Emission of N_2O from nitrogen-saturated, sub-tropical forest ecosystems in south China

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Abstract
Surplus reactive nitrogen (Nr) is one of China's most pressing environmental problems, and may cause a positive feedback to climate change through increased emission of N_2O, an important greenhouse gas. Forests in sub-tropical south China receive high atmogenic N loads (4g $N/m^2/yr$), primarily as ammonium (NH_4^+). Having limited ability to retain Nr, these forests may become N-saturated. Here we present mass balance data for the TieShanPing (TSP) forested catchment, near Chongqing, P.R. China; a site with acidic alisols and monsoonal climate (wet, hot summers). Data confirm N-saturation and show that much of the atmogenic N load is nitrified and leached as nitrate (NO_3^-) from the root zone. Yet, only a fraction of the NO_3^- flux leached from the soil is detected in catchment runoff, suggesting that denitrification may be a major sink of excess N. Initial data are presented showing elevated N_2O emission rates during the monsoon with rates varying between about 100 µg N_2O-$N/m^2/hr$ in soils with permanently high groundwater table to > 500 µg N_2O-$N/m^2/hr$ in soils with variable groundwater table at the foot of hill slopes.

Key Words
N_2O, nitrogen deposition, denitrification, *Pinus massoniana*, haplic alisol, monsoon.

Introduction
Population growth, intensification in agriculture and burning of fossil fuels lead to ever increasing emissions of reactive nitrogen (Nr) in the biosphere (Galloway *et al.* 2008). In China, fertiliser N input alone has increased from 0.54 Tg in 1961 to 28 Tg in 2005 (Xiong *et al.* 2008), giving rise to substantial emissions of ammonia-nitrogen (NHy). Together with NOx from combustion of fossil fuel this leads to a high atmospheric N load resulting in increasing N-deposition rates, particularly in forest, one of the dominant land-cover types in sub-tropical south China. Surplus of reactive nitrogen (Nr) is one of China's most pressing environmental problems, and may cause a positive feedback to climate change through increased emission of N_2O, an important greenhouse gas (Schlesinger, 2009). Recently, we found that forests in sub-tropical China receiving high atmogenic N loads (> 4g $N/m^2/yr$) have a limited ability to retain Nr and that forests may become N-saturated. Earlier mass balance studies suggest that denitrification may be a major sink for excess N (Chen & Mulder, 2007). Rapid N-cycling, in systems with acidic alisols and monsoonal climate (wet, hot summers) imply potentially high N_2O emissions from multiple sources. N_2O is produced both during the microbial processes of nitrification and denitrification and is a potent greenhouse gas (Firestone and Davidson, 1989). To date, no systematic assessment of N_2O emissions on a watershed level has been conducted. Here, we present annual mass balances of nitrogen TieShanPing forest (Chongqing, China; Larssen *et al.* 2006) from 2001 to 2004. Nitrogen mass balances were assessed both at the plot scale and integrated at the catchment level. In addition, initial data will be presented on N_2O emission rates in different landscape positions in relation to the availability of Nr and hydrological conditions. The overall goal is to estimate N_2O emission rates in humid sub-tropical south China and to understand the environmental controls at a watershed level. Ultimately, these data are needed to facilitate upscaling approaches for N_2O emissions and to predict the effect of changing hydrological conditions on N_2O emissions at a regional scale.

Methods
The study site
TieShanPing (TSP), 25 km northeast of Chongqing, is a 16.3 ha protected forest catchment, typical for southwest Chinese subtropical forest. The climate is monsoonal, with relatively dry winters and wet summers. Situated at 450 – 500 m asl, soil temperatures reach about 24°C in early summer, whereas in winter values decrease to about 9°C. Annual precipitation is about 1200 mm, and most precipitation occurs in the wet season between April and September. Only in the last part of the summer, when precipitation decreases, soil water potentials reach strongly negative values, indicating dry soils. By contrast, spring, early

