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Soil nitric oxide emissions from two subtropical humid forests in south China

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[1] Due to the dense population, rapid industrialization, and intensified agricultural activities, some regions in Asia are hot spots of airborne nitrogen oxides and also areas with increasing nitrogen deposition. Therefore the cycling of nitrogen gases in Asia might be of increasing importance on both a regional and a global scale for atmospheric chemistry and budgets of nitrogen. Yet, to date, knowledge of soil NO emission is quite limited in Asia, particularly in forest ecosystems. In this study, soil NO emissions in two subtropical humid forests, a broadleaf forest in climax successional stage and a pine forest in primary successional stage, were measured throughout the year 2005 in Dinghushan Biosphere Reserve, south China. In the broadleaf forest, mean NO emission in wet season (14.9 ng N m^{-2} s^{-1}) was lower than in dry season (23.8 ng N m^{-2} s^{-1}). In the pine forest, however, mean NO emission in wet season (17.1 ng N m^{-2} s^{-1}) was higher than in dry season (7.9 ng N m^{-2} s^{-1}). In both forests, soil water content was the dominant factor controlling the seasonal patterns of NO emissions, and soil NO emission was significantly correlated to percent water filled pore space (%WFPS) in a quadratic manner ($p < 0.001$). Annual NO emissions in the broadleaf forest and the pine forest were preliminarily estimated to be 6.1–6.9 and 4.0–4.3 kg N ha^{-1} yr^{-1}, respectively, by using three upscaling methods.

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1. Introduction

[2] Nitrogen oxides (NO_x = NO + NO_2) play a crucial role in atmospheric chemistry [*Crutzen*, 1979; *Logan et al.*, 1981]. In the stratosphere, NO_x mainly from the oxidation of nitrous oxide there could catalyze the destruction of ozone (O_3) [*Crutzen*, 1979]. In the troposphere, however, NO_x act as a key agent in the photochemical production of O_3, and are important in regulating many other oxidizing agents, especially hydroxyl radical (OH) [*Crutzen*, 1979; *Williams et al.*, 1992; *Intergovernmental Panel on Climate Change* (*IPCC*), 2001]. Thus they are important in controlling the oxidizing capacity of the troposphere, and impact the fate of carbon monoxide (CO), methane (CH_4) and nonmethane hydrocarbons (NMHC) [*Logan et al.*, 1981; *Liu et al.*, 1987]. The end photochemical product of NO_x, HNO_3 and NO_3^-, contribute to secondary aerosols and to the acidity of clouds and precipitation [*Liu et al.*, 1987].

[3] Soils are a major source of atmospheric NO_x [*Yienger and Levy*, 1995; *Delmas et al.*, 1997]. Although NO (the predominant form of NO_x emitted from soils) may come from abiotic processes (termed chemodenitrification) in acid soils especially those high in organic matter content, it is mainly produced by the biological processes of nitrification and denitrification [*Williams et al.*, 1992], which depend on physical, biological and chemical properties of the soil, e.g., soil porosity, soil water content, temperature and the nutrient status [*Ganzeveld et al.*, 2002]. *Firestone and Davidson* [1989] proposed a relatively simple conceptual model, called ''hole-in-the-pipe'', to describe the biogenic emissions of NO and N_2O in soils. According to this model, their emissions from soils were controlled mainly at two levels. The first level is the magnitude of flow through the pipe, i.e., rates of nitrification or denitrification. The second level is the size of the hole, i.e., soil properties such as soil water content, which determine how much of the produced nitrogen oxides can leak out to the atmosphere from soils.

[4] Although soils are a major source of atmospheric NO_x, there still exist huge uncertainties and disagreement about the source strength. The existing inventory models of soil biogenic NO emission showed a large range from <5 Tg N yr^{-1} to >20 Tg N yr^{-1} [*Yienger and Levy*, 1995; *Potter et al.*, 1996; *Davidson and Kingerlee*, 1997; *Ganzeveld et al.*, 2002]. If the soil source strength falls into the lower part of the range, soils might be the third largest source of atmospheric NO_x [*Delmas et al.*, 1997]. Nevertheless, if soils emitted 21 Tg N yr^{-1} of NO, then soil source would be similar in magnitude to fossil fuel emissions of NO_x [*Davidson and Kingerlee*, 1997]. The probable causes of this huge uncertainty include large variations of soil NO emissions either temporally [e.g., *Garcia-Montiel et al.*,

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Table 1. Characteristics of the Broadleaf Forest and Pine Forest (Means ± S.E.)

	Pine Forest	Broadleaf Forest
Biomass,[a] Mg C ha^{-1}	40.6	147.8
SOC,[a] Mg C ha^{-1}	105.2	164.1
Fine root biomass in topsoil,[a] Mg C ha^{-1}	1.9 ± 1.1	4.9 ± 3.0
Litter input,[a] Mg C ha^{-1} yr^{-1}	1.8	4.2
Microbial biomass,[b] μg C g^{-1} soil	551 ± 127	763 ± 73
Microbial amount,[b] 10^6 g^{-1} dry soil	1.17	2.09
pH[b]	4.3 ± 0.3	3.7 ± 0.2
Bulk density,[b] g cm^{-3}	1.50 ± 1.15	0.91 ± 0.12
Total C,[c] mg g^{-1}	22.7 ± 3.1	32.1 ± 2.7
Total N,[c] mg g^{-1}	1.3 ± 0.1	2.5 ± 0.2
C/N[c]	17.01 ± 1.35	12.84 ± 2.27
Available P,[c] mg kg^{-1}	3.59 ± 0.28	4.96 ± 0.16
Leaf area index (LAI)[d]	3.4 ± 0.6	6.1 ± 0.2
NH_4-N,[e] μg N. g^{-1} dry soil	3.1 ± 0.36[W]	2.9 ± 0.20[W]
	3.2 ± 0.49[D]	4.0 ± 0.57[D]
	2.3 ± 0.41[M]	3.0 ± 0.59[M]
NO_3^--N,[e] μg N. g^{-1} dry soil	2.6 ± 0.19 [W]	6.1 ± 0.42 [W]
	5.9 ± 1.34 [D]	11.5 ± 2.22 [D]
	6.4 ± 4.10 [M]	14.1 ± 6.45 [M]
Net mineralization rate,[e] μg N. g^{-1} dry soil day^{-1}	0.5 ± 0.06 [W]	1.6 ± 0.12 [W]
Net nitrification rate,[e] μg N. g^{-1} dry soil day^{-1}	0.4 ± 0.04 [W]	1.4 ± 0.11 [W]

