

Nitrous oxide dynamics in managed northern forest soil profiles: is production offset by consumption?

L. Kellman · K. Kavanaugh

Received: 18 January 2008 / Accepted: 15 August 2008 / Published online: 9 September 2008
© Springer Science+Business Media B.V. 2008

Abstract This study investigates soil N₂O dynamics in forest soils representing early (3-years) and late (>50 years) post-harvest succession in Atlantic Canada over a 9-month snow-free period in order to develop a better understanding of the role of managed forests as sources and sinks of N₂O. We couple measurement of surface flux with detailed measurements of subsurface N₂O concentrations at four mineral soil depths (0, 5, 20 and 35 cm) at 40 plots located within four sites. Median surface fluxes were similar at all sites regardless of the management stage (−5 to +19 $\mu\text{gN}_2\text{O-N/m}^2/\text{day}$), with all sites behaving as net sinks and sources of N₂O over the measurement period. Subsurface mineral soil N₂O concentrations at early (3-year) post-harvest succession sites, which ranged from median values of 362 ppbv at 0 cm to 1783 ppbv at 35 cm depth, were significantly higher than late post-harvest succession sites where median concentrations ranged from 329 ppbv at 0 cm to 460 ppbv at 35 cm depth. Examination of relationships between subsurface gas storage and surface flux magnitudes, suggested although recently harvested forest soils may be producing N₂O at a greater rate than mature forest soils, observed patterns are consistent with a strong sink for this gas that prevents its conservative

transport through the soil profile, and ultimate emission to the atmosphere through the majority of the measurement period.

Keywords Nitrous oxide · Forest management · Greenhouse gases · Climate change

Introduction

Globally, soils are the largest source of atmospheric nitrous oxide (N₂O) (Soloman et al. 2007), and N₂O soil emission studies to date have placed a major emphasis on quantifying emissions from anthropogenically altered agricultural soils, especially in the tropics (Goodroad and Keeney 1984; Matson and Vitousek 1990; Teepe et al. 2000; Keller and Reiners 1994; Keller et al. 1993). Recently, the issue of N₂O emissions from northern forest soils has been receiving more attention (Corre et al. 1999; Butterbach-Bahl et al. 2002; Schulte-Bisping and Brumme 2003; Wolf and Brumme 2003), but high uncertainties in northern forest soil N₂O budgets exist, due in large part to a lack of information about soil N₂O processes, high spatial and temporal variabilities in soil fluxes, and a paucity of field data (Ambus et al. 2006). Studies show that in intact forests, particularly those that are N-limited, soils tend to behave as both sources and sinks of N₂O (Castro et al. 1993; Corre et al. 1999; Bowden et al. 1991, 2000). A number of European ecosystem studies have documented

L. Kellman (✉) · K. Kavanaugh
Environmental Sciences Research Centre, St. Francis
Xavier University, Antigonish, NS, Canada B2G 2W5
e-mail: lkellman@stfx.ca

elevated N_2O fluxes due to N deposition (Butterbach-Bahl et al. 1997, 2002; Zechmeister-Boltenstern et al. 2002; Papen and Butterbach-Bahl 1999), although some exceptions to this observed trend apply (Borken and Beese 2005).

The aerobic and anaerobic processes of nitrification and denitrification that generate soil N_2O (Bouwman 1990), driven largely by soil moisture and temperature, often occur simultaneously in soils (Davidson et al. 2000), where there may exist strong spatial gradients in soil moisture and organic matter distribution (ex. Ambus and Christensen 1995). Overall, N_2O emissions from soils have been shown to be highly variable (Schlesinger 1997; Livingston and Hutchinson 1995; Ambus and Christensen 1995; Robertson and Tiedje 1984; Goodroad and Keeney 1984). In a recent review, Chapuis-Lardy et al. (2007) point out that efforts to examine the N_2O source potential of soils often results in attributing observations of negative fluxes to experimental noise. In fact, the potential of soils to behave as sinks for atmospheric N_2O (Cicerone 1989) may have important implications for global budgets. Chapuis-Lardy et al. (2007) suggest there is a strong likelihood that N_2O consumption is often masked by its production in soils. As a consequence, reported flux magnitudes of N_2O represent a balance between soil source and sink activity, rather than a gross estimate of soil N_2O production.

While increases in available soil N following forest harvesting activities such as clear cutting are well documented, there exists debate about the relative importance of reduced root competition versus increases in net soil N mineralization in making this N available (Burns and Murdoch 2005; Vitousek 1981; Vitousek and Matson 1985). The ‘hole-in-the-pipe’ conceptual model of N-cycling proposed by Firestone and Davidson (1989) would suggest that N_2O production in soils should also increase post-harvest with increased available inorganic N. A number of studies have confirmed this through documentation of increased N_2O soil emissions with the addition of inorganic N to the soil surface (Melillo et al. 1989; Brumme and Beese 1992; Matson et al. 1992; Papen et al. 2001). There exist only a few field studies that have investigated the effects of harvesting on soil N_2O emissions from northern forest soils however (Schiller and Hastie 1996; Zerva and Mencuccini 2005; Huttunen et al. 2003; Tate et al. 2006; Bowden and Bormann 1986),

with little consensus arising from these studies about flux magnitudes.

Process-based investigations of N_2O dynamics need to be carried out in-situ in order to advance our current understanding of the role soils play as sinks and sources of this gas. While surface flux studies provide essential information about the net balance between soils and the atmosphere, they provide limited information about the gross levels of N_2O production and consumption in soils (Chapuis-Lardy et al. 2007). The coupling of subsurface gas profile data with surface flux data in the field has the potential to further our understanding of the relative importance of these production and consumption processes in soils. This approach has been applied successfully in studies of subsurface greenhouse gas production dynamics in soils for other greenhouse gases, most notably CO_2 . Specifically, measuring subsurface gas concentration gradients and coupling these with estimates of gas transport rates (diffusivity) can yield more information about depths of gas production and quantitative relationships to soil environmental factors (Burton and Beauchamp 1994; Risk et al. 2002a, b, 2008). While vertically distinct gross soil gas production estimates can be obtained for gases such as CO_2 that are transported conservatively through the vertical soil profile, applying similar approaches to N_2O can only be carried out if it can be established that soils are not behaving as a significant sink for N_2O during transport. In cases where it is not possible to make this assumption, the subsurface gas concentration data can be used to qualitatively examine subsurface gas production and consumption dynamics. Since soil N_2O is generated by both aerobic and anaerobic processes, subsurface concentration data may yield information about where in the soil profile the gas is being generated and/or consumed and the mechanisms responsible for these processes.

The goal in this study is to examine the utility of the subsurface gas profile approach as a tool for understanding in-situ soil N_2O dynamics in managed northern forest soils. In order to accomplish this, we couple subsurface concentration profile data with surface flux data from 10 plots at each of 4 sites representing early stages (3 years after harvesting) and late stages (intact forest > 50 years of age) of post-harvest forest succession, typical of forests in the Acadian Forest Region of Atlantic Canada. The study

is carried out over a 9-month snow-free period in 2005. We examine relationships between surface flux, temperature and moisture, and use corresponding soil N₂O profile concentration data from each surface flux measurement plot to investigate: (1) the patterns of subsurface concentration gradients relative to atmospheric concentrations, (2) whether there is evidence that consumption plays an important role during N₂O transport to the soil-atmosphere interface in these soils, and (3), whether the assumption of conservative gas transport, necessary for the application of a gross gas production model in these soils, holds.

