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Soil-Atmosphere Exchange of CO₂, CH₄ and N₂O in Northern Temperate Forests: Effects of Elevated CO₂ Concentration, N Deposition and Forest Fire

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Abstract

Global environmental change and its causes and effects in relation to natural and anthropogenic activity have been the recent focus of concern. An important component of this issue is the role management of soils plays in contributing as a source or sink of greenhouse gases (GHGs), e.g. carbon dioxide (CO₂), methane (CH₄), nitrous oxide (N₂O), etc. In forest ecosystems, especially, the production and consumption of these three GHGs are biologically mediated, but also strongly controlled by natural or man-made disturbances and other climate changes influenced environmental variables, e.g. elevated atmospheric CO₂ concentrations (Chapter 2), nitrogen (N) deposition (Chapter 3) and fire (Chapter 4). In this study, such environmental changes in forest ecosystems had the potential impacts to change the soil-atmosphere exchange of GHGs. Changes in soil GHG fluxes, furthermore, is that it varied with the environmental changes. Firstly, reduced CH₄ consumption with CO₂ enrichment was observed with increased levels of soil moisture as a result of increased leaf stomatal closure and evaporative water loss from the forest floor. Secondly, simulated N addition, which was conducted to evaluate the effects of elevated atmospheric N deposition on soil GHG fluxes, inhibited soil CH₄ uptake and stimulated soil N₂O emission in response to increased inorganic-N concentration. Finally, our low-intensity surface fire reduced soil CO₂ flux by the combustion of understory vegetation and litter layer. Especially, soil N₂O flux was represented different seasonal pattern, depending on the existence of charcoal. We considered that although increased inorganic-N levels in the burned area as a result of the fire influence preferentially the soil N₂O emission, it might be suppressed through net immobilization of NH₄⁺ or interfere with nitrification by fire-produced charcoal.

Key words: Greenhouse gases, elevated CO₂ concentration, nitrogen deposition, forest fire, forest ecosystems

CHAPTER 1

GENERAL INTRODUCTION

1.1 Climate change and greenhouse gases

Long-term data-sets for global temperature indicate a clear and consistent increase in global temperature, particularly since the industrial revolution (IPCC 2007). As concentrations of the main greenhouse gases (GHGs) have risen in the atmosphere, global temperatures have also increased. A rapid increase in atmospheric concentrations of the three main GHGs – carbon dioxide (CO₂), methane (CH₄) and nitrous oxide (N₂O) – is currently one of the major environmental issues in the world and is clear from measurements over the past few decades. Ice-core records for these three gases show that their concentrations in the atmosphere are now higher than at any time in the last 650,000 years, and probably even in the last 20 million years. Since around the time of the industrial revolution, fossil fuel combustion, increasingly intensive agriculture and an expanding global human population have been the primary causes for this rapid increase.

An accumulation of GHGs in the atmosphere contributes to global climate change according to the latest data of Intergovernmental Panel on Climate

Change (IPCC) showing rapid increases in their atmospheric concentrations (IPCC 2007). Together these three GHGs are estimated to contribute approximately 70% of the total global warming potential (GWP) (Sommerfeld *et al.* 1993). After Kyoto Protocol (IGBP Terrestrial Carbon Working Group 1998), forest ecosystem is expected as a big carbon sink for moderating global warming by way of CO₂ absorption. However, we have not enough information of CH₄ and N₂O.

1.2 Three main GHGs and their atmospheric concentrations

Between 10,000 and 150 years ago, atmospheric concentrations of CO₂, CH₄, and N₂O were relatively stable. In the last 150 years, concentration of CH₄ and N₂O increased 148% and 18%, respectively (IPCC 2007). Carbon dioxide concentration in the atmosphere increased from approximately 280 parts per million by volume (ppmv) in pre-industrial times to 382 ppmv in 2006 according to the National Oceanic and Atmospheric Administration's (NOAA) Earth Systems Research Laboratory, a 36% increase. The rapid increase in CO₂ emissions observed during the last 250

years is expected to continue for several decades to come. Various scenarios have been examined, depending on factors like fossil fuel use and efficiency. Even the best case scenario predicts further increases in CO₂ emissions until at least 2040 (IPCC 2007).

Methane concentration in the atmosphere has been increasing rapidly in the last century or so. Peak concentrations were previously 0.715 ppmv, but since the beginning of the industrial era levels have more than doubled to their current high of about 1.774 ppmv (IPCC 2007). Although such concentrations are much lower than those of CO₂, CH₄ molecules are much more effective at trapping the infrared radiation reflected from the earth's surface. Indeed, the GWP of CH₄ on a mass basis is 25 times that of CO₂ over a 100-year time horizon and it increases at an annual mean rate of 1%, which is much faster than the rate of increase of CO₂ (Schlesinger 1997, IPCC 2007); thus it can have a large effect on climate change.

Nitrous oxide is known to be destructive of stratospheric ozone. Atmospheric concentration of N₂O has risen markedly in the last 200 years, increasing from 0.270 ppmv in the pre-industrial era to 0.319 ppmv currently, with the rate of increase estimated to be 0.25% per year. Nitrogen oxide is chemically rather inert, with an atmospheric lifetime of approximately 120 years, and each molecule of N₂O has a much greater radiative forcing potential than a molecule of CO₂. Although its concentration is very small relative to that of CO₂, N₂O has 298 times more impact per unit weight than CO₂ over a 100-year time horizon (IPCC 2007).

1.3 The role of forest soils as a source or sink of GHGs

Generally, forest soils are important to identifying and enhancing natural sink for carbon (C) sequestration to mitigate the climate change (Lal 2005). Among the forest biomes, soils in temperate forest have been considered as a major source of CO₂ and N₂O, while acting as a significant sink for CH₄ (Robertson *et al.* 2000, Le Mer and Roger 2001, Raich *et al.* 2002, Fest *et al.* 2009). In particular, temperate forest soils are identified to be a small N₂O source because the temperate forests have comparatively large C:N ratios in litter and topsoil (Butterbach-Bahl and Kiese 2005) responsible for between 2.3 and 13.5% of global non-anthropogenic N₂O emissions (IPCC 2007). Under well-drained aerobic soils, moreover, CH₄ is consumed by methane-oxidizing bacteria (methanotrophs), which use the CH₄ as a source of carbon in the process of biological CH₄ oxidation (Castro *et al.* 1995, Price *et al.* 2004). Globally, such soils are estimated to have consumed 30 Tg CH₄ yr⁻¹ (1 Tg = 10¹² g) during 2000 to 2004, equivalent to 5% of the annual load of CH₄ to the atmosphere (IPCC 2007).

1.3.1 Carbon dioxide: fluxes and processes

Soil is the largest carbon reservoir in terrestrial ecosystems, recently estimated at 1500 Pg C (1 Pg = 10¹⁵ g), about triple of the plant biomass or twice of the atmospheric CO₂-C pool (IPCC 2001). Changes in the

size of the soil C pool can significantly affect atmospheric CO₂ concentration (Raich and Potter 1995). As a major flux between atmosphere and land, soil CO₂ flux is estimated to be 50–78 Pg C yr⁻¹ (Raich and Schlesinger 1992, Raich and Potter 1995, IPCC 2001), accounting for about 25% of the global CO₂ exchange. Carbon dioxide flux from soil to the atmosphere is a major component greenhouse gas emission and is a crucial pathway of the C cycle. It is highly sensitive to temperature may have a great influence on the magnitude of soil CO₂ flux (Luo and Zhou 2006). Therefore, more accurate estimation of soil respiration becomes one of the important issues in the estimation of global C cycle.

Soil CO₂ flux is a combination of biological, chemical and physical processes and also a sensitive indicator of essential processes in ecosystems such as: metabolic activity of roots, mycorrhizae and other soil organisms, decomposition of plant residues in soil and conversion of soil organic C to atmospheric CO₂ (e.g. Ewel *et al.* 1987a, Rochette *et al.* 1997). There are different estimates of autotrophic respiration for different forests and in temperate regions there are estimates ranging from 33% to 51% in broad-leaved forests, and 35% to 62% in coniferous forests (Edwards and Sollins 1973, Nakane *et al.* 1983, Ewel *et al.* 1987b, Bowden *et al.* 1993, Nakane *et al.* 1996, Striagl and Wickland 1998, Ohashi *et al.* 2000, Lee *et al.* 2003, Yang and Wang 2006, Wang *et al.* 2008).

Several factors can affect soil CO₂ flux rate. The most important ones are soil temperature and soil moisture content (Keith *et al.* 1997, Law *et al.* 1999, Janssens *et al.* 2001, Jiang *et al.* 2005, Kim *et al.* 2009). In forest ecosystems, such as tropical rainforests, temperate forests, and grasslands, soil CO₂ flux usually follows seasonal patterns in soil temperature, and soil temperature has been proposed as the most important factor in controlling soil CO₂ flux (Hanson *et al.* 1993, Billings *et al.* 1998, Epron *et al.* 2001). Soil CO₂ flux is driven largely by changes in soil temperature. Its seasonal pattern is positively correlated with changes in soil temperature when soil water is not limited (Curiel Yuste *et al.* 2003, Drewitt *et al.* 2002). However, a few studies have reported a weak correlation between soil CO₂ flux and soil surface temperature during drought conditions (Parkin and Kaspar 2003, Xu and Qi 2001b).

In arid and semiarid ecosystems, soil moisture content is the main factor in limiting soil CO₂ efflux. Thus, seasonal patterns of soil CO₂ flux are closely related to soil moisture content (Davidson *et al.* 2000), and strong inhibition of soil CO₂ efflux has often been observed in low precipitation regions (Griffis *et al.* 2004, Harper *et al.* 2005). Moreover, Xu and Qi (2001a) stated that the relationship between soil CO₂ flux and soil temperature was affected by soil moisture content in a young ponderosa pine plantation. The main limiting factors in seasonal patterns of soil CO₂ flux may depend on ecosystem and climate types. Other environmental variables could influence soil CO₂ flux, including the seasonal changes in soil microclimate (Raich and Potter 1995), quantity and quality of soil C substrates (Coûteau *et al.* 1995), soil C inputs

(Trumbore *et al.* 1996), gas diffusivity (Davidson and Trumbore 1995), plant root activity (Bowden *et al.* 1993, Raich and Tufekcioglu 2000), root and microbial biomass (Ryan *et al.* 1996), and stand productivity (Raich and Schlesinger 1992, Davidson *et al.* 2002). On a global scale, mean soil CO₂ fluxes correlate positively with net primary production (NPP) across different vegetation biomes (Raich and Schlesinger 1992).

Furthermore, annual soil CO₂ fluxes correlate positively with aboveground litter production in forest ecosystems (Schlesinger 1977, Raich and Schlesinger 1992). These studies indicate a tight linkage between plant productivity and soil CO₂ flux, due to the fact that primary production provides the organic fuel that drives soil metabolic activity.

1.3.2 Methane: fluxes and processes

Methane (CH₄) emission to the atmosphere mainly derives from natural wetlands, rice paddies (Hatano and Lipiec 2004, Naser *et al.* 2007), enteric fermentation in ruminant livestock, landfills, fossil fuel production and biomass burning, which result in total emissions of about 600 Tg CH₄ yr⁻¹, with about half of this coming from natural sources and half from human activities (IPCC 2001). Of all of these natural sources, though, 70% of emissions come from the soils (Ritchie *et al.* 1997). Methane can be both produced and consumed depending on microsite conditions in the belowground environment. Actually, between 43 and 90% of the CH₄ produced in the soil is consumed before it can be emitted to the atmosphere (Liebner and Wagner 2007).

There are two significant sinks for atmospheric CH₄: (i) the reaction with OH radicals in the troposphere (506 Tg yr⁻¹) (IPCC 2001); around 90% of the removal of CH₄ from the atmosphere occurs through this reaction (Lelieveld *et al.* 1998). (ii) The oxidation of CH₄ in aerobic soils, which is estimated to be between 29 and 38 Tg CH₄ yr⁻¹ (Ridgwell *et al.* 1999, Smith *et al.* 2000). Methane is predominantly used by methanotrophic bacteria (methanotrophs) in well-drained conditions, which use the CH₄ as a source of carbon in the process of biological CH₄ oxidation. The key as to whether a soil acts as a sink or source of CH₄ tend to be water condition. Forest soils are often strong sinks for CH₄, because transpiration by the trees keeps the soil moisture content from becoming too high. Temperate and boreal forest soils are considered to be a major sink for atmospheric CH₄ with a consumption of up to 9.3 Tg C yr⁻¹ (Stuedler *et al.* 1989). Yavitt *et al.* (1990) estimated that forest soils in the temperate region consume 2.4 Tg CH₄ yr⁻¹. However, the net soil CH₄ consumption can be reduced by land use change and/or climate change. Soil moisture content is the major factors controlling CH₄ production and oxidation.

In addition to water content of the soil, factors such as pH, soil temperature and inorganic N concentrations can be crucial in determining whether a particular soil will act as a sink for CH₄ or not (Hütsch *et al.* 1994). Bowden *et al.* (1998) in incubation experiments with soil from mixed hardwood stands, found that soil CH₄ uptake was strongly related to soil moisture content in

the forest floor, with a maximum uptake at approximately 70% of field capacity, and lower values as soil moisture content approached 100% of capacity. Also, CH₄ fluxes measured in tropical regions during a rainy season tend to be higher than those measured during a dry season (Reiners *et al.* 1998). Besides, soils receiving high N input from the atmosphere or through fertilization consume often less CH₄ than undisturbed soils (Stuedler *et al.* 1989, Castro *et al.* 1993, Butterbach-bahl *et al.* 1998, Kim *et al.* 2012). Finally, some authors noted a relationship between CH₄ consumption rates and soil temperature (Boeckx and Van Cleemput 1997, MacDonald *et al.* 1997, Savage *et al.* 1997), whereas others found a rather weak influence (Crill 1991, Dunfield *et al.* 1993).

1.3.3 Nitrous oxide: fluxes and processes

Soils are the dominant source of nitrous oxide (N₂O) emissions to the atmosphere, accounting for approximately 9 Tg N yr⁻¹ (Batjes 1996), which is about 50% of the total annual global emissions of around 17.7 Tg N yr⁻¹ (IPCC 2001). The most significant natural sources of N₂O in the atmosphere are tropical soils, contributing between 2.2 and 3.7 Tg N yr⁻¹, while temperate forest soils are estimated to contribute between 0.1 and 2.0 Tg N yr⁻¹. Agricultural soils also significantly contribute (Takakai *et al.* 2006, Toma *et al.* 2010), releasing 4.2 Tg N yr⁻¹ to the atmosphere (IPCC 2001).

Nitrous oxide emissions from soils have increased in the last few decades due to nitrogen (N) inputs by fertilizers to agricultural soils and N deposition into forest soils (Skiba and Smith 2000). Nitrogen oxide is produced in soils by nitrification and denitrification reaction. Nitrification is an aerobic microbial processes that converts ammonium (NH₄⁺) to nitrate (NO₃⁻) in the presence of oxygen. During denitrification, nitrates are transformed into nitrogen gas (N₂). Denitrification requires anoxic conditions, but denitrifying bacteria are facultative anaerobes (Schlesinger 1997). These two processes contribute two thirds of all emissions and they vary with climate, soil conditions and soil management (Skiba and Smith 2000). Nitrification is promoted by low rainfall, good drainage, and coarse soil texture while denitrification is promoted by high rainfall, poor drainage, fine soil texture and high organic matter (Groffman *et al.* 2000). Nitrous oxide production through nitrification or denitrification is not static and it can switch rapidly between the two processes, as soil aeration state changes (Skiba and Smith 2000). Denitrification rate is controlled by the rate of nitrification, which supplies NO₃ as a substrate (Schlesinger 1997). In general, N₂O production during nitrification is smaller than that produced during denitrification (Sahrawat and Keeney 1986).

Forest soils are a source of N₂O to the atmosphere but the source strength of different forests is still uncertain. Annual emissions range from 0 to 20 kg N ha⁻¹, depending on atmospheric N deposition, forest type and management practices (Tietema *et al.* 1991, Papen and Butterbach-Bahl 1999, Bowden *et al.* 2000, Beier *et al.* 2001). Several soil physical, chemical and

biological factors control microbial N_2O production in the soil. It has been very difficult to establish strong predictive relationships between N_2O flux and soil variables. In forest ecosystems, soil moisture content, soil temperature and N availability are the key factors regulating N_2O emissions (Papen and Butterbach-Bahl 1999, Butterbach-Bahl *et al.* 2002, Schindlbacher *et al.* 2004, Kim *et al.* 2012). Increase in soil moisture content, temperature or inorganic N availability usually stimulates soil microbial processes and consequent N_2O production. Nitrogen oxide production appears to be higher in wet but not completely water-saturated soils, because of lack of limiting nitrification (Granli and Bockman 1994). Dobbie *et al.* (1999) suggested that maximum N_2O emissions occur at the water-filled pore space (WFPS) of 80–85% in agricultural soils.

In forest soils, however, there is often no apparent relationship between soil N_2O fluxes and soil moisture. Although Papen and Butterbach-Bahl (1999) found weak coefficients of determination between soil N_2O fluxes and WFPS in a Norway spruce forest in Germany, Henrich and Haselwandter (1997) found no relationship between soil N_2O fluxes and soil moisture content in a spruce forest soil. But even if there is no apparent relationship, soil moisture content is still a main factor regulating soil N_2O flux. Also, temperature is known to be an important control of microbial processes. Although it has been shown that soil N_2O emissions are not always directly related to changes in temperature, temperature is considered to be an important factor in conjunction with other environmental variables (Yates *et al.* 2006). Moreover, Papen and Butterbach-Bahl (1999) continuously measured soil N_2O fluxes in a spruce and beech forests for 3 years. They found high inter-annual variation, which was controlled by the annual climate.

1.4 Objectives and structure of study

In forest ecosystems, the production and consumption of these three GHGs are biologically mediated (Conrad 1996), but also strongly controlled by disturbances resulting from fire, drought, wind damage, nitrogen deposition, logging and other climate change influenced environmental variables (Kim and Tanaka 2003, Ambus and Robertson 2006, Sullivan *et al.* 2008, Yashiro *et al.* 2008, Goldberg and Gebauer 2009). Recently, disturbances are getting increasingly serious and relate to an enormous shift in the global climate change. In this study, therefore, changes in soil GHG fluxes were measured in forest ecosystems under various environmental changes which have the potential ability to change the soil-atmosphere exchange of GHGs, e.g. elevated atmospheric CO_2 , N deposition and fire (Fig. 1.1).

1.4.1 Objectives of this study

The main concerns of this study were to determine:

1) How to change the soil-atmosphere exchange of GHGs in forest ecosystems following various environmental changes?

2) What is the main controlling factor influencing the

changes in soil GHG fluxes under each environmental change?

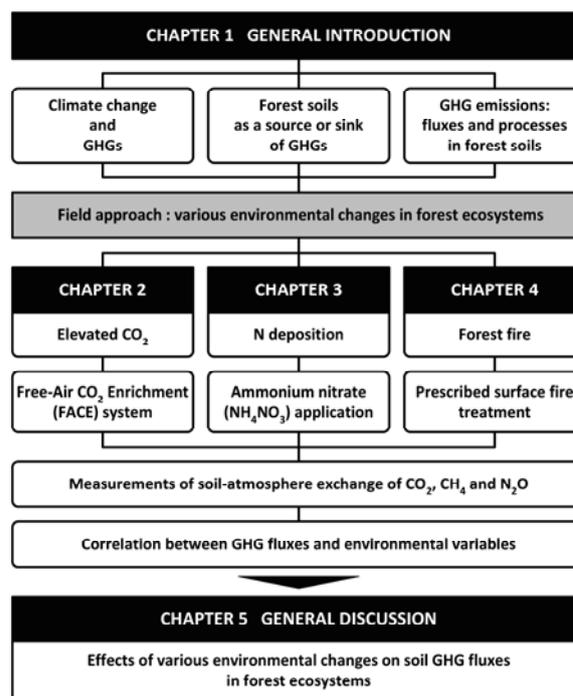


Fig. 1.1. Structure of the study.

1.4.2 Structure of this study

Chapter 1: This chapter introduces the important of forest soils for C storage and fluxes of GHGs. It represents the emission or uptake process of each GHG in forest and states the importance of environmental factors on their fluxes in forest ecosystems.

Chapter 2: This chapter presents the effects of elevated atmospheric CO_2 concentration on the soil CH_4 flux in forest ecosystems and the relationship between soil moisture and soil CH_4 flux. To determine this issue, field measurements were conducted using a Free Air CO_2 Enrichment (FACE) system under two forest soils for two growing seasons. The chapter has been published in the January issue of the Journal of Japan Society for Atmospheric Environment in 2011.

