Importance of soil NO emissions for the total atmospheric NO$_x$ budget of Saxony, Germany

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HIGHLIGHTS

- LandscapeDNDC reproduced soil NO emissions across various ecosystem types.
- On average soils are significant (8%, uncertainty 6–13%) source of tropospheric NO$_x$ in Saxony.
- Soil NO emissions can dominate tropospheric NO$_x$ budgets in rural areas.

ARTICLE INFO

ABSTRACT

Soils are a significant source for the secondary greenhouse gas NO and assumed to be a significant source of tropospheric NO$_x$ in rural areas. Here we tested the LandscapeDNDC model for its capability to simulate magnitudes and dynamics of soil NO emissions for 22 sites differing in land use (arable, grassland and forest) and edaphic as well as climatic conditions. Overall, LandscapeDNDC simulated mean soil NO emissions agreed well with observations ($r^2 = 0.82$). However, simulated day to day variations of NO did only agree weakly with high temporal resolution measurements, though agreement between simulations and measurements significantly increased if data were aggregated to weekly, monthly and seasonal time scales. The model reproduced NO emissions from high and low emitting sites, and responded to fertilization (mineral and organic) events with pulse emissions. After evaluation, we linked the LandscapeDNDC model to a GIS database holding spatially explicit data on climate, land use, soil and management to quantify the contribution of soil biogenic NO emissions to the total NO$_x$ budget for the State of Saxony, Germany. Our calculations show that soils of both agricultural and forest systems are significant sources and contribute to about 8% (uncertainty range: 6–13%) to the total annual tropospheric NO$_x$ budget for Saxony. However, the contributions of soil NO emission to total tropospheric NO$_x$ showed a high spatial variability and in some rural regions such as the Ore Mts., simulated soil NO emissions were by far more important than anthropogenic sources.

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1. Introduction

Nitric oxide (NO) is a short-lived chemically active gas and a key component in photochemistry, thereby significantly contributing to the formation of tropospheric ozone (O_3). The most important sources of NO_x are fossil fuel combustion (electricity and transport), biomass burning and emissions from soils of terrestrial ecosystems (Denman et al., 2007). Estimates on the relative contribution of soil biogenic NO emissions to the total tropospheric NO_x budget range from 4 to 40% (Denman et al., 2007). Butterbach-Bahl et al. (2009) hypothesized that soils from agricultural, natural and semi-natural ecosystems might dominate as sources of tropospheric NO_x in rural areas during spring and summer periods, but this still remains to be proven. Several studies (e.g. Davidson and Kingerlee (1997); Steinkamp and Lawrence (2011); Hudman et al. (2012)) reported global soil NO emissions ranging from 13 to 27 Tg NO-N a^{-1}, which are substantially higher than the estimates by IPCC (8.9 Tg NO-N; Denman et al. (2007)). However, based on a top-down approach involving tropospheric NO_2 column measurements Vinken et al. (2014) estimated that global NO emissions are approx. 12.9 Tg NO-N in 2005. The obvious discrepancies are due to a) divergences of methodologies (upscaling of a few site measurements vs. upscaling by highly averaged emissions factors or top-down approaches using atmospheric observations) and b) lack of understanding of processes mediating the production and consumption of NO across different terrestrial ecosystem types.

Soil emitted NO results from the net balance of simultaneously occurring microbial-mediated production and consumption processes mainly via nitrification (Laville et al., 2005), denitrification (Betlach and Tiedje, 1981; Conrad, 2002), and the abiotic process of chemodenitrification (Kesik et al., 2005). All of these processes are controlled by environmental conditions such as soil moisture, temperature, nitrogen (N) and carbon (C) availability, porosity and pH and thus are affected by land use history and management practices (Butterbach-Bahl et al., 2009; Kesik et al., 2005; Luo et al., 2012; Medinets et al., 2015; Pilegaard, 2013). Prevailing pathways responsible for soil NO emissions in agricultural and forest ecosystems are supposed to be nitrification and chemodenitrification (Kesik et al., 2005; Medinets et al., 2015). Reported soil NO emissions vary largely across a range of 0.2–23 kg NO-N ha^{-1} a^{-1} (see e.g. reviews by Davidson and Kingerlee (1997); Ludwig et al. (2001); Stehfest and Bouwman (2006)), with agricultural soils being assumed to be stronger sources than soils of semi-natural and natural ecosystems that have been found to emit up to a maximum of 11.4 kg NO-N ha^{-1} a^{-1} (Luo et al., 2012). Soil NO flux measurements at site scale are tedious and require substantial investment in instrumentation, so that the number of available field measurements is still limited (Stehfest and Bouwman, 2006; Medinets et al., 2015). In order to improve existing estimates, process based biogeochemical models capable of simulating soil emissions in high spatial and temporal resolution, have been used at site and regional scales (e.g. Kesik et al. (2005); Butterbach-Bahl et al. (2009)).