summer and winter are characterised by moist to wet soils, as indicated by soil water potentials close to zero (Chen 2006). The vegetation is dominated by Masson pine mixed with broad-leave sub-tropical forest and a well developed understory of evergreens. Typical for the region, the forest soils on the hill slopes are well-drained acidic Haplic Alisols (pH_{H2O} 3.5-3.8 in the O/A horizon; values increasing to about 4.5 in the B horizon) with a large nitrification potential; however, at the foot of the slopes in the groundwater discharge zone, the soils have developed aquic properties, so that they may act as important zones for denitrification. Soils have high clay contents; Low hydraulic conductivities may increase the residence time of NO_3^- on its way to the streams, possibly exposing it to denitrification conditions in hypoxic sub-soils. During intensive precipitation events, runoff from surface soil horizons may give rise to direct input of surface soil water and NO_3^- (but not NH_4^+) into the stream. Soils on the hill slopes have very thin litter layers and A horizons with low organic matter contents (Chen 2006).

Field and laboratory investigations
Atmogenic input of Nr is calculated as wet-only deposition and throughfall deposition. Ammonium (NH_4^+) and nitrate (NO_3^-) are analysed as described in ISO14911 and ISO10304-1, using ion chromatography. Occasional ring-tests indicate that the internal quality of the analyses is satisfactory.
Wet-only deposition, collected using a standardized wet-only collector (Dept. of Meteorology, Stockholm University, Sweden), is measured and sampled for chemical analysis at weekly intervals. Throughfall deposition is collected below the canopy of the ground vegetation at four plots in the catchment, using four collectors per plot. Throughfall collectors, made of plastic funnels, are located at permanent positions in the plots. At weekly intervals, the collected throughfall in all four collectors is pooled and weighed. Chemical analysis is done at monthly intervals in pooled samples. Pooled samples are stored in the refrigerator and filtered (0.45 μm membrane filter) prior to analysis.
Leaching of Nr is calculated at the plot-scale, based on concentrations in soil water and at the catchment-scale using concentrations in streamwater. Soil water is collected at monthly intervals below the root zone (at about 50 cm soil depth) using ceramic suction cups (P80). The suction applied is approximately 50 kPa. Streamwater discharge is recorded continuously using a V-notch weir, stage sensor and data-logger and streamwater samples are collected weekly for chemical analysis. Weekly and monthly fluxes of Nr in streamwater and soil water, respectively, are calculated using recorded water fluxes in the stream, thus assuming that soil water flux at 50 depth equals the flux measured as catchment runoff.
Emission of N_2O at a large number of landscape positions in the catchment is assessed using the closed chamber method. Concentrations of N_2O are determined in gas samples taken at fixed time intervals during a 30-minute period. Hot spots (selected for further N_2O monitoring) are characterised with respect to landscape position (hydrological setting, vegetation). Measured N_2O emissions at the plot-scale are combined with measuring soil physical conditions, biological process rates and N_2O product stoichiometries. For the period 2010 - 2012, we plan to study N_2O emission rates in response to the addition of ^{15}N labelled NH_4^- and NO_3^-. Supplementary studies will include determination of natural abundance (15N, 18O) and laboratory incubation experiments.

Results
The mass balance for inorganic nitrogen at TSP is shown in Figure 1. Median values of the annual flux of Nr in wet-only and throughfall deposition are about 1.2 keq/ha/yr (1.7 g/m^2/yr) and 2.5 keq/ha/yr (3.5 gm^2/yr), respectively. In both cases NH_4^+-N deposition contributes about 70% to total N. This illustrates the importance of agricultural sources for atmogenic N in this part of China. The doubling of the Nr flux in throughfall compared with the flux in wet-only deposition suggests that dry deposition is equally important as wet-only deposition. Possibly, the Nr flux in throughfall is underestimated, due to direct N uptake in the canopy of the trees and ground vegetation, but we have no data to quantify this.
The median annual flux of NH_4^+-N in soil water and streamwater is close to zero. All NH_4^+ is effectively converted, mostly to NO_3^-, in the upper few centimetres of the mineral soil. The median flux of NO_3^--N in soil water at 50 cm depth is about 2.9 keq/ha/yr (4 g/m^2/yr), which indicates that virtually all atmogenic N is leached from the root zone and little net retention occurs at TSP. This is in line with the small rates of growth of Masson pine at TSP (Wang YH, pers. comm.). The annual flux of NO_3^--N in streamwater is about 0.5 keq/ha/yr (0.7 g/m^2/yr). Thus, only 20% of the annual atmogenic input of Nr leaves the catchment with streamwater. Since the trees and ground vegetation on the well-drained soils on the hillslopes do not seem to be a major sink of reactive N at TSP, it is likely that the groundwater discharge zones in the catchment, converting groundwater to stream, constitute an important sink, probably through denitrification.