Chapter 3: This chapter describes the effects of atmospheric N deposition on soil CH_4 and N_2O fluxes in a brown forest soil and the relationship between soil variables and soil CH_4 and N_2O fluxes. To evaluate this issue, simulated N addition was initiated at the onset of this experiment and added as an ammonium nitrate (NH_4NO_3) solution distributed in four occasions per year during two growing seasons. The chapter has been published in the January issue of the Atmospheric Environment in 2012.

Chapter 4: This chapter explains the effects of forest fire on soil GHG fluxes in forest ecosystems and the relationship between soil variables and soil GHG fluxes. To assess this issue, prescribed low-intensity surface

fire, which is most common in the region, was conducted in white birch stands and the soil GHG fluxes and soil variables were determined during two growing seasons. The chapter has been published in the November issue of the European Journal of Forest Research in 2011.

Chapter 5: This chapter, finally, gives a summary of the main conclusion derived from the integration of the entire body of this study.

CHAPTER 2

REDUCED CH₄ CONSUMPTION IN TWO FOREST SOILS UNDER ELEVATED CO₂ CONCENTRATION

2.1 Introduction

The atmospheric concentration of CH₄ increased sharply throughout the 20th century and is now 2.48 times its pre-industrial level. Atmospheric CH₄ has recently reached a concentration not exceeded in the past 650,000 years, and it continues to increase (IPCC 2007). Methane is an important greenhouse gas, contributing roughly 20% of total non-H₂O radiative forcing (Menyailo and Hungate 2003). Its potential contribution to global warming is 25 times that of CO₂ when normalized by mass on a 100 year horizon (IPCC 2007). The only biological sink yet identified for atmospheric CH₄ is microbial CH₄ oxidation by methanotrophs in well-drained aerobic soils. These soils are estimated to have consumed 30 Tg CH₄ yr⁻¹ globally during 2000 to 2004, corresponding to 5% of the annual load of CH₄ to the atmosphere (IPCC 2007). Temperate forest soils are responsible for between 30–50% of the terrestrial CH₄ sink (e.g. Ojima *et al.* 1993).

The atmospheric CO₂ concentration has risen from around 280 μL L⁻¹ at the start of the industrial revolution to more than 385 μL L⁻¹ today (IPCC 2007); this increase, which is continuing, will have direct effects on physiological processes of plants and will feed back indirectly into the soil system in forest ecosystems. Such interactive feedback could alter the C and N cycles and the functioning of the ecosystem. Elevated CO₂ consistently stimulates the assimilation of photosynthetic carbon, and biomass accumulation, litterfall, and also fine root production and turnover (Karnosky 2003). Previous studies suggest that plant C/N ratios, nitrogen use efficiency and water use efficiency increase at elevated CO₂ concentrations (Rogers *et al.* 1994, Drake *et al.* 1997, Eguchi *et al.* 2008b). In particular, a higher efficiency of water use by plants and a slower transpiration rate resulting from the closure of leaf stomata should increase soil moisture, which is obviously one of the most important factors influencing atmospheric CH₄ consumption in well-drained aerobic soils.

Methane consumption often diminishes in response to elevated CO₂, an effect which has been related to increased soil moisture (Ambus and Robertson 1999, McLain *et al.* 2002, McLain and Ahmann 2008). Although a certain amount of soil water is required for methanotrophs to oxidize CH₄, influx of CH₄ into soils

may be impeded above a certain soil moisture content because no gas transport pores remain (Koschorreck and Conrad 1993, Del Grosso *et al.* 2000, McLain and Ahmann 2008, Dijkstra *et al.* 2010). Also, CO₂-induced increases in soil moisture under macroscopically aerobic soil conditions are likely to enhance methanogenesis (Castro *et al.* 2000, Eguchi *et al.* 2005, McLain and Ahmann 2008) and inhibit methanotroph activity (Ambus and Robertson 1999, Phillips *et al.* 2001). The CH₄ consumption rate in soil at high atmospheric CO₂ concentration may therefore be reduced.

To investigate the effects of elevated CO₂ on atmospheric CH₄ consumption by well-drained forest soil, we monitored changes in the soil CH₄ flux, soil moisture content and soil temperature at the atmospheric CO₂ enrichment facility in the Sapporo Experimental Forest maintained by Hokkaido University in northern Japan.

2.2 Materials and Methods

2.2.1 Site description

Field measurements were conducted at the Sapporo Experimental Forest of Hokkaido University in northern Japan (43°06' N, 141°20' E), using a Free Air CO₂ Enrichment (FACE) system in an experimental nursery under two soil types (brown forest soil × volcanic ash soil) for the two growing seasons 2008 and 2009. The FACE experimental design was based on the system at Davos, Switzerland (Hättenschwiler *et al.* 2002). Our site consists of three control and three treatment circular plots, each 6.5 m in diameter, surrounded by 5 m tall dispersal pipes. Since fumigation began in June 2003, CO₂ has been supplied so as to maintain the concentration at 130 μL L⁻¹ above ambient, or 500 μL L⁻¹, in three treatment plots, while the three control plots were subjected to ambient concentration of 370 μL L⁻¹ as of 2003 (Eguchi *et al.* 2005, 2008a, 2008b).

In October 2002, we divided each FACE ring into two and set two soil types: brown forest soil and volcanic ash soil, which are distributed over wide areas of northern Japan. Brown forest soil is native to the Sapporo Experimental Forest, and immature volcanic ash soil was taken from the Tomakomai Experimental Forest of Hokkaido University (42°40' N, 141°37' E) and mixed homogeneously with the soil at the depth of 20 cm at September 2002 and kept them for successive 6 years. Although fine soil density (< 2 mm in diameter) was significantly higher in volcanic ash soil than in brown forest soil, coarse soil density (> 2 mm in diameter) in brown forest soil was slightly higher than that in volcanic ash soil (Table 2.1). Volcanic ash soil had a lower solid phase and a higher gaseous phase than brown forest soil. However, there was no significant difference in maximum soil moisture content between two soil types (Eguchi *et al.* 2008a). A total of twelve treatment plots were established at the site: two CO₂ levels × two soil types × three replications.

In May 2003, after soil foundation, eleven types of two-year-old deciduous tree seedlings that are native to

the cool-temperate region in Japan were planted in each soil type in each FACE ring. These tree species are alder, two types of birch, larch, basswood, kalopanax, Manchurian ash, elm, maple, beech, and oak. Further details of the site and the experimental design are described by Eguchi *et al.* (2008b). Snowcover lasts for about 5 months from late December to April.

2.2.2 Flux measurement

Soil-atmosphere fluxes of CH₄ were determined by a closed-chamber method, with gas samples collected on four occasions to a monthly schedule during the growing season from July to October in 2008 and 2009. One selected sampling point was installed in each soil type in each FACE ring. To facilitate transportation of the chambers to and from the field, the chambers were constructed so that six chambers having slightly different sizes could fit nested within each other. The height of each chamber is the same (25 cm), but the diameters vary from 18.5 cm to 21.0 cm at 0.5 cm regular intervals. The chambers were installed to a depth of 3 cm below the soil surface. Flux measurements were generally done between mid-morning and noon. At 0, 10, 20, 40 and 60 minutes after the lid was put in place, a 20-mL gas sample from the headspace of the chamber was drawn into a 10-mL vacuum vial sealed with a butyl rubber stopper (SVF-10, Nichiden-Rika, Kobe, Japan). The CH₄ concentration in gas samples was analyzed using a gas chromatograph (GC-8A and GC-14B, Shimadzu, Kyoto, Japan) equipped with a flame ionization detector (Morishita *et al.* 2003, Takakai *et al.* 2008). The CH₄ concentration was calibrated with standard gases containing 0 and 2.03 ppmv. Soil CH₄ flux was calculated as follows:

$$F = \rho \times V/A \times \Delta c/\Delta t \times 273/(273 + T) \times \alpha \quad (1)$$

where F is the flux ($\mu\text{g C m}^{-2} \text{ h}^{-1}$), ρ is the gas density of CH₄ at STP ($0.716 \times 10^9 \mu\text{g m}^{-3}$), V is the volume of the chamber (m^3), A is the cross-sectional area of the chamber (m^2), $\Delta c/\Delta t$ is the rate of change of the gas concentration (c) inside the chamber per unit time (t) during the sampling period ($\text{m}^3 \text{ m}^{-3} \text{ h}^{-1}$), T is the air temperature ($^{\circ}\text{C}$), and α is a conversion factor to transform CH₄ into C. The CH₄ flux was calculated using linear regression between the gas concentration inside the chamber and the measured time. More than 71% of all flux measurements had a regression coefficient $R^2 > 0.7$. Positive values represent net production from the soil, and negative values indicate net uptake by the soil.

During gas sampling, the soil temperature and soil moisture content at 10 cm depth were measured adjacent to each chamber. The temperature was measured using a thermistor thermometer (CT-220, Custom, Tokyo, Japan), and gravimetric water content was determined by weighting the samples before and after drying at 105 $^{\circ}\text{C}$ for 48 hours, after collection with a core sampler.

2.2.3 Statistical analysis

All statistical analyses were performed using SAS 8.1 software (SAS 2001). The distributions of the measured soil moisture content, soil temperature and soil CH₄ flux were tested for normality with the Shapiro-Wilk test. Mann-Whitney U-test, a non-parametric ANOVA, was used to examine the statistical significant differences in all parameters between two treatments in each soil type. Two-way repeated measures analysis of variance was undertaken to determine the changing patterns of the variables under atmospheric CO₂ enrichment. Also, correlation analysis and stepwise linear regression analysis were used to quantify the relationship between soil CH₄ flux and environmental factors, e.g. soil moisture content and soil temperature.

Table 2.1. Initial physical properties of brown forest soil and volcanic ash soil (Eguchi *et al.* 2008a)

	Brown forest soil	Volcanic ash soil	x^2	P
Fine soil density ($\times 10^6 \text{ g m}^{-3}$)	1.88 \pm 0.20	2.42 \pm 0.16	4.62	*
Coarse soil density ($\times 10^6 \text{ g m}^{-1}$)	2.07 \pm 0.11	1.82 \pm 0.16	2.24	NS
Maximum volumetric water content (%)	35.67 \pm 2.76	31.51 \pm 1.39	0.25	NS
Solid phase (%)	52.58 \pm 3.55	40.84 \pm 2.86	23.64	***
Liquid phase (%)	35.67 \pm 1.33	18.49 \pm 2.18	13.93	***
Gaseous phase (%)	11.74 \pm 4.20	40.67 \pm 3.29	16.91	***

Values indicate the mean \pm SE ($n = 6$). Differences between the two soil types were evaluated using the maximum-likelihood ratio method with a GLMM procedure. NS means that the values are not significantly different. Asterisks denote significant differences at * $P < 0.05$ and *** $P < 0.001$.

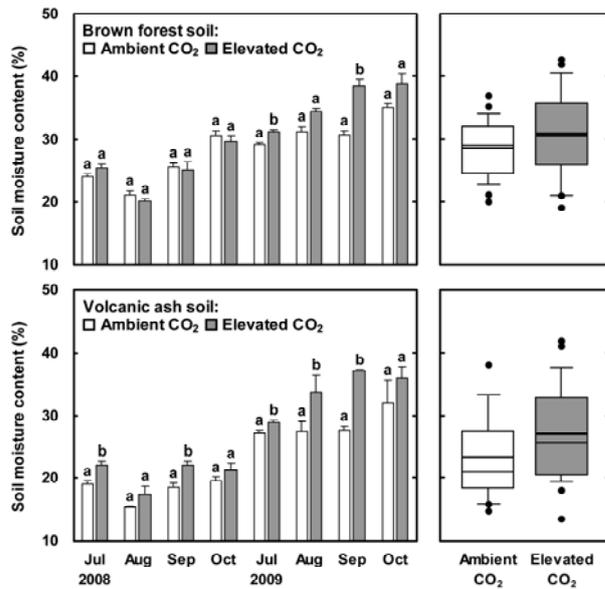


Fig. 2.1. Changes in soil moisture content under atmospheric CO₂ enrichment during two growing seasons. Values with different small letters indicate significant differences between two treatments in each measurement date (Mann-Whitney *U*-test, $P < 0.05$). Bold and thin lines in each box plot correspond to the mean and median values ($n = 24$). (Developed from Kim *et al.* 2011b, with kind permission of Japan Society for Atmospheric Environment)

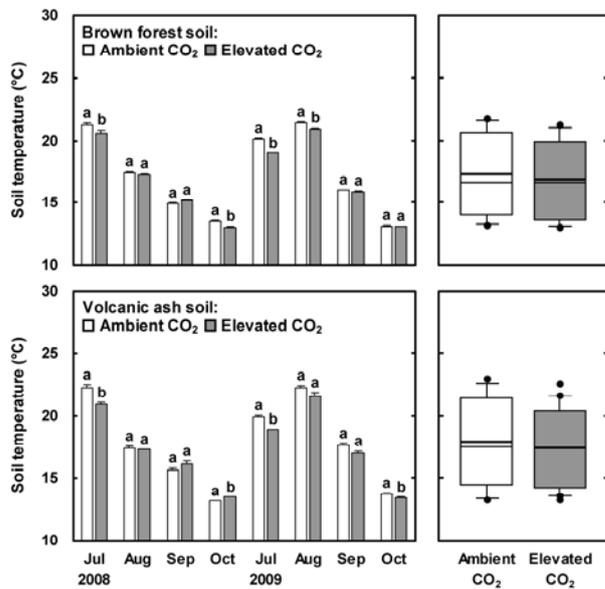


Fig. 2.2. Changes in soil temperature under atmospheric CO₂ enrichment during two growing seasons. Values with different small letters indicate significant differences between two treatments in each measurement date (Mann-Whitney *U*-test, $P < 0.05$). Bold and thin lines in each box plot correspond to the mean and median values ($n = 24$).

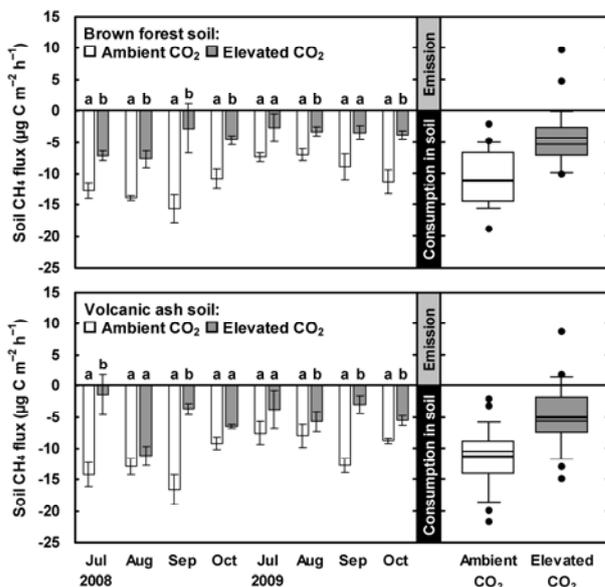


Fig. 2.3. Changes in soil CH₄ flux under atmospheric CO₂ enrichment during two growing seasons. Values with different small letters indicate significant differences between two treatments in each measurement date (Mann-Whitney *U*-test, $P < 0.05$). Bold and thin lines in each box plot correspond to the mean and median values ($n = 24$). (Developed from Kim *et al.* 2011b, with kind permission of Japan Society for Atmospheric Environment)

2.3 Results

Soil moisture was greater in the elevated CO₂ plot than in the ambient CO₂ plot during the two growing seasons (Fig. 2.1, Table 2.2). The soils in the elevated CO₂ plot consistently contained significantly higher soil moisture than at of ambient CO₂, by 7% in brown forest soil and 17% in volcanic ash soil.

The elevated CO₂ concentration decreased soil temperature in the elevated CO₂ plot as compared with the ambient CO₂ plot during the two growing seasons (Fig. 2.2, Table 2.2). The soils in the elevated CO₂ plot consistently contained significantly lower soil temperature than at of ambient CO₂, by 2% in brown

forest soil and 3% in volcanic ash soil.

Moreover, soil CH₄ consumption rates in ambient CO₂ plot were 10.94 $\mu\text{g C m}^{-2} \text{h}^{-1}$ in brown forest soil and 11.23 $\mu\text{g C m}^{-2} \text{h}^{-1}$ in volcanic ash soil. Comparable values for elevated CO₂ plot were reduced by more than 50%, to 4.45 and 5.12 $\mu\text{g C m}^{-2} \text{h}^{-1}$ respectively (Fig. 2.3). However, there was no significant difference in soil CH₄ consumption between the two soil types (Table 2.2). Regardless of the soil type, soil CH₄ consumption was observed at all measurement points in the ambient CO₂ plot, but approximately 13% of all sampling points in the elevated CO₂ plot gave net CH₄ production (Fig. 2.4).

Table 2.2. Repeated measures analysis of variance for the effects of atmospheric CO₂ enrichment on soil moisture content, soil temperature and soil CH₄ flux during two growing seasons

Source	df	Soil moisture content			Soil temperature			Soil CH ₄ flux		
		MS	<i>F</i> -value	<i>P</i> > <i>F</i>	MS	<i>F</i> -value	<i>P</i> > <i>F</i>	MS	<i>F</i> -value	<i>P</i> > <i>F</i>
<i>Between subjects</i>										
Soil type	1	393.05	28.11	0.0007	6.30	16.28	0.0038	5.44	0.13	0.7254
CO ₂ level	1	213.52	15.27	0.0045	3.76	9.71	0.0143	951.11	23.14	0.0013
Soil × CO ₂	1	23.63	1.67	0.2298	0.02	0.05	0.8241	0.88	0.02	0.8873
<i>Within subjects</i>										
Date	7	427.71	43.93	<0.0001	126.29	1538.8	<0.0001	47.4	2.53	0.0249
Date × Soil	7	19.01	1.95	0.1067	0.78	9.55	<0.0001	5.04	0.27	0.9636
Date × CO ₂	7	22.14	2.27	0.0652	0.68	8.3	<0.0001	33.27	1.77	0.1110
Date × Soil × CO ₂	7	1.71	0.18	0.9704	0.18	2.16	0.0515	13.03	0.69	0.6765

(Modified from Kim *et al.* 2011b, with kind permission of Japan Society for Atmospheric Environment)

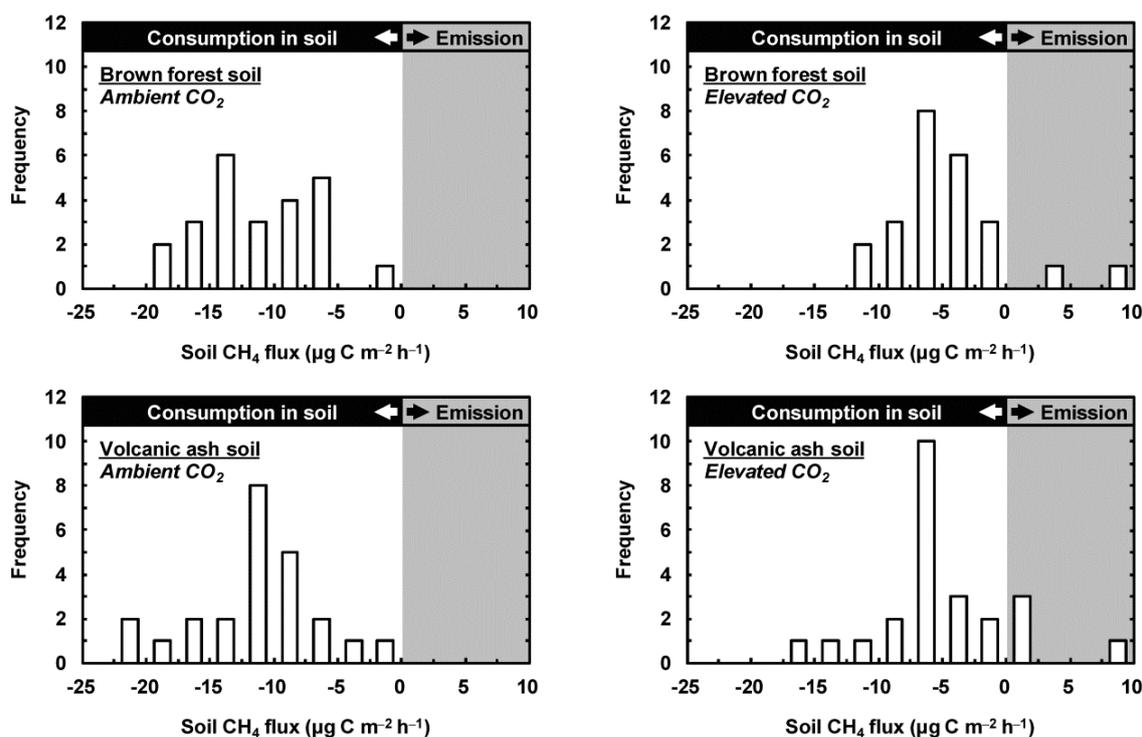


Fig. 2.4. Frequency distribution of soil CH₄ flux in four treatment plots during two growing seasons. (Reprinted from Kim *et al.* 2011b, with kind permission of Japan Society for Atmospheric Environment)

2.4 Discussion

During the measurement period, increased soil moisture content and decreased soil temperature was found in the elevated CO₂ plot for both soil types, as compared with the ambient CO₂ plot. In a previous study at the same experimental location, Eguchi *et al.* (2005) reported that volumetric soil moisture content in the elevated CO₂ plot was approximately 28% higher than that in the ambient CO₂ plot during two growing seasons in 2003 and 2004. The increase in soil moisture induced by elevated CO₂ is due to greater efficiency of water use by the plant as a result of increased leaf stomatal closure. Eguchi *et al.* (2008b) found that elevated CO₂ reduced water flow, reduced leaf-specific hydraulic conductivity and reduced the total vessel area of the petiole in sun leaves, regardless of species or soil type. They concluded that plants grown at elevated CO₂ concentrations generally have lower stomatal conductance or greater water use efficiency, indicating acclimation of hydraulic structure. Eguchi *et al.* (2005) also suggested that evaporative water loss from the forest floor decreased significantly under the elevated CO₂ plot, because soil surface temperature fell in both soils as a result of increase in leaf area index that blocked direct sunlight at the soil surface. Greater soil moisture in the elevated CO₂ plot is consistent with the findings of Eguchi *et al.* (2005, 2008b). In particular, the findings of Eguchi *et al.* (2005, 2008b) could explain why soil moisture became higher in the elevated CO₂ plot in the present study. Our result was comparable with previous field studies (McLain and Ahmann 2008) which found that volumetric soil moisture was on average 17% higher in the 0–30 cm layer of soils at elevated CO₂ compared to ambient, at the Free Air Carbon Transfer and Storage (FACTS)-1 site in a 25-year-old loblolly pine plantation in the Duke Forest, NC, USA. A similar result has been reported, that CO₂ fumigation leads to greater soil moisture content, on average by 11% over ambient CO₂, in aspen stands in Michigan, USA (Ambus and Robertson 1999).