[a] *Tang et al.* [2006]. Fine root in topsoil refers to root (diameter less than 6 mm) biomass in 0–20 cm depth of soil.
[b] *Yi et al.* [2007].
[c] *Mo et al.* [2006].
[d] LAI was measured at a height of about 80 cm above forest floor. (These data were provided by Yuanwen Kuang from South China Botanical Garden, the Chinese Academy of Sciences.)
[e] This study. W, D, and M represent wet season, dry season, and March, respectively.

2001; *Butterbach-Bahl et al.*, 2002] or spatially (as was clearly demonstrated by *Davidson and Kingerlee* [1997]), and the uncertain importance of the canopy in absorbing NO_x from soil sources [*IPCC*, 2001].

[5] Considering the large spatial variation of NO emissions, previous studies are imbalanced in regions under investigation. As to the forests, for example, most studies about NO fluxes were carried out in Europe, USA, tropical America and Africa [*Willams et al.*, 1992, and references therein; *Davidson and Kingerlee*, 1997, and references therein; *Holtgrieve et al.*, 2005], as well as in Australia [*Butterbach-Bahl et al.*, 2004]. Only one report on soil NO emission from Asian forests, to our knowledge, is available to date [*Purbopuspito et al.*, 2006]. Considering the rather diverse forest types in Asia, more studies are undoubtedly needed.

[6] In this study, we measured NO emissions from two forests in different successional stages in Dinghushan Biosphere Reserve in south China. Objectives were to determine (1) seasonal patterns of NO emissions in the two forests, (2) controls on NO emissions, and (3) annual NO emission rates from these subtropical humid forests.

2. Materials and Methods

2.1. Site Description

[7] The experiment was carried out in a broadleaf forest (BF) in climax successional stage and a pine forest (PF) in primary successional stage in Dinghushan Biosphere Reserve (23°09′21″–23°11′30″N, 112°30′39″–112°33′41″E). The reserve is located in the subtropical humid forest life zone with a monsoon climate. Annual mean relative humidity is about 80%. The averaged annual rainfall is about 1927 mm with a distinct seasonal pattern. Typically the period from April to September is wet season, and that from October to March is dry season. March and October are transition periods from dry season to wet season and from wet season to dry season, respectively. Annual mean air temperature is about 21°C, with monthly means the lowest in January (13°C) and the highest in July (28°C).

[8] The broadleaf forest, about 250–300 m above sea level, has been protected without direct human interference for more than 400 years. The pine forest, about 50–200 m above sea level, has been evolved under human disturbance, like collecting litters, since pine trees were first planted in 1930s. The two forests vary not only in their stages of succession, but also in their species composition. The major species in the broadleaf forest are *Castanopsis chinensis*, *Schima superba*, *Cryptocarya chinensis*, *C. concinna*, *Machilus chinensis* in the tree layer and *Hemigramma decurrens* in the understory layer. Tree heights range from 4 to 30 m and diameters from 5 to 163 cm. The pine forest is dominated by *P. massoniana* with densities of 100–1000 trees ha^{-1}, diameters of 4–32 cm and heights of 3–11 m. Understory species included grasses, ferns, vines and shrubs for a total of 43 species [*Mo et al.*, 2006].

[9] The soil in the two forests is lateritic red earth formed from sandstone, but the soil depths vary in each site. Some important characteristics of the two forests were presented in Table 1.

[10] Since the reserve is in the north edge of the Pearl River Delta, one of the most densely populated and industrialized regions in China, annual total N input from atmosphere is relatively high; N deposition through rainfall alone reached 38.4 kg N ha^{-1} yr^{-1} in 1998–1999 [*Zhou and Yan*, 2001].

2.2. NO Flux Measurement

[11] At the beginning of the experiment, three 1 m × 1 m plots (the aims to enclose three plots in each forests were to fix the sampling areas and to protect them from human disturbance) in each forest were chosen and marked. Distance between plots was about 10 m. Field measurements were conducted in the three plots in March (1), April (1), May (1), June (2), July (6 for broadleaf forest and 4 for pine forest), August (4 for broadleaf forest and 7 for pine forest), September (2) and December (3) in 2005 (numbers in the parentheses denoted sampling days within that month). At each sampling date, flux measurements were conducted from 0800 to 1800, during which 3–6 fluxes per plot were obtained at each forest.

