

N₂O emissions and carbon sequestration in a nitrogen-fertilized Douglas fir stand

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[1] This study investigated how nitrogen (N) fertilization with 200 kg N ha⁻¹ of a 58-year-old West Coast Douglas fir stand influenced its net greenhouse gas (GHG) global warming potential (GWP) in the first year after fertilization. Effects of fertilization on GHG GWP were calculated considering changes in soil N₂O emissions, measured using the static chamber technique and the soil N₂O gradient technique; eddy covariance (EC) measured net ecosystem productivity (NEP); and energy requirements of fertilizer production, transport, and its aerial spreading. We found significant N₂O losses in fertilized plots compared to a small uptake in nonfertilized plots. Chamber-measured N loss in the fertilized plots was about 16 kg N₂O ha⁻¹ in the first year, which is equivalent to 10 kg N ha⁻¹ or 5% of the applied fertilizer N. Soil N₂O emissions measured using the gradient technique, however, exceeded the chamber measurements by about 50%. We also compared a polymer-coated slow-release urea with regular urea and found that the former delayed N₂O emissions but the year-end total loss was about the same as that from regular urea. Change in NEP due to fertilization was determined by relating annual NEP for the nonfertilized stand to environmental controls using an empirical and a process-based model. Annual NEP increased by 64%, from 326 g C m⁻², calculated assuming that the stand was not fertilized, to the measured value of 535 g C m⁻² with fertilization. At the end of the year, net change in GHG GWP was -2.28 t CO₂ ha⁻¹ compared to what it would have been without fertilization, thereby indicating favorable effect of fertilization even in the first year after fertilization with significant emissions of N₂O.

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1. Introduction

[2] Forests play a key role in the natural carbon (C) cycle. Each year forests absorb billions of tons of CO₂, a high proportion of which is lost when trees respire and also in the decomposition of soil organic matter and forest floor litter. In an east-west transect study of Canadian forests and peatlands, intermediate-aged stands (35–60 years old) showed the highest maximum net ecosystem productivity (NEP) and gross primary productivity (GPP) [Coursolle *et al.*, 2006]. Our recent research shows that a 58-year-old West Coast Douglas fir sequesters 2–3 t C ha⁻¹ y⁻¹ [Humphreys *et al.*, 2006; Jassal *et al.*, 2007]. Weather can have a significant impact on C exchange. It was found that

warmer temperatures associated with El Niño caused an increase in C emissions, reducing the net amount of C sequestered [Morgenstern *et al.*, 2004]. In the coastal regions of British Columbia, which have very little nitrogen (N) deposition from pollution sources owing to their remote location, and soils deficient in N [Hanley *et al.*, 1996], Douglas fir stands respond to N fertilization [Brix, 1991; Fisher and Binkley, 2000; Chapin *et al.*, 2002]. The standard forest fertilization application rate in West Coast forests is 200 kg N ha⁻¹ from prilled urea [Hanley *et al.*, 1996]. While fertilization of stands of midrotation trees (i.e., of commercial thinning size, 20–40 years old) can result in additional merchantable timber volumes, fertilization late in the rotation may be the most attractive alternative economically. A single application 8–10 years before the final harvest of near-end-of-rotation (50–60-year-old) Douglas fir stands provides an attractive financial return with a volume growth increase of about 20% on average sites [Hanley *et al.*, 1996]. Also, fertilization is one of the eligible management practices for C sequestering and hence reducing CO₂ emissions under Article 3.4 of the Kyoto Protocol.

[3] Simulated chronic N deposition has been shown to increase C storage in northern temperate forests [Pregitzer *et al.*, 2008]. In a meta-analysis, Johnson and Curtis [2001]

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found that N fertilization was the only forest management practice that had a clearly positive effect on the soil C pool. Steadily rising atmospheric CO₂ may likely stimulate the effect of N deposition and fertilization resulting in increased forest biomass [Nadelhoffer *et al.*, 1999]. Leggett and Kelting [2006] found that N fertilization of loblolly pine plantations not only increased aboveground and belowground biomass but also significantly increased soil C. The increase in soil C storage with N fertilization has been attributed, in part, to an increase in litter inputs as a result of higher plant production and partly to reduced decomposition rates of soil organic matter and humus [Magill and Aber, 1998; Prescott, 1995]. Olsson *et al.* [2005] found that fertilization of a boreal Norway spruce stand led to a threefold increase in aboveground productivity, possibly because of decreased C allocation to roots in response to higher nutrient availability.

[4] Another major concern with N fertilization is the potential loss of applied fertilizer N, e.g., NH₃ volatilization, nitrate leaching, and denitrification, resulting in possible negative environmental impacts. Losses of N in ammonia volatilization, when fertilizer is applied to well-drained and acidic forest soils (pH ~ 5) under wet and cool conditions, are likely to be small [Rachhpal-Singh and Nye, 1986]. Chappell *et al.* [1999] found that net nitrification rates following repeated (8–10 years) N fertilization of coastal Douglas fir stands were higher than those in the unfertilized stands. N losses in denitrification have been extensively studied in agricultural crops [e.g., Pattey *et al.*, 2007] and in grazed pastures [e.g., Liebig *et al.*, 2006], but few studies have been conducted on denitrification losses in forest soils following fertilizer N application. Pang and Cho [1984] reported negligible denitrification loss of N when forest soils from Shawnigan Lake, British Columbia, were incubated with different rates of fertilizer N. Schiller and Hastie [1996], using static chambers, measured N₂O losses from lowland, drained lowland, clear-cut, and upland boreal forests near Cochrane, Ontario, and found an emission of 3.1 μg N₂O m⁻² h⁻¹ from an unvegetated clear-cut site compared to an uptake of 7.7 μg N₂O m⁻² h⁻¹ at a drained lowland site. Liebig *et al.* [2006] reported that fertilized crested wheatgrass enhanced deep storage of soil organic C but resulted in greater N₂O emissions relative to native nonfertilized pastures in the northern Great Plains. However, little is known about denitrification losses in N-fertilized forest soils. The current Intergovernmental Panel on Climate Change (IPCC) guidelines assume that 1.25% of fertilizer N is lost as N₂O and NO. These estimates, taken from National Greenhouse Gas Inventory Committee (NGGIC) [2001], are based on fertilizer application in farming systems. In coniferous forest soils, highly mobile NO₃⁻ may get converted into less mobile N because of microbial assimilation of NO₃⁻ followed by a release of biomass N as organic N and NH₄⁺ [Stark and Hart, 1997], thereby minimizing NO₃⁻ leaching and denitrification losses of N.