In field measurements, decreases in CH₄ consumption with CO₂ enrichment have been associated with increased levels of soil moisture (Ambus and Roberson 1999, McLain *et al.* 2002, McLain and Ahmann 2008), but no change in CH₄ consumption has been reported in a field study (Mosier *et al.* 2002) or in laboratory incubation (Dijkstra *et al.* 2010). They suggested that, rather than influence methanotroph activity, soil moisture reduced CH₄ diffusion into the soil, inhibiting CH₄ consumption, and stimulated CH₄ production by methanogens (Ambus and Roberson 1999, McLain *et al.* 2002, McLain and Ahmann 2008). Although the present mean soil CH₄ consumption rate of 10.94–11.23 μg C m⁻² h⁻¹ in the ambient CO₂ plot was within the range of 2.9–214.7 μg C m⁻² h⁻¹ reported in Japanese forests (Morishita *et al.* 2007, Ishizuka *et al.* 2009), and was comparable to the range of 11–43 μg C m⁻² h⁻¹ found for temperate forests (Smith *et al.* 2000), we considered that our mean value in experimental nursery was decreased the level as compared with the values in forest areas in same

temperate zone. Five years after fumigation began in June 2003, we noticed evidence of depressed CH₄ consumption, relative to the ambient CO₂ plot, by the two soil types in the elevated CO₂ plot during the two growing seasons. McLain and Ahmann (2008) suggested that soil moisture is a key regulator of diffusive transport of atmospheric CH₄ into soils. They found that the negative correlation between water holding capacity and soil CH₄ consumption in FACTS-1 soil incubations from both ambient and elevated CO₂ rings, with decreasing CH₄ oxidation correlated with increasing soil moisture. In this study, also, negative linear relationship was found between soil CH₄ consumption and soil moisture content in the ambient CO₂ ($r = 0.34$, $P < 0.05$) and elevated CO₂ ($r = 0.64$, $P < 0.001$) plots during two growing seasons (Fig. 2.5, Table 2.3). A similar relation between soil CH₄ consumption and soil moisture has been observed in European and American forest soils (Koschorreck and Conrad 1993, McLain *et al.* 2002). However, we observed no significant difference between soil temperature and soil CH₄ consumption in both soil types. Regardless of soil type, only soil moisture content was the dominant factors that controlled soil CH₄ consumption in our experiment from stepwise linear regression analysis (Table 2.3).

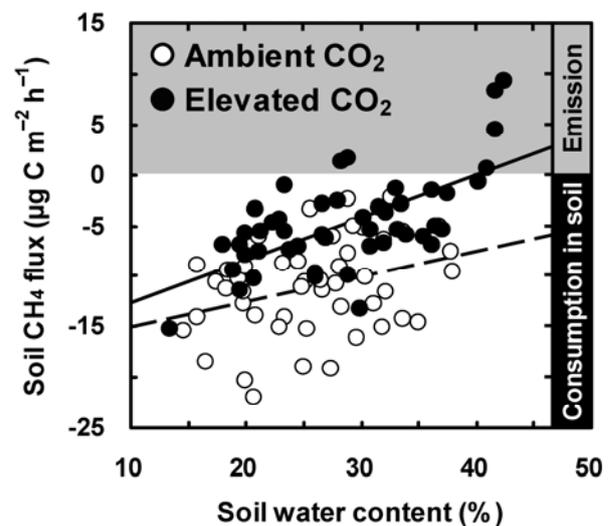


Fig. 2.5. Relationship between soil CH₄ flux and soil moisture content during two growing seasons. Linear regression lines are shown for ambient (dotted line) and elevated (solid) CO₂ plot. (Reprinted from Kim *et al.* 2011b, with kind permission of Japan Society for Atmospheric Environment)

Table 2.3. Relationship between soil CH₄ flux and soil variables in a brown forest nursery soil during two growing seasons

Treatment	Pearson correlation coefficient		Stepwise linear regression	
	SMC	ST	Equation	Adj. R^2
Ambient CO ₂	0.34*	0.12	$y = 0.25 \text{ SMC} - 17.67$	0.09*
Elevated CO ₂	0.64***	0.05	$y = 0.42 \text{ SMC} - 16.79$	0.40***

SMC and ST indicate soil moisture content and soil temperature, respectively. Asterisks with values represent the level of significance of correlations and regression models (* $P < 0.05$ and *** $P < 0.001$).

In particular, soil CH₄ consumption was observed at all measurement points in the ambient CO₂ plot, whereas around 13% of all sampling points in the elevated CO₂ plot produced CH₄ overall, regardless of soil type. Although the FACE site provides fundamentally well-drained aerobic soil conditions, localized anaerobic conditions due to increasing plant water use efficiency or decreasing transpiration rate, which stimulates the activity of methanogens, may increase in response to environmental factors (e.g. increase in soil moisture). According to McLain and Ahmann (2008), more than 6% of the 360 activity assays showed net CH₄ production, and of these, 80% were collected from elevated CO₂ plots. Net CH₄ production indicates that methanogenesis can take place in macroscopically aerobic soil conditions, because long term repeated exposure to elevated CO₂ causes fundamental changes in these soils, including increases in moisture. Eventually, elevated CO₂ can exert an influence in reducing the CH₄ sink in forest ecosystems. Our results suggest that net CH₄ production should be considered as a contributor to reduced CH₄ consumption at elevated CO₂ concentrations in well-drained forest soils.

2.5 Conclusions

Theoretically, increase in soil moisture induced by elevated CO₂ concentration is due to greater efficiency of water use by the plant as a result of increased leaf stomatal closure (e.g. Eguchi *et al.* 2005). Moreover, evaporative water loss from the forest floor decrease under the elevated CO₂ plot, because soil temperature fell as a result of increase in aboveground biomass and leaf area index that blocked direct sunlight at the soil surface. In field measurement, reduced CH₄ consumption with CO₂ enrichment has been associated with increased levels of soil moisture (Ambus and Roberson 1999, McLain *et al.* 2002, McLain and Ahmann 2008) including this study (Fig. 2.6), but no change in CH₄ consumption has also been reported in field or laboratory experiments (Mosier *et al.* 2002, Dijkstra *et al.* 2010).

Most researchers suggest that increased soil moisture reduced CH₄ diffusion into the soil, inhibiting CH₄ consumption and stimulated CH₄ production by methanogens rather than influence methanotroph activities. McLain and Ahmann (2008) found a negative correlation between soil CH₄ oxidation and soil moisture and they suggested that soil moisture is a key

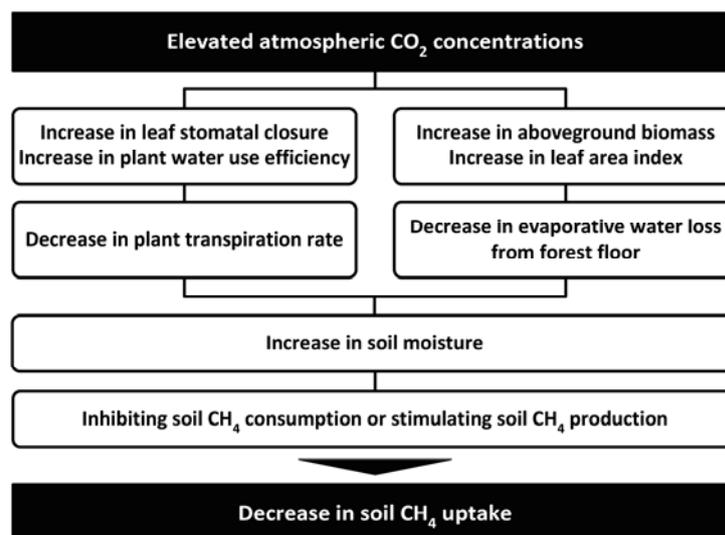


Fig. 2.6. Overall effects of the elevated CO₂ concentrations on soil CH₄ flux.

regulator of diffusive transport of atmospheric CH₄ into soils. In this study, also, a negative linear relationship was found between soil CH₄ consumption and soil moisture content in both soil types. Especially, soil CH₄ consumption was observed at all measurement points in the ambient CO₂ plot, whereas around 13% of all sampling points in the elevated CO₂ plot produced CH₄ overall, regardless of soil type. Although the FACE site provides fundamentally well-drained soil conditions, localized anaerobic conditions, which stimulates the methanogen activities, may increase in response to higher soil moisture. In conclusion, elevated CO₂ can exert an influence in reducing the CH₄ sink in forest ecosystems.

CHAPTER 3

SOIL CH₄ AND N₂O FLUXES IN A YOUNG LARCH PLANTATION

RECEIVING SHORT-TERM NITROGEN INPUTS

1 Introduction

Increases in N deposition in broad areas of Europe and North America, and parts of Asia, over the last 50 years have resulted in adverse environmental effects (Galloway *et al.* 2004). Recent modeling projects have predicted increased rates of atmospheric N deposition over the next decades in most regions of the globe. Atmospheric N deposition rates in the temperate zones of North America, Europe, and East Asia have increased dramatically during recent decades due to emissions of nitrogen oxides (NO_x) and ammonia (NH₃) from combustion processes and agricultural activities (Vitousek 1994, Galloway *et al.* 1995, Holland *et al.* 1999).

Between 1860 and the early 1990s, the amount of reactive nitrogen (Nr) created by natural terrestrial processes decreased by 15% while Nr creation by anthropogenic processes increased tenfold. Globally, the quantity of reactive atmospheric N inputs increased from 41 Tg N yr⁻¹ in 1950 to 103 Tg N yr⁻¹ in 2000, with a proportional increase in deposition on land (Galloway *et al.* 2004). One-third of global nitrogen inputs entered terrestrial ecosystems and one-tenth entered forests (Churkina *et al.* 2007). For example, atmospheric N deposition has exceeded 25 kg N ha⁻¹ yr⁻¹ in industrially developed regions of Europe (Binkley *et al.* 2000). In the Netherlands, N deposition in throughfall, overall, exceeded 50 kg N ha⁻¹ yr⁻¹, and even reached 100 kg N ha⁻¹ yr⁻¹ in some regions (Wright and Rasmussen 1998). In eastern North America, the atmospheric N deposition rate has increased 10–20 times (Magill *et al.* 1997).

Recently, East Asia has received attention in terms of N mobilization (e.g. Galloway 2000). Its growing population and rapid economic development have markedly increased Nr emissions in the latest two decades. During 2003–2007, total annual N deposition at 12 monitoring sites throughout Japan ranged from 3.1 to 18.2 kg N ha⁻¹ yr⁻¹ (Japanese Ministry of the Environment 2009). Nitrate deposition increased in the mid-1990s and leveled off thereafter, with some slightly fluctuations in the period from 1991 to 2007. Japan has been receiving long-range transport of N compounds

from eastern Asia, where NO_x emissions have increased by 2.8 times during 1980–2003 (Ohara *et al.* 2007).

According to the projections of Galloway *et al.* (2004), the large increase in NO_x and NH₃ emissions is drastically changing the pattern and magnitude of total inorganic N deposition. Asia is experiencing the largest changes. Relative to the early 1990s, deposition has increased significantly, from >10 kg N ha⁻¹ yr⁻¹ to >20 kg N ha⁻¹ yr⁻¹, and large regions of South and East Asia will receive >50 kg N ha⁻¹ yr⁻¹ by 2050. Under the SRES A2 emissions scenario, worldwide Nr deposition will increase by between 50% and 100% by 2030 relative to 2000, with the largest absolute increases occurring over East and South Asia (Reay *et al.* 2008). The current emissions legislation also suggests that nitrogen oxide (NO) emissions in East Asia will grow by around 50% and N deposition in Japan will increase by approximately 30% (Ichikawa *et al.* 2001).

In forest ecosystems, increased N deposition usually leads to increased NPP and fixation of CO₂ by vegetation (e.g. Kenk and Fischer 1988, Townsend *et al.* 1996, Hungate *et al.* 2003). Increased N availability has probably contributed to the increase in forest growth rates observed across Europe (e.g. Hunter and Schuck 2002). Increased productivity may in turn increase C sequestration in soils due to increased litterfall (e.g. Nadelhoffer *et al.* 1999, De Vries *et al.* 2006). However, N₂O emissions tend to increase when soil C increases due to the positive relationships among soil C content, the nitrogen turnover rate, and N₂O production (Six *et al.* 2004, Li *et al.* 2005). Moreover, increased N deposition can directly increase N₂O emissions by increasing N availability for microbial processes that are directly involved in N₂O production (Bowden *et al.* 1991, Butterbach-Bahl *et al.* 1997, 2002). Increased NH₄⁺ concentrations, however, rely on N deposition and may decrease the oxidation capacity of soils for atmospheric CH₄, thereby decreasing the net influx of CH₄ from the atmosphere to the biosphere (Stuedler *et al.* 1989, Sitaula *et al.* 1995a, van den Pol-van Dasselaar *et al.* 1999). However, inverse effects have also been reported (Bodelier and Laanbroek 2004).

Future increases in N deposition have the potential to change nutrient dynamics in forest ecosystems and can alter soil–atmosphere exchanges of CH₄ and N₂O, which are the third and fourth greatest contributors to the greenhouse effect after water vapor and CO₂ (IPCC 2007). Therefore, to investigate the effects of N deposition on soil CH₄ and N₂O fluxes, these gas fluxes and various soil properties were measured and quantified in an experimental forest following simulated N inputs.

2 Materials and Methods

2.1 Site description and experimental design

The study was conducted in the Sapporo Experimental Forest of Hokkaido University in northern Japan (43°06' N, 141°20' E). The forest had a mean annual temperature of 9.5°C and mean precipitation of 995.0 mm during the measurement period from 2008 to 2009 (Japan Meteorological Agency 2009). Meteorological data showed no

difference between 2008 and 2009 in the mean annual temperature, but annual precipitation in 2009 increased by 1.3 times compared to 2008.

To investigate changes in soil CH₄ and N₂O fluxes following a N application, we established research plots in brown forest soil in the experimental field, which contained 3-year-old seedlings of hybrid larch F₁ (F₁: *Larix gmelinii* var. *japonica* × *Larix kaempferi*); the initial density was 4600 trees ha⁻¹. Larch species have been planted intensively over a large area in northern Asia and are still being planted in many places (Zhang *et al.* 2000). Among these species, Japanese larch (*Larix kaempferi*) is a typical coniferous afforestation species in Hokkaido, northern Japan. Its capacity for rapid growth in cold climates has made it a key afforestation species, and it has been planted on 451,000 ha in the region, approximately 30% of the total plantation (Kajisa *et al.* 2007). However, this species has a high susceptibility to diseases and grazing damage by insects and fungi (Hayashi *et al.* 1998, Koike *et al.* 2003). For this reason, the hybrid larch F₁ was developed to address these problems (Ryu *et al.* 2009), and approximately 700,000 seedlings of this hybrid are planted annually in the region (Kita *et al.* 2009).

In spring 2008, eight 10 × 10-m research plots were randomly installed at the site. Nitrogen input was initiated at the onset of our experiment and included two treatments, a 0 kg N ha⁻¹ yr⁻¹ treatment (Zero-N) and a 50 kg N ha⁻¹ yr⁻¹ treatment (50 kg-N), with four latticed replicates of each treatment. The N was added as an ammonium nitrate (NH₄NO₃) solution and was distributed on four occasions during each growing season, which began in April and ended in November.

During each N application, 50 kg-N addition plots received 3.57g L⁻¹ of the NH₄NO₃ solution, while Zero-N control plots were treated with distilled water. All applications were spread evenly over the canopy and surface soil in each plot using a watering sprayer because the site was planted with 3-year-old seedlings under the height of 1 m. Additional N applications were administered on 25 June, 11 July, 1 August, and 22 August in the first year, and on 18 May, 27 June, 6 August, and 18 September in the second year. Gas

sampling was conducted at least 5–10 days before and after each N input event. In mid-June 2008, before the N application, soil pH values in the 10-cm soil layer (by depth) were 4.17–4.21, organic-C values were 37.97–39.10 g kg⁻¹, and total-N values were 3.05–3.08 g kg⁻¹. Moreover, inorganic-N concentrations at the site were 3.44–4.11 mg kg⁻¹ for ammonium nitrogen (NH₄-N) and 9.37–10.00 mg kg⁻¹ for nitrate nitrogen (NO₃-N). No significant differences in general soil properties were observed between the two treatments before N application ($P > 0.05$) (Table 3.1).

2.2 Soil CH₄ and N₂O fluxes

Soil–atmosphere exchanges of CH₄ and N₂O were determined using the closed-chamber method (Morishita *et al.* 2003, Takakai *et al.* 2008, Kim *et al.* 2011a, 2011b), with gas samples collected on ten occasions per year during the growing seasons in 2008 and 2009. Two sampling points were installed in each plot in both treatments. The chamber, which consisted of aluminum cylinders (inside diameter 20 cm, height 25 cm) that were open at the top and bottom, were inserted into the soil to a depth of 3 cm to make a gastight seal (Kim *et al.* 2011a, 2011b). An acrylic sheet with a compressive rubber seal acted as a lid at the top of each chamber.

After each N application, to minimize the effects of the disturbance caused by inserting the sampler into the soil, the chambers were installed at locations more than 1 day before the measurements were taken. Moreover, every time we measured soil CH₄ and N₂O fluxes, gas samples were always collected in the same place in each plot to reduce the spatial heterogeneity in fluxes as much as possible. Flux measurements were generally taken between midmorning and noon on all dates. The chambers were sealed with the lids for 1 h. At 0, 10, 20, 40 and 60 min after the lids were put in place, 20-mL air samples were taken from the headspace inside the chambers with a 25-mL syringe and transferred to 10-mL vacuum vials that had been evacuated with a butyl rubber stopper (SVF-10, Nichiden-Rika, Kobe, Japan). To determine concentrations of CH₄ and N₂O, 1-mL gas samples were extracted from the sample vials and analyzed using a gas chromatograph (GC-14B,

Table 3.1. Initial soil properties in study site

	Zero-N control	50 kg-N addition	<i>U</i>	<i>P</i>
Soil pH (1:2.5 KCl)	4.21 ± 0.07	4.17 ± 0.09	27.0	0.598
Organic-C (g 100g ⁻¹)	3.91 ± 0.58	3.80 ± 0.61	31.0	0.916
Total-N (g 100g ⁻¹)	0.31 ± 0.04	0.30 ± 0.04	30.5	0.875
C:N ratio	12.40 ± 0.34	12.15 ± 0.37	25.5	0.495
NH ₄ -N (mg kg ⁻¹)	4.11 ± 0.41	3.44 ± 0.26	20.0	0.207
NO ₃ -N (mg kg ⁻¹)	10.00 ± 3.51	9.37 ± 2.62	29.0	0.753

Values indicate the mean ± SE ($n = 8$). Significant differences (P -value) between Zero-N control and 50 kg-N addition plots were evaluated by Mann-Whitney U -test. (Adapted from Kim *et al.* 2012, with kind permission of Atmospheric Environment).

