REPORTS – RAPPORTS

Estimation of N₂O and NO emission from a tropical highland forest in Rwanda[1]

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The research of this work fits within the broad subject of greenhouse gas emissions and climate change. More specifically, this work focuses on the contribution of tropical forest soils to the atmospheric N_2O and NO budgets. Nitrous oxide has a global warming potential of 310 relative to CO_2 and is one of the main greenhouse gases covered by the United Nations Framework Convention on Climate Change (UNFCCC). Nitrous oxide also plays a key role in modulating the stratospheric ozone layer. Tropical forest soils are considered to be the largest natural source of N_2O. Additionally, they can produce considerable amounts of NO. Estimates of the contribution of tropical rainforest ecosystems, which cover 1150×10^6 ha of the global land surface, to atmospheric N_2O and NO have a high uncertainty. Despite the fact that tropical forest soils are considered to be a key source of N_2O and NO, only a relatively small number of detailed studies investigating the temporal and spatial variability of the N_2O and NO soil-atmosphere exchange are available, particularly for Africa. Therefore the general objective of this research was to improve N2O and NO emission predictions from tropical forest. The research has been carried out in the Nyungwe forest in Rwanda for which a large number of legacy data are available. The results of this thesis led to (i) a better understanding of the N_2O and NO source strength of a central African tropical forest soil, (ii) improved NO and N_2O emission estimates from a central African tropical highland forest and more insight into the importance of soil properties controlling N_2O and NO emissions and (iii) a better insight in those parameters that preferentially should be monitored to allow better global simulations of N_2O and NO emissions from tropical forests.

Key words: ForestDNDC-tropica, nitrous oxide, nitric oxide, sensitivity analysis, tropical forest, Nyungwe, Rwanda

Introduction

Nitrous oxide (N_2O) is a powerful greenhouse gas (GHG) contributing to climate change. It is also a major stratospheric ozone depleting substance. Nitric oxide (NO) is a so-called indirect greenhouse as it contributes to the formation of tropospheric ozone

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(O_3), which is a greenhouse gas. Further, NO contributes to acidifying deposition (Meehl et al., 2007; Delmas et al., 1997). Worldwide, soils have been identified as a major source of N_2O and NO and tropical forest soils count for the largest part of the natural terrestrial N_2O and NO source (Matson et al., 1990; Bouwman et al., 1993; Mosier et al., 1998; Breuer et al., 2000; Kiese et al., 2002,2003; Butterbach-Bahl et al., 2004; Werner et al., 2007b). In contrast to the importance of tropical forest ecosystems as one the major sources of N_2O and NO, spatial and temporal estimates of N_2O and NO emission are still limited and highly uncertain. To overcome this problem the ForestDNDC-tropica model coupled to Geographic Information Systems (GIS) is considered a promising tool for the calcula-tion of N_2O and NO emission on a regional, national and even global scale. The model is able to simulate processes in tropical forest soils involved in N-trace gas production and emission and their dependency on changing soil properties, vegetation and climate conditions (Werner et al. 2007b). However, detailed information on spatial variation of tropical soil properties (e.g. sand, silt and clay fraction, gravel, soil organic carbon, total nitrogen, mineral soil pH, and bulk density) and vegetation, as well as daily climate data are required for the calculation of N_2O and NO emission on a regional scale. However, this type of information is largely lacking for tropical forests. Moreover, respective long-term field based N_2O and NO validation datasets at this scale are virtually absent. The latter two constraints are especially true for Africa.

To better characterize spatial and temporal patterns of greenhouse gas emissions from tropical forests, the biogeochemical model ForestDNDC-tropica has been developed, calibrated and tested and applied at various scales (site, regional, national and global). Even though ForestDNDC-tropica has been significantly tested, model output has still a high de-gree of uncertainty. However, reducing of uncertainty strongly depends on the availability of more spatially explicit data to parameterize the model. However, acquiring detailed spatial information on soil, climate and vegetation data for tropical forests, especially for remote areas, is challenging.