[5] While it is necessary to determine and quantify the effects of N fertilization on stand C sequestration (i.e., NEP), it is also important to address environmental concerns by measuring N₂O emissions to determine the net greenhouse gas (GHG) global warming potential (GWP). The GWP of N₂O is 296 times (100-year time horizon) greater than that of CO₂ [Ehhalt and Prather, 2001], yet

there is little information on its net radiative forcing as a result of forest fertilization.

[6] We report the effects of N fertilization of a 58-year-old West Coast Douglas fir stand with 200 kg N ha⁻¹ on soil N₂O emissions and stand NEP. We also calculate the net change in GHG GWP resulting from forest fertilization by accounting for N₂O emissions and energy costs of fertilizer production, transport, and application. We also compare polymer-coated slow-release urea (Environmentally Smart Nitrogen (ESN), Agrium Inc., Calgary, Alberta, Canada) to regular urea for its potential effectiveness in reducing N₂O emissions from the forest floor.

2. Materials and Methods

2.1. Site Description

[7] Measurements were made in a 58-year-old Douglas fir stand (~130 ha) located about 10 km southwest of Campbell River (49°52′7.8″N, 125°20′6.3″W, flux tower location), on the east coast of Vancouver Island, Canada. The soil is a humo-ferric podzol (Quimper sandy loam) with a variable surface litter-fermenting-humified organic layer 0–6 cm thick and underlain with a dense compacted till at a depth of 1 m [Jungen, 1986]. Below the organic layer, soil texture gradually changes to gravelly loamy sand in the upper 40 cm and to gravelly sand with increasing depth. The mean annual temperature and precipitation at the site are 8.6°C and 1450 mm, respectively, and the site is occasionally subjected to a soil water deficit in August, September, and October. Further details on soil and vegetation characteristics can be found in the work by Humphreys *et al.* [2006].

2.2. Climate and Eddy Covariance Measurements

[8] Climate and eddy covariance (EC) instrumentation and measurements at this site are described in detail by Jassal *et al.* [2007], Humphreys *et al.* [2006], and Morgenstern *et al.* [2004]. Briefly, EC fluxes were measured at the 43-m height using a three-axis sonic anemometer (model R3, Gill Instruments Ltd., Lymington, UK) and an infrared gas analyzer (IRGA) (model LI-6262, LI-COR Inc., Lincoln, Nebraska, USA) in a temperature-controlled housing with a heated 4-m-long air sampling tube. CO₂ concentrations were measured at four heights using a manifold, pump, and LI-840 IRGA to estimate half-hourly changes in CO₂ storage in the air column beneath the EC sensors. Half-hourly measurements of soil water content and soil temperature profiles were made continuously near the EC flux tower. At two locations, soil volumetric water content (θ) was measured at 1–2-, 10–12-, 35–48-, and 70–100-cm depths using 30-cm-long water content reflectometers (model CS-615, Campbell Scientific Inc., Logan, Utah, USA). Soil temperature (T_s) measurements were made at the 5-, 10-, 20-, 50-, and 100-cm depths with copper-constantan thermocouples. Downwelling photosynthetically active radiation (Q) was measured at the canopy height with a quantum sensor (model LI-190SB, LI-COR Inc.). Air temperature (T_a) and relative humidity were measured at the height of the EC instrumentation (43 m above the ground surface) using a temperature and humidity probe (model HMP-35C, Vaisala Oyj, Helsinki, Finland). Precipitation (P) was measured using two tipping-bucket rain gauges

(model 2501, Sierra Misco, Berkeley, California, USA) mounted on the flux tower at the 25-m height and a precipitation gauge (model I-200B, Geonor A. S., Oslo, Norway) (for determining the water equivalent of snowfall in winter) installed in a young Douglas fir plantation about 3 km from the site.

[9] Net ecosystem exchange (NEE) was calculated as the sum of the half-hourly EC-measured flux of CO₂ (F_C) and the rate of change in CO₂ storage (F_S) in the air column between the ground and the EC measurement level. F_C was calculated using $F_C = \overline{\rho_a w' s'_c}$, where $\overline{\rho_a}$ is the mean molar density of dry air at sensor height and $w' s'_c$ is the covariance between the vertical wind velocity (w) and the mole mixing ratio of CO₂ (s_c), after making a three-axis coordinate rotation so that the mean vertical and lateral components of wind velocity and the covariance between them were equal to zero [Humphreys *et al.*, 2006]. The overbar and prime denote time average (half hour) and fluctuation from the average, respectively. F_S was approximated by $F_S = h_m \overline{\rho_a} \Delta s_c / \Delta t$ [Hollinger *et al.*, 1994; Morgenstern *et al.*, 2004], where h_m is the EC measurement height (i.e., 43 m), Δs_c is the difference between the average (half-hourly mean) CO₂ mixing ratio measured at the EC level ($\overline{s_c}$) of the following and previous half hours, and $\Delta t = 3600$ s. NEP, which is the net C sequestered by the ecosystem, was calculated as $NEP = -NEE$.

2.3. Stand Fertilization

[10] On the West Coast, fertilizer is usually applied aerially to forest stands during late fall, winter, or early spring with cool, wet, and windless weather in fog-free conditions. Under these weather and nongrowing conditions, (1) losses of applied N through ammonia volatilization are minimal, (2) intercepted fertilizer does not stay long on the foliage and gets washed to the ground, and (3) direct foliar uptake of N is small. An area of 1115 ha of Douglas fir forest on TimberWest Forest Corp. land on the east coast of Vancouver Island was aerially fertilized with urea at 200 kg N ha⁻¹ during 11–15 January 2007 using a Eurocopter SA315B helicopter (Western Aerial Applications Ltd., Chilliwack, British Columbia, Canada) with an in-house-engineered hydraulic-driven spreader bucket and a GPS-assisted guidance system. A block of 390-ha forest fertilized on 13 January included the DF49 EC flux tower footprint area. A nonfertilized area of about 17 ha (200 m × 850 m) on the southeast side of the fertilized block (500 m from the flux tower) served as a control for comparing differences in tree growth, C stocks, and greenhouse gas emissions. Location of the control area was chosen to minimize the number of times the tower footprint included winds blowing over the control area (winds from this direction accounted for only 5% of wind directions during the year). We observed that on the day of fertilizer application, about half of the fertilizer was retained in the snow-laden foliage, which was washed to the ground surface with the melting of intercepted snow in the following days.