We expect that if these soils are producing N₂O we should observe a positive subsurface concentration gradient (i.e. concentrations greater than atmospheric), and interpret a negative concentration gradient relative to atmospheric, as evidence that N₂O consumption is the dominant process in these soils. We expect to observe a positive relationship between quantities of N₂O stored in the soil profile and surface flux magnitudes (based upon the assumption that gas transport is driven by a positive concentration gradient) if gas transport is conservative and consumption not a process that dominates the gas exchange signal. Non-conservative transport of N₂O would be demonstrated by a disconnection between these two variables.

Study site and methods

The study was conducted from March to November 2005 at two paired sites representing early (3 years) and late stages (>50 years) of post-harvest succession typical of the Acadian Forest Region in Northeastern Nova Scotia, Canada. These sites are representative of forest soils in the regions, that are predominantly managed, with clear cutting as the primary harvesting mechanism. The paired sites, Lakevale (45°45'6" N, 61°56'46" W) and Pomquet (45°39'22" N, 61°50'32" W), are located at a 20 km distance from each other, while at each site the forest-clear cut pair is separated by several hundred meters to several kilometers. Both sites are within 60 m of sea level and encompassing minimum areas of 5 hectares. These sites are situated in part of the Acadian Forest Region of Atlantic Canada that were cleared for agriculture during early European settlement, but which have since undergone reconversion to forest

(Johnson 1986). The Lakevale paired site soils (LF—mature forest; LCC—3 year post-clearcut forest) are classified as Millbrook soils with brown loam over reddish brown gravely clay loam formed on a parent material of brown shales and sandstone (Cann and Hilchey 1955). The Pomquet paired site soils (PF—mature forest; PCC—3 year post-clearcut forest) are classified as Queens soils with light brown clay loam over reddish brown clay loam formed on a parent material of dark reddish brown clay loam till derived from brown shale (Cann and Hilchey 1955). This soil contains distinct gray and yellow mottles, indicating some fluctuation in water table depth annually. At the specific study locations, soil textural analysis confirmed a sandy textured soil at Lakevale and clayey textured soil at Pomquet. The finer textured Pomquet soils are more poorly drained than the sandy Lakevale soils (Bekele et al. 2007). All soils are seasonally wet, with water tables commonly within the upper 0.5 m of the soil surface during the cooler part of the year, and typically within 1m of the soil surface through the majority of the year. The pH of the Millbrook soils is between 3.7 and 4.6, while the pH of the Queens soils is between 4.6 and 4.7 (Cann and Hilchey 1955). Soil nitrogen supply rates were measured in surface soils at the sites using Plant Root Simulator Probes (PRS-Probes, Western Ag Innovations). Significant differences between N supply rates of young vs. old sites was not observed with supply rates ($\mu\text{g}/10\text{ cm}^2/55\text{ day}$ fall incubation period; 3 sets of 4 cation/anion pairs at each site) for nitrate of 3 ± 0.5 , 11 ± 6 , 13 ± 6 and 4 ± 0.4 at LCC, LF, PCC and PF respectively, and supply rates of ammonium of 3 ± 0.3 , 14 ± 4 , 14 ± 9 and 5 ± 0.5 respectively.

The forest at LF is approximately 85 years old and consists of Balsam fir (*Abies balsamea* (L.) Mill, 38%), Red spruce (*Picea rubens* Sarg., 35%) and White spruce (*Picea glauca* Moench Voss, 11%). The LCC site was clear cut in the spring of 2002 and sprayed with a herbicide (Vision glyphosate (N-phosphonomethyl glycine), Monsanto Corp., St. Louis, MO) in late summer 2003 to hinder growth of deciduous plants. The new growth consists of a mixture of Raspberry (*Rubus idaeus* L.), Red Maple (*Acer rubrum* L.), and Trembling Aspen (*Populus tremuloides* Michx.). The stand at PF is approximately 55 years old, and consists of mainly Red spruce (86%). Other plants at PF include Trembling Aspen (5%), Sugar Maple (*Acer saccharum* Marsh,

4%) and Paper Birch (*Betula papyrifera* Marsh, 4%). The PCC site was clear cut in the spring of 2002 and was beginning to regenerate with ash (*Froxinus*) and spruce (*Picea*) seedlings. No herbicide was applied to this site.

Both paired sites receive mean annual precipitation of 1290 mm and have mean annual air temperatures of 5.5°C. The region is described as humid to perhumid with a deficiency of moisture during the summer months with potential evapotranspiration rates often exceeding precipitation from May to August (Cann and Hilchey 1955).

Meteorological stations

Each site (LF, LCC, PF and PCC) was equipped with a Campbell Scientific meteorological station continuously monitoring meteorological variables including air temperature at 2 m above the soil surface, soil temperatures at 5 cm depth below the organic–mineral soil interface and soil moisture (using 30 cm long TDR probes installed on an angle to obtain volumetric soil water content) in the upper soil profile (Beltrami and Kellman 2003). The meteorological data were collected immediately adjacent to the grids of individual plots at each site and assumed representative of the site characteristics. Analyses of the plot-specific fluxes and subsurface concentrations that yielded non-significant relationships to microtopography at the site (data not shown), suggested this was a reasonable assumption. Volumetric soil moisture was converted to percent water filled pore space (%WFPS) (Davidson et al. 2000) based upon total pore space estimates for the purpose of quantifying relationships between moisture, temperature and surface flux between sites.

Gas sampling, design and analysis

At each of the 4 sites, gas sampling was carried out on a grid of 10 fixed plots with adjacent plots separated by a distance of approximately 10 m. Topography at the sites was relatively flat and plot grids were located on flat or slightly sloping terrain within each site, covering a representative range of microtopographic positions. Surface flux gas samples were collected in N₂ purged and evacuated Labco Exetainers from non-steady state vented chambers (diameter 45 cm; volume 0.025 m³) deployed on

permanently installed collars (to 5 cm depth) over a period of 60 min at all 4 sites. The collars were installed directly above the subsurface samplers described below. On several occasions at the beginning of the sampling period (April and May), collars were not used and chambers were inserted directly into the soil. Concern that flux magnitudes might be underestimated due to a poor seal between the chambers and soil which could result in a dilution of headspace gas concentrations with atmospheric air, led us to install permanent collars in May 2005.

Subsurface soil gas samples were collected at each plot immediately below the surface flux chambers by sampling air from individual gas samplers that were permanently installed at depths of 0, 5, 20 and 35 cm within the mineral soil (0 cm represents the organic–mineral interface) for a total of 40 samplers per site and 160 across all 4 sites. These soil gas samplers are constructed of 50 cm long PVC tubes with a 1.2 cm inner diameter and internal volume of 56.5 cm³ covered in a water resistant porous membrane. The samplers were installed horizontally within the mineral soil after digging soil pits and drilling 50 cm long holes into the side of each pit at each of the 4 depths. The individual samplers were therefore located within a 50 cm wide undisturbed area of the soil profile and were installed along the same vertical section of the pit profile. The soil was immediately replaced in the soil pits in the same order it was removed. Lengths of microbore tubing (2 set of tubing per sampler in order to ensure the presence of a backup should one fail) sealed to the subsurface gas samplers, connected subsurface samplers to the soil surface, where they were secured to a post and housed in water tight boxes. The ends of the microbore tubing were fitted with 3-way valves that allowed sampling of subsurface gases at the soil surface. During subsurface gas sample collection, a volume of gas greater than the volume of microbore tubing was extracted and purged, with samples subsequently collected in N₂ purged and evacuated Labco Exetainers. Valves were kept closed between sampling in order to ensure subsurface gas samplers were not contaminated with atmospheric air. Plot specific square meter soil profile N₂O storage estimates ($\mu\text{gN}_2\text{O-N/m}^2$) were generated from subsurface gas concentration profiles and estimates of fractions of air filled pore space obtained from moisture probes. These estimates provided a single