[12] NO fluxes were measured by a dynamic flow-through chamber technique, which was similar to that used by *Pilegaard et al.* [1999]. Briefly, the chambers were made of stainless steel (inner walls coated with Teflon films), each covering an area of 30 cm × 30 cm with a total volume of 9 L. Each chamber has one inlet port, one exhaust port and one outlet port for sampling. Inside each chamber, a thermosensor was fixed to measure air temperature, and a fan attached to ensure sufficient mixing of air within the

Table 2. Monthly Averaged NO Fluxes, Percent Water Filled Pore Space, and Soil Temperature at 5 cm Depth[a]

	Broadleaf Forest				Pine Forest			
	NO Fluxes	% WFPS	T5	N	NO Fluxes	% WFPS	T5	N
Mar	45.0 ± 12.2	52.0 ± 0.7	17.0 ± 0.1	9	26.7 ± 8.6	43.5 ± 1.6	20.9 ± 0.7	9
Apr	8.5 ± 0.5	60.2 ± 0.9	24.4 ± 0.4	15	17.2 ± 1.1	39.8 ± 1.7	26.7 ± 0.4	15
May	15.7 ± 1.1	51.0 ± 1.3	25.9 ± 0.2	36	20.5 ± 0.9	40.3 ± 1.9	26.7 ± 0.2	15
Jun	20.7 ± 0.5	45.1 ± 1.1	25.1 ± 0.3	15	18.6 ± 0.7	58.5 ± 1.2	28.0 ± 0.3	33
Jul	17.8 ± 0.6	39.8 ± 1.5	27.1 ± 0.1	75	11.9 ± 0.8	17.6 ± 1.2	29.9 ± 0.3	48
Aug	13.2 ± 0.7	52.7 ± 1.3	27.3 ± 0.2	36	18.2 ± 0.6	36.9 ± 1.9	29.6 ± 0.2	63
Sep	13.3 ± 0.9	56.1 ± 1.1	27.4 ± 0.2	18	16.4 ± 0.5	44.6 ± 2.0	28.8 ± 0.3	18
Dec	23.8 ± 1.2	26.5 ± 0.6	18.8 ± 0.4	36	7.9 ± 0.5	9.3 ± 0.5	21.9 ± 0.3	36

[a]NO fluxes are in ng N m^{-2} s^{-1}. % WFPS, percent water filled pore space. Soil temperature at 5 cm depth, T_5, is in °C. Values in the table are means ± standard errors, and N denotes the number of NO flux measurements.

chamber. During field measurement, the sampling chambers were put onto the forest floor without soil frames to avoid disturbing the soils. Instead they were sealed against the outward atmosphere with Teflon foil pinned down by sand bags [*Gut et al.*, 2002]. An additional reference chamber, closed at the bottom with Teflon sheet, was employed for in situ quantification of chemical reactions and chamber wall deposition effects [*Kirkman et al.*, 2002]. Ambient air was pumped into the chambers at a rate of 4 L min^{-1} through 10 m long Teflon tubes with inner diameters of 4.8 mm, and the sample air was taken in through tubes of the same dimension. The residence time of air in the chambers was about 2.25 min. After about 15 min (over 5 cycles of residence time) when a steady state was reached inside the chambers, NO was analyzed by a model 42C chemiluminescence NO-NO2-NOx analyzer (Zero noise 25 ppt and detection limit 50 ppt, Thermo Electron Corporation, USA). By the contrast of sampling chambers and the reference chamber, net fluxes from the soils could be obtained [*Pilegaard et al.*, 1999].

[13] No corrections were made for the possible removal of NO by chemical reactions taking place in the chambers or by absorption on the chamber wall due to reasons also described by *Pilegaard et al.* [1999]. First, measurements with zero-air and ambient air showed no difference in the calculated fluxes, probably due to low concentrations of O_3 at the forest floor; In addition, very small changes in the concentrations of NO_2 were observed, and on average there was a deposition of NO_2 to the soil, and the NO_2 flux was found to be independent of the NO emission, which indicated that the NO + O_3 reaction did not play a significant role in the chambers.

2.3. Soil Parameter Measurement

[14] When measuring NO fluxes, soil temperatures at 5 cm depths, and volumetric soil water content at 0–5 cm depth were also determined. Soil temperature was measured with soil temperature probes (TES, Ltd., China). Volumetric soil water content was measured with MPA-160 Moisture Probe Meter (ICT international, Australia). In this paper, the volumetric water content was converted to percent water filled pore space (%WFPS) on the basis of averaged soil bulk density in each forest [*Davidson*, 1993].

[15] At selected sampling days, soil samples (0–5 cm) were collected with a 2.5 cm diameter soil corer at each plot for determination of KCl-extractable NH_4^+-N and NO_3^--N (plus NO_2^--N) [*National Standard Bureau of China*, 1987], net rates of N mineralization and nitrification [*Piccolo et al.*, 1994]. In each forest inorganic N pools were analyzed for 11 samples, and net rates of N mineralization and nitrification were analyzed only for 9 samples.

2.4. Statistics and Data Analysis

[16] The statistical software package SPSS 10.0 (SPSS Inc. USA) was used for ANOVA testing (post-hoc LSD analysis) of NO fluxes of different months, and for independent-samples t testing between NO fluxes in the two forests, or between NO fluxes in dry season and wet seasons within a forest. The statistical software package SigmaPlot 9.0 (SPSS Inc. USA) was used for regression analysis.

3. Results and Discussion

3.1. Measured NO Fluxes and Their Temporal Patterns

[17] Measured NO fluxes were listed in Table 2. As mentioned above, in this region the period from April to September was wet season and that from October to March was dry season, among which March and October were transition period between wet and dry seasons. In the broadleaf forest, mean flux in wet season (14.9 ng N m^{-2} s^{-1}) was lower than that in dry season (23.8 ng N m^{-2} s^{-1}) ($p < 0.05$); in the pine forest, however, mean flux in wet season (17.1 ng N m^{-2} s^{-1}) was higher than that in dry season (7.9 ng N m^{-2} s^{-1}) ($p < 0.05$). In wet season, soil NO emissions from the two forests had no significant difference, while in dry season NO fluxes in the broadleaf forest were significant higher than those in the pine forest ($p < 0.05$).