2.4. Measurement of Soil N₂O Effluxes

[11] To study the effect of fertilization on soil N₂O emissions, and to compare regular urea to slow-release urea (ESN), we established a randomized complete block design (RCBD) experiment with a control and the two types of

urea, with four replications, on twelve 100-m² plots in the unfertilized (control) area. Fertilizer treatment at 200 kg N ha⁻¹ with urea and ESN of these plots was done manually on 11 April 2007, and N₂O efflux measurements began on 12 April and continued every 2–3 weeks until the end of the year.

[12] To determine soil N₂O efflux, we used the static chamber technique following the procedure described by Schiller and Hastie [1996]. PVC cylindrical collars of 21-cm diameter and 10-cm length were installed and firmly secured up to 5 cm deep in the soil. Before each measurement, Plexiglas circular covers, each with a vent tube and silicon rubber septum (Soil Moisture Systems, Tucson, Arizona, USA) at the top, fitted with a small fan underneath (with a 9-V battery secured at the top), were placed on the collars. Each cover had two circular rubber seals that allowed the cover to fit firmly to the collar, thus avoiding leaks, while the fan, directed upward, ensured proper mixing of the chamber headspace during efflux measurement. Gas samples of 20 cm³ from the chamber headspace were drawn with a syringe from near the soil surface immediately before placing the cover and at 3, 10, 20, and 30 min after placing the cover and were transferred to preevacuated 12-cm³ vials (Exetainers, Labco Limited, Buckinghamshire, UK). These samples were analyzed in the laboratory on a gas chromatograph (model 3800, Varian Inc., Palo Alto, California, USA) fitted with an electron capture detector using 5% methane balance argon as the carrier gas within 24 h of sampling. The N₂O mixing ratio (s_{N_2O}) of the samples was determined by comparing with zero and 1-ppbv standards, which were run twice, once before and once after the samples. The agreement between replicate samples at ambient concentrations was very good, differing by less than 1%. The Exetainers tested by storing gas samples of varying concentrations up to 6 weeks maintained a vacuum quite well. While fresh supplies of Exetainers shipped by air directly from the manufacturers in the United Kingdom were exceptionally well sealed, about 8% of the vials from the previous year had lost vacuum. However, we discarded any vials not showing enough suction at the time of injection of samples in the field.

[13] The increase in chamber headspace s_{N_2O} over the sampling time (up to 30 min) was generally nonlinear, especially when emissions were high, so the rates of change in s_{N_2O} (ds_{N_2O}/dt , $\mu\text{mol mol}^{-1} \text{h}^{-1}$) were obtained from either a linear increase up to 10 min or a logarithmic fit up to 30 min. The N₂O efflux (F_{N_2O} , $\mu\text{mol m}^{-2} \text{h}^{-1}$) was calculated using $F_{N_2O} = (\rho_a V/A) ds_{N_2O}/dt$, where ρ_a is the molar density (mol m^{-3}) of dry air, V is the volume (m^3) of the chamber headspace, and A is the cross-sectional area (m^2) of the collar.

[14] We also compared the chamber N₂O effluxes with those obtained using the soil N₂O gradient technique. On 11 July 2007, we inserted pointed 2-mm outer-diameter, 35-cm-long stainless steel gas-sampling tubes, each fitted with a silicone rubber septum at the top, into the ground near each collar. The tubes were bent at ~60° and pushed at this angle from the vertical so that the 10-cm-long perforated end of each tube was positioned horizontally 5 cm below the surface. Immediately after insertion, we pumped air into each tube to remove any soil particles blocking the perforations that could have occurred when the tubes were

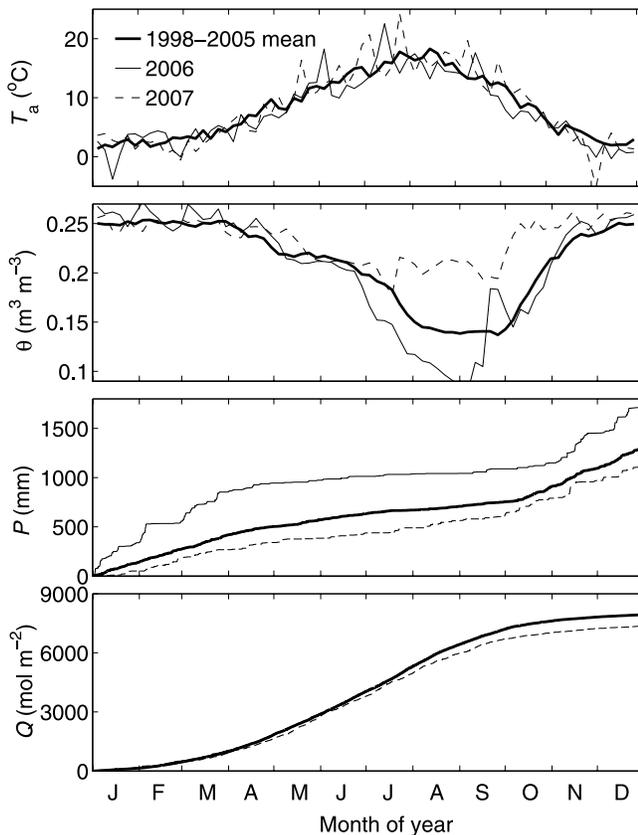


Figure 1. Interannual variations in 5-day mean air temperature (T_a), 0–30-cm soil water content (θ), cumulative precipitation (P), and cumulative total photosynthetically active radiation (Q) at the 58-year-old West Coast Douglas fir stand.

pushed into the ground. Gas samples from the 5-cm depth and the soil surface were also taken and injected into the Exetainers, generally in duplicate, before and after the chamber samplings and analyzed as above. Soil N_2O effluxes were calculated using Fick's first law as $F_{N_2O} = D_s dC_{N_2O}/dz$, where D_s , the diffusivity of the N_2O in the surface soil layer ($m^2 h^{-1}$), was obtained from measured soil water content and bulk density values and dC_{N_2O}/dz is the N_2O concentration gradient between 5-cm depth and the soil surface ($\mu mol m^{-4}$), with C_{N_2O} calculated from $\rho_a s_{N_2O}$. We used the relationship $D_s = 1.18 D_m \varepsilon^{2.28}$ obtained from diffusivity measurements on soil cores taken from this site but at a location about 200 m away from these measurements [Jassal et al., 2005], where D_m is the molecular diffusivity of N_2O ($0.051 m^2 h^{-1}$ at $20^\circ C$) and ε is the fractional air-filled porosity.