integrated measure of profile subsurface N₂O concentration information to 35 cm depth in the mineral soil for the purposes of examining relationships to surface flux. For a layered soil profile, storage (S_{N_2O}) ($\mu\text{gN}_2\text{O-N/m}^2$), was calculated according to:

$$S_{N_2O} = \sum_{i=1}^{\max} (\Theta_{V_{\max}} - \Theta_{V_{\text{obs}}})_i \times M_i \times z_i, \quad (1)$$

where for each layer, i , $\Theta_{V_{\max}}$ and $\Theta_{V_{\text{obs}}}$ are the maximum (saturated) and observed volumetric water contents (v/v), M is the mass of N₂O–N in a known volume ($\mu\text{gN}_2\text{O-N/m}^3$) obtained from point measurements of N₂O–N concentrations within this layer, and z is the layer depth (m).

Surface flux and subsurface soil air samples were collected on a minimum of seven to nine occasions at each site between March and November 2005. Paired sites were sampled on the same day at approximately the same time of day (between 10 am and 2 pm) with the exception of two occurrences where subsurface soil air samples were taken the day after surface flux (June 3rd and July 12th, 2005) at LCC and LF. All gas samples were returned to the Environmental Sciences Research Centre and analyzed on a Shimadzu Gas Chromatograph 8A fitted with an electron capture detector (ECD) within approximately 5 days of sample collection. Standard curves were established for samples run at the start of each day (Matheson Tri-Gas N₂O standards). Repeatability of gas standards was typically represented by a coefficient of variation of less than 5%. In order to ensure there was no instrument drift, multiple gas standards were run every 10–15 samples and a full set of standards were run again at the end of the day. It is estimated that approximately 30% of the instrument analysis time was spent running standards on a typical day. The combined sampling errors associated with gas sample collection, and transfer from vials to GC were estimated to be on the order of 10%.

Data analysis

Exploratory data analysis (EDA) for surface flux and subsurface gas data was performed using histograms and box plots. This analysis along with the test for normality using the Shapiro-Wilk statistic (SAS 2004) indicated the data were highly skewed and non-normally distributed. We therefore used a

nonparametric test based on ranked data to test differences in N₂O concentration due to depth and recent harvesting.

The Kruskal–Wallis test (Desu and Raghavarao 2004) was used to evaluate the within site variability of N₂O concentration due to depth for each site. The median was used as a measure of the center value rather than the mean for skewed data (Zar 1999), and the 95% confidence intervals for the medians (computed as outlined in Desu and Raghavarao (2004) using the Proc Univariate procedure of SAS (2004) were used to construct error bars and to determine which sample medians did not differ significantly from each other. The medians being compared were considered statistically different when the 95% confidence intervals of the medians did not overlap.

Regression models examining relationships amongst soil moisture, temperature and surface flux were carried out using Sigmaplot 8.0 (SPSS Inc., Chicago Illinois, USA). Analyses examining relationships between soil gas storage and surface flux were carried out using SPSS 14.0 (SPSS Inc., Chicago, Illinois, USA). All statistical significance was judged at the 5% probability level.

Results

While there are no differences in mean daily air temperatures between sites, recently clearcut site soil temperatures are often several degrees higher than mature forest soil temperatures during warmer periods (Fig. 1). Soil moisture minimums are observed during the period of maximum daily temperatures in the July through October period (Fig. 1), with recently harvested sites characterized by greater volumetric soil water contents than their respective intact forest pairs. Relationships between shallow moisture (as %WFPS) and soil temperature at clearcut and forest sites on gas sampling dates show that significant inverse linear relationships exist between temperature and %WFPS at three of these sites (LF: $R^2 = 0.93$, $P < 0.05$; LCC: $R^2 = 0.53$, $P < 0.05$; PF: $R^2 = 0.62$, $P < 0.05$). The clearcut sites were characterized by a smaller moisture range than forested sites, with site PCC exhibiting a non-significant relationship with temperature ($R^2 = 0.15$, $P > 0.05$), indicating some decoupling between these two variables with the removal of vegetation.

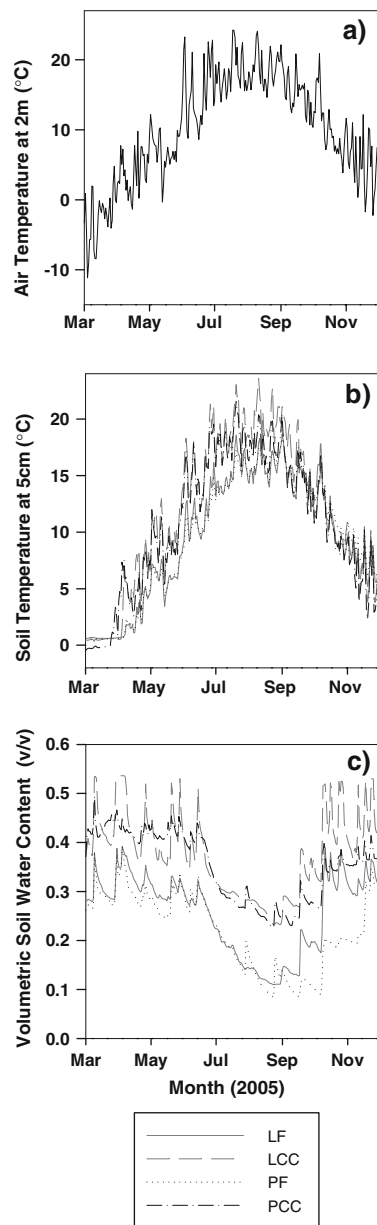


Fig. 1 Mean daily (a) air temperature (°C) at 2 m (b) soil temperature at 5 cm depth within the mineral soil (°C) and (c) volumetric soil water content (v/v) in the upper mineral soil (0–15 cm) at the LF, LCC, PF and PCC sites for the March–November 2005 gas sampling period

Surface flux: temporal patterns and relationships to soil temperature and moisture

Surface flux variability over the measurement period (Table 1) shows a large range of positive and negative fluxes from the soils of these sites, with

Table 1 Nitrous oxide surface flux ($\mu\text{gN}_2\text{O-N/m}^2/\text{day}$) summary statistics for the measurement period at each site

	Surface flux			
	LF	LCC	PF	PCC
Mean	−42	−7	11	11
Std Dev	140	184	316	179
Std Error	16	21	38	21
Median	−5	19	17	−1
Max	145	583	1367	515
Min	−614	−432	−811	−629
<i>n</i>	77	78	68	73

median values close to zero for all sites. Temporal patterns in median N_2O fluxes from each site for each measurement date are variable, particularly at the Pomquet sites (Fig. 2), with the lowest fluxes generally observed in the early and late portions of the sampling period. We note this dataset is complicated by the fact that initial measurements were made using a slightly different chamber design.

