (IPCC 1997, 2006) and measured crop attributes used for the prediction of soil N_2O emissions from biological N_2 fixation and the decomposition of pulse crop residues

Crop attribute	IPCC (1997) value*	IPCC (2006) value†	Measured value
Residue to crop ratio (R, kg residue kg^{-1} crop grain)	2.1	1.98	1.35 (0.03)
Dry matter content (DM, kg dry weight kg^{-1} crop residue)	0.8	na	nd
Mass fraction of C in aboveground crop residue (CC, kg C kg^{-1} crop residue)	0.4	na	0.45 (0.22)
Mass fraction of N in aboveground crop residue (N, kg N kg^{-1} crop residue DM)	na	0.008	0.010 (0.00)
N : C in crop residue (NC)	0.05	na	0.022 (0.00)
Fraction of the crop residue removed (FFOD, kg dry weight kg^{-1} crop residue)	0.09	na	0.0
Fraction of the aboveground residue removed ($Frac_{Remove}$, kg residue N kg^{-1} crop N)	na	0	0
Ratio of belowground residue to harvest yield (R_{BG}, kg residue DM kg^{-1} crop grain DM)	na	0.38	nd
N content of belowground residues (N_{BG}, kg residue N kg^{-1} residue DM)	na	0.008	nd

*Derived from the IPCC method published in 1997 (IPCC 1997), which is the methodology currently utilised by Australia.
†Derived from IPCC method published in 2006 (IPCC 2006), which is the most recently published methodology and omits N_2O emissions from biological N_2 fixation.
na, not applicable; nd, not determined.

crops grown in similar environments. Greatest hourly fluxes followed summer and autumn rainfall, coinciding with elevated soil water contents, greatest inorganic N and DOC soil solution concentrations, and mild to warm (i.e., $>25\,^{\circ}C$) soil temperatures in the surface 100 mm, conditions universally understood to promote N_2O fluxes. Correlation analysis shows daily N_2O fluxes to be positively influenced by WFPS (Table 1), which is often the key factor in temperate agricultural soils (e.g., Dobbie *et al*., 1999). However, the magnitude of hourly N_2O fluxes were not necessarily proportional to the size of the rainfall event, with the greatest flux occurring following the first of the summer rains (Fig. 5), and is consistent with our previous findings and that of others (Davidson *et al*., 1993; Scholes *et al*., 1997; Barton *et al*., 2008, 2010). This suggests that N_2O fluxes following summer rainfall events may also be driven by substrate availability and not soil WFPS alone. Despite the N_2O response to summer rainfall, hourly fluxes were still relatively low in comparison with fluxes reported for temperate agriculture systems; which can probably be attributed to the low biological activity and the coarse texture of the soil. Sandy soils tend to dry rapidly following rainfall, and conditions conducive to microbial activity, and thus N_2O production would be expected occur for a limited period of time after each rainfall event.

Cumulative N_2O emissions from summer and autumn accounted for 58% of the annual emission from the legume treatment, with 31% of the annual rainfall occurring during this time. A similarly pronounced N_2O flux response to summer rainfall was observed at the same site when planted to wheat and canola (Barton *et al*., 2008, 2010), as well as in a semiarid soil in Mali cropped to a grain–legume (Dick *et al*., 2008), and has been discussed in a review of trace gas fluxes from semiarid and arid zones (Galbally *et al*., 2008). We hypothesized previously that the rapid increase in soil N following summer–autumn rainfall, and a subsequent increase in N_2O emissions, can be attributed to the mineralization of readily decomposable organic matter following the wetting of dry soil (van Gestel *et al*., 1993; McNeill *et al*., 1998; Murphy *et al*., 1998). Mineralized N is especially at risk of being emitted as N_2O in the present environment, as there is no active plant growth to compete with soil microorganisms for inorganic N during summer and early autumn. Laboratory-based research has also shown that N mineralization is likely to exceed N immobilization rates at the soil temperatures recorded in summer (Hoyle *et al*., 2006), further increasing the availability of inorganic N for nitrification and denitrification. Developing strategies that retain or utilize mineralized N before it is emitted as N_2O requires a better understanding of soil N dynamics following summer rainfall.

Developing mitigation strategies for N_2O emissions from the soil surface in semiarid, rain-fed crop may not be considered to be a high priority given the magnitude of the losses; however, semiarid regions represent a significant global land area. Most approaches to

decrease N_2O emissions from cropped soils focus on improving N fertilizer use efficiency, by fine-tuning plant growth-limiting factors and improving the synchrony between plant N uptake and N supply from all sources (e.g., fertilizer, mineralization, and fixation). These approaches are unlikely to have a significant impact on N_2O emissions from legume-cropped soils in semiarid regions of Australia, as negligible amounts of N fertilizer is applied to these crops, and a significant proportion of the annual N_2O is emitted postharvest, and when the soil is fallow. We expect that nitrification activity is the main source of N_2O emissions in the present study as soil WFPS did not exceed values expected to promote denitrification in a sandy textured soil (>80% WFPS; Barton *et al*., 1999). If nitrification is the main source of N_2O following summer rainfall, then losses may be lowered by either decreasing the availability of NH_4^+, the portion of nitrified N emitted as N_2O, or decreasing nitrification activity overall. Developing strategies that retain or utilize mineralized N, and before it is nitrified, may prove challenging.

Conclusions

This study reports daily N_2O fluxes from a rain-fed, legume crop grown in a semiarid region. Daily N_2O fluxes were low (-0.5 to $24\,g\,N_2O\text{-}N\,ha^{-1}\,day^{-1}$), not different between the legume cropped and bare soil, and culminated in an average annual loss of $127\,g\,N_2O\text{-}N\,ha^{-1}$. Greatest daily N_2O fluxes occurred following harvest when the soil was fallow, and following a series of summer rainfall events. However, N_2O fluxes following summer rainfall were still relatively low in comparison with fluxes reported for temperate agriculture systems, which is attributed to the low biological activity and the coarse texture of the soil. The contribution of the biological N_2 fixation process to N_2O emissions appeared negligible in the present study, further supporting its omission as a source of N_2O from the currently recommended IPCC methodology (IPCC, 2006). N_2O emissions from the decomposition of legume crop residue were also low, and less than that predicted using the current IPCC methodology (IPCC, 2006). Further research is needed to refine the contribution of legume residues to N_2O and NO fluxes in semiarid environments, especially when tillage is minimized and the residue is not incorporated into the soil following harvest.

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