This research aimed to fill severe knowledge gaps on spatial variability of N_2O and NO emissions and their control factors for tropical forest soils and to derive estimates of regional fluxes of N_2O and NO as well as a sensitivity analysis for input parameters to run the ForestDNDC-tropica model.

Experimental work

N_2O and NO emissions from Nyungwe forest soil were investigated in Chapter 4 us-ing a laboratory incubation technique (Gharahi Ghehi et al. (2012a)). Through laboratory experiments determining N_2O and NO emissions from 31 soil samples from different soil types and climate conditions in the Nyungwe forest, we found a strong dependency of N_2O and NO fluxes on soil moisture. For all sites, significant positive correlations be-tween N_2O and soil moisture were found. In contrast to N_2O emissions, NO emissions decreased with increasing soil moisture, with significant differences between the soil moisture levels.

The average N_2O emission from the Nyungwe forest (ffiranging from 46.3 to 98.8 µg N m^{-2} h^{-1}) was in line with measured N_2O emissions from the Mayombe forest soils in the Democratic Republic of Congo (mean flux in the rainy season: 19.6 µg N m^{-2} h^{-1} and mean flux at the end of the rainy season: 207 µg N m^{-2} h^{-1}) by Serca *et al.* (1994).

The N_2O emission rates we measured during laboratory incubations ranged from 4.5 to 400.5 µg N m^{-2}h^{-1} and were comparable in magnitude with N2O fluxes from field measurements at the Kakamega forest sites in Kenya (1.1–324.8 µg N m^{-2} h^{-1}; Werner *et al.*, 2007b). In addition, our measured individual N2O fluxes up to 400 µg N m^{-2}h^{-1} were similar in magnitude as the reported maximum N2O fluxes of 492.1 µg N m^{-2}h^{-1} and 570.8 µg N m^{-2}h^{-1} for Australian rain forest soils following periods of intensive rainfalls (Breuer *et al.*, 2000; Kiese and Butterbach-Bahl, 2002). Compared to N_2O emission data for tropical forest ecosystems in Australia, the observed spatial variability of N_2O emission in the Nyungwe forest was high. Kiese and Butterbach-Bahl (2002) and Breuer *et al.* (2000) reported coefficients of variation for the N_2O emission rate in tropical forest ecosystems in Australia, between 36.5-60.2% and 16.6-67.6%, respectively. The coefficient of variation for N_2O emission rates at the Nyungwe forest between different sampling sites was 89.9-133.3% and this was in good agreement with results from Werner *et al.* (2007b), who reported values of 52.8-147.9% for the spatial variation of N_2O fluxes for the Kakamega forest in Kenya.

With mean NO emission rates ranging from 36.1 to 69.3 µg N m^{-2}h^{-1} the Nyungwe soil emitted higher levels of NO as reported by Serca *et al.* (1994) (14.4 µg N m^{-2}h^{-1} at the end of a rainy season – mean flux rainy season 4.1 µg N m^{-2}h^{-1}) for the Mayombe forest in DR Congo.

Compared to other NO emission data for other tropical forest ecosystems our observed maximum NO emission rates (up to 265.5) were higher than previously reported maximum NO emissions of 60.2 µg N m^{-2}h^{-1} (Bakwin *et al.*, 1990) in the Amazon, 16.9 µg N m^{-2}h^{-1} for the Amazon tropical forest site at Biológica do Jaru, Brazil (Gut *et al.*, 2002). However, the observed NO emission rates for the Nyungwe forest were lower than reported fluxes (mean NO emission = 207.1 µg N m^{-2}h^{-1}, range: 0.1-773.8 µg N m^{-2}h^{-1}) for rain forests in Queensland, Australia at the onset of the rainy season (Butterbach-Bahl *et al.*, 2004).

The differences in N_2O and NO emitted from different locations could be explained by differences in clay content, organic carbon, total nitrogen, soil pH and iron of the topsoil. Furthermore, our study revealed a significantly higher N_2O emission level at higher altitude sites, where micaschists dominate the soilscape, areas with a high free iron content (iron > 3%) and low soil pH (pH < 4). High NO emissions were distributed among sampling sites with high organic carbon and total nitrogen content. Our result indicates that the factor controlling N_2O and NO emission varies spatially.