2.5. Calculation of Net GHG GWP

[15] We calculated the net GHG budget in the first year after fertilization by considering the CO_2 equivalence of the net change in soil N_2O emissions, the net change in NEP after adjusting for climate variability, and the GHG equivalence of energy needed for the manufacturing, transport, and aerial application of fertilizer urea. The CO_2 equivalent of soil N_2O emissions was calculated by multiplying the net N_2O loss by 296 [Ehhalt and Prather, 2001]. The net

change in NEP due to fertilization was computed using an empirical model and a process-based model. The former was developed by relating (using multiple-linear regression) the annual NEP values prior to fertilization to climatic factors. The latter was the EASS-BEPS, a land surface and ecosystem model, [Chen et al., 2007a, 2007b] coupled with the PnET-CN C balance model [Aber et al., 1997] and was run at half-hourly time steps for 1998–2007. Key parameters were estimated using an inverse algorithm with measured meteorological and EC data for 2001–2006. Since any change in soil CO_2 efflux is included in the measurement of NEP, there was no need to account this for the purpose of the calculation of net change in GHG GWP following fertilization.

[16] The Eurocopter SA315B consumed 8 L of JetA fuel per hectare, which is equivalent to $20 kg CO_2 ha^{-1}$ using the conversion in the IPCC guidelines for national greenhouse gas inventories. The CO_2 equivalent of the fertilizer was calculated from the energy requirement of fertilizer production and its transport at $40 MJ kg^{-1} N$ [Kongshaug, 1998; Ozkan et al., 2004]. Any change in soil CO_2 efflux due to fertilization, including that released upon urea hydrolysis ($(NH_2)_2CO + 3H_2O \rightarrow 2NH_4^+ + 2OH^- + CO_2$) and reduction of N_2O to N_2 (i.e., $2N_2O + CH_2 \rightarrow CO_2 + H_2O + 2N_2$), in the surface labile-C-rich layer is accounted for in the measurement of NEP. The energy equivalent of human labor, calculated as the CO_2 equivalent of 4 man hours per hectare of forest fertilization at $2 MJ h^{-1}$ [Ozkan et al., 2004], was negligible compared to fossil fuel energy involved in the manufacturing, transport, and application of fertilizer.

[17] While the focus of this study was to report change in calculated GHG GWP, arising because of changes in soil N_2O emissions and NEP, at DF49, we also report results on the effect of fertilization on NEP at two other nearby Douglas fir stands, a 19-year-old stand (HDF88) and a 7-year-old plantation (HDF00) (for site details, see Humphreys et al. [2006]). While the 58- and 19-year-old stands were similarly fertilized, the 7-year-old stand, because of its young age and competing understory, was fertilized manually at 80-g urea per tree along the tree's drip line.

3. Results

3.1. Weather

[18] The weather variables T_a , θ , P , and Q followed the same general seasonal trend between years (Figure 1). While the 5-day mean T_a during 2007 varied about the 9-year mean, P was well distributed, so that summertime (July–September) θ was the highest in the 10-year site record, indicating the absence of the generally observed summer drought, which limits productivity at this site [Jassal et al., 2007]. However, because of cloudy weather, annual Q was about 7% lower than the 9-year mean, suggesting possible growth limitation during 2007 due to low Q [Morgenstern et al., 2004].

3.2. C Exchange Between the Atmosphere and the Forest Ecosystem

[19] Figure 2 shows that fertilization increased annual NEP at all three sites in this West Coast Douglas fir chronosequence. It also suggests that a near-end-of-rotation

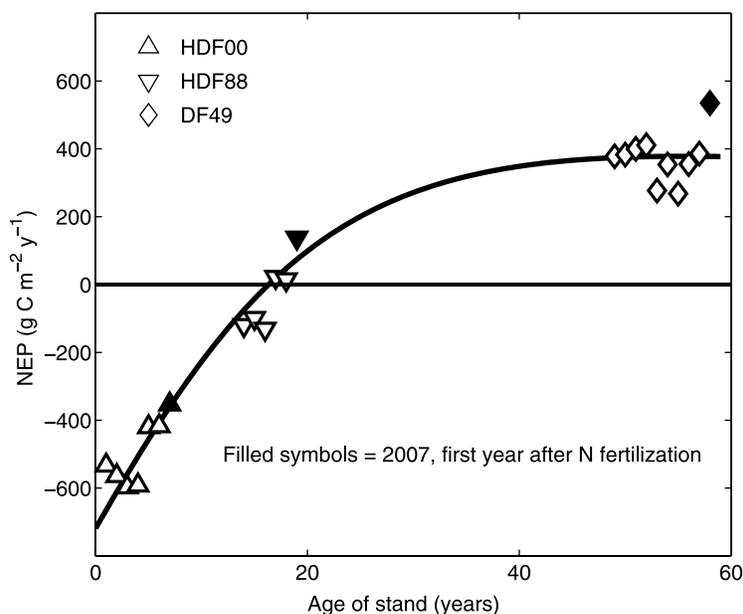


Figure 2. Effect of stand age and N fertilization on NEP in a chronosequence of West Coast Douglas fir stands.