Shimadzu, Kyoto, Japan) equipped with a flame ionization detector and an electron capture detector, respectively (Morishita *et al.* 2003, Takakai *et al.* 2008, Kim *et al.* 2011a). The concentrations were calibrated with standard gases containing 0 and 2.03 ppmv CH₄ and 0, 0.286, 0.630, and 0.955 ppmv N₂O. The minimum detectable concentrations of CH₄ and N₂O were ± 0.1 and ± 0.007 ppmv, respectively. Gas fluxes were calculated as follows:

$$F = \rho \times V/A \times \Delta c/\Delta t \times 273/(273 + T) \times \alpha \quad (1)$$

where F is the gas flux ($\mu\text{g C m}^{-2} \text{h}^{-1}$ for CH₄ and $\mu\text{g N m}^{-2} \text{h}^{-1}$ for N₂O), ρ is the gas density as air at STP (CH₄ = $0.716 \times 10^9 \mu\text{g m}^{-3}$ and N₂O = $0.978 \times 10^9 \mu\text{g m}^{-3}$), V is the volume of the chamber (m^3), A is the cross-sectional area of the chamber (m^2), $\Delta c/\Delta t$ is the rate of change of the gas concentration (c) inside the chamber per unit time (t) during the sampling period ($\text{m}^3 \text{m}^{-3} \text{h}^{-1}$), T is the air temperature ($^{\circ}\text{C}$), and α is a conversion factor to transform CH₄ and N₂O into C and N (CH₄ = 12/16 and N₂O = 28/44). Methane and N₂O fluxes were calculated using linear regression between the gas concentration inside the chamber and the measured time. More than 84% of the CH₄ and 100% of the N₂O flux measurements had a correlation coefficient of determination with an R^2 value over 0.7. Positive values represent net production from the soil and negative values indicate net uptake by the soil.

2.3 Soil variables

During gas sampling, which occurred on ten occasions per year during two growing seasons, soil temperature and soil moisture content at a depth of 10 cm were measured at points adjacent to each chamber. Temperature was measured using a thermistor thermometer (CT-220, Custom, Tokyo, Japan) and gravimetric water content was determined by weighing samples collected with a corer before and after drying at 105 $^{\circ}\text{C}$ for 48 h. Soil samples were collected adjacent to each chamber at three randomly selected points using a core sampler. The three samples collected at soil depths of 0–10 cm were combined to generate a single sample for analysis. The samples were sieved (2 mm mesh), subsampled for 2 M KCl extraction (1:5, KCl), pH analysis (1:2.5, 1 M KCl), gravimetric water content (105 $^{\circ}\text{C}$ for 48 h), and then air-dried.

To determine soil inorganic-N contents, such as NH₄-N and NO₃-N, KCl extracts were filtered and frozen prior to analysis. Concentrations of NH₄-N and NO₃-N in the extracts were measured using an autoanalyzer (AACS-4, BL-TEC Inc., Osaka, Japan). Samples were air-dried and total C and N contents were determined using a NC analyzer (Sumigraph NC-1000, Sumika Chemical Analysis Service Ltd., Osaka, Japan).

2.4 Statistical analysis

All statistical analyses were performed using the software SAS 8.1 (SAS 2001). The distributions of the measured soil GHG fluxes and environmental variables were tested for normality with the Shapiro-Wilk test. The Mann-Whitney U -test, a nonparametric ANOVA,

was used to test for statistically significant differences in all parameters between the treatments. Repeated measures ANOVA was also used to examine changes in the effect of the N treatment; measurement date was included as a repeated factor. Correlation analysis and stepwise linear regression analysis were employed to examine the relationships between soil GHG fluxes and environmental variables.

3 Results

3.1 General conditions and soil properties

Mean air temperature in the region was 9.5 $^{\circ}\text{C}$ during 2008 and 9.4 $^{\circ}\text{C}$ during 2009. For the same periods, annual precipitation reached 843.0 and 1147.0 mm, respectively. The greater amount of precipitation during 2009 was reflected in higher soil moisture conditions compared to 2008. Seasonal patterns in climatic conditions at the site are shown in Fig. 3.1, accompanied by the soil moisture content and soil temperature. Soil moisture content varied from 12.6% to 41.2%, with seasonal variations following the pattern of precipitation, and somewhat dry soil conditions were maintained throughout the measurement periods. Mean soil moisture content was 25.8% for Zero-N control and 25.6% for 50 kg-N addition plots; no significant difference was observed between the two treatments ($P > 0.05$).

Soil temperature ranged from 10.3 $^{\circ}\text{C}$ to 29.1 $^{\circ}\text{C}$ at the site during the two growing seasons, with a clear seasonal pattern of increase during the early growing season, a peak in August, and a decrease from early fall to November. Mean soil temperature was 19.9 $^{\circ}\text{C}$ for Zero-N control and 19.8 $^{\circ}\text{C}$ for 50 kg-N addition plots; soil temperature did not differ significantly between the two treatments.

During the two growing seasons, in the Zero-N control and 50 kg-N addition plots, soil pH was 4.21–4.25, organic-C was 37.05–37.37 g kg⁻¹, and total-N was 3.00–3.02 g kg⁻¹, respectively. Organic-C and total-N remained constant throughout the measurement period while soil pH increased slightly in August (Fig. 3.2), but no significant differences were detected between the two treatments ($P > 0.05$) (Table 3.2). Before the N application, no significant differences in any of the soil variables were observed between the two treatments.

In contrast, NH₄-N and NO₃-N concentrations increased immediately after N inputs in the 50 kg-N addition plots (Fig. 3.3). Mean NH₄-N was 3.78 mg kg⁻¹ in the Zero-N control and 4.80 mg kg⁻¹ in the 50 kg-N addition plots during the two growing seasons. Mean NH₄-N in the 50 kg-N addition plots increased by 27% compared to the control as a result of the N application ($P = 0.007$) (Table 3.2). The N inputs also increased the NO₃-N concentrations by an average of 50%. Mean NO₃-N in the Zero-N control and 50 kg-N addition plots were 8.14 and 12.21 mg kg⁻¹, respectively. Although the mean values were not significantly different between the two treatments, repeated measures ANOVA found a significant interaction between treatment and time ($P = 0.004$) (Table 3.2).

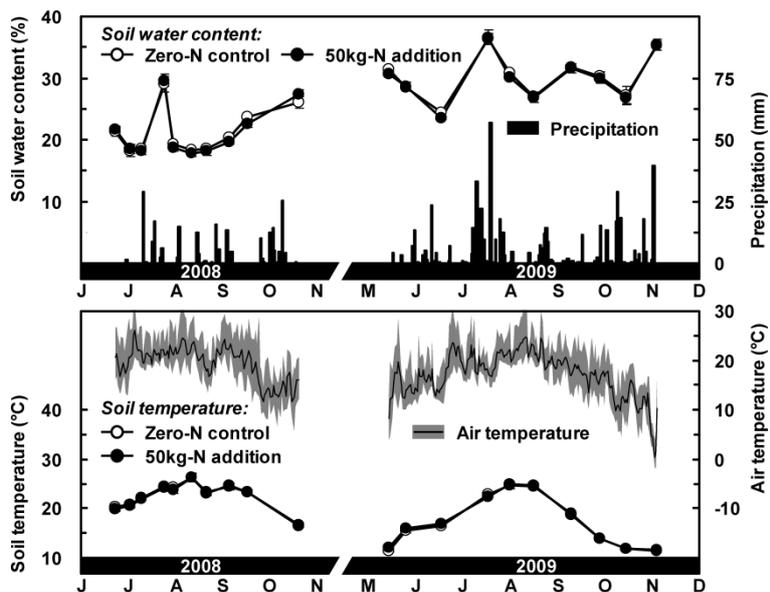


Fig. 3.1. Seasonal dynamics in air temperature, soil temperature, soil moisture content and precipitation during two growing seasons. Error bars indicate 1 SE ($n = 8$) of the mean. (Reprinted from Kim *et al.* 2012, with kind permission of Atmospheric Environment)

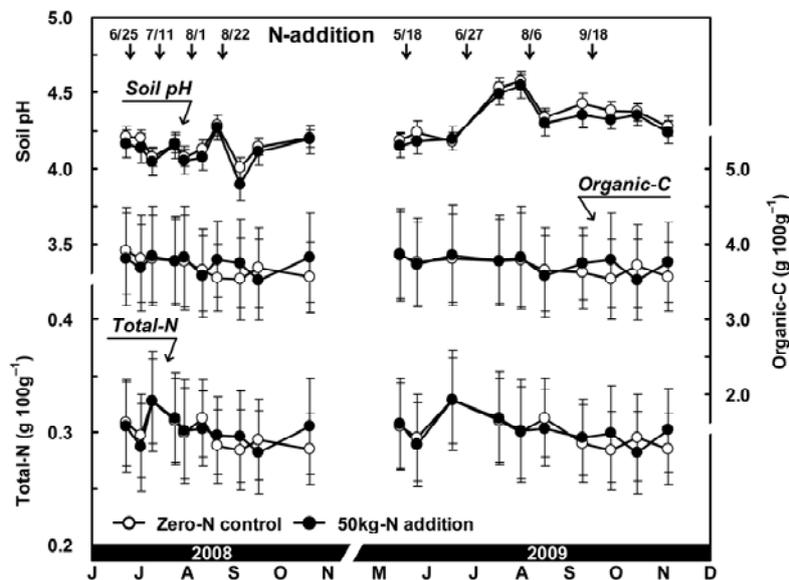


Fig. 3.2. Seasonal dynamics of soil pH, organic-C and total-N in a brown forest nursery soil following N inputs during two growing seasons. Error bars indicate 1 SE ($n = 8$) of the mean. (Reprinted from Kim *et al.* 2012, with kind permission of Atmospheric Environment)

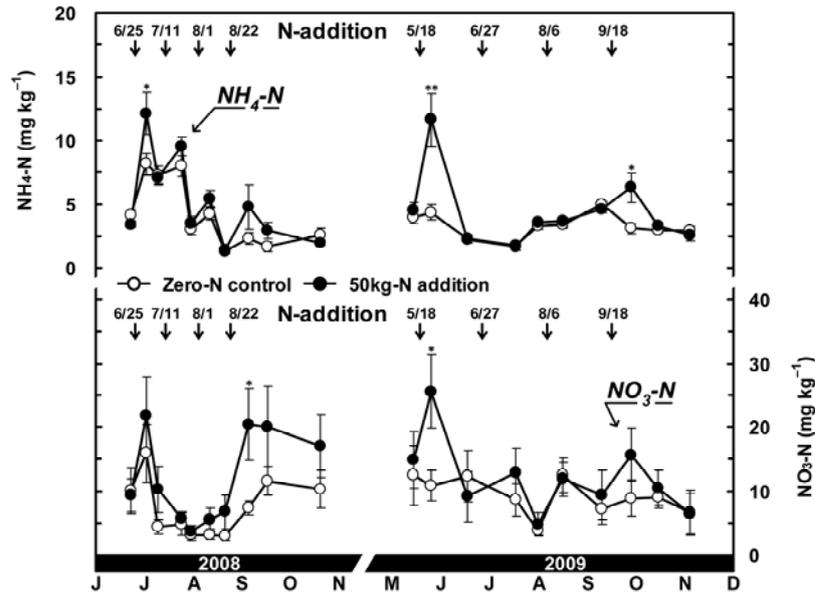


Fig. 3.3. Seasonal dynamics of $\text{NH}_4\text{-N}$ and $\text{NO}_3\text{-N}$ in a brown forest nursery soil following N inputs during two growing seasons. Error bars indicate 1 SE ($n = 8$) of the mean. Values with asterisks represent significant differences between the control and N addition plots (Mann-Whitney U -test, * $P < 0.05$ and ** $P < 0.01$). (Reprinted from Kim *et al.* 2012, with kind permission of Atmospheric Environment)

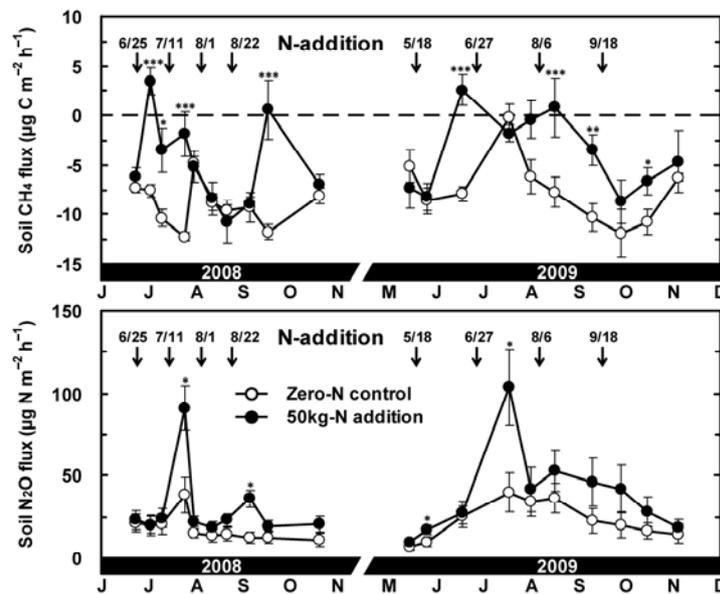


Fig. 3.4. Seasonal dynamics of CH_4 and N_2O fluxes in a brown forest nursery soil following N inputs during two growing seasons. Error bars indicate 1 SE ($n = 8$) of the mean. Values with asterisks represent significant differences between the control and N addition plots (Mann-Whitney U -test, * $P < 0.05$, ** $P < 0.01$ and *** $P < 0.001$). (Reprinted from Kim *et al.* 2012, with kind permission of Atmospheric Environment)

3.2 Soil CH₄ and N₂O fluxes

Throughout the measurement period, Zero-N control soils absorbed atmospheric CH₄, except in July 2009, whereas 50 kg-N addition soils both absorbed and emitted CH₄ (Fig. 3.4). Mean CH₄ consumption rates in 50 kg-N addition plots generally tended to be less than in Zero-N control plots, except in August 2008 and July 2009. We observed that mean CH₄ uptake into the soil was 8.25 $\mu\text{g C m}^{-2} \text{h}^{-1}$ for Zero-N control and 4.26 $\mu\text{g C m}^{-2} \text{h}^{-1}$ for 50 kg-N addition plots during the two growing seasons, and found that soil CH₄ uptake in the 50 kg-N addition plot decreased by 48% compared to the control. In particular, reduced CH₄ uptake or CH₄ emissions were observed with 50 kg-N addition soils immediately after N input events. Repeated measures ANOVA found significant effects of N, time, and the treatment \times time interaction (Table 3.2).

The mean N₂O emission rate was 20.26 $\mu\text{g N m}^{-2} \text{h}^{-1}$ for Zero-N control and 34.41 $\mu\text{g N m}^{-2} \text{h}^{-1}$ for 50 kg-N addition plots. The N inputs increased N₂O emissions by an average of 69% during the two growing seasons. Soil N₂O emission rates in 50 kg-N addition plots were one to three times greater than in Zero-N control plots throughout the measurement period. Soil N₂O emissions were observed at all sites, with peak emissions occurring during the summer (Fig. 3.4). Soil N₂O emissions generally did not exceed 50 $\mu\text{g N m}^{-2} \text{h}^{-1}$ in the Zero-N control plots. In the control treatment, minor peaks in N₂O emissions were observed in mid-July in 2008 and 2009. Both of these peaks were characterized by substantial amounts of rainfall during the preceding weeks, with 34.0 mm falling in 2007 and 66.5 mm falling in 2009. In contrast, soil N₂O emissions in the 50 kg-N addition plots appeared to increase occasionally in response to N inputs with precipitation events, e.g. in July 2008 and July 2009.

Peak N₂O emissions from the 50 kg-N addition plots were measured in early September 2008 and found to be linked to the N treatment, not to specific precipitation events. Although the mean value was not significantly different between the two treatments, repeated measures ANOVA found a significant interaction between treatment and time ($P < 0.001$) (Table 3.2).

4 Discussion

4.1 Effects of N deposition on soil CH₄ and N₂O fluxes

Atmospheric N deposition can increase the abundance of NH₄⁺-oxidizing bacteria, which significantly inhibit methanotrophic activities in soils, especially in surface soil layers (King and Schnell 1994, Whalen and Reeburgh 2000). Three mechanisms have been posited for the partial inhibition of CH₄ uptake by well drained soils in response to increased N inputs: competitive inhibition of CH₄ monooxygenase (MMO), toxic inhibition by hydroxylamine (NH₂OH) and nitrite (NO₂⁻) produced via NH₄⁺ oxidation, and osmotic stress due to high concentrations of NO₃⁻ and/or NH₄⁺ (Schnell and King 1994, Bradford *et al.* 2001, Bodelier and Laanbroek 2004, Reay and Nedwell 2004).

Although inverse effects have also been reported, most studies, including ours, have reported an inhibition of atmospheric CH₄ oxidation in N-fertilized forest soils (Castro *et al.* 1995, Sitaula *et al.* 1995a, Steinkamp *et al.* 2001, Gullledge *et al.* 2004, Chan *et al.* 2005, Maljanen *et al.* 2006, Zhang *et al.* 2008b, Basiliko *et al.* 2009). According to a literature review with meta-analysis, Liu and Greaver (2009) reported that soil CH₄ uptake across all ecosystems was significantly reduced by an average of 38% under N application, ranging from 10 to 560 kg N ha⁻¹ yr⁻¹.

Table 3.2. Repeated measures analysis of variance of soil GHG fluxes and environmental variables as affected by N inputs in a brown forest nursery soil during two growing seasons

Source	df	Soil moisture content			Soil temperature			Soil pH		
		MS	F-value	P > F	MS	F-value	P > F	MS	F-value	P > F
<i>Between subjects</i>										
N treatment	1	4.20	0.62	0.45	0.02	0.00	0.96	0.12	0.15	0.705
<i>Within subjects</i>										
Date	19	558.96	152.59	<0.001	384.77	555.35	<0.001	0.37	117.05	<0.001
Date \times N treatment	19	1.28	0.35	0.995	0.39	0.57	0.926	0.00	1.01	0.449
Source	df	Organic-C			Total-N			NH ₄ -N		
		MS	F-value	P > F	MS	F-value	P > F	MS	F-value	P > F
<i>Between subjects</i>										
N treatment	1	8.15	0.00	0.97	0.01	0.00	0.98	83.63	10.06	0.007
<i>Within subjects</i>										
Date	19	12.04	0.50	0.96	0.23	2.18	0.004	93.50	25.88	<0.001
Date \times N treatment	19	7.86	0.32	1.00	0.04	0.35	0.995	15.08	4.17	<0.001
Source	df	NO ₃ -N			Soil CH ₄ flux			Soil N ₂ O flux		
		MS	F-value	P > F	MS	F-value	P > F	MS	F-value	P > F
<i>Between subjects</i>										
N treatment	1	1328.04	1.64	0.222	1269.38	28.22	<0.001	15778.15	2.66	0.126
<i>Within subjects</i>										
Date	19	346.58	7.75	<0.001	115.21	6.03	<0.001	4370.31	17.88	<0.001
Date \times N treatment	19	95.77	2.14	0.004	88.33	4.62	<0.001	1130.84	4.63	<0.001

(Adapted from Kim *et al.* 2012, with kind permission of Atmospheric Environment)

Furthermore, Gulledge *et al.* (2004) observed that fertilization decreased *in situ* CH₄ consumption in pine and hardwood sites in the Harvard Forest by 86% and 49% relative to a control plot, respectively.

Sitaula *et al.* (1995a) found that N fertilization with NH₄NO₃ significantly reduced atmospheric CH₄ uptake in a 100-year-old Scots pine forest in Norway. They reported that 30 kg N ha⁻¹ yr⁻¹ resulted in a CH₄ uptake rate that was 85% of the control, while the uptake rate with 90 kg N ha⁻¹ yr⁻¹ was 62% of the control. Castro *et al.* (1995) also found that N-fertilized soils, which received 50 and 150 kg N ha⁻¹ yr⁻¹, had annual average CH₄ consumption rates that were 15% and 64% lower than annually averaged (over 6 years) CH₄ consumption by control soils located in the Harvard Forest, MA, USA.

Moreover, Chan *et al.* (2005) found that *in situ* CH₄ consumption rates were 0.19 for a control and 0.12 mg C m⁻² h⁻¹ for a N treatment, indicating that consumption had been reduced by 35% after 8 years of N applications in a productive temperate deciduous forest in northwestern Pennsylvania. To investigate the CH₄ uptake response to simulated N deposition, Zhang *et al.* (2008b) created four N treatment levels in the Dinghushan Biosphere Reserve (DHSBR) in southern China and observed that average CH₄ uptake rates in a mature forest decreased by 6%, 14% and 32% when compared to the control for 50, 100 and 150 kg N ha⁻¹ yr⁻¹ plots, respectively. They suggested that the response in soil CH₄ uptake to N deposition at the site could vary directly depending on the soil N status.