[18] The seasonal pattern of NO emissions from the broadleaf forest (Figure 1) was similar to those observed in some tropical rain forests [*Kaplan et al.*, 1988; *Bakwin et al.*, 1990; *Keller and Reiners*, 1994; *Verchot et al.*, 1999; *Garcia-Montiel et al.*, 2001]. In a primary rain forest in eastern Amazonia, *Verchot et al.* [1999] measured mean net NO flux of 3.3 ± 0.4 ng N m^{-2} s^{-1} in the wet season and 5.9 ± 0.7 ng N m^{-2} s^{-1} in dry season. In a humid tropical forest, *Garcia-Montiel et al.* [2001] observed mean NO fluxes of 1.4 and 9.2 ng N m^{-2} s^{-1} in wet and dry season of 1998 and 1999, respectively. However, opposite seasonal pattern was also observed in some rain forests [*Serca et al.*, 1994, 1998] and seasonally dry tropical forest [*Davidson et al.*, 1991], just similar to the case in the pine forest (Figure 1) of this study.

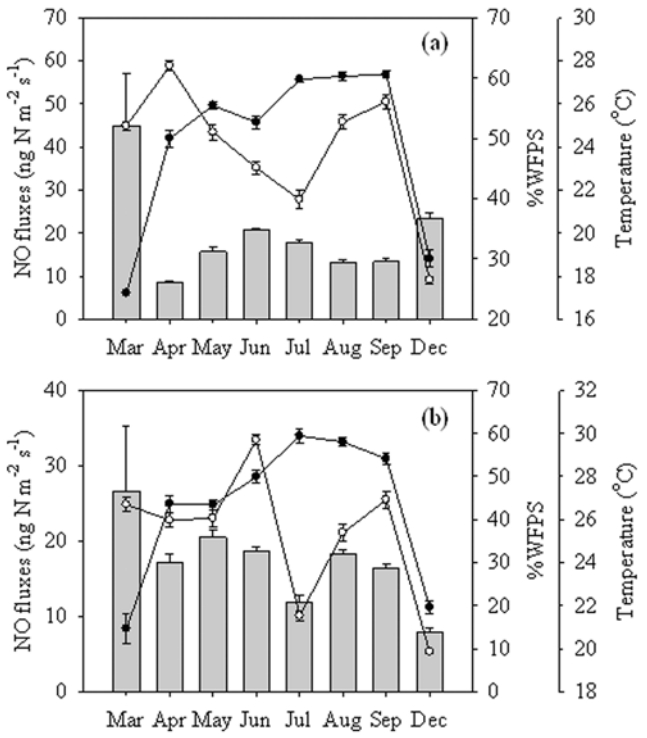

Figure 1. Monthly averaged NO fluxes (vertical bars), percent water filled pore space (open circles), soil temperature at 5 cm depth (solid circles) for (a) the broadleaf forest and (b) the pine forest. Each value is the mean of fluxes from the three plots within a month, and error bars represent standard errors.

[19] Relatively higher NO fluxes observed in March (Figure 1) in the present study should be largely caused by "pulsing" emission, which was characteristic for the transition period from dry season to wet season. Just the day before the sampling day there was a 33 mm rainfall after a long time of dryness. These NO emission pulses had been also observed previously in other forests [*Bakwin et al.*, 1990; *Davidson et al.*, 1991; *Davidson*, 1993; *Meixner et al.*, 1997; *Garcia-Montiel et al.*, 2003; *Butterbach-Bahl et al.*, 2004], and they were probably resulted from a quick use of the accumulated inorganic N during the prolonged soil dryness by soil microbes when wetting soils [*Garcia-Montiel et al.*, 2003].

3.2. Soil Moisture and NO Fluxes

[20] Averaged soil %WFPS in the broadleaf forest was greater than in the pine forest either in wet season or in dry season (Table 2). In wet season, monthly averaged %WFPS varied from 39 to 60% with a mean value of 51% in the broadleaf forest, and from 17 to 59% with a mean value of 40% in the pine forest. In dry season, the averaged %WFPS in the broadleaf forest and the pine forest lowered to 26.5% and 9.3%, respectively.

[21] When excluding the fluxes measured in March (they were probably pulses as discussed above), NO fluxes were found to be significantly correlated with %WFPS in a quadratic manner in both forests (Figure 2). For the broadleaf forest $r^2 = 0.48$, $p < 0.001$, $n = 230$; and for the pine forest, $r^2 = 0.56$, $p < 0.001$, $n = 228$). The fitted regression equations for the two forests were as follows:

$$\text{For BF} \qquad F_{NO} = -1.55 \times 10^{-2} \times W^2 + 1.05 \times W + 4.08 \quad (1)$$

$$\text{For PF} \qquad F_{NO} = -6.66 \times 10^{-3} \times W^2 + 0.670 \times W + 2.82 \quad (2)$$

where F_{NO} is the soil NO emission flux (positive value means flux direction from soil to atmosphere), and W is the %WFPS. According to the above equations, there existed optimum soil moistures for NO emissions in the two forests, and they were calculated to be about 34% WFPS for the broadleaf forest and 50% WFPS for the pine forest.

[22] Due to the strong correlation between NO fluxes and soil %WFPS, NO fluxes can be predicted on the basis of the above equations using available moisture data. The predicted NO fluxes in comparison with those field measurements in the two forests were presented in Figure 3. We can see that large deviations only occurred in March due to pulse emissions.