stand, DF49, has reached a nearly constant annual growth rate with small interannual variability in NEP arising because of variations in seasonal and annual climate [Jassal *et al.*, 2007; Schwalm *et al.*, 2007; Morgenstern *et al.*, 2004] (see also this study). Uncertainty associated with annual estimates of NEP was addressed by assigning a random error of 20% to half-hourly measurements, which were then resampled 100 times using the bootstrap Monte Carlo method, and annual sums were calculated at 95% confidence levels. Such random error in the estimates of annual NEP at DF49 was found to be within ± 30 g C m⁻² [see also Morgenstern *et al.*, 2004; Schwalm *et al.*, 2007]. As fertilization of DF49 appreciably increased NEP over the previous year and the 9-year mean, part of this increase may be attributed to interannual climate variability as stated above. To account for the variation in climate, we fitted an empirical model to our 9 years of prefertilization annual NEP values:

$$\text{NEP} = 128 - 46T_{\text{MJ}} - 143\theta_{\text{AO}} + T_{\text{M}} + 0.107Q_{\text{MO}},$$

where T_{MJ} , θ_{AO} , T_{M} , and Q_{MO} are mean T_a (°C) for May–June, mean 0–30-cm θ (m³ m⁻³) for August–October, mean T_a (°C) for March, and total photosynthetically active radiation (mol m⁻²) for May–October, respectively. This model described our measurements fairly well ($R^2 = 0.83$) (Figure 3) with Q_{MO} explaining as much as 20% of the variance. Using this model and the above-noted climatic variables for 2007, we calculated NEP for 2007 assuming that the stand had not been fertilized. Fertilization increased measured NEP to 535 g C m⁻² compared to the calculated value of 326 g C m⁻² (Table 1), resulting in a 64% increase in the first year after fertilization. This was confirmed by the simulation results using the process-based model, which indicated that NEP in 2007 without fertilization would have been 312 g C m⁻² (Table 1). We also compared annual totals of daytime NEP values and found that fertilization

increased annual (2007) daytime NEP to 1258 g C m⁻² from 1162 ± 58 (plus or minus standard deviation) and 1163 g C m⁻² for 1998–2005 mean and 2006, respectively.

3.3. Soil N₂O and CO₂ Emissions

[20] Chamber measurements made 1 day after fertilizer treatments showed a mean uptake of about 0.06 $\mu\text{mol N}_2\text{O m}^{-2} \text{ h}^{-1}$ in the control plots compared to zero flux in the urea- and ESN-treated plots, with high plot-to-plot variability, especially in the fertilized plots. N₂O emissions in the fertilized plots started increasing slowly with the mean loss

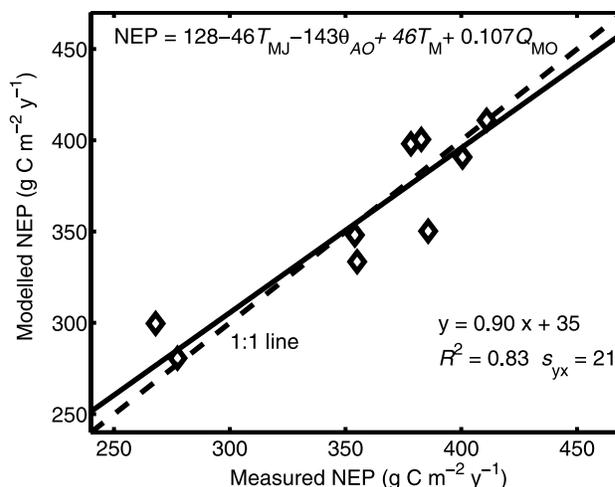


Figure 3. A comparison of EC-measured annual NEP values with those calculated using an empirical model used to determine the effect of climate variability. T_{MJ} is mean air temperature for May–June (°C), θ_{AO} is mean 0–30-cm soil water content (m³ m⁻³) for August–October, T_{M} is mean air temperature (°C) for March, and Q_{MO} is cumulative photosynthetically active radiation (mol m⁻²) for May–October.

Table 1. Effect of Nitrogen Fertilization on EC-Measured NEP in a 58-Year-Old West Coast Douglas Fir Stand and Comparison With an Empirical and a Process-Based Model^a

Year	Measured	Empirical Model ^b	EASS-BEPS Model ^c
1998–2005 mean	353 ± 51 ^d	358 ± 49 ^d	354 ± 61 ^d
2006	386 ± 22 ^e	356	364
2007	535 ± 31 ^e	326	312

^aUrea at 200 kg N ha⁻¹ aerially applied on 13 January 2007. NEP is in g C m⁻² y⁻¹.

^bUsing an empirical model fitted to 9-year (1998–2006) measured NEP and climate variables (see text and Figure 3).

^cUsing the EASS-BEPS model coupled with the PnET-CN model (see text).

^dPlus or minus standard deviation.

^ePlus or minus random error (see text).

peaking at 26 $\mu\text{mol N}_2\text{O m}^{-2} \text{h}^{-1}$ in the urea-treated plots on 24 July (about 3 months after fertilization) and then slowly declining (Figure 4a) probably because of decreasing soil temperature. From 15 August onward, N losses from ESN-treated plots were higher than those from urea-treated plots, indicating that most of the fertilizer N from the urea likely moved down and was distributed in the soil profile, while the ESN was still able to supply enough N for nitrification and subsequent denitrification in the active near-surface soil layer. Similar results were obtained with the gradient technique (Figure 4b), though the gradient fluxes were consistently higher (1.5 times on average) than those measured using the chamber technique (Figure 5). By the end of the first year of fertilization (i.e., 2007), chamber-measured cumulative N losses were about 37 and 35 mmol N₂O m⁻² in the urea- and ESN-treated plots, respectively, compared to an uptake of about 0.5 mmol N₂O m⁻² in the nonfertilized plots.

3.4. Net GHG GWP

[21] Table 2 shows that while the energy used in the production, transport, and aerial spreading of fertilizer at

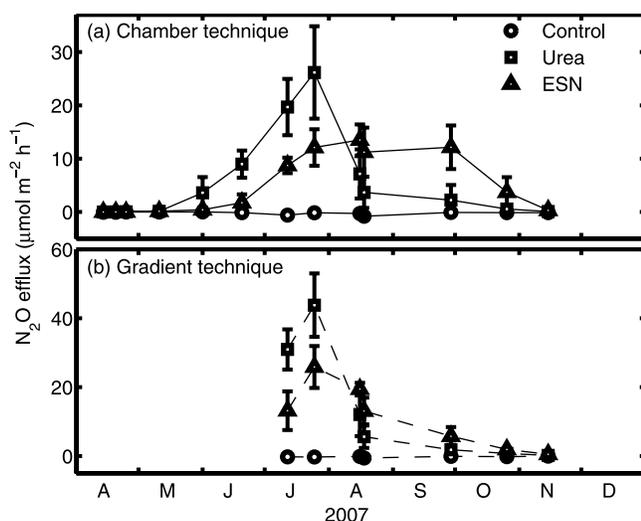


Figure 4. Effect of N fertilization using regular urea and a slow-release urea (ESN) on soil N₂O efflux (a) measured using static chambers and (b) calculated using the soil N₂O concentration gradient technique. Vertical bars indicate ±1 standard deviation.