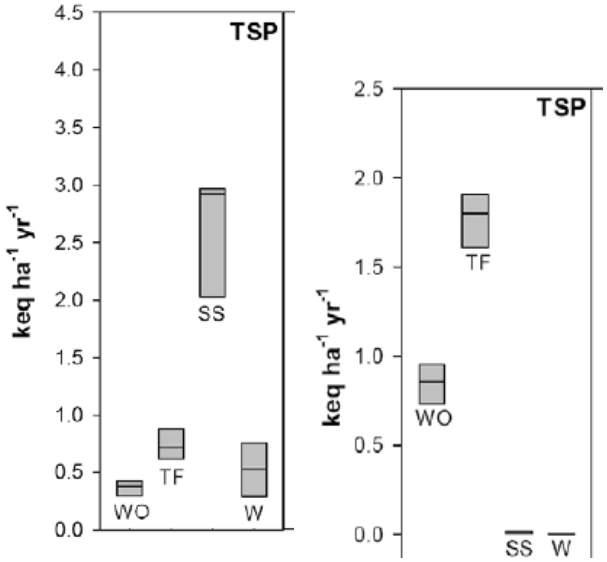

Figure 1. Medians, 10-percentiles and 90-percentiles of annual fluxes of inorganic nitrogen at TSP (2001-2004). Left panel is NO_3^-, right panel NH_4^+. Values are in keq/ha/yr. Abbreviations indicate NO_3^- and NH_4^+ fluxes in wet-only deposition (WO), throughfall (TF), soil water below the root zone (SS) and streamwater (W). The nitrogen flux in soil water refers to values at 50 cm soil depth, which were estimated, multiplying the monthly water flux, measured as streamwater discharge, with the nitrogen concentration in soil water.

The data in Figure 1 suggests that both nitrification and denitrification are important processes in the TSP catchment. N_2O emission may result from both processes and its importance is confirmed by initial measurements for July and August 2009, which indicate elevated N_2O emission rates throughout the catchment. Values, although variable in space and time, are particularly high (> 500 µg N_2O-N/m^2/hr) at the foot of the hill slopes, where the groundwater table is most variable. Thus, these positions may constitute important hot spots for N_2O emission in the TSP catchment. In the groundwater discharge zone, where the soils are permanently close to saturation, the N_2O emission rates are about 100 µg N_2O-N/m^2/hr. Similar values are found at the well drained soils on the hill slope. Currently, additional data on N_2O emissons, and N_2O concentrations in the soil atmosphere are collected also during the drier and cooler part of the year. N_2O emission data will be related to measurements of groundwater table (piezometers) and soil water contents (TDR).

Conclusion

As many other forest ecosystems in south China, the TieShanPing catchment, near Chongqing receives large amounts of atmogenic reactive nitrogen (3.5 g/m^2/yr), in particular as NH_4^+. Complete nitrification and little retention in the forest results in large nitrate fluxes from the root zone. Denitrification is the major sink of reactive N in the TieShanPing catchment, responsible for the removal of about 80% of the annual input. The TieShanPing catchment shows elevated emission rates of N_2O during the monsoon, most notably at the interface of the foot of the hill slopes. and the groundwater discharge zone. The project will continue until the end of 2012.

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