Independent regressions of all N_2O surface flux with temperature and %WFPS for each site were significant for LF, LCC and PF. Relationships with PCC were not significant. When N_2O surface fluxes were fit to regression models that contained both temperature and %WFPS as independent variables (Table 2), R^2 values for LF, LCC and PF were greatest, although never in excess of 0.55.

Patterns of subsurface soil N_2O concentrations

Distribution of the subsurface concentration data shows it is highly positively skewed (Fig. 3), particularly at the 3-year post-harvest sites (LCC; PCC). Subsurface concentration patterns at each site and depth (Table 3) show median subsurface concentrations at the intact forest sites that are lower than those of the corresponding 3-year post-clearcut pairs. Significant increases in median subsurface concentrations were not observed through depth at either LF or PF (Fig. 4), while significant differences between the shallow and deep concentrations were observed at LCC (both 0 cm and 5 cm were significantly different than 20 cm and 35 cm concentrations) and at PCC (0 cm concentrations were significantly different than both 20 cm and 35 cm concentrations).

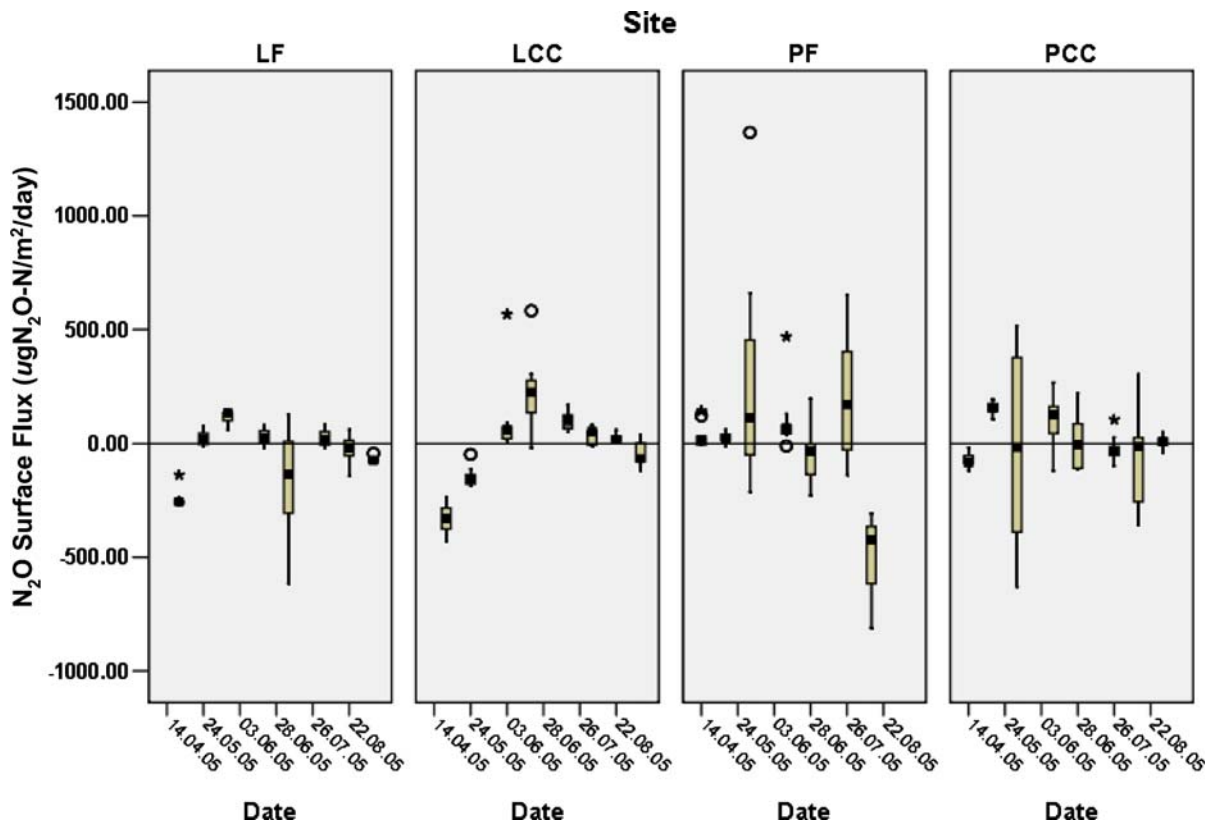


Fig. 2 Daily N_2O surface fluxes ($\mu\text{gN}_2\text{O-N/m}^2/\text{day}$) from the 2005 sampling period. The median is represented by the line in the middle of the box and the limits of the box represent the 25th and the 75th percentiles. The whiskers represent the

smallest and largest values that are not considered outliers. The open circles represent the mild outliers (cases with values of more than 1.5 time the box length) and the asterisks represent extreme values (cases with greater than 3 times the box length)

Table 2 Polynomial equations relating nitrous oxide surface flux ($\text{SF}_{\text{N}_2\text{O}}$ in $\mu\text{gN}_2\text{O-N/m}^2/\text{day}$) to temperature ($^{\circ}\text{C}$), and moisture (%WFPS)

Site	$\text{SF}_{\text{N}_2\text{O}} = a + b(\text{Moisture}) + c(\text{Temperature}) + d(\text{Moisture})^2 + e(\text{Temperature})^2$						
	R^2	P -value	a	b	c	d	e
LF	0.39	<0.0001	126.67	11.42*	-21.02	-0.1634*	0.3089
LCC	0.54	<0.0001	-1308.95*	44.01*	43.95*	-0.5270*	-0.8947
PF	0.33	<0.0001	-2034.38*	52.88*	-48.62	-0.3202*	5.2090*
PCC	ns	—	—	—	—	—	—

Statistically significant model components ($P < 0.05$) are indicated by an asterisk

Coupled N_2O surface flux and subsurface concentration data

Relationships between N_2O surface flux and subsurface storage at individual sites, examined by fitting a linear regression model to these two variables for all plots within a site on each sampling date, showed that through the majority of the measurement period, no

significant correlations were observed (Table 4). Positive correlations were observed more frequently at 3-year post-harvest sites (LCC 38%; PCC 25%) than forest sites (LF 11%; PF 14%). This illustrates a lack of connection between soil N_2O storage and surface flux through much of the measurement period. The majority of the positive correlations were observed within the June–August period, although

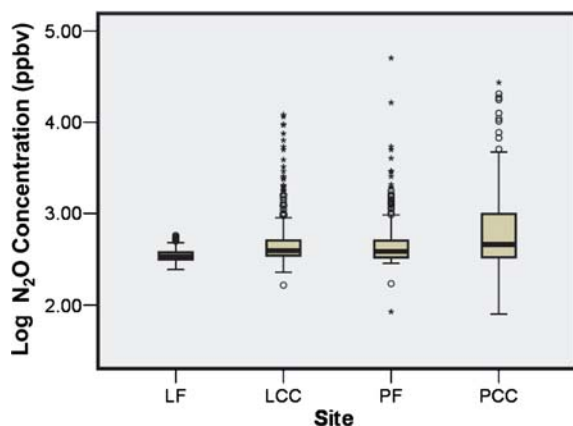


Fig. 3 Log transformed subsurface soil air N₂O concentrations (ppbv) for all depths combined for each site for the 2005 measurement period. The median is represented by the line in the middle of the box and the limits of the box represent the 25th and the 75th percentiles. The whiskers represent the smallest and largest values that are not considered outliers. The open circles represent the mild outliers (cases with values of more than 1.5 time the box length) and the asterisks represent extreme values (cases with greater than 3 times the box length)

not all sampling dates during this period yielded a significant relationship.