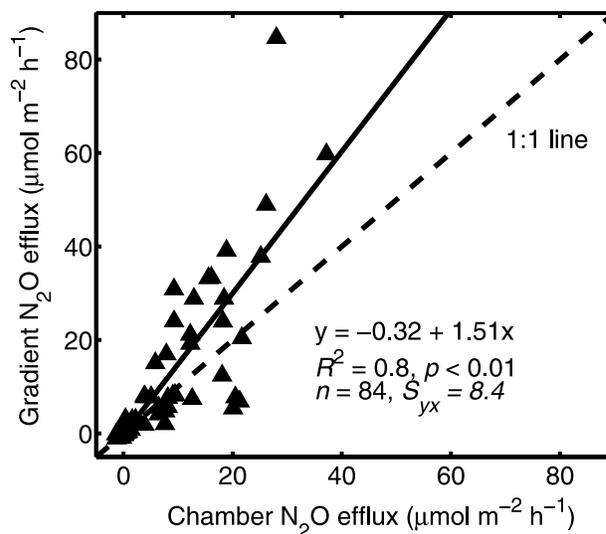


Figure 5. A 1:1 graph showing the relationship between chamber-measured and gradient N₂O effluxes.

200 kg N ha⁻¹ accounted for GHG emissions equivalent to 0.63 t CO₂ ha⁻¹, the major GHG GWP, equivalent to 4.75 t CO₂ ha⁻¹, was caused by soil N₂O emissions. However, a substantial increase in NEP, i.e., an additional uptake of atmospheric CO₂ by the trees, which was equivalent to a GHG GWP of -7.66 t CO₂ ha⁻¹, not only neutralized the increased GHG emissions due to N₂O emissions and the CO₂ equivalent of fertilizer application but resulted in an appreciable decrease in the net GHG GWP of -2.28 t CO₂ ha⁻¹ in the first year after fertilization.

4. Discussion

4.1. Effect of N Fertilization on C Sequestration

[22] The soil in this 58-year-old West Coast Douglas fir stand with an average C:N ratio of 44 in the 0–15-cm soil layer is deficient in available N. The only previous fertilizer application to this stand, 200 kg urea N ha⁻¹, was made in 1994 [Morgenstern et al., 2004]. Such nutrient-limited stands are likely to respond to fertilizer application such that increased nutrient availability stimulates aboveground net primary production [Fisher and Binkley, 2000; Chapin et al., 2002]. Foliar N analysis on current-year needles sampled at the end of the growing season on similar trees

Table 2. Change in Net Greenhouse Gas Global Warming Potential, in Terms of CO₂ Equivalent, in the First Year After Fertilization of a 58-Year-Old Douglas Fir Stand With 200 kg N ha⁻¹

Cause of Change in GWP	ΔCO ₂ Equivalence ^a (t CO ₂ ha ⁻¹)
Change in NEP	-7.66
Change in soil N ₂ O emission	+4.75 ^b
Fertilization	+0.63 ^c
Net change in GWP	-2.28

^aMinus indicates uptake of CO₂ by the ecosystem, whereas plus indicates release of CO₂ to the atmosphere.

^bCalculated by multiplying 16 kg N₂O ha⁻¹ with 296, the GWP of N₂O (see text).

^cOn the basis of energy consumed in the production, transport, and application of fertilizer urea.

Table 3. Effect of N Fertilization on Dry Needle Mass and N Content in Current Year Needles After the First Growing Season at HDF49^a

Position in the Crown	Needle Mass (mg/100 dry needles)		Percent N (dry needle basis)	
	Control	Fertilized	Control	Fertilized
Lower (1/6)	328	437	1.06	1.58
Middle (3/6)	436	568	1.14	1.65
Upper (5/6)	577	692	1.25	1.59
Mean	445	559	1.15	1.61

^aSampled 6 December 2007 and needles dried at 70°C.

in the unfertilized and fertilized areas showed that current-year needles in the unfertilized trees were severely deficient in N and that fertilization resulted in increased needle mass and N content (Table 3). Improved nutritional status is also known to increase leaf area index and favor net photosynthesis rate in Douglas fir [Brix, 1991] and to result in increased aboveground C storage by decreasing C allocation to fine roots [Teskey et al., 1995]. Our results showed that N fertilization of three West Coast Douglas fir stands of different ages resulted in increased NEP (i.e., C sequestration) in all the stands, with a substantial increase of about 64% in the near-end-of-rotation stand (DF49). This was confirmed with significant increases in the annual NEP values measured during the daytime, when confidence in the quality of EC measurements is much higher.

[23] Our empirical model indicated that annual NEP was sensitive to early spring and early summer temperatures, late summer soil water content, and extended summer (May–October) photosynthetically active radiation. The results suggested that while future climate with early springs would tend to increase C sequestration, warmer early summers would likely result in its decrease. Higher May–October photosynthetically active radiation would be expected to result in increased C sequestration. However, an increase in late summer soil water content would tend to decrease C sequestration, possibly because of a greater increase in respiration than photosynthesis.