In contrast, Basiliko *et al.* (2009) measured soil CH₄ uptake in fertilized western Canadian plantation forests. On average, the control and N-treatment plots oxidized 30 and 26 μg CH₄ m⁻² h⁻¹, respectively, at the Douglas-fir site, but repeated measures ANOVA found no significant effects of fertilizer, time, or the treatment × time interaction.

The positive relationship between N deposition and N₂O emissions from forest soils has mainly been attributed to increased soil N availability for the microbial processes of nitrification and denitrification (Matson *et al.* 2002, Venterea *et al.* 2003). Various studies have reported that N deposition is a key site parameter influencing the soil–atmosphere exchange of N₂O, and can increase soil N₂O emissions in forest ecosystems, including our study site, although inverse effects have also been reported (Butterbach-Bahl *et al.* 1997, Magill *et al.* 1997, Nadelhoffer *et al.* 1999, Sitaula *et al.* 1995b, Maljanen *et al.* 2006, Ullah and Zinati 2006, Zhang *et al.* 2008a). These studies have shown that temperate forests can function as significant sources for N₂O, especially if these forests are affected over decades by high rates of atmospheric N deposition. Nitrogen addition, ranging from 10 to 562 kg N ha⁻¹ yr⁻¹, significantly increased soil N₂O emission by an average of 216% across all ecosystems according to a literature review that involved a meta-analysis (Liu and Greaver 2009).

Nadelhoffer *et al.* (1999) found that soil N₂O emissions exhibited positively linear relationships with N input rates in nine temperate forests in Europe. The

results of a simulated N deposition experiment in DHSBR, southern China, indicated that N additions significantly increased soil N₂O fluxes in a mature forest and that the increases were variable following four N input levels (Zhang *et al.* 2008a). Sitaula *et al.* (1995b) found that N fertilization with NH₄NO₃ increased N₂O emissions by a factor of 1.2–6 with an application of 30 kg N ha⁻¹ yr⁻¹ (Medium-N) and 4–7 with additions of 90 kg N ha⁻¹ yr⁻¹ (High-N) in a 100-year-old Scots pine forest in Norway. Also, they observed that the average soil N₂O concentration in fertilized soils was 1.1–2.2 times greater in Medium-N and 2–7 times greater in High-N treatment plots compared to a control.

Magill *et al.* (1997) found that the average rate of N₂O emissions from a high-N plot in a pine stand, 9.43 μg N m⁻² h⁻¹, was significantly greater than the average rate from a control plot, 4.17 μg N m⁻² h⁻¹, and was twice the maximum summer flux rate observed in these plots in the Harvard Forest during the first 2 years of their study (Bowden *et al.* 1991). Butterbach-Bahl *et al.* (1997) showed that 1.3–5.0% of N deposition was translated into N₂O in regions with high atmospheric N deposition in Germany. In addition to atmospheric N deposition, many commercial forests receive applications of fertilizer with N in the form of urea or NH₄NO₃. Nitrous oxide emission rates from forest soils could potentially be enhanced several-fold by such fertilization.

In contrast, Matson *et al.* (1992) measured soil N₂O fluxes in a Douglas-fir forest in northeastern New Mexico. Despite the increase in soil N availability because of the N application, they considered that global N deposition into forests was not currently contributing significantly to increasing atmospheric N₂O concentrations.

4.2 Temporal and spatial correlations between soil GHG fluxes and soil variables

The brown forest nursery soil in this study acted as a relatively small sink of CH₄. Soil CH₄ fluxes in both treatments showed no clear seasonal variation (Fig. 3.4). During the two growing seasons, we could find no temporal correlations between soil CH₄ uptake and environmental variables. In Zero-N control plots, soil moisture content and organic-C and NH₄-N concentration were the dominant factors that controlled soil CH₄ consumption in our experiment, according to a stepwise linear regression analysis (Table 3.3).

However, these factors could only explain 27% of the temporal variability in soil CH₄ uptake ($P < 0.05$). Moreover, we observed very high spatial variability in soil CH₄ flux within the research plots, with an average coefficient of variation (CV) of 114% in Zero-N control and 283% in 50 kg-N addition plots. These values were obviously larger than those for soil N₂O flux: 83% in Zero-N control and 65% in 50 kg-N addition plots. This suggests that we need to increase the number of spatial replicates for field measurements to accurately estimate soil CH₄ flux and to evaluate the effect of N deposition more accurately.

Table 3.3. Temporal correlation between soil GHG fluxes and soil variables in a brown forest nursery soil during two growing seasons

Soil GHG flux	Treatment	Pearson correlation coefficient						
		SMC	ST	pH	OC	TN	NH ₄ -N	NO ₃ -N
CH ₄ flux	Zero-N	0.30	0.02	0.29	0.35	0.18	-0.27	0.07
	50 kg-N	0.08	0.23	0.20	-0.14	0.13	0.11	0.03
N ₂ O flux	Zero-N	0.34	0.43*	0.53**	0.25	0.41	0.19	-0.07
	50 kg-N	0.48*	0.33	0.43	0.05	0.19	-0.01	-0.19

Soil GHG flux	Treatment	Stepwise linear regression	
		Equation	Adj. R ²
CH ₄ flux	Zero-N	y = 0.16 SMC + 13.66 OC - 0.64 NH ₄ -N - 59.64	
	50 kg-N	-	
N ₂ O flux	Zero-N	y = 0.83 SMC + 1.21 ST + 21.79 pH + 268.45 TN - 198.15	
	50 kg-N	y = 3.32 SMC + 3.54 ST + 281.33 TN - 205.72	

SMC, ST, OC and TN indicate soil moisture content, soil temperature, organic-C and total-N, respectively. Asterisks with values represent the level of significance of correlations and regression models (* $P < 0.05$, ** $P < 0.01$ and *** $P < 0.001$). (Adapted from Kim *et al.* 2012, with kind permission of Atmospheric Environment)

Generally, soil moisture content is the major factor controlling CH₄ production and oxidation (Stuedler *et al.* 1989, Koschorreck and Conrad 1993, van den Pol-van Dasselaar *et al.* 1998, Stein and Hettiaratchi 2001, Dijkstra *et al.* 2010, Kim *et al.* 2011b). In addition to the water content of the soil, factors such as pH, soil temperature, and inorganic N concentrations can be crucial in determining whether a particular soil will act as a sink for CH₄ (Hütsch *et al.* 1994). In incubation experiments with soil from mixed hardwood stands, Bowden *et al.* (1998) found that soil CH₄ uptake was strongly related to soil moisture content in the forest floor. Maximum uptake occurred at approximately 70% of field capacity and lower values were observed as soil moisture content approached 100% of capacity. Also, Maljanen *et al.* (2003) measured CH₄ flux in birch forest soil and found that CH₄ uptake was positively correlated with soil temperature.

In our study, however, soil CH₄ flux was not correlated with soil moisture content, which ranged from 12.60% to 41.21%, or soil temperature, which ranged from 10.30°C to 29.10°C, in either treatment during the measurement period. A lack of a significant relationship between CH₄ flux and water content has been commonly reported (Sjögersten and Wookey 2002, Maljanen *et al.* 2003). Plausible explanations are that maximum rates of CH₄ oxidation occur at soil depths of 10–20 cm (Whalen *et al.* 1992), where soil moisture content was measured, and that methanotrophs are insensitive to these levels of soil moisture content. Furthermore, Castro *et al.* (1995) observed that temperature was a strong controller of soil CH₄ consumption at low temperatures, but that CH₄ consumption became independent of soil temperature between 10°C and 20°C.

Increased atmospheric N deposition increases NH₄⁺ in the soil and usually decreases CH₄ uptake by well drained soils (Stuedler *et al.* 1989, Gullledge and

Schimel 1998, van den Pol-van Dasselaar *et al.* 1999, Le Mer and Roger 2001), although inverse effects have also been reported (Borken *et al.* 2002, Basiliko *et al.* 2009). Whalen and Reeburgh (2000) concluded that N inputs did not influence CH₄ uptake until they significantly increased soil N availability in boreal forest soils. During two growing seasons in this study, mean NH₄-N concentrations in the 50 kg-N addition plots increased by 27% compared to the control as a result of the N application ($P = 0.007$) (Table 3.2). With the change in NH₄-N between the two treatments, we found a negative spatial relationship between soil CH₄ uptake and the NH₄-N concentration at the site ($R^2 = 0.85$, $P < 0.001$) (Fig. 3.5). This result indicates that N deposition can inhibit soil CH₄ uptake through the three mechanisms mentioned above.

Soil N₂O fluxes did not show clear seasonal variations in any of the treatment plots. Two major peaks were seen in soil N₂O emissions in the two growing seasons, in July 2008 and July 2009 (Fig. 3.4). During the two growing seasons, we observed a positive temporal correlation between soil N₂O emissions and soil moisture content ($P < 0.05$), soil temperature ($P < 0.05$), or soil pH ($P < 0.01$) (Table 3.3). Among all of the soil variables, soil moisture content, soil temperature, soil pH, and total-N were the dominant factors that controlled soil N₂O emissions in our experiment, according to a stepwise linear regression analysis, and they explained 59–71% of the temporal variability ($P < 0.001$).

Generally, in forest ecosystems, soil moisture content, soil temperature, and N availability are the key factors regulating N₂O emissions (Papen and Butterbach-Bahl 1999, Butterbach-Bahl *et al.* 2002, Schindlbacher *et al.* 2004). Increases in soil moisture content, temperature, or inorganic N availability usually stimulate soil microbial processes and consequently N₂O production. Although soil moisture content has been shown to be the dominant factor controlling soil N₂O emission rates

in many terrestrial ecosystems, Henrich and Haselwandter (1997) observed no relationship between soil N₂O flux and soil temperature or soil moisture in an 80-year-old Norway spruce forest in Austria.

Also, soil temperature is known to be an important controller of microbial processes. Changes in soil temperature, coupled with the available substrate, have probably favored increased N₂O production. Although soil N₂O emissions are not always directly related to changes in temperature, temperature is considered an important factor in conjunction with other environmental variables (Yates *et al.* 2006). Soil pH can also affect the soil N₂O flux and an increase in soil pH can often increase the nitrification rate in forest ecosystems (Tietema *et al.* 1992, Smolander *et al.* 1998). Furthermore, a weak positive correlation between soil N₂O flux and soil moisture content, and

the correlation between two N₂O emission peaks and increases in soil moisture content, suggest that precipitation was a significant variable controlling soil N₂O emissions and that denitrification might have been the main source of N₂O production at our site. However, establishing a strong predictive relationship between soil N₂O flux and a single parameter, such as soil temperature, soil moisture content, and soil inorganic-N concentrations is very difficult in this field study, perhaps because the process of soil N₂O production is very complex and influenced by many factors (Groffman *et al.* 2000).

The lack of dependence between soil N₂O flux and soil temperature, and the poor relationships with soil moisture content in both treatments, indicate that emissions were probably dependent on the available substrate, instead of abiotic factors. Thus, soil N₂O emissions depended primarily on the availability of inorganic-N in the soil because soil N₂O production is mainly dominated by microbial nitrification and denitrification. Nitrous oxide production correlates with nitrogen availability in the soil because nitrification and denitrification depend on NH₄⁺ and NO₃⁻, respectively (Bouwman 1990).

In this study, we found a positive spatial correlation between soil N₂O emissions and inorganic-N concentrations. In particular, a strong positive spatial relationship was observed between soil N₂O emissions and NO₃-N concentrations ($R^2 = 0.94$, $P < 0.001$) (Fig. 3.5). Soil NO₃⁻ concentrations represent an index of N availability because nitrification supplies and denitrification uses NO₃⁻ in the soil. Therefore, this result suggests that N deposition increases soil nitrogen availability and can thereby favor soil N₂O production, at least over a short term.

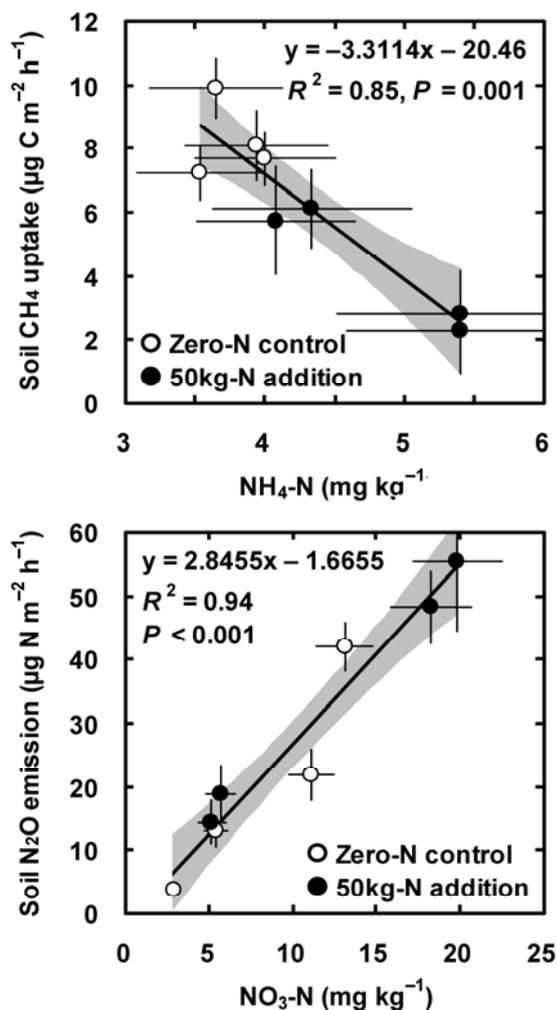


Fig. 3.5. Spatial relationship between soil GHG fluxes and inorganic N concentrations in a brown forest nursery soil. Values indicate the mean \pm SE ($n = 40$), which was measured in each research plot during two growing seasons. The solid line represents a linear regression between the subjects, with gray shaded 95% confidence limits. (Reprinted from Kim *et al.* 2012, with kind permission of Atmospheric Environment)

5 Conclusions

Simulated N additions, which were conducted to evaluate the effects of elevated atmospheric N deposition on soil GHG fluxes, inhibited soil CH₄ uptake and stimulated soil N₂O emissions in response to increased inorganic-N status in a brown forest soil in a hybrid larch F₁ plantation (Fig. 3.6). During two growing seasons, we found a negative relationship between soil CH₄ uptake and NH₄-N concentrations, and a positive relationship between soil N₂O emissions and NO₃-N concentrations. These results showed that NH₄-N and NO₃-N concentrations were the environmental factors that most influenced or controlled the spatial variation in soil CH₄ and N₂O fluxes, respectively. Overall, as N deposition-induced changes in inorganic-N are slow in the present stage, the additional impact on soil CH₄ consumption probably remains small. However, if the increase in N deposition continues, we may not be able to expect well drained forest soils to act as sinks for CH₄. Also, soil N₂O emissions seem to be largely dependent on whether the ecosystem is N-limited at the time of the N inputs. N cycling in forest ecosystems, which already exhibit large N₂O emissions, responds strongly to added N, whereas an ecosystem that has been limited by N rapidly uses up the added N and soil N₂O

emissions are only elevated for a short period of time. However, continuous elevated-atmospheric N deposition is expected to stimulate soil N₂O emissions from forest soils, which are identified as a small N₂O source. Finally, the uncertainties about the effects of other environmental changes and their interactions on soil GHG fluxes, and the coupling between N and C cycles, underlines the need for further study. Large uncertainties remain about the *in situ* effects of various environmental changes on soil GHG fluxes in different forest ecosystems.

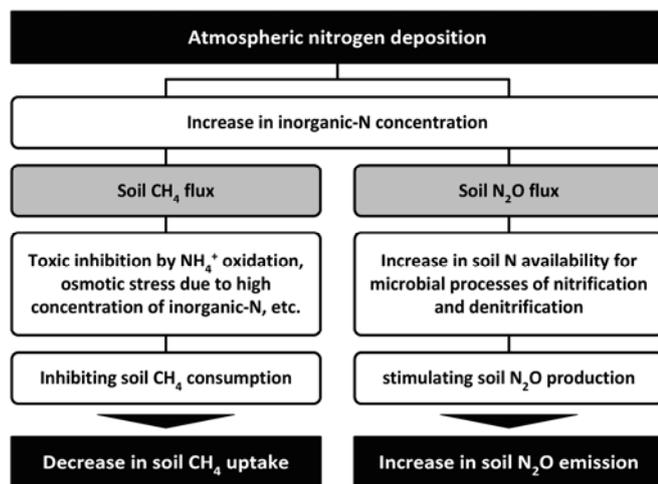


Fig. 3.6. Overall effects of the atmospheric N deposition on soil CH₄ and N₂O fluxes.

CHAPTER 4

SOIL GHG FLUXES AFTER A PRESCRIBED FIRE IN WHITE BIRCH STANDS

4.1 Introduction

Forest ecosystems are generally, an important natural sink for C sequestration that could mitigate climate change (Lal 2005). The production and consumption of these GHGs (mainly CO₂, CH₄ and N₂O) are mediated biologically (Conrad 1996), but are also strongly affected by disturbances due to fire, drought, wind damage, nitrogen deposition, logging and other environmental variables relating to climate change (Kim and Tanaka 2003, Ambus and Robertson 2006, Sullivan *et al.* 2008, Yashiro *et al.* 2008, Goldberg and Gebauer 2009). Such disturbances are becoming increasingly serious and relate to major shifts in the global climate.

Fires, unlike other disturbances, are critical natural or man-made disturbances that directly affect forest carbon cycling by the immediate combustion of biomass. Fires are a significant driver of forest structure and composition in most forest ecosystems (Auclair 1985, Fisher and Binkley 2000). The effects of fire depend mainly on the peak temperature and duration of the fire (Neary *et al.* 1999, Dale *et al.* 2001, Certini 2005). Environmental change in burned areas is liable to affect the exchange of GHGs between the soil and the atmosphere.

During most forest fires, charcoal is created by the partial combustion of organic matter. Charcoal may influence physical, chemical, and biotic processes in soil. In particular, since charcoal influences the transformation and retention of C and N in soils (Deluca *et al.* 2006, Forbes *et al.* 2006), it could influence soil GHG fluxes, directly or indirectly, through its high adsorbing capacity and other actions. Charcoal is also a highly stable form of C and has the potential to act as an effective C sink.

Recently, charcoal has been observed to mitigate GHG emissions; several studies reported that charcoal reduces CH₄ and N₂O emissions from soils following the application of biochar in both field and laboratory incubations (Rondon *et al.* 2005, Rondon *et al.* 2006, Yanai *et al.* 2007, Spokas and Reicosky 2009, Van Zwieten *et al.* 2009). There is only very limited understanding of the processes through which charcoal influences the exchange of GHGs between soil and the atmosphere, and further research is needed.

To determine the effects of forest fires on soil GHG fluxes *in situ*, through the presence of charcoal, soil GHG fluxes (CO₂, CH₄ and N₂O) were measured in a white birch forest in northern Japan, following experimental forest burning and removal of charcoal after the fire. We discuss the role of charcoal on GHGs dynamics in surface soil.

4.2 Materials and Methods

4.2.1 Site description

This study was conducted in the Teshio Experimental Forest (TEF) of Hokkaido University, in the northernmost part of Japan (44°57' N, 142°01' E). This study area is set in a mixed coniferous-broadleaved forest in cool temperate climate. The mean annual precipitation and temperature were around 1100 mm and 5–6°C (Takagi *et al.* 2009). Snowmelt in this region is completed by late April, and snow covers the ground in late November or early December. The dominant overstory species is white birch (*Betula platyphylla* var. *japonica*), mixed with some Japanese larch (*Larix kaempferi*), ash (*Fraxinus mandshurica* var. *japonica*) and elm (*Ulmus laciniata*); the stand constituents are set out in Table 4.1. In all four sites, the forest floor is densely covered by understory dwarf bamboo (*Sasa senanensis*), almost 100% coverage, and the aboveground biomass of bamboo was not significantly different among treatment plots (Table 4.2).

This same stand composition covers large areas of the TEF. White birch, which is broadly distributed across the Eurasian continent, is a typical light-demanding early successional tree species, and usually forms a pure stand after forest fires (Koike 1995). Its rapid capability for recovery is useful for storing CO₂ and for moderating the emission of CO₂ and other GHGs after fires. As a pioneer, white birch often grows in infertile habitats, such as areas with volcanogenous regosols which are distributed widely in northern Japan (Kitao *et al.* 2007).