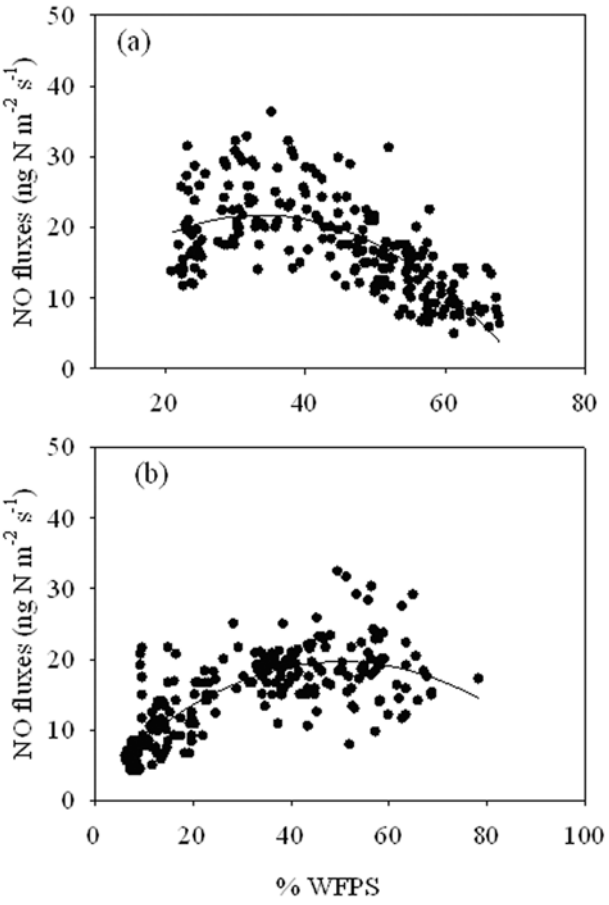

Figure 2. Correlation of soil NO emission fluxes and %WFPS based on field measurements in (a) the broadleaf forest ($r^2 = 0.48$, $p < 0.001$, $n = 231$) and (b) the pine forest ($r^2 = 0.56$, $p < 0.001$, $n = 228$). NO fluxes in March were excluded as they were pulses.

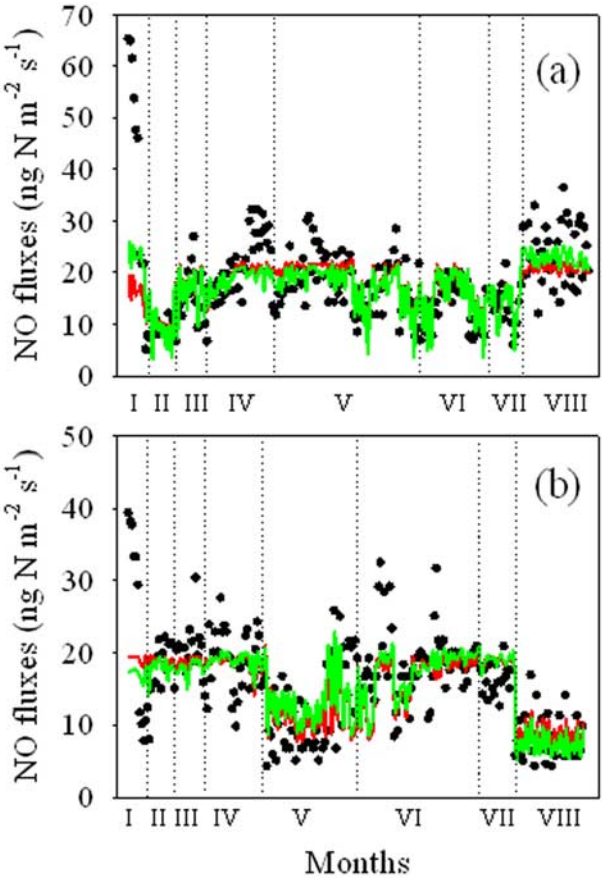

Figure 3. Observed NO fluxes (solid circles) versus predicted NO fluxes by soil moisture alone (red lines) or by both soil moisture and temperature (green lines) for (a) the broadleaf forest and (b) the pine forest. I, March; II, April; III, May; IV, June; V, July; VI, August; VII, September; VIII, December.

[23] Soil moisture regulated NO production through its role in the stimulation of microbial activity, in the delivery of electron donors ($NH4^+$, dissolved organic carbon) and acceptors (O_2, NO_3^-), and in the diffusion of NO from soils [*Firestone and Davidson*, 1989; *Stark and Firestone*, 1995; *Holtgrieve et al.*, 2005]. Soil moisture, especially %WFPS, as an important regulating factor on soil NO emissions has been observed in quite a few studies [*Davidson et al.*, 1991; *Keller and Reiners*, 1994; *Yang and Meixner*, 1997; *Otter et al.*, 1999; *Verchot et al.*, 1999; *Butterbach-Bahl et al.*, 2004], although the effects of increased soil moisture on soil NO emissions were diversified in these studies. However, there were studies, in which no clear relationship between soil moisture and NO emissions was observed [*Ludwig et al.*, 2001].

[24] Many studies also showed that optimum soil moisture existed for soil NO emissions [*Yang and Meixner*, 1997; *Otter et al.*, 1999; *Verchot et al.*, 1999], but the value varied according to soils. For example, a laboratory study showed the optimum soil moisture of about 20% WFPS by *Yang and Meixner* [1997], but in another study conducted in a seasonally dry forest of eastern Amazon, maximum NO fluxes was observed at approximately 50% WFPS [*Verchot et al.*, 1999]. In our study, optimum soil moisture for the two forests significantly varied, which was probably due to their different soil properties. The different seasonal patterns of NO emissions in the broadleaf forest and pine forest of this study were also largely to be resulted from their different responses to the seasonal variations of soil water contents.