4.2. Effect of Fertilization on N₂O Emissions

[24] Fertilizer N application from both urea and ESN resulted in soil N₂O emissions with significant soil N₂O effluxes after 1 month following fertilizer application. The emissions slowly increased thereafter, possibly with increased nitrification as a result of increasing soil temperature. Soil N₂O efflux was initially higher in urea-treated plots than ESN-treated plots likely because of faster hydrolysis, nitrification, and subsequent denitrification, but N₂O emission from ESN-treated plots exceeded that from urea-treated plots at 4 months after fertilizer application since by then most of the NO₃⁻ from urea either had moved deeper in the less active soil layer, where denitrification may be limited by lack of readily available C [McCarty and Bremner, 1992; Yeomans et al., 1992], or was fixed in microbial biomass, while ESN continued to dissolve and release NO₃⁻ substrate for denitrification. The highest soil N₂O efflux measured was 26 μmol N₂O m⁻² h⁻¹ in the urea-treated plots on 24 July. However, in nonfertilized plots we found consistent uptake of N₂O, except on 31 May, when the soil water content reached a minimum

(Figure 1), with efflux from individual plots reaching -1.3 μmol N₂O m⁻² h⁻¹. Total loss in both urea- and ESN-treated plots was about 16 kg N₂O ha⁻¹ in the first year, which is equivalent to 10 kg N ha⁻¹ or 5% of the applied fertilizer N. This contrasts with 1.25% assumed by the IPCC, on the basis of losses in farming systems [NGGIC, 2001], and 1.3–5.5% observed in onion fields in Japan [Toma et al., 2007]. Goossens et al. [2001] studied N₂O losses in Belgian soils under different land use and found that N₂O losses from arable and grass lands that received 325 kg (249 kg in manure plus 75 kg in fertilizer) N ha⁻¹ y⁻¹ were 1–3 and 10–36 kg N ha⁻¹, respectively, compared to an uptake of about 1 kg N ha⁻¹ in a forest soil. In dry and wet meadows that received 500 kg N ha⁻¹ for 2 consecutive years, Neff et al. [1994] found soil N₂O effluxes up to 8.5 μmol N₂O m⁻² h⁻¹ in dry meadows and 13 μmol N₂O m⁻² h⁻¹ in wet meadows. In a 100-year-old Norway spruce with yearly N addition of 35 kg in fertilizer and 12 kg in wet deposition, N₂O losses were 0.1 and 0.25 kg N ha⁻¹ y⁻¹ [Klemedtsson et al., 1997]. Flechard et al. [2005] measured N₂O emissions up to 36 μmol N₂O m⁻² h⁻¹ and N₂O uptake up to 8 μmol N₂O m⁻² h⁻¹ following fertilization of agricultural soils at 110 kg N ha⁻¹ y⁻¹, both showing marked diurnal patterns.

[25] The process of denitrification is capable of producing and consuming N₂O and NO [Firestone and Davidson, 1989]. However, in wet soils, any NO produced during the oxidative process of nitrification generally gets reduced before escaping from the soil such that N₂O is the dominant end product [Davidson et al., 2000]. It has also been reported that nitrifiers consume N₂O in denitrification, reducing it to N₂. The availability of organic C in soils has been correlated with the production of N₂ in soil cores [Mathieu et al., 2006], suggesting anaerobic denitrification in anaerobic microsites resulting from increased respiration. Also, common heterotrophic nitrifying bacteria like *Alcaligenes faecalis* and *Thiosphaera pantotropha* are often able to denitrify under aerobic conditions [Robertson et al., 1995], which occur under low soil NO₃⁻ but high soil C contents [Wrage et al., 2001], as may be expected in the surface litter layer. Our results showed that soil N₂O losses calculated from soil N₂O concentration gradient measurements consistently exceeded our chamber measurements (Figure 5). We also found that N₂O concentrations at the 5-cm depth were slightly higher than at the surface though chamber fluxes were zero and even negative. These results suggest that N₂O was reduced to N₂ in the surface (0–5 cm) layer. The N₂:N₂O ratio depends on the availability of C and NO₃⁻. With very low NO₃⁻ and high availability of labile C near the surface, N₂O produced in the subsoil may be reduced to N₂ as it diffuses to the soil surface. In a recent laboratory study using the recirculating gas (N₂:Ar) flow core technique on sealed intact forest soil cores, Dannenmann et al. [2008] have shown that N₂ emissions were substantially higher than N₂O, indicating that the dominant end product of denitrification was N₂ rather than N₂O.

[26] Chapuis-Lardy et al. [2007] reviewed the literature on soil N₂O fluxes with special interest in potential N₂O uptake and found that low-mineral N and high soil water content favor N₂O consumption. Rosenkranz et al. [2006] attributed lack of response of N₂O fluxes to simulated rainfall to simultaneous increases in N₂O production and

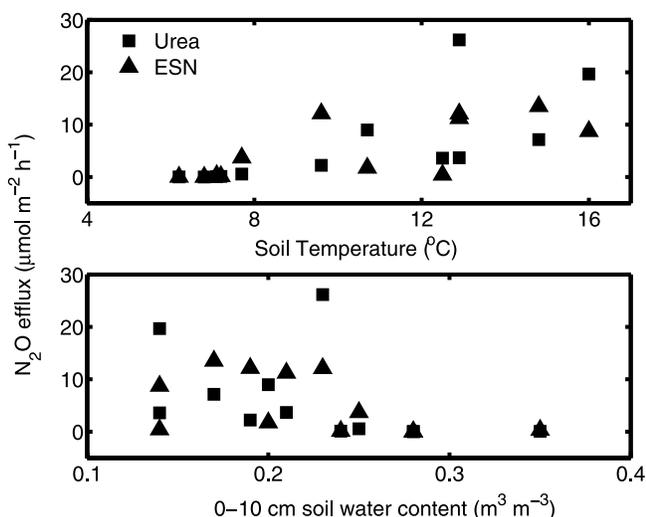


Figure 6. Effects of soil temperature and soil water content on soil N₂O efflux following N fertilization of a 58-year-old West Coast Douglas fir stand.

consumption. *Li and Kelliher* [2005] studied N₂O emissions in poorly and freely drained grazed grasslands in New Zealand using the chamber and the soil gas gradient techniques. Calculations using Fick's law underestimated N₂O emissions in poorly drained soils but overestimated them in freely drained soils. The authors attributed the former to a violation of the steady state assumption due to rapid changes in soil water and attributed the latter to the possible occurrence of gas convection. *Maljanen et al.* [2003] found good agreement between chamber and gradient N₂O fluxes in dry organic soils but not in wet soils. Some disagreement between chamber- and gradient-measured N₂O effluxes may be due to the difficulty in accurately parameterizing diffusivity values. We used diffusivity values measured on undisturbed soil cores taken from this site about 200 m away from the location of N₂O measurements [*Jassal et al.*, 2005].