Four 20 m × 30 m research sites were established in white birch forests in the TEF. The four sites were

Table 4.1. Stand structure of each experimental site in study forest

	Site1	Site2	Site3	Site4
Density (stem ha ⁻¹)	900	1060	1380	1100
<i>Betula platyphylla</i> var. <i>japonica</i>	540	980	960	840
<i>Flaxinus mandshurica</i>	320	60	180	80
<i>Ulmus laciniata</i>	20	20	220	160
<i>Larix kaempferi</i>	20	0	20	20
Mean DBH (cm)	14.4	13.1	13.2	12.1
Mean height (m)	12.4	12.9	15.9	11.3
Basal area (m ² ha ⁻¹)	23.3	19.8	29	20.5

(Adapted from Kim *et al.* 2011a, with kind permission of European Journal of Forest Research)

Table 4.2. Aboveground biomass of understory vegetation and soil profiles in three treatments before and after the experimental burning

	CON		BURN		BURN-CHA	
	Before	After	Before	After	Before	After
Understory (kg m ⁻²)	1.3 ± 0.1	1.3 ± 0.2	1.3 ± 0.2	–	1.3 ± 0.2	–
Litter layer (cm)	5.5 ± 0.5	5.4 ± 0.3	6.2 ± 0.4	0.1 ± 0.2	5.2 ± 0.2	0.3 ± 0.2
Charcoal layer (cm)	–	–	–	3.0 ± 0.2	–	0.1 ± 0.1
F-H layer (cm)	2.7 ± 0.5	2.6 ± 0.2	3.1 ± 0.2	2.6 ± 0.4	3.2 ± 0.3	3.5 ± 0.4
A layer (cm)	5.6 ± 1.0	5.0 ± 0.1	6.4 ± 0.7	4.5 ± 0.6	6.9 ± 0.4	5.3 ± 0.6

Data are given as the mean ± SE. CON, BURN and BURN-CHA indicate a control plot, a burned plot and a plot that had been burned and the charcoal removed, respectively. Before and after represent that the values of 1 week before and after the experimental burning. Understory in this forest is almost completely covered by dwarf bamboo. (Adapted from Kim *et al.* 2011a, with kind permission of European Journal of Forest Research)

distributed in three different small watersheds and separated from each other by at least 50 m to 300 m. To reduce interference due to topographical variation, the elevations of the sites were similar (about 20 m a.s.l.), with flat topography. All of the sites were set in well-drained rusty brown forest soils, classified as Dystric Cambisols according to the FAO-UNESCO system, which had developed from tertiary sedimentary rock.

In early July 2007, before the experimental burning, the soil pH in the soil layer of depth 5–10 cm was 5.5 ± 0.0 (mean ± SE), organic-C was 37.4 ± 1.6 g kg⁻¹, and the total-N was 3.4 ± 0.2 g kg⁻¹. Before the fire, the soil GHG fluxes were 0.13 ± 0.00 g C m⁻² h⁻¹ for CO₂, -31.44 ± 2.05 µg C m⁻² h⁻¹ for CH₄, and 2.21 ± 0.24 µg N m⁻² h⁻¹ for N₂O. There were no significant differences in general soil properties or soil GHG fluxes between the four research sites ($P > 0.05$).

4.2.2 Experimental design

In mid-July 2007, experimental burning was conducted in the white birch forests of the TEF at the

end of the fire season in the Hokkaido region. Three treatment-plots were installed within each site: a control plot (CON), a burned plot (BURN), and a plot that had been burned and the charcoal removed (BURN-CHA). In the Hokkaido region, forest fires are usually surface fires, which are of low intensity and burn mainly the dominant dwarf bamboo in the understory and part of the canopy stems (Masaka *et al.* 2000). The square comprising each plot was limited to 5 m × 5 m in order to demonstrate the low intensity surface fire by controlling the amount of fuel.

The soil temperature during burning was monitored by thermocouples in the surface of the litter layer and at 0 cm depth of soil layer (i.e. the boundary between humus layer and mineral soil layer). Four thermocouples at each depth were set about 50 cm in from the edge opposite the fire ignition. The maximum temperature at the forest floor was about 500–530°C, and the duration above 400°C was 2–4 minutes, but the temperature at the surface soil increased by only 3–5°C during the burning. Two days after the burning respectively for each site, charred materials in

BURN-CHA were carefully removed by hand sorting. Small particles that were inevitably remained on the surface floor were not easily visible. Therefore, the effects of charcoal removal observed in this study were, to be exact, those of the removal of visible charcoal.

4.2.3 Soil GHG fluxes

Greenhouse gas fluxes from the soil at each plot were measured on four or five occasions per year, using a closed-chamber method, over almost two entire growing seasons, from July 2007 to November 2008. To facilitate transport of the chambers to the field, the chambers were constructed such that six chambers of differing sizes could fit concentrically within each other (Morishita *et al.* 2003, Takakai *et al.* 2008). The heights of each chamber are the same at 25 cm, but the diameters vary from 18.5 cm to 21.0 cm at regular intervals of 0.5 cm. All understory plants within the chamber were clipped close to ground level and were removed before the chamber was installed, because the forest floor was almost completely covered by dwarf bamboo with its subterranean stem. The chambers were installed to a depth of 3 cm below the soil surface in each plot. After the experimental burning, the chambers were set up again at least one day before the second measurement on August 2007 so as to limit the disturbance caused by their installation. Two chosen sampling points for each treatment were installed in the four study sites, giving a total of twenty-four measurement points. The fluxes were generally measured between mid-morning and noon.

For CO₂ analysis, a 250-mL gas sample was collected in a 500-mL sampling bag (Tedlar bag, OMI Odorair Service, Shiga, Japan) from the headspace of the chamber, using a 25-mL polypropylene syringe at 0 and 6 minutes after the chambers were closed (Toma and Hatano 2007, Takakai *et al.* 2008). Each chamber remained open for at least 10 minutes after the CO₂ flux measurement was complete, to allow complete exchange of air inside the chamber; the N₂O and CH₄ gas samples were then collected. For the CH₄ and N₂O analyses, a 20-mL gas sample was withdrawn at 0, 10, 20, 40 and 60 minutes after the top of the chamber was resealed. These samples were placed in 10-mL vacuum vials sealed with a butyl rubber stopper (SVF-10, Nichiden-Rika, Kobe, Japan) (Morishita *et al.* 2003, Takakai *et al.* 2008).

The CO₂ concentration was measured immediately after sampling, using an infrared CO₂ analyzer (ZFP9, Fuji Electronic, Tokyo, Japan). For the CH₄ and N₂O concentrations, a 1-mL gas sample was taken from the sampling vial and analyzed using a gas chromatograph (GC-8A and GC-14B, Shimadzu, Kyoto, Japan) equipped with a flame ionization detector and an electron capture detector, respectively (Takakai *et al.* 2008). The concentrations were calibrated using standard gases containing 0 and 1810 pmv CO₂ or 2.03 ppmv CH₄ or 0.286, 0.630 or 0.955 ppmv N₂O. The minimum detectable concentrations of CO₂, CH₄ and N₂O were ± 1.0, ± 0.1 and ± 0.007 ppmv, respectively. Gas fluxes were calculated from the formula:

$$F = \rho \times V/A \times \Delta c/\Delta t \times 273/(273 + T) \times \alpha \quad (1)$$

where F is the flux (mg C m⁻² h⁻¹ for CO₂, µg C m⁻² h⁻¹ for CH₄ and µg N m⁻² h⁻¹ for N₂O), ρ denotes the gas density as air at the standard conditions of 0°C and pressure 101.325 kPa (CO₂ = 1.976 × 10⁶ mg m⁻³, CH₄ = 0.716 × 10⁹ µg m⁻³ and N₂O = 0.978 × 10⁹ µg m⁻³), V is the volume of the chamber (m³), A is the cross-sectional area of the chamber (m²), $\Delta c/\Delta t$ is the rate of change in the gas concentration (c) inside the chamber during the sampling period (m³ m⁻³ h⁻¹), T is the air temperature (°C) and α is a conversion factor to transform CO₂ and CH₄ into C, N₂O into N (CO₂ = 12 / 44, CH₄ = 12 / 16 and N₂O = 28 / 44). The methane and N₂O fluxes were estimated by linear regression between the gas concentrations inside the chamber and the measured time. 98% for CH₄ and 77% for N₂O of all flux measurements had the regression coefficients of $R^2 > 0.7$. Positive values represent net emission from the soil, and negative values indicate net uptake into the soil.

During the gas sampling, soil temperature and soil moisture content at 10 cm depth were measured adjacent to each chamber. The temperature was measured using a thermistor thermometer (CT-220, Custom, Tokyo, Japan), gravimetric water content was determined by weighting the samples before and after drying at 105°C for 48 hours after collecting soil samples with a core sampler.

4.2.4 Understory vegetation and soil profile

One week before and after experimental burning, 1 m × 1 m subplots were randomly established within each plot to survey losses of understory biomass due to the fire. For biomass sampling of the understory, all of the understory plants within each subplot were harvested by clipping the vegetation close to the ground. The collected samples were oven dried in the laboratory at 65°C for at least 48 hours, and weighed. To examine changes in the upper soil layer due to the fire, the thickness of each layer was measured in each plot at the time we collected the soil samples using the soil core sampler.

4.2.5 Soil sampling and analysis

Samples of organic and soil (0–10 cm depth) layer were collected on five occasions during the first growing season 2007, using the soil core sampler. The samplings of organic and soil layer were conducted at three points adjacent to the chamber that were determined randomly within the each plot. At each plot, three collected sub-samples for organic and soil layer were combined and mixed to generate a single sample for analysis, respectively. These samples were sieved (2-mm mesh) and subdivided for 2 M KCl extraction (1:5, KCl), pH measurement (1:10, H₂O) and gravimetric analysis of water content (105°C for 48 hours). They were then air-dried. To quantify NH₄⁺ and NO₃⁻ from the soil solution, KCl extracts were filtered, and were frozen prior to analysis. The concentrations of NH₄-N and NO₃-N in the extracts were measured using an auto analyzer (AACS-4, BL-TEC Inc., Osaka,

Japan). The total C and N contents in air-dried samples were measured using an NC analyzer (Sumigraph NC-1000, Sumika Chemical Analysis Service Ltd., Osaka, Japan). One week before and after the fire, moreover, bulk density was determined in each plot at the time we measured the soil moisture content. We also calculated water-filled pore space (WFPS) using the bulk density and soil moisture content.

4.2.6 Statistical analysis

All statistical analyses were performed using SAS 8.1 software (SAS 2001). Distributions of the data for soil GHG fluxes and environmental conditions were tested for normality using the Shapiro-Wilk test. Kruskal-Wallis test, a non-parametric ANOVA, was used to examine statistically significant differences in parameter values between the three treatments. Repeated measures ANOVA was also undertaken to determine the changing patterns of soil GHG fluxes following the fire with measurement date as the repeated factor. Correlation analysis and stepwise linear regression analysis were also employed to examine the relationships between soil GHG fluxes and environmental variables. Furthermore, the van't Hoff equation ($y = a \exp(b ST)$) was established to calculate the temperature sensitivity ($Q_{10} = \exp(10 b)$) of soil CO₂ flux to the change of soil temperature (ST).

4.3 Results

4.3.1 General conditions and soil properties

Surface fire removed almost all of the aboveground biomass of understory vegetation, which is completely dominated by dwarf bamboo, and the litter layers (Table 4.2). The depths of Fermentation-humus (FH) layers were not significantly affected by the burning in the BURN and BURN-CHA. As a result of the burning, about 3.0 cm of the charcoal layer was established in the BURN. However, we observed no changes in bulk density and WFPS under three treatments before and after the fire (Table 4.3). In organic layer after the fire, the soil pH in BURN and BUR-CHA was slightly higher than in CON. Inorganic N pools were dominated by NH₄-N, rather than NO₃-N, in all three treatments. We observed an increase of inorganic-N concentration in organic layer in the burned area after the fire (Fig.

4.1). In particular, NH₄-N concentration in BURN and BURN-CHA soils was about three times higher immediately after the fire than before. And then, one month after the fire, the increased NH₄-N concentrations declined drastically in BURN and BURN-CHA to values similar to or lower than those of CON. No significant effects on NO₃-N concentration in organic layer were measured between CON and BURN soils throughout the first growing season 2007, while significantly higher concentrations were detected in BURN-CHA than in the CON and BURN one month after the fire (Fig. 4.1). Although inorganic-N pools in the organic layer were increased as a result of the fire, the organic-C and total-N concentrations in the organic layer were not significantly different after the fire from beforehand (Fig. 4.1). In fact, there were no changes in any property of the soil layer among three treatments due to the fire (Fig. 4.2). In this study, the surface fire has a positive effect on inorganic-N in the organic layer or no effect in the soil layer.

4.3.2 Soil GHG fluxes

The soil CO₂ flux in the three treatments showed a clear seasonal change, which coincided with changes in the soil temperature: an increase during the early growing season, a peak in August, and a decrease toward autumn (Fig. 4.3). The surface fire significantly decreased soil CO₂ flux in BURN and BURN-CHA during two growing seasons ($P = 0.049$) (Table 4.4). Soil CH₄ uptake was observed in all research plots throughout the measurement period (Fig. 4.3). In July and August 2008, soil CH₄ uptake in BURN and BURN-CHA tended to be greater than in CON. However, no difference was observed in the overall trends of soil CH₄ uptake between the three treatments ($P = 0.235$) (Table 4.4). The N₂O flux from BURN-CHA soil was slightly greater than in CON or BURN. In BURN-CHA the soil N₂O flux peaked in August, whereas there was no peak in BURN (Fig. 4.3). Although the mean values of soil N₂O flux were not significantly different from three treatments ($P = 0.129$), repeated measures ANOVA indicated significant interaction between the treatment and time ($P = 0.012$) (Table 4.4).

Table 4.3. Physical properties at 0–10 cm depth of soil layer in three treatments before and after the experimental burning

Bulk density (g cm ⁻³)	CON	BURN	BURN-CHA
Before	0.61 ± 0.02	0.62 ± 0.01	0.62 ± 0.01
After	0.61 ± 0.03	0.62 ± 0.02	0.61 ± 0.02
WFPS (%)	CON	BURN	BURN-CHA
Before	31.98 ± 2.32	31.98 ± 2.32	30.23 ± 0.74
After	30.10 ± 1.89	32.66 ± 1.67	30.22 ± 1.17

Data are given as the mean ± SE. (Adapted from Kim *et al.* 2011a, with kind permission of European Journal of Forest Research)

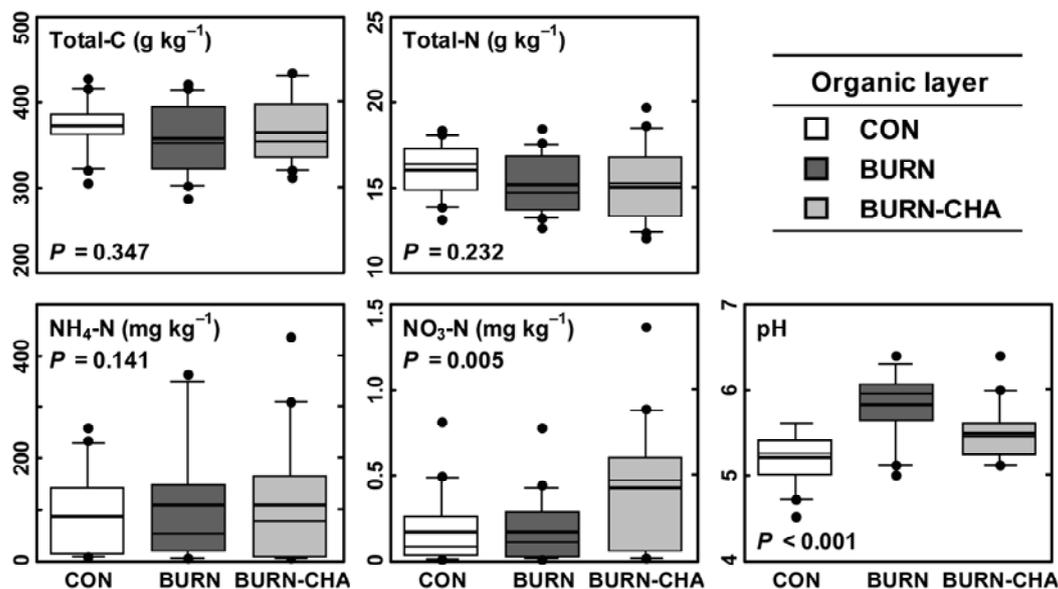


Fig. 4.1. Changes in soil variables at the organic layer in three treatments during the first growing season 2007. Bold and thin lines in each box plot correspond to the mean and median values, respectively. P -values represent significant differences among the treatments using the repeated measures ANOVA. (Reprinted from Kim *et al.* 2011a, with kind permission of European Journal of Forest Research)

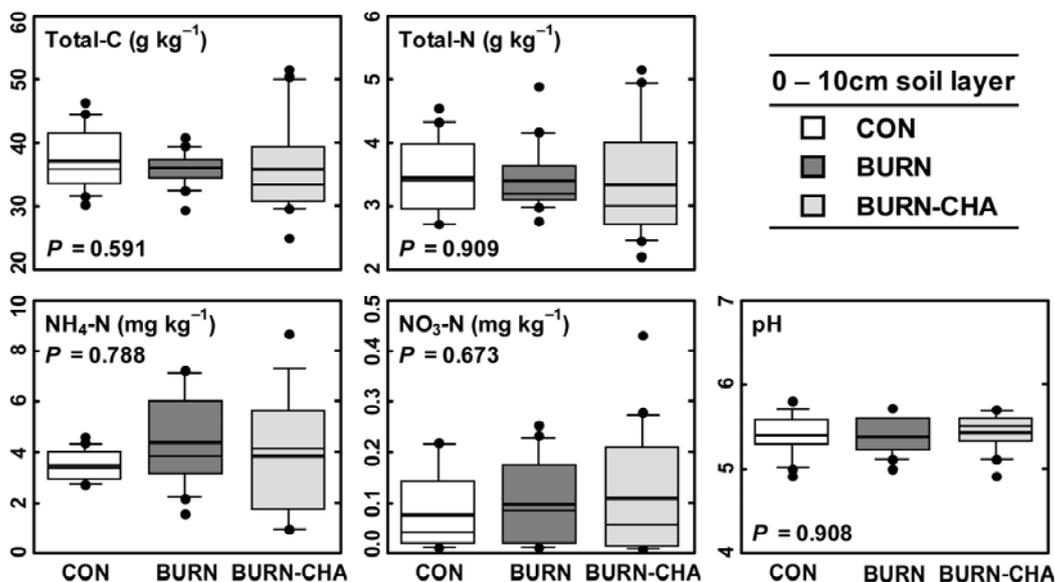


Fig. 4.2. Changes in soil variables at the 0–10 cm depth of soil layer in three treatments during first growing season 2007. Bold and thin lines in each box plot correspond to the mean and median values, respectively. P -values represent significant differences among the treatments using the repeated measures ANOVA.