[25] Due to its higher content of soil organic matter and clay, the soil in the broadleaf forest had a better water holding capacity (Table 1), which is defined as the ability of a soil to retain water against the pull of gravity and is positively correlated with content of soil organic matter and clay [*Khaleel et al.*, 1981; *Zhang and Zhuo*, 1985]; on the contrary, water is easier to be drained out of the soil due to its coarse particles in the pine forest. For this reason and other properties like much more developed root systems in the broadleaf forest (Table 1), during the dry season, the broadleaf forest still kept a considerably higher soil water contents, but water content in the pine forest become so low that microbial activity was reduced for restricted substrate supply. As pores within solid matrices drain and water films coating surfaces become thinner, diffusion path lengths become more tortuous, and the rate of substrate diffusion to microbial cells declines [*Stark and Firestone*, 1995]. Therefore the soil NO emissions would be influenced much more by the dryness in the pine forest than in the broadleaf forest. During wet season, for the broadleaf forest, frequent rains would cause a much higher percent of water-filled pore space, so NO emissions decreased due to the reduced NO diffusibility [*Ludwig et al.*, 2001], and also due to the increasing consumption of NO by denitrifying bacteria under more anoxic conditions [*Krämer and Conrad*, 1991; *Ye et al.*, 1994]. For the pine forest soil, however, increased soil water content in wet season would benefit the microbial activity and thus increased the production of NO; and due to its poorer water holding capacity %WFPS would not reach a high level at which NO diffusion would impeded as it did in the broadleaf forest. NO emissions from the pine forest floor were thus greatly raised during the wet season while those from the broadleaf forest decreased. So the impacts of water content on soil NO emission depend on the balance between its role in NO biogenic production and its role in NO diffusion from soil to the atmosphere.

3.3. Soil Temperature and NO Fluxes

[26] Monthly mean soil temperature at 5 cm depth ranged from 17 to 28 °C in the broadleaf forest and from 20 to 30°C in the pine forest, respectively (Table 2). Exponential relationship existed between NO fluxes and soil temperature at 5 cm depth in the pine forest ($\boldsymbol{F_{NO}} = 3.48\exp^{0.05T}$, $r^2 = 0.19$, $p < 0.001$, $n = 228$), but no significant relationship was found between NO fluxes and soil temperature in the broadleaf forest.

[27] Many previous studies observed that soil temperature was an important regulator on soil NO fluxes [*Williams and Fehsenfeld*, 1991; *Otter et al.*, 1999; *Gut et al.*, 2002]. However, other studies found no obvious effects of soil temperature on NO fluxes [*Cardenas et al.*, 1993; *Pilegaard*, 2001]. Considering the dominance of soil microbial processes for the production of NO, the dependence of NO emissions on soil temperature is reasonable, since enzymatic

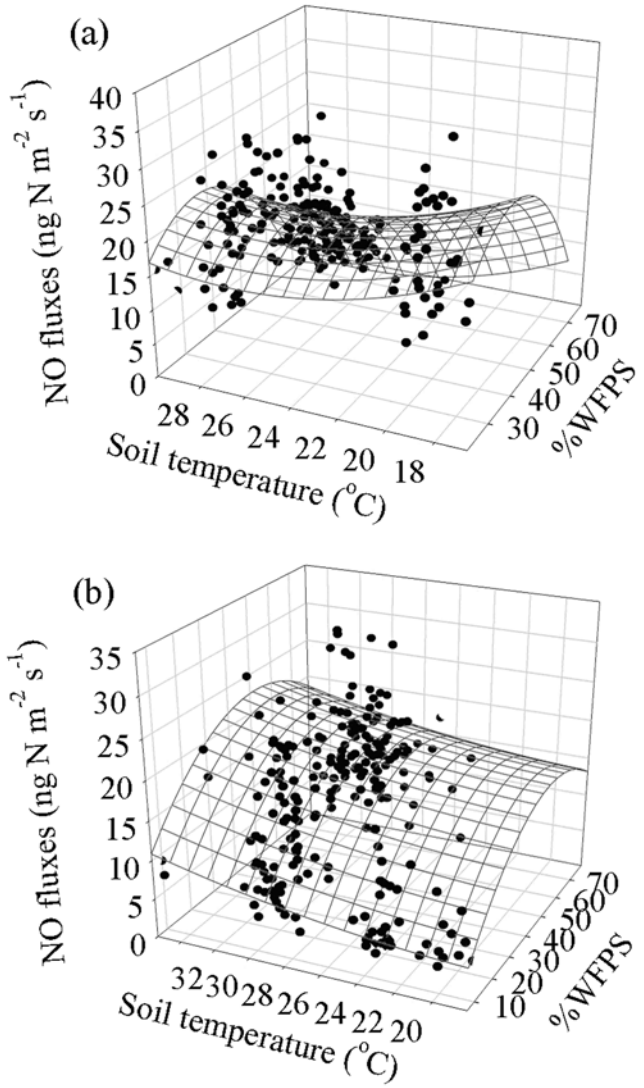

Figure 4. NO emission as a function of soil moisture (% WFPS) and temperature (°C) in (a) the broadleaf forest (r^2 = 0.52, $p < 0.001$, n = 231) and (b) the pine forest (r^2 = 0.58, $p < 0.001$, n = 228).

processes generally increase exponentially with temperature within a certain range, as long as other factors (substrate or moisture availability) are not limiting [*Ludwig et al.*, 2001]. The lack of correlation between soil temperature and NO fluxes in the broadleaf forest was probably because the effect of soil water content greatly overrode that of soil temperature.