In addition, tropical forest soils are often characterized by low soil pH, high crystalline reactive Fe and fluctuating redox conditions which are likely to support chemo-denitrification (auto-decomposition of nitrites) for NO and N_2O production in addition to microbial processes (Van Cleemput and Baert, 1984). In general, high Fe concentrations and soil pH below 4 providing such conditions were found frequently for the Nyungwe.

We performed a preliminary experiment to determine whether chemo-denitrification could be of importance in the Nyungwe (Chapter 4, supplementary material). We performed a laboratory experiment, where ^{15}N was added as NO_2^- to mercury-treated soils, and ^{15}N recoveries were determined as well as N_2O and NO emissions. Evidence from our preliminary experiment and data from Chapters 4 and 5 suggest that abiotic processes might be an important production pathway for NO and less for N_2O. However, we do not have evidence for the contribution of the abiotically produced NO to microbially produced N_2O from our experiment.

Modeling approach

In the scope of this study, the process-based ForestDNDC-tropica model, which simulates process involved in N-trace gas production and emission in tropical forest soils on a daily scale, was used to predict N_2O and NO emissions from Nyungwe forest soil (Chapter 5). Before ForestDNDC-tropica was applied for the calculation of regional N_2O and NO emissions, several improvements for model data input were performed.

In order to predict N_2O and NO emission from Nyungwe forest, soil data (Verdoodt and Van Ranst, 2006a, 2006b) (texture, organic carbon and total nitrogen, mineral soil pH and bulk density), vegetation parameters (aboveground biomass, wood lead and floor mass) and climate data (daily precipitation, minimum and maximum temperature) were integrated into a GIS database covering the Nyungwe forest, with a spatial resolution of 0.025° x 0.025°.

In Chapter 2 the spatial distribution of soil properties of the Nyungwe forest have been derived. A comparison between two interpolation methods showed that both methods gave similar results in terms of accuracy. Thus, IDW was used for preparing the spatial distribution of soil input parameters for grid cells where no observational data was available. Since soil bulk density, an important parameter, was largely missing in the original survey, bulk density was derived using specific pedo-transfer functions (PTF) for tropical highland forest soils (Gharahi Ghehi et al., 2012b; Chapter 3).

In Chapter 3 we developed BRT and k-NN estimation techniques that provide an estimation of bulk density for tropical forest soils using the available soil data. Models that used soil particle size distribution, pH, CEC and organic carbon content as input and limiting factors, such as land-use type (forest and non-forest) and master horizon notation were tested. Adding particle size distribution, pH and CEC as input variables did not enhance the predictive capacity significantly compared to a model that used OC content as the sole input. The model solely based on organic carbon resulted in an error of 0.17 to 0.19 g cm^{-3} for bulk density estimation, which was reasonable and comparable with those reported Benites et al. (2007) (0.19–0.28 g cm^{-3} for Brazilian soils), Tranter et al.

(2007) (0.153–0.195 g cm^{-3} foı Australian soils), Jalabert *et al.* (2010) (0.13–0.21 g cm^{-3} for French forest soils) and Nemes *et al.* (2010) (0.17 g cm^{-3} for soils from United States). So we suggested that using organic carbon as a single predictor is parsimonious and easy to use for predicting topsoil bulk density tropical forest soils. Furthermore, the k-NN and BRT techniques showed a comparable performance and predicted bulk density equally for an independent data set compared to multiple linear regression developed via tropical nonforest general dataset. However, BRT and k-NN methods have several advantages that make them more flexible statistical learning methods.

All above 0.025° x 0.025° resolution data sets were finally converted to a ForestD-NDC-tropica soil, climate and vegetation input file format. Linking this overall raster GIS database covering Nyungwe forest with ForestDNDC-tropica resulted in regional estimates of N$_2$O and NO emissions (Chapter 5).