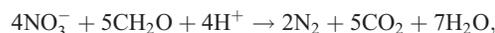
[27] It is well known that soil temperature and water regimes play a key role in the dynamics of N₂O production, reduction, and transport. We found that N₂O efflux generally increased with the increase in soil temperature (Figure 6), likely because of increased nitrification. However, the effect of soil water content was somewhat erratic; it showed either a very little or a somewhat negative effect on N₂O efflux (Figure 6). On one hand, high soil water content stimulates denitrification and thus increases the production of N₂O, but on the other hand, it decreases the diffusive transport of N₂O in the soil. Consequently, the residence time of N₂O in the soil increases, allowing its microbial reduction to N₂. Thus, under water-logged field conditions, N₂O emissions are low [*Firestone and Davidson*, 1989]. However, the influences of soil water content and temperature should be interpreted cautiously as soil water content is often negatively correlated to soil temperature [e.g., *Davidson et al.*, 1998; *Jassal et al.*, 2008]. *Klemmedtsson et al.* [1997] found that neither soil temperature nor soil water content was well correlated with N₂O emissions in Swedish forest soils following fertilization at 47 kg N ha⁻¹ y⁻¹. Thus, estimating total gaseous N losses requires measurement of

both N₂O and N₂ effluxes. We hypothesize that the gradient method provides estimates of net N loss in denitrification, i.e., in N₂O and N₂ together. That soil N₂O emissions in this ecosystem were highly correlated to soil temperature, and not to soil water content, suggests that the possibility of missing some episodic pulses due to rain events was low. It also justifies the linear interpolation between sampling dates for the purpose of computing cumulative losses. The results further suggest that had the fertilization in this RCBD plot experiment in the control area been implemented in January as in the tower footprint area, perhaps the total first year N₂O emissions would have been somewhat lower because of the movement of applied N to deeper soil depths before the increase in soil temperature.

4.3. Net GHG GWP

[28] Our results showed that at the end of the first year after fertilization of this 58-year-old Douglas fir stand with 200 kg N ha⁻¹, the net change in GHG GWP of the stand was -2.28 t CO₂ ha⁻¹, i.e., an additional sequestration of 2.28 t CO₂ ha⁻¹ compared to what it would have been without fertilization. Judging from the substantial increase in NEP in the first year, and as the effect of fertilization is expected to last for several years, it appears that increases in NEP in the following years will significantly increase C sequestration with likely reduction in N₂O emissions, thereby further decreasing the net GHG GWP. If, as stated in section 4.2, the January application of fertilizer in this plot experiment had resulted in lower N₂O emissions, this could have resulted in still greater reduction in net GHG GWP. In the absence of a second EC tower in the control area, we modeled the NEP in the flux tower footprint area how it would have been during 2007 were the stand not fertilized and assumed that the N₂O emissions in the flux tower footprint area were the same as those measured in the fertilized plots of the RCBD experiment in the control area.

[29] Although N fertilization may decrease CH₄ oxidation due to suppression of methanotrophic bacteria [*Mosier et al.*, 1991], possibly because of elevated NH₄⁺ concentrations [*Bodelier and Laanbroek*, 2004]. *Neff et al.* [1994] found that fertilization at 500 kg N ha⁻¹ showed no effect on soil CH₄ efflux in a dry meadow but significantly decreased CH₄ uptake in the wet meadow. Assuming that CH₄ dynamics in this rapidly draining soil were of insignificant consequence and that fertilization had little effect on possible uptake of CH₄, we did not include CH₄ in our analysis. However, preliminary estimates of soil CH₄ efflux measurements in 2006 at this site indicated an uptake of about 1.23 kg CH₄ ha⁻¹ y⁻¹ (K. Lee, personal communication, 2007), which with a GWP of 23 is equivalent to 0.03 t CO₂ ha⁻¹ y⁻¹ and is insignificant compared to other fluxes shown in Table 2. Any CO₂ emitted during denitrification according to the reaction



like that from urea hydrolysis and from a change in soil CO₂ efflux, would have been taken into account in the NEP measurements. We do not take into account fixation of atmospheric CO₂ during fertilizer manufacturing when ammonia is converted to urea. Regarding the possibility of any change in GWP due to a change in water vapor

concentration in the troposphere as a result of forest fertilization, we found little change in evapotranspiration.

[30] In addition to direct effects of N fertilization in decreasing the net GHG GWP, increased foliar N content of the forests has been shown to increase shortwave albedo [Ollinger *et al.*, 2007] with the potentially additional benefit of reducing shortwave radiative forcing. Measurements at our site, however, showed that although N fertilization appreciably increased N content of current-year needles by 0.46% (Table 3), it did not increase albedo. In fact, albedo during 2007 was slightly lower than the mean for 1998–2006 and was likely due to 2007 being the wettest of the 10 years with wet canopy conditions occurring during most of the year [Oguntunde and van de Giesen, 2004]. However, since foliar N is tightly linked to soil N availability, the former measurements, direct-field-sampling-based or remotely assessed, should be strong predictors of denitrification [Kulkarni *et al.*, 2008].

5. Conclusions

[31] 1. Fertilization of the 58-year-old West Coast Douglas fir stand with 200 kg N ha⁻¹ increased NEP by 64%, from 326 g C m⁻² to 535 g C m⁻², in the first year.

[32] 2. Fertilization resulted in significant N losses in denitrification with total N₂O loss by the end of first year amounting to about 5% of the applied N.

[33] 3. Initially, slow-release urea (ESN) looked promising in limiting N₂O emissions, but later in the growing season emissions exceeded those from regular urea, with the result that total N₂O emissions from ESN-treated plots by the end of the year were almost the same as from regular urea-treated plots.

[34] 4. Soil N₂O effluxes calculated using Fick's law (i.e., the gradient technique) exceeded the chamber measurements by 1.5 times, likely indicating that significant amounts of the N₂O produced in the soil below 5-cm depth were reduced to N₂ during its diffusion through the surface 0–5-cm labile-C-rich soil layer.

[35] 5. Compared to what GHG GWP would have been without fertilization, N fertilization decreased net GHG GWP by 2.28 t CO₂ ha⁻¹ at the end of first year, thereby indicating a favorable effect of fertilization despite significant N₂O emissions.

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