At the time of writing, a direct comparison of soil NO emissions with anthropogenic NO_x emissions from energy related processes has not yet been carried out, especially not in high spatial and temporal resolution to determine soil contributions in dependence on land use. Here we establish such an inventory for a region with significant industrial and agricultural emission sources to investigate if soil NO emissions are indeed a significant source for tropospheric NO_x. To do so, we evaluate the biogeochemical LandscapeDNDC model (Haas et al., 2013) for various land uses and a sufficient range of environmental boundary conditions, in order to test the reliability of simulations at local scale. In the following, we calculate a soil NO emission inventory for the state of Saxony, Germany, and compare these results with published NO_x emissions from energy related processes. We hypothesized that in rural areas NO emissions from agricultural and forest soils are stronger sources than energy production related NO_x emissions, while the opposite is found in regions with a mixture of industry and agriculture.

2. Material and methods

2.1. Model description

LandscapeDNDC (Haas et al., 2013) is a simulation framework for ecosystem models including various sub-models for, e.g., plant growth, micrometeorology, water fluxes and biogeochemistry. LandscapeDNDC has been successfully applied for a multitude of ecosystem simulation studies on, e.g., crop growth (Kim et al., 2014, 2015; Molina-Herrera et al., 2016), tree growth (Grote et al., 2011; Molina-Herrera et al., 2015), greenhouse gas emissions (Kim et al., 2014, 2015; Kraus et al., 2015; Molina-Herrera et al., 2016; Zhang et al., 2015) and NO_3 leaching (Dirnböck et al., 2016; Kiese et al., 2011). In this study, the selected sub-model setup included the micrometeorological ECM model (Grote et al., 2009), the DNDC crop model (Li et al., 1992), the PnET-II forest model (Aber et al., 1995), the DNDC hydrology model (Li et al., 2000) and the soil biogeochemical MeTr^x model (Kraus et al., 2015). Implemented processes cover all major soil C-, N-, turnover processes, though the focus of this work is on soil biogenic NO emissions. Hence, here only the main functionalities that are directly involved in the production and consumption of soil NO emissions are further explained.

All NO- production, consumption and transport processes are calculated by the MeTr^x model, e.g., nitrification, denitrification, chemodenitrification as well as gas transport processes within the soil. Nitrification and denitrification are both explicitly simulated via microbial metabolism (Kraus et al., 2015). During the aerobic process of nitrification a varying fraction of nitrified NH_4 (r_{NH_4}) is transformed to NO (r_{NH_4-NO}), depending on modifying factors such as pH (f_{pH}), temperature and moisture (f_{tm}) and the specific rate constant (K_{NO}) for NO reductase:

$$r_{NH_4-NO} = K_{NO}\, r_{NH_4} f_{pH} f_{tm}\, K_{NO} \qquad \text{Equation 1}$$

During anaerobic conditions NO_3^- is stepwise denitrified to N_2 via the intermediate products NO_2^-, NO and N_2O. The MeTr^x model calculates denitrification of each substrate depending on its relative abundance (Kraus et al., 2015). Both, aerobic and anaerobic produced NO_2^- is subject to chemodenitrifcation, which is calculated as abiotic chemical reaction depending on pH (f_{pH}), temperature (f_t), the reaction constant (K_{chemo}) and the NO_2^- content $c_{NO_2^-}$.

$$r_{NO_2^--NO} = K_{chemo} f_{pH} f_t\, c_{NO_2^-} \qquad \text{Equation 2}$$

All biotic and abiotic processes are simulated spatially explicit for defined layers throughout the complete soil profile. The actual amount of emitted NO finally depends on the net balance of production and consumption processes as well as superimposed transport via diffusion and advection into the atmosphere following concentration gradients. Functions used in the computation of NO can be found in the Appendix.

2.2. Local scale model evaluation

Twelve different forest stands, including the most dominant tree species across Europe (Spruce, Beech, Sitka spruce, Silver birch, Douglas fir and Oak), and ten agricultural sites (six arable and four grasslands) with sufficient soil NO flux data, auxiliary information