Data from sites expected to generate elevated surface fluxes based upon strong positive subsurface concentration gradients (LCC; PCC) showed a large range in surface flux values (ranging from positive to negative) within a single day. Additionally, specific plots yielded elevated subsurface concentrations that persisted over a minimum period of several months. Plots with elevated subsurface concentrations only occasionally showed corresponding surface fluxes that were also elevated at the sites. A subset of

subsurface N₂O concentration profiles within each early post-harvest successional site (Fig. 5) show in detail, examples that capture the observed range of subsurface concentration patterns. At plots of elevated subsurface concentrations (LCC-M9; PCC-M10), the strongest gradients were observed on the late-June sampling date. Shallow soil concentrations (0 and 5 cm) also peak when elevated surface fluxes are observed. In contrast, the lower concentration plots (LCC-M6; PCC-M2) show only weak positive concentration gradients, with maximum concentrations also observed on the late-June sampling date at depth at both sites.

Discussion and conclusions

N₂O Surface fluxes: magnitudes and relationships to temperature and moisture

Surface fluxes from the forested sites (Table 1) are comparable to or slightly below means reported in other temperate North American studies (Castro et al. 1993; Bowden et al. 1991, 2000; Corre et al. 1999), and well below those often observed in European ecosystems (Butterbach-Bahl et al. 1997, 2002; Zechmeister-Boltenstern et al. 2002; Papen and Butterbach-Bahl 1999) where N deposition is often elevated. Soils at these sites had more negative minimum N₂O fluxes than reported in most other studies, although within the range of negative fluxes reported in the literature for soils, including agricultural soils (Chapuis-Lardy et al. 2007). While extremes in surface flux magnitudes were observed

Table 3 Subsurface soil air N₂O concentration (ppbv) summary statistics for each site and mineral soil depth

	Site															
	LF				LCC				PF				PCC			
	0 cm	5 cm	20 cm	35 cm	0 cm	5 cm	20 cm	35 cm	0 cm	5 cm	20 cm	35 cm	0 cm	5 cm	20 cm	35 cm
Mean	346	352	365	371	437	749	773	948	702	1157	588	607	453	577	1533	4293
Std Dev	58	53	54	50	307	1515	1219	1709	1858	5467	452	610	259	439	2023	6539
Std Error	6	5	6	6	32	152	129	196	204	593	54	74	28	49	263	1075
Median	329	339	358	373	362	387	436	502	370	383	450	460	370	424	875	1783
Max	583	542	563	536	2511	12110	9310	11499	16422	50667	2901	4995	1480	2565	12607	27323
Min	284	290	286	246	285	253	166	284	295	85	173	295	108	80	206	178
n	97	98	90	61	95	99	89	76	83	85	69	68	87	79	59	37

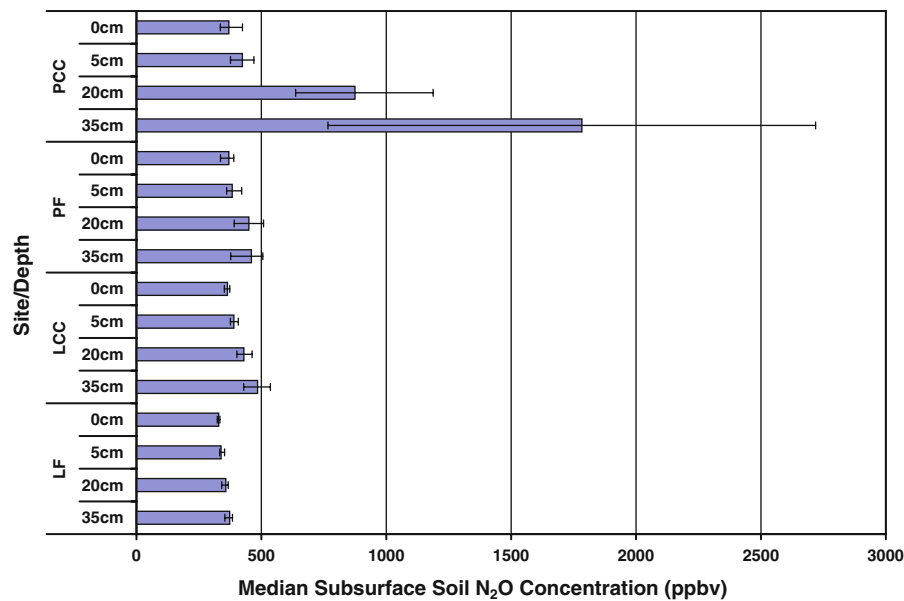


Fig. 4 Median subsurface soil air N₂O concentrations (ppbv) at each site per measurement depth. Error bars represent the 95% confidence intervals

Table 4 Relationships between subsurface N₂O storage ($\mu\text{gN}_2\text{O-N/m}^2$) to 35 cm mineral soil depth and corresponding surface flux ($\mu\text{gN}_2\text{O-N/m}^2/\text{day}$) for all plots (10) in each site on a given sampling day in 2005

Site	Date	R^2	P -value	Site	JD	R^2	P -value
LF	Apr 27	0.032	0.623	LCC	Apr 27	0.019	0.705
	May 24	0.000	0.980		May 24	0.000	0.960
	Jun 2	0.021	0.709		Jun 3	0.417*	0.044
	Jun 23	0.097	0.381		Jun 23	0.617*	0.007
	Jul 11	0.000	0.971		Jul 11	0.284	0.112
	Aug 8	0.455*	0.046		Aug 8	0.004	0.867
	Aug 29	0.001	0.928		Aug 29	0.066	0.506
	Oct 4	0.006	0.949		Oct 4	—	—
PF	Nov 13	0.026	0.656	PCC	Nov 13	0.450*	0.034
	Apr 14	0.122	0.322		Apr 14	0.365	0.151
	May 5	0.092	0.395		May 5	0.211	0.214
	May 31	0.006	0.825		May 31	0.002	0.915
	Jun 15	0.965*	0.000		Jun 15	0.023	0.746
	Jun 28	0.165	0.244		Jun 28	0.479*	0.027
	Jul 26	0.036	0.654		Jul 26	0.083	0.420
	Aug 16	0.001	0.938		Aug 16	0.697*	0.003
	Sep 14	—	—		Sep 14	0.019	0.744