Based on the above approach the annual (mean of the 13 year period 1981-1993) N$_2$O and NO emissions from the Nyungwe forest soil was on average 3.8 ± 0.52 kg N$_2$O-N ha^{-1} yr^{-1} (2.8-5.5 kg N ha^{-1} yr^{-1}) and 2.0 ± 0.87 kg NO-N ha^{-1} yr^{-1} (0.8-5.1 kg N ha^{-1} yr^{-1}). The average model estimates in our study were supported by field measurements from other tropical rain forest ecosystems in Australia, southwest China and West Africa (Kiese *et al.*, 2005; Werner *et al.*, 2007a). There was also agreement between our laboratory N$_2$O and NO measurements (Chapter 4) and the model predictions though the ForestDNDC-tropica resulted in considerable lower emissions for few sites. Low similarity between the simulated and laboratory measured N$_2$O and NO emissions was specifically found for acidic soil with high clay content and reduced metals, indicating that model improvements are needed to increase the reliability of simulated N$_2$O and NO emission for such soil conditions.

The small area investigated (1113 km2) was estimated to emit ca. 439 ± 50 t N$_2$O-N yr^{-1} and 244 ± 16 t NO-N yr^{-1}. These estimates underpin the potential importance of tropical rainforest soils as a source of atmospheric N$_2$O and NO on a regional, national and even global scale.

The spatial variation in N$_2$O and NO emissions mainly resulted from differences in the spatial distribution of physicochemical soil properties and indicated that the clay content, pH and predominantly soil organic carbon are the major drivers for N$_2$O and NO emissions. Moreover, the observed two-fold increase of total annual NO emission for 2007-2008 years (384-489 t NO-N yr^{-1}) compared to 1981-1993 years (244 ± 16 t NO-N yr^{-1}) also indicates that climate variability plays a major role for soil N dynamics.

The sensitivity analyses demonstrated that the ForestDNDC-tropica model was most sensitive to change in bulk density and pH With model validation with measured N-trace gases fluxes from chapter 4 and also with results from our preliminary experiment (supplemental material, chapter 4) we argue that abiotic denitrification can be an important source of N$_2$O and especially NO under certain circumstances, i.e. low pH and presence of reduced iron.

Although uncertainties remain in our model results, we are confident that process-based models in combination with spatially explicated non-clustered site-specific input

data are very useful, especially for those attributes that ForestDNDC-tropica are most sensitive to (i.e. soil-pH, bulk density, organic carbon and rainfall can be used to improve (global) estimates of N_2O and NO emission from tropical forests and give us an accurate insight in the spatial and temporal variability in N_2O and NO dynamics).

Limitations and perspectives for further research

The ForestDNDC-tropica simulations for the Nyungwe forest as well as simulations for other tropical forests rely on historical soil and climate and simulated vegetation data. Here soil properties have been obtained at least three decades ago and these may have changed in time (e.g. pH, organic carbon). Thus, these uncertainties affect the overall uncertainty of N_2O and NO inventories. On the other hand other soil properties are more stable and change little over time (e.g. clay content), however, these effects were not (and could not) be taken into account in the simulations.

In this study, changes in clay, organic carbon, total nitrogen and pH values over time have been shown by selecting 29 locations out of the 147 sites in the Nyungwe forest, for which a soil sampling was performed during September 2009 (Chapter 5). However, since the recent sampling has not been taken place at the exact same locations as for the historical sites in the Nyungwe forest (due to global positioning system "GPS" error <~20 m) data were not fully comparable.

The parameters that changed most (~985 to 2009) were total nitrogen, organic carbon and pH. The probable reasons for these changes in the Nyungwe forest (Wallin G, personal communication) could be the following:

(i) forest fires in the eastern part of the forest in 1999, (ii) traditionally there have been more disturbances in the eastern part than in the western part of the forest, although locally some mining and logging activities have been done in the west and (iii) disturbance caused by people normally has occurred where the forest is accessible, thus likely in the same areas where the most extensive soil sampling has been carried out.