3.4. Combined Effects of Soil Moisture and Temperature

[28] As biogenic NO production and emission were affected by soil moisture, temperature and many other factors, fitting NO emission with these factors within an acceptable range of confidence would benefit prediction of NO emission. Soil moisture and temperature are mostly available among soil parameters; in the present study they are combined to see their relations to the NO emissions. Using measurements in the two forests, NO emission fluxes were found to be significantly related to soil %WFPS and temperature in a quadratic way (for the broadleaf forest, r^2 = 0.52, $p < 0.001$, n = 231; for the pine forest, r^2 = 0.58, $p < 0.001$, n = 228):

$$\text{For BF} \quad F_{NO} = 51.9 + 1.41W - 4.40T - 1.90 \times 10^{-2}W^2 + 8.41 \times 10^{-2}T^2 \tag{3}$$

$$\text{For PF} \quad F_{NO} = 4.62 + 0.585W - 0.390T - 5.63 \times 10^{-3}W^2 + 1.33 \times 10^{-2}T^2 \tag{4}$$

where $\boldsymbol{W}$ is the % WFPS and $\boldsymbol{T}$ is the soil temperature (°C) at 5 cm depth. These relations are also shown in Figure 4. Predicted NO fluxes based on equations (3) and (4) in comparison with those measured in the field are presented in Figure 3. In the two forests, incorporating $\boldsymbol{T}$ into the formulation of fluxes does not significantly improve the prediction of fluxes, implying the dominant influence of soil moisture.

3.5. Soil N Availability and NO Emission

[29] Throughout the year, total soil inorganic N pools, especially NO_3^--N pools, were greater in the broadleaf forest than in the pine forest (Table 1). NO_3^--N pools in both forests were greater in dry season than in wet season, implying accumulation of NO_3^--N during dry season. For example, the NO_3^--N pools in the broadleaf forest during the dry season were nearly 2 times those during the wet season (Table 1).

[30] In the broadleaf forest NO fluxes were significantly correlated with NO_3^--N pools (r^2 = 0.73, $p < 0.001$, n = 11), net rates of N mineralization (r^2 = 0.75, $p < 0.01$, n = 9) and nitrification (r^2 = 0.67, $p < 0.01$, n = 9). However, no significant relationships were found between NO fluxes and NH_4^+-N pools in the broadleaf forest, or between NO fluxes and indices of N availability in the pine forest.

[31] Previous studies also found strong correlation between soil NO emissions and indices of N availability in temperate forests [*Regina et al.*, 1998; *Stark et al.*, 2002] and tropical forests [*Davidson et al.*, 1991; *Verchot et al.*, 1999; *Garcia-Montiel et al.*, 2001]. The existence of strong correlation between NO emission and soil N availability may be explained by the fact that available N serves as substrate for nitrifying and denitrifying bacteria, which are responsible for soil NO production [*Ludwig et al.*, 2001]. *Stark et al.* [2002] concluded that net rates, rather than gross rates of N cycling might be better predictors of soils NO fluxes. But other studies failed to find any significant relationships between soil NO fluxes and indices of N availability [*Keller and Reiners*, 1994; *Veldkamp et al.*, 1999]. The lack of clear correlation between soil NO fluxes and N availability in the pine forest in the present study also confirmed that N availability might do a poor job of predicting the fluxes of individual NO or N_2O, although it was probably a good predictor for the combined N oxide emissions [*Verchot et al.*, 1999].

3.6. Preliminary Estimates of Annual NO Emissions

[32] Many previous studies estimated annual NO emission from a specific site by extrapolating the mean fluxes

Table 3. Reported Annual NO Emissions From Forests of Different Regions

Location	Forest	Annual Emission, kg N ha^{-1} yr^{-1}	Reference
		Tropical Forests	
Brazil	moist tropical forest	1.7	*Nepstad et al.* [2002]
Brazil	primary forest	1.5	*Verchot et al.* [1999]
	secondary forest	0.3	
Brazil	humid tropical forest	2.4	*Garcia-Montiel et al.* [2003]
Puerto Rico	subtropical wet forest	0–0.6	*Erickson et al.* [2001]
Puerto Rico	older forest	0.5–0.8	*Erickson et al.* [2002]
	younger forest	2.5–11.9	
Costa Rica	old growth forest	0.9	*Keller and Reiners* [1994]
	secondary forest	0.4	
Mexico	dry tropical forest	0.5–1.0	*Davidson et al.* [1991]
Congo	equatorial rain-forest	0.7	*Serca et al.* [1994]
West Africa	Gallery forest	0.5	*Le Roux et al.* [1995]
China	Broadleaf forest	6.1–6.9	this study
	Pine forest	4.0–4.4	
		Temperate Forests	
USA	Deciduous forest	0.05	*Williams and Fehsenfeld* [1991]
USA	Deciduous forest	0.2	*Williams et al.* [1988]
Sweden	Pine forest	0.04	*Johansson* [1984]
France	Pine forest	0	*Jambert et al.* [1994]
		Temperate N Affected Forest	
Germany	Spruce forest	6.4–9.1	*Butterbach-Bahl et al.* [2002]
	Beech forest	2.3–3.5	
USA	Hardwood forest (Low N)	0.16	*Venterea et al.* [2003]
	Hardwood forest (High N)	4.7	
	Pine forest (Low N)	4.8	
	Pine forest (High N)	5.9	
USA	Deciduous forest	1.9	*Valente and Thornton* [1993]

measured in specific time intervals to a whole year [*Verchot et al.*, 1999; *Erickson et al.*, 2001, 2002; *Butterbach-Bahl et al.*, 2002; *Venterea et al.*, 2003; *Purbopuspito et al.*, 2006]. Some of these studies had time resolutions from several measurements within one year [e.g., *Erickson et al.*, 2002] to once per month [e.g., *Erickson et al.*, 2002; *Venterea et al.*, 2003; *Purbopuspito et al.*, 2006]. Nevertheless, improved time resolution of field measurements would better the flux estimates. In the present study, annual totals were calculated by stratifying the year into wet season (April-September) and dry season (October-March) and multiplying the mean flux for the season by the number of days in the respective season [*Verchot et al.*, 1999; *Garcia-Montiel et al.*, 2003]. It turned out that annual NO emission was 6.1 kg N ha^{-1} yr^{-1} in the broadleaf forest and 4.0 kg N ha^{-1} yr^{-1} in the pine forest.