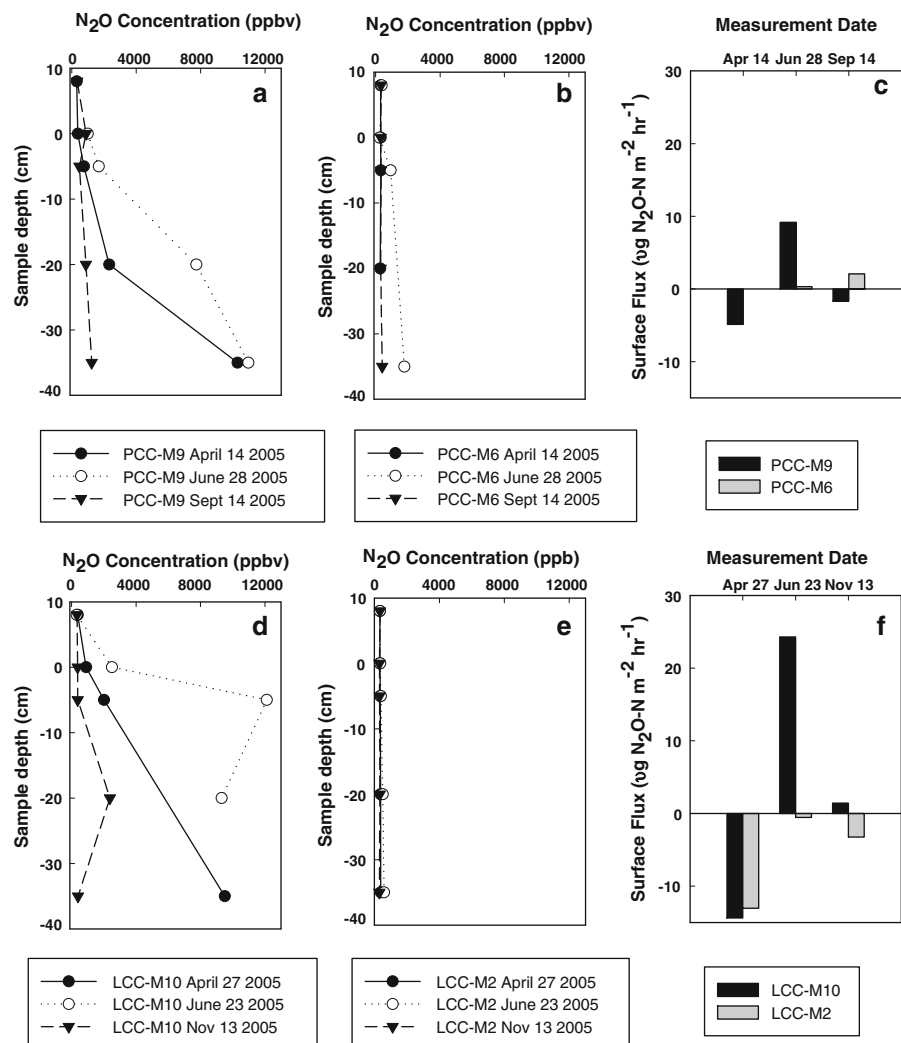
Statistically significant relationships ($P < 0.05$) are indicated by an asterisk

across all sites (Table 1), medians over the study period were close to zero for all sites.

The early post-successional sites suggest that three years following clearcut harvesting, soils at these forest sites are in the balance, neither a significant net source nor sink of N₂O relative to the late post-

harvest succession soils (Table 1). The observed median fluxes are in the range of N₂O surface fluxes reported from other forest soil harvest studies in northern climates (Schiller and Hastie 1996; Huttunen et al. 2003) although, not unexpectedly, lower than typical observations made during the first year

Fig. 5 Examples of vertical subsurface soil air N_2O concentration profiles (a, b, d, e) and associated surface fluxes (c, f) for plots characterized by high and low subsurface concentrations at (a) PCC-M9, (b) PCC-M6, (d) LCC-M10 and (e) LCC-M2 for early (April), mid (June) and late (Sept/Nov) growing season dates. Significant relationships between subsurface N_2O storage ($\mu\text{gN}_2\text{O-N/m}^2$) and surface flux ($\mu\text{gN}_2\text{O-N/m}^2/\text{day}$) ($P < 0.05$) at the PCC and LCC sites were observed only on the June sampling dates



following clearcutting when availability of inorganic N is expected to be higher (Zerva and Mencuccini 2005; Huttunen et al. 2003; Tate et al. 2006).

While higher N_2O fluxes at clearcut sites 3-years post-harvest were expected as a result of the mobilization of N following clear cutting of the forests due to increased rates of N mineralization and/or reduced competition from roots post-harvest (Hornbeck et al. 1986; Pennock and van Kessel 1997; Vitousek and Matson 1985; Johnson 1995; Frazer et al. 1990), this study did not pick up a significant difference. Consistent with other studies, we documented a high degree of spatial variability in N_2O surface fluxes (Fig. 2) (Livingston and Hutchinson 1995; Ambus and Christensen 1995; Robertson and Tiedje 1984; Goodroad and Keeney 1984; Parkin 1987).

Despite the high variability in fluxes within sites, relationships were observed between N_2O surface fluxes, temperature and moisture, which were significant at 3 of the 4 sites (LF, LCC, PF). Temperature and moisture combined provided the best predictive model for surface flux (Table 2), although a maximum of 54% of the variability in surface flux was explained by these two variables. The exception (PCC) was a site where moisture varied little during the growing season (generally $> 40\%$ WFPS) and no significant relationships between moisture and temperature were observed for the sampling dates. This site and its forest pair, PF, were also characterized by more finely textured and more poorly drained soils where variability in N_2O surface fluxes were greater and clear temporal patterns less evident (Fig. 2).

Subsurface soil air N₂O concentration trends

The subsurface soil air N₂O concentrations observed in this study span a large concentration range (80–50667 ppb) (Table 3). Large concentration ranges are consistent with observations from other studies measuring subsurface soil air N₂O concentrations at a range of non-forested sites (Burton and Beauchamp 1994; Burton et al. 1997; Clark et al. 2001). In addition to the higher subsurface concentrations observed at the early post-harvest successional sites than their respective late post-harvest forest pairs (Table 3), a pattern of a strong positive skew to the concentration data is evident (Fig. 3). The occasional observation of gas concentrations below atmospheric (Table 3), does indicate a consumption which appears, on occasion, to exceed the supply of N₂O to the soil.

Significant positive concentration gradients are observed in subsurface soil air N₂O concentration patterns at the 3 year post-harvest sites, with no significant increases in concentration through depth observed at late successional stage forested sites (Fig. 4). Differences in observed subsurface N₂O concentrations between Lakevale (sandy soils) and Pomquet (clayey soils) (Bekele et al. 2007) sites are likely due to the differences in soil texture that result in lower diffusion rates in the Pomquet soils. Overall, these subsurface observations that document differences in soil N₂O dynamics between paired forest and clearcut sites stand in contrast to surface flux observations at the same sites.

Coupling subsurface soil air concentrations and surface fluxes

To our knowledge there have been no attempts to use N₂O concentration profile data in an effort to improve current understandings of N₂O production–consumption dynamics in northern forest soils. In general, positive subsurface gas concentration gradients indicate that soils are net sources of N₂O. However, observations of subsurface concentrations below atmospheric (Table 3), the strong sink behavior of these soils (Figs. 2, 5), and lack of a consistent quantitative relationship between soil profile gas storage and surface flux (Table 4), suggests N₂O is not transported conservatively in these soils. This precludes the application of a gas production model for

N₂O concentration profile studies at these sites to calculate gross production, allowing only a qualitative assessment of consumption–production processes.

At the 3 year post-harvest sites, the vertical concentration profiles typically show the most elevated subsurface concentrations at depth in the soil profile, while shallow subsurface soil air N₂O concentrations are more consistently close to atmospheric values. This pattern suggests the shallow soil is not acting as an important source of N₂O to the atmosphere during much of the measurement period. The largest gradients are, however, observed through the deeper part of the measured profile, raising the possibility that this zone may act as an important source of this gas at more recently disturbed sites.

There are indications of seasonal behavior with respect to soil N₂O dynamics at these sites during the snow free period of the year. Plots with elevated subsurface concentrations that were observed early in the sampling period were maintained through a large proportion of this sampling period (Fig. 5). It is interesting to note that the strongest relationships between subsurface storage and surface fluxes were observed in the early summer (i.e. late June).