As a result, changes over time of the ForestDNDC-tropica input parameters might be a crucial factor for prediction uncertainty and will be an obvious source of bias.

Historical data are, however, the best data, which are available and obviously the uncertainty in the estimations will increase with older data. Therefore, in addition to high-resolution soil data that represent the spatial variability and distribution of soil properties, monitoring of soil changes is recommended and the establishment of a soil monitoring network across the tropical forest on regional scale is needed. However, on a regional scale, data monitoring via field sampling and direct measurement are not feasible (especially not for remote areas). Therefore, to be able to obtain the multi-resolution of spatial and temporal patterns of the soil, vegetation and climate data with a high degree of accuracy for tropical forest ecosystems, the following tools and projects might be helpful:

– *Climate data (daily minimum and maximum temperature and precipitation)*
    Using European Center for Medium-range Weather Forecasts
    (ECMWF http://data-portal.ecmwf.int) is recommended.

- Soil *texture and OC (as direct parameter and indirect parameter to generate DB data) and pH, tool.*
  Near-infrared(NIR)and mid-infrared(MIR)reflectancespectroscopymayallowrapid and cost effective acquisition of soil data (African Soil Information Service (AfSIS): http://www.africasoils.net).

- *Vegetation data (to assess aboveground biomass)*
  LPJ-GUESS Ecosystem model (http://www.nateko.lu.se/lpj-guess/lpj_guess_ main.html) is recommended. Taking time series of climate data and, given constraints of latitude, topography, and soil characteristics, as input, LPJ-GUESS dynamically computes spatially explicit transient vegetation composition. So, obviously the uncertainty in the prediction of the vegetation parameters using LPJ-GUESS will decrease with recent input data.

In addition to LPJ-GUESS, high throughput analyses based on infrared (IR) spectroscopy might help in collecting vegetation parameters.

Though non-biological reaction, usually taking place at low pH if inorganic electron donors (e.g. Fe (II)/Fe (III)) are present in high concentrations, may play a significant role in N2O and NO production in soils, no spatial information is available for initializing ForestDNDC-tropica, which is also likely incomplete with regard to the description of chemo-denitrification and Feammox. Further studies that examine mechanisms and the importance of abiotic N_2O and NO production are needed to improve NO and N_2O estimates from tropical forests.

To achieve a narrowing of the uncertainty, in addition to the availability of detailed information on soil, vegetation and climate data, the ForestDNDC-tropica model requires future development to encompass all biotic (e.g. dissimilatory nitrate reduction to ammonium) and abiotic (e.g. chemo-denitrification, Feammox) sources of N_2O and NO emission. Thus, more effort should be put into the research of these pathways in highland tropical rain forest soils.

However, reducing the uncertainty in quantifying fluxes of N_2O and NO from tropical forests from site to regional scale requires a close cooperation of experimentalist and modellers. Further progress crucially depends on the availability of more comprehensive and long-term field measurements of N-gas fluxes for model performance validation as well as a detailed evaluation of controlling variables. Detailed datasets are one of the main requirements to guide further improvement and validation of ForestDNDC-tropica. The collection of comprehensive datasets can commonly not be tackled by normal research projects and thus clearly points towards the importance and need of an international monitoring program such as the CARBOAFRICA project (http://www.carboafrica.net) or the NITROEUROPE project in Europe. CARBOAFRICA has been established in Sub-Saharan Africa and it aimed to quantify and understand carbon cycling, and emission of other greenhouse gases. The project involved 11 developing countries with different ecosystem types, representative for Africa's biodiversity.

Again, a recent comprehensive database of African tropical soil parameters using the AfSIS project is essential for reducing the uncertainty in ForestDNDC-tropica results. Furthermore, we urge that current research and policy programs (UN-REDD) for tropical forest regions which, mainly focus on carbon (CO_2) budgeting include N_2O (and NO as an indirect greenhouse gas via tropospheric ozone formation) fluxes in an effort to cut back on greenhouse gas emissions.

Acknowledgments

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