[33] Empirical formulation was believed to be helpful in the annual flux estimating. *Williams et al.* [1992] derived an empirical formula using soil temperature as the only predictor variable, and used it to estimate the soil NO emission of the USA. *Kirkman et al.* [2001] developed a formula considering soil moisture effects by a first-order empirical function involving several parameters obtained through laboratory analysis to upscale soil NO emission in Zimbabwe. *Verchot et al.* [1999] also established an empirical formula using nitrification potential and %WFPS as variables. In the present study, since daily mean soil moisture and temperature data were available throughout the year, based on the strong correlation as shown in equations (1) to (4), we can roughly predicted NO fluxes each day by soil moisture or by soil moisture and temperature together. According to the computed results, if only soil moisture was considered, annual NO emission was calculated to be 6.1 kg N ha^{-1} yr^{-1} in the broadleaf forest and 4.4 kg N ha^{-1} yr^{-1} in the pine forest; if both soil moisture and temperature were considered as equations (3) and (4), annual NO emission was calculated to be 6.9 kg N ha^{-1} yr^{-1} in the broadleaf forest and 4.3 kg N ha^{-1} yr^{-1} in the pine forest. We can see that in the two forests, especially in the pine forest, taking soil temperature into consideration would not significantly change the annual NO flux estimation. Also we can see that these predictions agree very well with those calculated on the base of observed data (6.1 kg N m^{-2} s^{-1} in the broadleaf forest and 4.0 kg N m^{-2} s^{-1} in the pine forest), suggesting that soil moisture and temperature, especially soil moisture, were good predictors for soil NO emission in the studied sites.

[34] Annual NO emissions in this study were relatively higher than those from most tropical forests, which usually emit less than 5 kg N ha^{-1} yr^{-1} (Table 3) [see also *Davidson and Kingerlee*, 1997]. However, remarkably large NO emissions, with a total amounted to about 3 kg N ha^{-1} during a three-month period, were observed at a rain forest in Queensland, Australia [*Butterbach-Bahl et al.*, 2004]. The largest annual NO emission (11.9 kg N ha^{-1} yr^{-1}) from tropical forest was reported by *Erickson et al.* [2002]. Compared to annual NO emissions from temperate forests, where annual NO emissions were usually lower than 0.2 kg N ha^{-1} yr^{-1} (Table 3) [see also *Davidson and Kingerlee*, 1997], NO emissions in the subtropical humid forests from our studies were much higher.

[35] However, annual NO emissions in the present study were in line with those from temperate forests affected by high N deposition (Table 3). For example, NO emissions as high as 6.4–9.1 kg N ha^{-1} yr^{-1} was reported in a spruce forest experiencing nitrogen inputs of 30 kg N ha^{-1} yr^{-1} [*Gasche and Papen*, 1999; *Butterbach-Bahl et al.*, 2002]. N deposition as an important controller of soil annual NO emissions was clearly indicated by *Pilegaard et al.* [2006], who measured soil NO emissions at 15 European forest sites and found that significant positive correlations existed between N inputs and NO emissions. In this study substantially high annual NO emissions measured in the broadleaf forest and the pine forest were also likely due to the high N deposition in this region.

[36] For the difference of annual NO emissions between the broadleaf forest and the pine forest, there were several possible causes besides soil moisture. N status was better in the broadleaf forest than in the pine forest (Table 1) largely because the pine forest was under human disturbance, like collecting litters, since it was first planted in 1930s, but the broadleaf forest has been protected without direct human interferences for more than 400 years. Another reason is that N inputs in throughfall to the broadleaf forest floor were higher than that to the pine forest floor due to the higher leaf area index (Table 1), which resulted in better filtering capacity of the air [*Pilegaard et al.*, 2006]. Litter quality and structure probably also played a role in the difference of annual NO emissions between these two forests, since litter quality and structure were found to affect soil emissions of NO [*Erickson et al.*, 2002], N_2O [*Brumme et al.*, 1999] and other trace gases like carbonyl sulfide [*Yi et al.*, 2007].

4. Conclusions

[37] Due to the dense population, rapid industrialization and intensified agricultural activities, some regions in Asia are hot spots of airborne nitrogen oxides and also areas with increasing nitrogen deposition, therefore the cycling of nitrogen gases in Asia might be of increasing importance in both regional and global scale for atmospheric chemistry and global budgets of nitrogen. Yet, to date, data about NO emissions from forest soils are quite limited in Asia. In the present study, measurements of soil NO emissions were conducted in two subtropical humid forests in south China. The investigated forests included a broadleaf forest in climax successional stage and a pine forest in primary successional stage. Annual total soil NO emission was estimated to be 6.1–6.9 kg N ha^{-1} yr^{-1} in the broadleaf forest and 4.0–4.4 kg N ha^{-1} yr^{-1} in the pine forest with three upscaling methods. These values were relatively higher than those reported in most tropical forests according to the previous studies, and were in line with those from temperate forests affected by high N deposition.

[38] Soil water content was found to be a very important factor controlling the seasonal patterns of soil NO emissions from the two forests, but its influence varied with forest types due to different soil properties. In the broadleaf forest, mean NO emission in wet season (14.9 ng N m^{-2} s^{-1}) was lower than in dry season (23.8 ng N m^{-2} s^{-1}). In the pine forest, however, mean NO emission in wet season (17.1 ng N m^{-2} s^{-1}) was higher than in dry season (7.9 ng N m^{-2} s^{-1}).

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