This study suggests that there may exist a disconnection between soil profile N₂O production processes and soil-atmosphere N₂O exchanges from northern forest soils. In this study, concentration profile data is consistent with a greater production of N₂O at the early post-harvest successional sites, (evidenced by strong positive concentration gradients and greater net soil N₂O storage), yet this does not necessarily translate into positive surface fluxes. This pattern is consistent with consumption of N₂O during vertical transport through the soil profile. If the factors that limit N₂O diffusion in soils do in fact increase its consumption in soils as concluded by Chapuis-Lardy et al. (2007), increases in soil moisture accompanying forest clear cutting at these sites should act as a negative feedback on the production process by increasing N₂O consumption. While there may be a greater potential for these sites to act as elevated sources of N₂O during the period immediately following clear cutting, it appears that the potential for these soils to also behave as a sink for this N₂O is quite high and could act to strongly dampen their source potential. Consequently, we would view the predictive relationships of surface flux with soil moisture and temperature (Table 2), as

representing the combined influences of both production and consumption on soil-atmosphere N_2O exchanges, even when positive fluxes are documented. Gross rates of production of soil N_2O and consumption of both soil and atmospheric sourced N_2O are therefore likely underestimated. We caution that soils at these study sites are quite wet and may represent conditions that would maximize in-situ consumption during soil transport. The extent to which this would also be a dominant process in other regions would require further investigation.

This study represents an initial step in attempting to resolve questions about N_2O soil profile production and consumption dynamics in managed northern forest soils. The results of this study, while not allowing quantification of gross production and consumption of soil N_2O at these sites, does provide evidence that a greater emphasis should be placed upon questioning the role of N_2O consumption processes in managed forest soils. This is consistent with the conclusions of the recent review by Chapuis-Lardy et al. (2007). A lack of a quantitative relationship between the magnitudes of surface flux and subsurface soil gas storage through the majority of the measurement period is consistent with a significant fraction of the soil profile N_2O pool not reaching the atmosphere. This appears to be particularly significant at early post-harvest successional sites. The gross sink potential of these soils will be a combination of atmospheric uptake (i.e. negative fluxes) and consumption of N_2O produced in-situ in the soil. Subsequent efforts need to work towards combining the N_2O gas profile approach with other advanced tools (such as stable isotope signatures of N_2O) that would allow quantification of these consumption–production processes in soil profiles. Combined with efforts to resolve the individual roles of gas storage and transport processes, these approaches could yield valuable information about in-situ rates of gross production and consumption within soil profiles.

We conclude that consumption during vertical transport may obscure gross soil N_2O production values and lead to underestimates of gross soil N_2O production in some soils. A strong sink during vertical transport, if most N_2O is produced at depth in the mineral soil, or if a reduction in soil diffusion rates due to changes in environmental conditions occurs, could provide an explanation for why some

conceptual models don't predict soil N_2O emissions as well as might be expected (ex. Davidson et al. 2000). Consequently, care needs to be exercised in differentiating between soil N_2O emissions and soil N_2O production in the soil as these two values may in fact differ substantially.

Acknowledgments This research was supported by the Natural Sciences and Engineering Research Council of Canada (NSERC), the Atlantic Innovation Fund (AIF). Vegetation site descriptions were provided by Amanda Diochon. Thanks to all members of the Environmental Sciences Research Centre at StFXU who contributed to the collection of this data, and particularly to A. Bekele for discussions related to data analysis.

References

- Ambus P, Christensen S (1995) Spatial and seasonal nitrous oxide and methane fluxes in Danish forest-, grassland-, and agroecosystems. *J Environ Qual* 24:993–1001
- Ambus P, Zechmeister-Boltenstern S, Butterbach-Bahl K (2006) Sources of nitrous oxide emitted from European forest soils. *Biogeochemistry* 3:135–145
- Beltrami H, Kellman L (2003) An examination of short- and long-term air–ground temperature coupling. *Global Planet Change* 38:291–303
- Bekele A, Kellman L, Beltrami H (2007) Soil profile CO_2 concentrations in forested and clear cut sites in Nova Scotia, Canada. *Forest Ecol Manage.* doi:10.1016/j.foreco.2007.01.088
- Borken W, Beese F (2005) Control of nitrous oxide emissions in European beech, Norway spruce and Scots pine forests. *Biogeochemistry* 76:141–159
- Bouwman AF (1990) Exchange of greenhouse gases between terrestrial ecosystems and the atmosphere. In: Bouwman AF (ed) *Soils and the Greenhouse Effect*. Wiley
- Bowden WB, Bormann FH (1986) Transport and loss of nitrous oxide in soil water after forest clear-cutting. *Science* 233:867–869
- Bowden RD, Melillo JM, Steudler PA (1991) Effects of nitrogen additions on annual nitrous oxide fluxes from temperate forest soils in the Northeastern United States. *J Geophys Res* 96:9321–9328
- Bowden RD, Rullo G, Stevens GR, Steudler PA (2000) Soil fluxes of carbon dioxide, nitrous oxide, and methane at a productive temperate deciduous forest. *J Environ Qual* 29:268–276
- Brumme R, Beese F (1992) Effects of limon and nitrogen fertilization on emissions of CO_2 and N_2O from a temperate forest. *J Geophys Res* 97:12851–12858
- Burns DA, Murdoch PS (2005) Effects of a clearcut on the net rates of nitrification and N mineralization in a Northern Hardwood Forest, Catskill Mountains, New York, USA. *Biogeochemistry* 72:1–24
- Burton DL, Beauchamp EG (1994) Profile nitrous oxide and carbon dioxide concentrations in a soil subject to freezing. *Soil Sci Soc Am J* 58:115–122