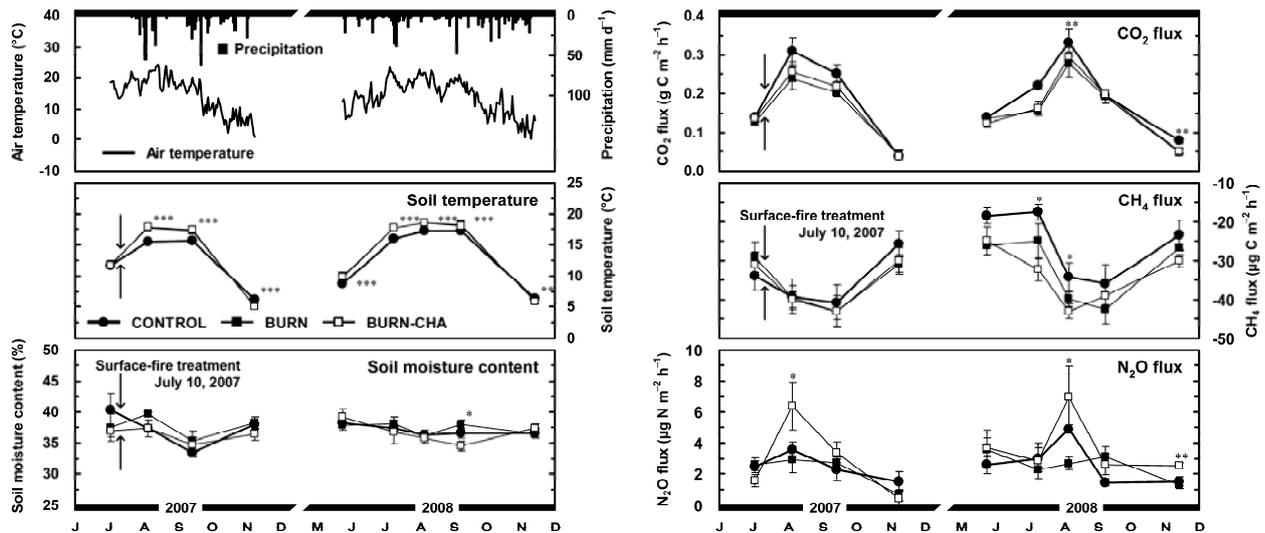


Fig. 4.3. Seasonal changes in soil temperature, soil moisture content, and soil GHG fluxes in three treatments following the experimental burning. Error bars indicate 1 SE ($n = 8$) of the mean. Values with asterisks indicate significant differences among the treatments in each measurement point (Kruskal-Wallis test, * $P < 0.05$, ** $P < 0.01$ and *** $P < 0.001$). (Reprinted from Kim *et al.* 2011a, with kind permission of European Journal of Forest Research)

Table 4.4. Repeated measures analysis of variance for the effects of experimental burning on soil GHG fluxes

Source	df	Soil CO ₂ flux			Soil CH ₄ flux			Soil N ₂ O flux		
		MS	F-value	P > F	MS	F-value	P > F	MS	F-value	P > F
<i>Between subjects</i>										
Treatment	2	0.02	3.43	0.049	470.51	1.55	0.235	18.54	2.26	0.129
<i>Within subjects</i>										
Date	8	0.187	93.07	<0.001	1281.7	21.18	<0.001	35.02	8.88	<0.001
Date × Treatment	16	0.002	0.95	0.511	71.61	1.18	0.286	8.12	2.06	0.012

(Modified from Kim *et al.* 2011a, with kind permission of European Journal of Forest Research)

4.4 Discussion

4.4.1 General conditions and soil properties

Although the relation between fire and soil nutrients is complex, in view of the interactions among many environmental factors (Wright and Bailey 1982, Pritchett and Fisher 1987), fire intensity is usually the major factor driving post-fire nutrient dynamics, with greater nutrient losses occurring with higher fire intensity. According to the peak temperatures at the forest floor during the fire (500–530°C), our prescribed fire is classified as low intensity (Neary *et al.* 1999). This low-intensity surface fire caused a significant reduction in aboveground levels, including the organic layer, whereas the mineral soil layer did not undergo dramatic changes, because the increase in soil temperature during this fire was only 3–5°C and the duration of the burning was only half an hour.

Fires generally lead to higher soil temperatures in

subsequent months as a result of removal of the forest floor, darkening of soil surfaces, and opening of the canopy. At the same time, removal of above-ground biomass may increase soil moisture by reducing evapotranspiration (Fisher and Binkley 2000, Certini 2005). Our surface fire also increased the soil temperature in the burned area, BURN and BURN-CHA, as compared with CON by removing the understory vegetation and litter layer (Fig. 4.3). There was no difference in soil temperature between the burned plots. This is probably due to the presence of overstory trees at both burned sites. Moreover, we expected the soil moisture content in BURN and BURN-CHA to increase due to less transpiration by trees, but there was no significant difference in soil moisture content between the three treatments. It was considered that increased evaporation resulting from removal of the understory shield (increasing solar

radiation) and darkening of the soil surface (decreasing the albedo) outweigh the decreased transpiration by the understory vegetation.

The low-intensity surface fire slightly increased the pH in the organic layer, in the order: BURN > BURN-CHA > CON. However, soil pH in the soil layer did not change significantly among three treatments during the first growing season 2007. The large amount of ash deposited in the wake of the fire was probably the major cause of increased pH in the organic layer. The pH rise in BURN and BURN-CHA could be due to the release of extractable basic cations from organic matter during the fire (Makoto 2010), but also to increased ammonification immediately after the fire. The soil pH typically increases immediately after fire, and then declines to pre-fire levels over a period of months, years, or decades. Soil pH usually influences the availability of some nutrients, through direct geochemical effects and through indirect effects on microbial activity (Fisher and Binkley 2000).

Moreover, although the low-intensity surface fire appeared to have no effect on the inorganic N concentration in the soil layer (Fig. 4.2), the $\text{NH}_4\text{-N}$ concentration in the organic layer of the burned area, BURN and BURN-CHA, increased considerably immediately after the burning (Fig. 4.1). Both NH_4^+ and NO_3^- are inorganic forms of nitrogen that originate during the burning. Ammonium is a direct product of combustion, and NO_3^- is formed from NH_4^+ some weeks or months later as a result of nitrification (Covington and Sackett 1992).

During the first growing season 2007, the $\text{NO}_3\text{-N}$ concentration in the soil layer did not change significantly in any of the three treatments after the fire

(Fig. 4.2), while increased $\text{NO}_3\text{-N}$ concentration in the organic layer in BURN-CHA was observed after one month of the fire (Fig. 4.1). In contrast, the $\text{NO}_3\text{-N}$ concentration in the organic layer did not increase in BURN soil, despite the increased $\text{NH}_4\text{-N}$ concentration immediately after the fire. This implicates that charcoal presence suppress the increase of NO_3^- after fire. One possible mechanism is that charcoal affects the biological processes. Immobilization of NO_3^- which is the process potentially decreasing the temporal $\text{NO}_3\text{-N}$ concentration. The C/N ratio of charcoal was about 30 in this study (Makoto 2010), which can enhance the NO_3^- immobilization by microorganism and reducing the $\text{NO}_3\text{-N}$ pools (Lehmann *et al.* 2003). On the other hand, the inevitably disturbance humus layer during charcoal removal is possible confounding factor that could cause $\text{NO}_3\text{-N}$ concentration to increase because disturbance of the surface floor results in more aerobic conditions, which are suitable for nitrification (Makoto 2010). So far, we can only speculate the mechanism how charcoal affects biological process of nitrogen cycling as reported by previous studies (Berglund *et al.* 2004, Makoto 2010)

At any rate, there are often short-term fire induced changes in the nitrogen budget. Soil inorganic N pools typically increase, due to heat-induced NH_4^+ release from clay complexes, to deposition of organic N in ash, and enhanced ammonification (Grogan *et al.* 2000, MacKenzie *et al.* 2004). In this connection, Schoch and Binkley (1986) have found increased N mineralization from the forest floor following a low-intensity prescribed burn in a previously unburned loblolly forest in the North Carolina piedmont.

Table 4.5. Temporal correlation between soil GHG fluxes and soil variables measured in study stands during first growing season 2007

Soil GHG flux	Pearson correlation coefficient						
	SMC	ST	pH	OC	TN	$\text{NH}_4\text{-N}$	$\text{NO}_3\text{-N}$
CO_2 flux	-0.28	0.91***	-0.05	0.31	0.22	0.31	0.28
CH_4 flux	0.32	-0.87***	-0.28	-0.01	0.08	-0.02	-0.10
N_2O flux	-0.10	0.78**	0.09	0.05	-0.09	0.18	0.67*
Soil GHG flux	Stepwise linear regression						Adj. R^2
	Equation						
CO_2 flux	$y = 0.017 \text{ ST} - 0.050$						0.82***
CH_4 flux	$y = -1.323 \text{ ST} - 20.248$						0.73***
N_2O flux	$y = 0.302 \text{ ST} + 1.188 \text{ NO}_3\text{-N} - 2.495$						0.77***

SMC, ST, OC and TN indicate soil moisture content, soil temperature, organic-C and total-N, respectively. Asterisks with values represent the level of significance of correlations and regression models (* $P < 0.05$, ** $P < 0.01$ and *** $P < 0.001$). (Adapted from Kim *et al.* 2011a, with kind permission of European Journal of Forest Research)

Table 4.6. Q_{10} functions and van't Hoff equations of soil CO₂ flux on soil temperature at 10 cm depth in three treatments

Treatment	Q_{10}	van't Hoff equation	R^2	P
CON	4.0	$y = 0.0270\exp(0.1384 \text{ ST})$	0.81	< 0.01
BURN	3.1	$y = 0.0288\exp(0.1134 \text{ ST})$	0.88	< 0.001
BURN-CHA	3.3	$y = 0.0270\exp(0.1203 \text{ ST})$	0.89	< 0.001

ST indicates soil temperature at 10 cm depth of soil. (Adapted from Kim *et al.* 2011b, with kind permission of European Journal of Forest Research)

4.4.2 Soil GHG fluxes

The mean soil CO₂ flux rate of $0.19 \pm 0.01 \text{ g C m}^{-2} \text{ h}^{-1}$ in CON measured in our white birch forest is comparable to the range of $0.05\text{--}0.31 \text{ g C m}^{-2} \text{ h}^{-1}$ reported in a cool-temperate deciduous broad-leaved forest in central Japan (Lee *et al.* 2003), and is within the range of $0.12\text{--}0.20 \text{ g C m}^{-2} \text{ h}^{-1}$ found by four distinct types of measurement in a 45-year-old Japanese larch plantation in northern Japan (Liang *et al.* 2004). This value is of the same magnitude as the soil CO₂ fluxes in temperate forests of the northern hemisphere, ranging from $0.12\text{--}1.10 \text{ g C m}^{-2} \text{ h}^{-1}$ (e.g. Raich and Schlesinger 1992). The soil CO₂ flux in three treatments displayed a clear season variation: increasing during the early growing season, peaking in August, and decreasing from the early fall to November (Fig. 4.3). During the first growing season 2007, we observed that a positive temporal correlation between soil CO₂ flux and soil temperature ($P < 0.001$) (Table 4.5). The soil temperature was the dominant factor that controlled soil CO₂ emission in our experiment from the stepwise linear regression analysis and it could explain 82% of temporal variability ($P < 0.001$). Moreover, we found a weak positive correlation of spatial variation between soil CO₂ emission and total-C in study stands ($R^2 = 0.38$, $P < 0.025$) (Fig. 4.4).

The soil CO₂ flux is generally correlated positively with soil temperature (Luo and Zhou 2004, Kim *et al.* 2009). The soil temperature in the burned area, BURN and BURN-CHA, was clearly higher than that in CON. In contrast, soil CO₂ flux was significantly lower in BURN and BURN-CHA than in CON (Fig. 4.3, Table 4.4). An exponential regression of each CO₂ measurement against the corresponding soil temperature at 10 cm depth was highly significant in all three treatments ($R^2 = 0.81\text{--}0.89$, $P < 0.01$); this relationship was used to determine Q_{10} functions. In forest ecosystems, Q_{10} values were lower in burned sites than in intact controls, because fires usually reduce subsequent soil CO₂ flux; the more severe the fire, the greater the effect. In this study, the Q_{10} values in BURN and BURN-CHA were slightly lower than in CON (Table 4.6). The likely explanation is that most root respiration decreased in the burned area, because root activity was diminished due to the combustion of the aboveground biomass. It is also supposed that the reduced litter layer thickness reduces the soil CO₂ flux.

In particular, the declining ratio of emissions in BURN and BURN-CHA, compared with CON, throughout two growing seasons indicates greater root activity in CON during the summer period. The burned area might have lower root activity, and hence a smaller increase in CO₂, during the measurement period. This hypothesis is supported by the work of Sawamoto *et al.* (2000), who found that soil CO₂ flux in burned sites fell to 46–75% of that in intact forest at 1 and 5 years after the fire; they concluded that this was due to reduced respiration of larch roots. Moreover, some previous studies in black spruce forests in Alaska have found that soil CO₂ flux was reduced by 22–50% in a burned area at 0–2 years after a forest fire, and 37–63% at 1–10 years after (Kim and Tanaka 2003, O'Neill *et al.* 2006). However, Takakai *et al.* (2008) observed in the permafrost taiga region of eastern Siberia that the cumulative soil CO₂ flux in a burned site was similar to that of an intact forest site.

Aerobic well-drained forest soils, in particular temperate forest soils, act as a significant sink for CH₄, as a result of the high rate of CH₄ diffusion into such soils and subsequent oxidation by methanotrophs (e.g. Dalal *et al.* 2008). In the present study, all research plots were in a well-drain condition and generally consumed CH₄ before the experimental burning. The mean soil CH₄ flux rate of $-29.98 \pm 1.55 \mu\text{g C m}^{-2} \text{ h}^{-1}$ in CON during the measurement period in our white birch forest is within the range of -24 to $-66 \mu\text{g C m}^{-2} \text{ h}^{-1}$ reported in northern Japan (Morishita *et al.* 2004), and is comparable to the range of -11 to $-43 \mu\text{g C m}^{-2} \text{ h}^{-1}$ found for temperate forests (Smith *et al.* 2000). During two growing season, soil CH₄ uptake increased gradually from the early growing season, reached maximum uptake value in August or September and then decreased gradually (Fig. 4.3). We found that a negative temporal correlation between soil CH₄ flux and soil temperature, i.e. CH₄ consumption rate into the soil increases along with soil temperature ($P < 0.001$) (Table 4.5). The soil temperature was the main controlling factor for the temporal variation of soil CH₄ uptake and explained 73% of the temporal variability from the stepwise linear regression analysis ($P < 0.001$).

After the low-intensity surface fire, soil CH₄ uptake was observed in all research plots during the measurement period. Soil CH₄ uptake in the burned

area, BURN and BURN-CHA, tended to be greater than in CON. Our findings indicate that low-intensity fire has the potential to increase, not decrease, CH₄ consumption in forest soils, but there was no significant difference between the three treatments except in July and August 2009 (Fig. 4.3, Table 4.4). Although the effect of forest fire on CH₄ uptake by forest soil has been studied mainly in boreal regions, most previous studies have reported that forest fire enhances soil CH₄ uptake (Burke *et al.* 1997, Jaatinen *et al.* 2004, Takakai *et al.* 2008). In particular, Jaatinen *et al.* (2004) found that fire led to increased CH₄ oxidation rates and caused no detectable changes in the methanotroph community. The same authors also pointed out that increased pH after the fire, and ash, probably do not cause any change in the methanotroph community, and the increased soil pH is not directly responsible for increased oxidation rates after fire. Thus, increasing pH, along with other factors such as the combustion of the litter layer, may enhance soil CH₄ uptake in burned areas. For example, the removal of a thin organic layer in early June increased the CH₄ oxidation rates by 50% during the summertime in a boreal Scots pine forest in Finland (Saari *et al.* 1998). It was also similar to a previous study of spruce forest in Germany (Steinkamp *et al.* 2001). These authors concluded that the organic layer acts as a barrier to soil-atmosphere gas exchange by diffusion (e.g. CH₄, O₂), thereby limiting atmospheric CH₄ oxidation. Fires reduced the thickness of the organic layer and exposed the underlying mineral soil. Fires could therefore influence methanotroph activity through increased gas diffusion, and thereby increase the methanotroph biomass.

Recent studies have indicated that the incorporation of biochar within soil decreases N₂O emissions and increases CH₄ uptake from soil, which could assist in reducing GHG emissions (Rondon *et al.* 2006, Yanai *et al.* 2007, Spokas and Reicosky 2009). Biochar is charcoal created by pyrolysis of biomass for biosequestration or atmospheric C capture and storage; it is not used as a fuel, and is of increasing interest because of concerns about climate change due to GHG emissions. Evidence exists that charcoal decreases CH₄ emission from soil, but only very limited information is available. Rondon *et al.* (2005) observed that CH₄ emissions were totally suppressed when biochar was applied to tropical soils. Rondon *et al.* (2006) also demonstrated that mixing of 20 t ha⁻¹ of wood-derived biochar into unfertile tropical soils increased the annual CH₄ sinks in soil by approximately 200 mg CH₄ m⁻². However, in laboratory incubation, Spokas and Reicosky (2009) found that sixteen different biochars and soil combinations resulted in decreased or unaltered rates of CH₄ oxidation, and no increase was observed in CH₄ oxidation or production. Their findings do not appear to support the results of Rondon *et al.* (2005, 2006) on the suppression observed in CH₄ emissions from tropical soils, interpreted as an increase CH₄ oxidation activity. This effect could also be explained by a decrease in the activity of CH₄-producing bacteria (methanogens), yielding the same net effect in reducing the observed CH₄ emissions. We expected to find that

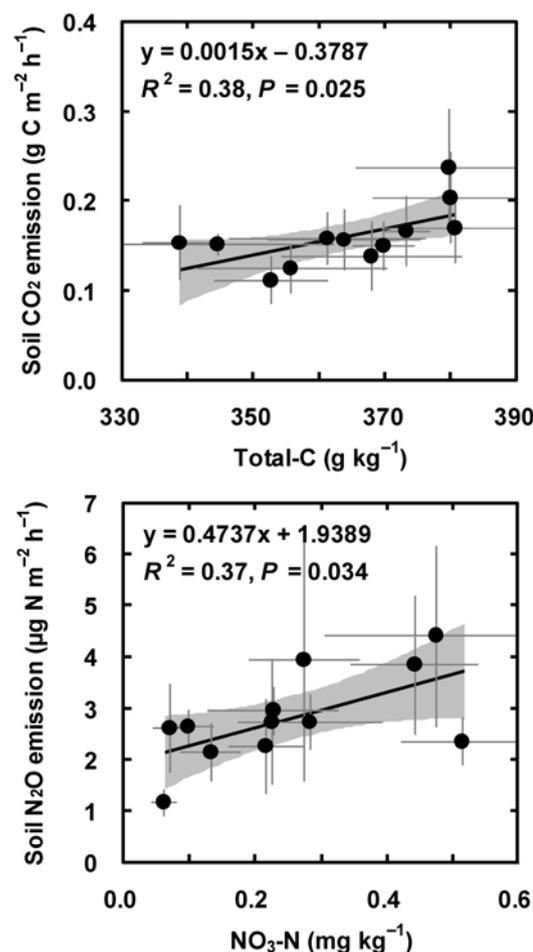


Fig. 4.4. Spatial relationship between soil GHG fluxes and soil variables in study stands. Values indicate the mean \pm SE ($n = 8$), which was measured in each research plot during first growing season 2007. (Reprinted from Kim *et al.* 2011a, with kind permission of European Journal of Forest Research)

charcoal produced in the fire in our study would influence the soil CH₄ flux, but no significant difference was observed between BURN and BURN-CHA. This result is explicable by the site conditions; since our experimental burning was conducted upon aerobic well-drained forest soil, CH₄ production would not be favored.

Consider now the soil N₂O flux. The mean value of $2.59 \pm 0.20 \mu\text{g N m}^{-2} \text{h}^{-1}$ in CON during the measurement period in our white birch forests was similar to the rate of $2.6 \mu\text{g N m}^{-2} \text{h}^{-1}$ measured in Japanese cypress forests in Japan (Morishita *et al.* 2007), and comparable to the rate of $3.3 \mu\text{g N m}^{-2} \text{h}^{-1}$ reported in a cool-temperate eucalypt forest in south-eastern Australia (Fest *et al.* 2009). However, our value is slightly above the range of $0.17\text{--}1.59 \mu\text{g N m}^{-2} \text{h}^{-1}$ determined in a northern Japanese forest (Morishita *et al.* 2007). Variation of soil N₂O flux did not show a clear seasonal pattern (Fig. 4.3). There were two major peaks of soil N₂O emission in BURN-CHA during two

growing season, e.g. in early August 2007 and 2008. We could find a positive temporal correlation between soil N₂O flux and soil temperature ($P < 0.01$) or NO₃-N ($P < 0.05$) during the first growing season 2007 (Table 4.5). Among the environmental variables, soil temperature and NO₃-N concentration were the dominant factors that controlled soil N₂O emission in this study from the stepwise linear regression analysis and they could explain 77% of the temporal variability ($P < 0.001$). In this study, a high spatial variability of soil N₂O flux among four study sites, with an average coefficient of variation (CV) of 58–98%, which was obviously larger than those of CO₂ (25–29%) and CH₄ (24–35%). The result suggests that it is necessary to increase the chamber of spatial replicates in field observation. However, we also found a weak positive correlation of spatial variation between soil N₂O emission and NO₃-N concentration ($R^2 = 0.38$, $P = 0.034$) (Fig. 4.4). In general, soil N₂O production correlates with N availability in the soil, because nitrification and denitrification depending on NH₄⁺ and NO₃⁻, respectively (Bouwman 1990). Therefore, we considered that increased inorganic-N levels in the soil as a result of the fire can influence the soil N₂O emission in forest ecosystems, at least for a short term.