- Burton DL, Bergstrom DW, Covert JA, Wagner-Riddle C, Beauchamp EG (1997) Three methods to estimate N_2O fluxes as impacted by agricultural management. *Can J Soil Sci* 77:125–134
- Butterbach-Bahl K, Gasche R, Breuer L, Papen H (1997) Fluxes of NO and N_2O from temperate forest soils: impact of forest type, N deposition and of liming on the NO and N_2O emissions. *Nutrient Cycl Agroecosyst* 48:79–90
- Butterbach-Bahl K, Rothe A, Papen H (2002) Effect of tree distance on N_2O and CH_4 -fluxes from soils in temperate forest ecosystems. *Plant Soil* 240:91–103
- Cann DB, Hilchey JD (1955) Soil survey of Antigonish County, Nova Scotia. E. Cloutier, Ottawa
- Castro MS, Steudler PA, Melillo JM, Aber JD, Millham S (1993) Exchange of N_2O and CH_4 between the atmosphere and soils in spruce-fir forests in the northeastern United States. *Biogeochemistry* 18:119–135
- Chapuis-Lardy L, Wrage N, Metay A, Chottes J, Bernoux M (2007) Soils, a sink for N_2O ? A review. *Global Change Biol* 13:1–17. doi:[10.1111/j.1365-2486.2006.01280.x](https://doi.org/10.1111/j.1365-2486.2006.01280.x)
- Cicerone R (1989) Analysis of sources and sinks of atmospheric nitrous oxide. *J Geophys Res* 94:18265–18271
- Clark M, Jarvis S, Malby E (2001) An improved technique for measuring concentration of soil gases at depth in situ. *Commun Soil Sci Plant Anal* 32:369–377
- Corre MD, Pennock DJ, Van Kessel C, Elliott DK (1999) Estimation of annual nitrous oxide emissions from a transitional grassland-forest region in Saskatchewan, Canada. *Biogeochemistry* 44:29–49
- Davidson EA, Keller M, Erickson HE, Verchot LV, Veldkamp E (2000) Testing a conceptual model of soil emissions of nitrous and nitric oxides. *BioScience* 50:667–680
- Desu MM, Raghavarao D (2004) Nonparametric statistical methods for complete and censored data. Chapman & Hall, CRC, Boca Raton, FL
- Firestone MK, Davidson EA (1989) Microbial basis of NO and N_2O production and consumption in soil. In: Exchange of trace gases between terrestrial ecosystems and the atmosphere. Wiley
- Frazer DW, McColl JG, Powers RF (1990) Soil nitrogen mineralization in a clearcutting chronosequence in a Northern California conifer forest. *Soil Sci Soc Am J* 54:1145–1152
- Goodroad LL, Keeney DR (1984) Nitrous oxide emission from forest, marsh, and prairie ecosystems. *J Environ Qual* 13:448–452
- Hornbeck JW, Martin CW, Pierce ES, Bormann FH, Likens GE, Eaton JS (1986) Clearcutting Northern Hardwoods: effects on hydrologic and nutrient ion budgets. *Forest Sci* 32:667–686
- Huttunen JT, Nykänen H, Martikainen PJ, Nieminen M (2003) Fluxes of nitrous oxide and methane from drained peatland following forest clear-felling in southern Finland. *Plant Soil* 255:457–462
- IPCC (2007) In: Solomon S, Qin D, Manning M, Chen Z, Marquis M, Averyt KB, Tignor M, Miller HL (eds) *Climate Change 2007: the physical science basis. Contribution of working group I to the fourth assessment report of the intergovernmental panel on climate change*. Cambridge University Press, Cambridge
- Johnson RS (1986) *Forests of Nova Scotia*. Four East Publications, Halifax NS, Canada
- Johnson CE (1995) Soil nitrogen status 8 years after whole-tree clear-cutting. *Can J Soil Res* 25:1346–1355
- Keller M, Reiners WA (1994) Soil-atmosphere exchange of nitrous oxide, nitric oxide, and methane under secondary succession of pasture to forest in the Atlantic lowlands of Costa Rica. *Global Biogeochem Cycles* 8:399–409
- Keller M, Veldkamp E, Weitz AM, Reiners WA (1993) Effect of pasture age on soil trace-gas emissions from a deforested area of Costa Rica. *Nature* 365:244–246
- Livingston GP, Hutchinson GL (1995) Enclosure-based measurement of trace gas exchange: applications and sources of error. In: Matson PA, Harriss RC (eds) *Biogenic trace gases: measuring emissions from soil and water*. Blackwell Science, Cambridge
- Matson PA, Vitousek PM (1990) Ecosystem approach to a global nitrous oxide budget. *Bioscience* 40:667–672
- Matson P, Gower ST, Volkman C, Billow C, Grier CC (1992) Soil nitrogen cycling and nitrous oxide flux in a Rocky Mountain Douglas-fir forest: effects of fertilization, irrigation and carbon addition. *Biogeochemistry* 19:101–117
- Melillo JM, Steudler PA, Aber JD, Bowden RD (1989) Atmospheric deposition and nutrient cycling. In: Andreae MO, Schimel DS (eds) *Exchange of trace gases between terrestrial ecosystems and the atmosphere*. Wiley, pp 263–280
- Papen H, Butterbach-Bahl K (1999) A 3-year continuous record of nitrogen trace gas fluxes from untreated and limed soil of a N-saturated spruce and beech forest ecosystem in Germany 1. N_2O emissions. *J Geophys Res* 104:487–503
- Papen H, Daum M, Steinkamp R, Butterbach-Bahl K (2001) N_2O and CH_4 -fluxes from soils of a N-limited and N-fertilized spruce forest ecosystem of the temperate zone. *J Appl Bot Angew Bot* 75:159–163
- Parkin TB (1987) Soil microsites as a source of denitrification variability. *Soil Sci Soc Am J* 51:1194–1199
- Pennock DJ, van Kessel C (1997) Clear-cut forest harvest impacts on soil quality indicators in the mixedwood forest of Saskatchewan, Canada. *Geoderma* 75:13–32
- Risk D, Kellman L, Beltrami H (2002a) Soil CO_2 Production and surface flux at four climate observatories in eastern Canada. *Global Biogeochemical Cycles* 16 doi:[10.1029/2001GB001831](https://doi.org/10.1029/2001GB001831)
- Risk D, Kellman L, Beltrami H (2002b) Carbon dioxide in soil profiles production and temperature dependence. *Geophys Res Lett* 29. doi:[10.1029/2001GL014002](https://doi.org/10.1029/2001GL014002)
- Risk D, Kellman L, Beltrami H (2008) A new method for in-situ soil gas diffusivity measurement and applications in the monitoring of subsurface carbon dioxide. *J Geophys Res—Biogeosciences* 113:G02018, doi:[10.1029/2007JG000445](https://doi.org/10.1029/2007JG000445)
- Robertson GP, Tiedje JM (1984) Denitrification and nitrous oxide production in successional and old-growth Michigan forests. *Soil Sci Soc Am J* 48:383–389
- SAS Institute (2004) *SAS/STAT user's guide, version 9.1*. SAS Institute, Cary, NC
- Schiller CL, Hastie DR (1996) Nitrous oxide and methane fluxes from perturbed and unperturbed boreal forest sites in northern Ontario. *J Geophys Res* 101:767–774

- Schlesinger W (1997) Biogeochemistry: an analysis of global change. Academic Press, London
- Schulte-Bisping H, Brumme R (2003) Nitrous oxide emission inventory of German forest soils. *J Geophys Res* 108(D4): 4132. doi:[10.1029/2002JD002292](https://doi.org/10.1029/2002JD002292)
- Tate KR, Ross DJ, Scott NA, Rodda NJ, Townsend JA, Arnold GC (2006) Post-harvest patterns of carbon dioxide production, methane uptake and nitrous oxide production in a *Pinus radiata* D. Don plantation. *Forest Ecol Manage* 228:40–50
- Teepe R, Brumme R, Beese F (2000) Nitrous oxide emissions from frozen soils under agricultural, fallow and forest land. *Soil Biol Biochem* 32:1807–1810
- Vitousek PM (1981) Clear-cutting and the nitrogen cycle. In: Clark FE, Rosswall T (eds) *Terrestrial nitrogen cycles*. *Ecol Bull*, 33. Stockholm, pp 631–642
- Vitousek PM, Matson PA (1985) Disturbance, nitrogen availability, and nitrogen losses in an intensively managed loblolly pine plantation. *Ecology* 66:1360–1376
- Wolf I, Brumme R (2003) Dinitrogen and nitrous oxide formation in beech forest floor and mineral soils. *Soil Sci Soc Am J* 67:1862–1868
- Zar JH (1999) *Biostatistical analysis*. Prentice Hall, Upper Saddle River, NJ
- Zechmeister-Boltenstern S, Hahn M, Meger S, Jandl R (2002) Nitrous oxide emissions and nitrate leaching in relation to microbial biomass dynamics in a beech forest soil. *Soil Biol Biochem* 34:823–832
- Zerva A, Mencuccini M (2005) Short-term effects of clearfelling on soil CO₂, CH₄, and N₂O fluxes in a Sitka spruce plantation. *Soil Biol Biochem* 37:2025–2036