On the other hand, Takakai *et al.* (2008) demonstrated that although there were no significant differences between the sites, cumulative N₂O emission from a burned site during the growing season was slightly lower, and emission from a clear-felled site was slightly higher, than at an intact forest site. In particular, they found substantial N₂O emissions only from the clear-felled site, although soil moisture increased at both burned and clear-felled sites. Also, forest fire decreased by 10–50% the N₂O emission from soil in mixed hardwood and black spruce forests in Alaska (Kim and Tanaka 2003). It can be expected that fire-produced charcoal suppresses N₂O emission from forest soils. We therefore conducted a charcoal removal treatment after the experimental burning, to verify the effect of charcoal on soil N₂O flux. In the present study, the soil N₂O flux in BURN-CHA and CON peaked in August 2008 and 2009, but there was no peak in BURN despite the immediate increase in NH₄-N concentration in the organic layer after the fire (see Fig. 4.3). We considered that charcoal produced by the low-intensity surface fire in BURN suppressed soil N₂O emission as a result of net immobilization of NH₄⁺ or interfere with nitrification. The charcoal has the capacity to absorb organic compound that might otherwise induce net immobilization of NO₃⁻ (Fierer *et al.* 2001) or interfere with nitrification (White 1994). Our result was consistent with previous field studies (Rondon *et al.* 2005, Rondon *et al.* 2006) and laboratory incubations (Yanai *et al.* 2007, Spokas *et al.* 2009). This is not always the case, however, because charcoal made from pine wood chip with high N compost increased N₂O emission across three distinct soil types: agricultural, forest nursery and landfill cover soil (Spokas and Reicosky 2009). Van Zwieten *et al.* (2009) pointed out that the NO₃⁻ concentration in Ferrosol soil modified with low temperature green waste biochar fell by 20%

over 47 days of incubation, concomitant with the increased N₂O that was also produced. These authors proposed that charcoal addition initially slows the soil microbial activity, but this claim is based on laboratory incubation. Alternatively, NO₂, NO and N₂O formed in soil through denitrification might be chemically adsorbed on charcoal surfaces and electrochemically reduced to N₂. High N₂O consumption rates on wood charcoal have been reported in 30 ppm N₂O mixtures (Terazoe *et al.* 2002), and the sorption behavior of N₂O on charcoal has been known for some time (McBain 1926). However, Spokas and Reicosky (2009) did not observe significant N₂O sorption with a range of charcoals at 0.3 ppm N₂O (ambient). It is important to understand which components of the N cycles, and which enzymes associated with nitrogen mineralization, nitrification and denitrification, are most affected by charcoal (Van Zwieten *et al.* 2009).

Finally, fire-produced charcoal as a tool for reducing GHG emissions from soil will require detailed understanding of the interactions between charcoal and soil conditions, of climate patterns, and of vegetation growth that alters the GHG source or sink capacity of soils. The processes by which charcoal interacts with these long-term controls so as to influence soil GHG fluxes are still unclear. In this study, we could recognize the potential possibility that charcoal remaining in soil can play a significant role in reducing emissions and increasing the sink capacity for GHGs. Charcoal remaining after forest fires can play an important role in regulating not only nutrients, but also GHG emissions from the forest floor of a white birch forest.

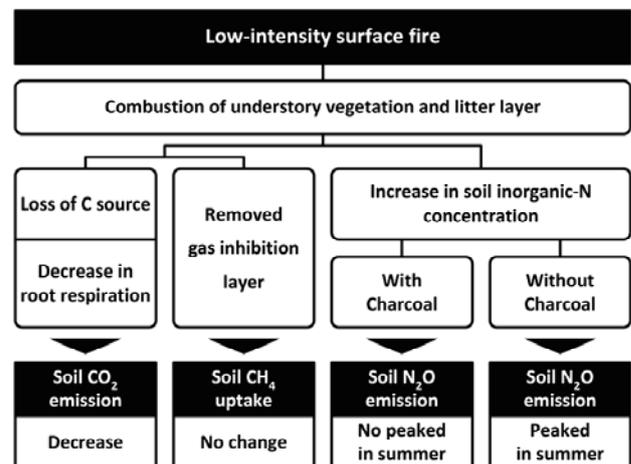


Fig. 4.5. Overall effects of the low-intensity surface fire on soil GHG fluxes.

4.5 Conclusions

Generally, forest fires affect nutrient cycling in forest ecosystems, and thereby influence the soil-atmosphere exchange of GHGs. Fires, unlike other forest disturbances, are critical natural or man-made disturbance that directly affect forest carbon cycling by the immediate combustion of biomass depending on the peak temperature and duration of the fire (Neary *et al.* 1999, Dale *et al.* 2001, Certini 2005). Finally, environmental change in burned areas is liable to affect the exchange of GHGs between the soil and the atmosphere. In this study, especially, our low-intensity surface fire increased $\text{NH}_4\text{-N}$ concentration in both burned area, BURN and BURN-CHA, immediately after the fire, while $\text{NO}_3\text{-N}$ concentration was only increased in BURN-CHA after one month of the fire (Fig. 4.5). Soil CO_2 flux was significantly lower in BURN or BURN-CHA than in CON. The plausible explanation is that most root respiration decreased in the burned area, because root activity was diminished due to the combustion of understory vegetation. It is also supposed that the combustion of litter layer reduces the soil CO_2 flux. The experimental burning removed the litter layer and exposed the underlying mineral soil.

As this result, we expected to increase in soil CH_4 uptake in burned area as a result of the combustion of litter layer which acts as a barrier to soil-atmosphere gas exchange by diffusion. However, we observed no significant difference in soil CH_4 uptake between the three treatments. Moreover, soil N_2O flux in BURN-CHA was slightly greater than in CON or BURN. In BURN-CHA, especially, the soil N_2O flux peaked in August, but there was no peak in BURN. According to this point, we considered that although increased inorganic-N levels in the burned area as a result of the fire influence preferentially the soil N_2O emission, it might be suppressed through net immobilization of NH_4^+ or interfere with nitrification by fire-produced charcoal. In conclusion, our results suggest that environmental changes following fire, including the increased availability of N and the disappearance of the litter layer, have the potential to change soil GHG fluxes. Fire-produced charcoal could be significant in reducing soil N_2O flux in temperate forests.

CHAPTER 5 GENERAL DISCUSSION

Since the industrial revolution, the atmospheric concentrations of CO_2 , CH_4 and N_2O have been increasing at mean annual rates over last two decades (IPCC 2007). Because of the potential effects of increased atmospheric concentrations of CO_2 , CH_4 and N_2O on the global energy budget, considerable efforts have been recently made to quantify the strength of terrestrial sources and sinks for these gases. Forests are key components of global CO_2 , CH_4 and N_2O budgets (Schlesinger 1997).

Current estimates of GHG budgets of forests indicate that these ecosystems act as carbon sinks, regardless of climatic region. The sequestering of GHGs by forests is

the result of the balance between a substantial uptake of CO_2 by biomass and CO_2 release through soil respiration, oxidation of CH_4 by methanotrophs, and a more or less significant emission of N_2O from soils, a by-product of nitrification and denitrification reactions. According to recent budgets, close to 20% of the CO_2 found in the atmosphere would transit annually through forests, and around 2 Pg C yr^{-1} (excluding deforestation) would have been sequestered in the biomass and forest soils in the 1980s, reducing the impact of anthropogenic GHG emissions, such as those from fossil fuel burning (Schlesinger 1997, IPCC 2007). The importance of forest soils as methane sinks are significant, with a consumption estimated at around 5% ($30 \text{ Tg CH}_4 \text{ yr}^{-1}$) of the CH_4 emitted worldwide (Schlesinger 1997). It is difficult to accurately assess the net contribution of forests to global climate change due to the complexity of the processes involved. This evaluation is intimately linked to local environmental conditions and controlled by spatio-temporal factors.

Generally, forest soils are important to identifying and enhancing natural sink for C sequestration to mitigate the climate change (Lal 2005). Among the forest biomes, soils in temperate forest have been considered as a major source of CO_2 and N_2O , while acting as a significant sink for CH_4 (Robertson *et al.* 2000, Le Mer and Roger 2001, Raich *et al.* 2002, Fest *et al.* 2009). In particular, temperate forest soils are identified to be a small N_2O source because the temperate forests have comparatively large C/N ratios in litter and topsoil (Butterbach-Bahl and Kiese 2005). However, the higher atmospheric CO_2 concentration and the increase in atmospheric N deposition could have transformed these systems, by having changed their physico-chemical soil conditions. Furthermore, natural disturbances are expected to increase in frequency and extent under conditions of a changing climate (IPCC 2007). The forest GHG budgets do not, however, take into account the influence of natural disturbances (fire, drought, wind damage, etc.) on large-scale carbon dynamics of Asian forests. Over wide areas, these natural disturbances, especially forest fire, can result in significant short and long-term emissions of GHG from soils. As a result, the production and consumption of these three GHGs are biologically mediated in forest ecosystems, but also could change by natural or man-made disturbances and other climate change influenced environmental variables.

In this study, therefore, changes in soil GHG fluxes were measured in forest ecosystems under various environmental changes which have the potential ability to change the soil-atmosphere exchange of GHGs, e.g. elevated atmospheric CO_2 , N deposition and fire. Such environmental changes in forest ecosystems have the potential impacts to change the soil-atmosphere exchange of GHGs (Fig. 5.1). Changes in soil GHG fluxes, furthermore, is that it varied with the environmental changes.

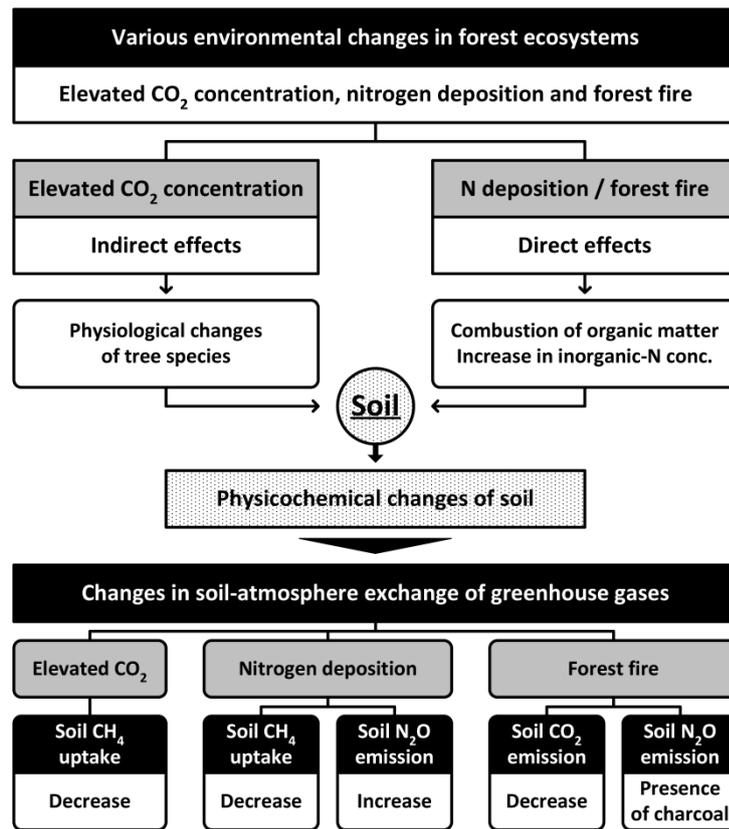


Fig. 5.1. Overall effects of various environmental changes on soil GHG fluxes.

Firstly, reduced CH_4 consumption with CO_2 enrichment was observed with increased soil moisture as a result of increased leaf stomatal closure and evapo-transpiration water loss from the forest floor. Secondly, simulated N addition, which was conducted to evaluate the effects of elevated atmospheric N deposition on soil GHG fluxes, inhibited soil CH_4 uptake and stimulated soil N_2O emission in response to increased inorganic-N concentration. Finally, our low-intensity surface fire reduced soil CO_2 flux by the combustion of understory vegetation and litter layer. Especially, depending on the existence of charcoal, soil N_2O flux was represented different seasonal pattern. We considered that although increased inorganic-N levels in the burned area as a result of the fire influence preferentially the soil N_2O emission, it might be suppressed through net immobilization of NH_4^+ or interfere with nitrification by fire-produced charcoal. According to the study, our results suggest that environmental changes following elevated CO_2 concentration, increasing atmospheric N deposition and surface fire have the potential to change soil GHG fluxes through the various processes in forest ecosystems.

Overall, the slow increases of soil moisture content or soil inorganic-N concentration may not markedly influence CH_4 consumption rate in soils under aerobic conditions, the decreased in soil CH_4 consumption of well-drained forest soils will evidently be of minor

importance to the global atmospheric CH_4 concentrations. As the climate change induced changes in the soil moisture or inorganic-N are slow, the additional impact on soil CH_4 consumption probably remains small. However, if the elevated atmospheric CO_2 concentration or increasing N deposition continues, we may not be able to expect the role of well-drained forest soils as a sink for CH_4 .

Also, soil N_2O emission seems to largely depend on whether the ecosystem N limited or not at the time of N inputs. Nitrogen cycling in forest ecosystems, which already exhibits large N_2O emission, responds strongly to the added N, whereas an ecosystem that has been limited by N uses up the added N rapidly and soil N_2O emission is elevated only for a short period of time. However, it is expected that continuous elevated-atmospheric N deposition will lead to stimulate soil N_2O emission from forest soils, which is identified to be a small N_2O source.

Moreover, although fires influence the soil-atmosphere exchange of GHGs, fire-produced charcoal could be significant in reducing soil N_2O flux in forest ecosystems, at least for a short term. Fire-produced charcoal as a tool for reducing GHG emissions from soil will require detailed and plausible understanding of the interactions between charcoal and environmental condition in forest ecosystems. However, it is considered to keep an eye on the potential possibility that charcoal remaining in soil can play a

significant role in reducing emissions and increasing the sink capacity for GHGs, as well as a contributor for soil C storage or a reservoir for nutrients.

Finally, the uncertainties about the effects of individual environmental changes and their interactions on soil GHG fluxes, and coupling between N and C cycles underline the need for further studies. Large uncertainties still remain concerning the *in situ* effect of environmental changes on soil GHG fluxes in different forest ecosystems. Accordingly, I hope these results of the study will help for the development of forest management to reduce the atmospheric GHG concentrations.

SYNOPSIS

Globally, soil is the largest reservoir in terrestrial ecosystems, recently estimated at 1500 Pg C, about triple of the plant biomass or twice of the atmospheric CO₂-C pool. Because of the potential effects of increased atmospheric concentrations of CO₂, CH₄ and N₂O on the global energy budget, considerable efforts have been recently made to quantify the strength of terrestrial sources and sinks for these gases. Forests are key components of global CO₂, CH₄ and N₂O budgets. Current estimates of GHG budgets of forests indicate that these ecosystems act as carbon sinks, regardless of climatic region. The sequestering of GHGs by forests is the result of the balance between a substantial uptake of CO₂ by biomass and CO₂ release through soil respiration, oxidation of CH₄ by methanotrophs, and a more or less significant emission of N₂O from soils, a by-product of nitrification and denitrification reactions. These two processes contribute two thirds of all N₂O emissions. However, it is difficult to accurately assess the net contribution of forests to global climate change due to the complexity of the processes involved. This evaluation is intimately linked to local environmental conditions and controlled by spatio-temporal factors. In forest ecosystems, especially, the production and consumption of these three GHGs are biologically mediated, but also strongly controlled by natural or man-made disturbances and other climate changes influenced environmental variables, e.g. elevated atmospheric CO₂, nitrogen deposition and fire.

At elevated CO₂ concentration, firstly, this increase will have direct effects on physiological processes of plants and will feed back indirectly into the soil system in forest ecosystems, and thereby influence the soil-atmospheric exchange of GHGs. To investigate the effect of elevated CO₂ on atmospheric CH₄ consumption by well-drained forest soil, we measured the soil CH₄ flux at the Free Air CO₂ Enrichment (FACE) site of Hokkaido University during two growing seasons (2008 and 2009). Fumigation with CO₂ began in June 2003, since when the plots have either been exposed to ambient levels or enriched with CO₂ at 130 μL L⁻¹. We also studied two soil types, brown forest soil and volcanic ash soil, which are widely distributed in northern Japan. The soil CH₄ flux, soil moisture content and soil temperature were measured simultaneously on four occasions on a monthly schedule during each growing season. During

the measurement period, the elevated CO₂ plot consistently contained significantly higher soil moisture than at of the ambient CO₂, by 7% in brown forest soil and 17% in volcanic ash soil. In both types of soil, the elevated CO₂ plot experienced an approximately 50% lower soil CH₄ consumption rate than in the ambient CO₂ plot. However, there was no significant difference in soil CH₄ consumption between two soil types. The negative linear relationship was found between soil CH₄ consumption and soil moisture in this study. Regardless of the soil type, soil CH₄ consumption was observed at all measurement points in the ambient CO₂ plot, but approximately 13% of all sampling points in the elevated CO₂ plot experienced net CH₄ production.

Secondly, increased atmospheric N deposition can lead to changes in soil N-status in forest ecosystems, and thereby can affect the exchange of GHGs between soil and atmosphere. To clarify the changes in soil CH₄ and N₂O fluxes following N application, N input was initiated at the onset of our experiment, and included two treatments with four replications, a zero and 50 kg N m⁻² yr⁻¹, in a brown forest nursery soil in the Sapporo Experimental Forest of Hokkaido University during two growing seasons (2008 and 2009). The N was added as an ammonium nitrate (NH₄NO₃) solution simulated to acid deposition distributed in four times during each growing season. Soil GHG fluxes and soil variables were determined on ten occasions for each growing season. Before the N application, no significant differences were observed on all soil variables between two treatments. However, NH₄-N and NO₃-N concentrations increased immediately after N inputs in 50 kg-N addition plot. With the increasing inorganic-N pools, we found soil CH₄ uptake in 50 kg-N addition plot decreased by 48% when compared to Zero-N control plot. Especially, reduced CH₄ uptake or CH₄ emission at 50 kg-N addition soils were observed immediately after N input events. Moreover, the N input increased soil N₂O emission by an average of 69% in 50 kg-N addition plot as compared with the control during two growing seasons. In this experiment site, there was a negative correlation between soil CH₄ uptake and NH₄-N concentration, and a positive correlation between soil N₂O emission and NO₃-N concentration, despite the soil CH₄ flux had a high inherent spatial variability.

Finally, forest fires affect nutrient cycling in forest ecosystems, and thereby influence the soil-atmosphere exchange of GHGs. Fires, unlike other forest disturbances, are critical natural or man-made disturbance that directly affect forest nutrient cycling by the immediate combustion of biomass depending on the peak temperature and duration of the fire. To determine changes in the soil GHG fluxes following a forest fire, we arranged a low-intensity surface fire in a white birch forest in the Teshio Experimental Forest of Hokkaido University. We established three treatments, having four replications each: a control plot (CON), a burned plot (BURN), and a plot burned with removal of the resulting charcoal (BURN-CHA). Soil GHG fluxes and various properties of the soil were determined on four or five occasions during a period that spanned two

growing seasons (2007 and 2008). We observed increased concentrations of $\text{NH}_4\text{-N}$ in BURN and BURN-CHA immediately after the fire, while $\text{NO}_3\text{-N}$ concentration was only increased in BURN-CHA after one month of the fire. The soil CO_2 flux was significantly higher in CON than in BURN or BURN-CHA, but there was no difference in soil CH_4 uptake between the three treatments. Moreover, the N_2O flux from BURN-CHA soil was slightly greater than in CON or BURN. In BURN-CHA, the soil N_2O flux peaked in August, but there was no peak in BURN. We found temporal correlations between soil GHG fluxes and soil variables, e.g. soil temperature or $\text{NO}_3\text{-N}$.

In this study, such environmental changes in forest ecosystems had the potential impacts to change the soil-atmosphere exchange of GHGs. Changes in soil GHG fluxes, furthermore, is that it varied with the environmental changes. Firstly, reduced CH_4 consumption with CO_2 enrichment was observed with increased levels of soil moisture as a result of increased leaf stomatal closure and evaporative water loss from the forest floor. Secondly, simulated N addition, which was conducted to evaluate the effects of elevated atmospheric N deposition on soil GHG fluxes, inhibited soil CH_4 uptake and stimulated soil N_2O emission in response to increased inorganic-N concentration. Finally, our low-intensity surface fire reduced soil CO_2 flux by the combustion of understory vegetation and litter layer. Especially, soil N_2O flux was represented different seasonal pattern, depending on the existence of charcoal. We considered that although increased inorganic-N levels in the burned area as a result of the fire influence preferentially the soil N_2O emission, it might be suppressed through net immobilization of NH_4^+ or interfere with nitrification by fire-produced charcoal. According to the study, our results suggest that environmental changes following elevated CO_2 concentration, atmospheric N deposition and fire have the potential to change soil GHG fluxes through the various processes in forest ecosystems. If the elevated atmospheric CO_2 concentration continues, we may not be able to expect the role of well-drained forest soils as a sink for CH_4 . Also, continuous atmospheric N deposition could lead to stimulate soil N_2O emission from forest soils, which is identified to be a small N_2O source. Moreover, although fires influence the soil-atmosphere exchange of GHGs, fire-produced charcoal could be significant in reducing soil N_2O flux in forest ecosystems, at least for a short term.

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Appendix

FACE System in experimental nursery



Young larch plantation in experimental nursery



White birch forest in Teshio experimental forest



Experimental burning in Teshio experimental forest



Soil GHG sampling after experimental burning



Closed-chamber method