Human nitrogen fixation and greenhouse gas emissions: a global assessment

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Abstract
The net impact of human nitrogen (N) fixation on climate (ignoring short-lived components) mainly depends on the magnitude of the warming effect of (direct and indirect) nitrous oxide (N_2O) emissions and the cooling effect of N-induced carbon dioxide (CO_2) uptake. N-induced CO_2 uptake is caused by anthropogenic N deposition which increases net primary production (NPP) in N-limited ecosystems and thus CO_2 sequestration. Nitrogen oxide (NO_x) emissions, however, also induce tropospheric ozone (O_3) formation, and elevated O_3 concentrations reduce NPP and thus plant C sequestration. We estimated global-scale impacts of anthropogenic N fixation on net greenhouse gas emissions using recent data and modelling approaches with respect to N inputs to various ecosystems, N_2O emissions in response to N inputs, and C exchange in responses to N inputs (C–N response) and O_3 exposure (C–O_3 response). The estimated impact of human N fixation is dominated by an increase in N_2O emissions equal to 1.02 (0.89–1.15) Pg CO_2-C equivalent (eq) yr^{-1}. CO_2 uptake due to N inputs to terrestrial and aquatic ecosystems corresponds to net emissions of -0.75 (-0.97 to -0.56) Pg CO_2-C_{eq} yr^{-1}, while the reduction in CO_2 uptake by N-induced O_3 exposure corresponds to net emissions of 0.14 (0.07–0.21) Pg CO_2-C_{eq} yr^{-1}. Overall, human N fixation causes an increase in net greenhouse gas emissions of 0.41 (-0.01–0.80) Pg CO_2-C_{eq} yr^{-1}. Even considering all uncertainties, it is likely that N inputs lead to a net increase in greenhouse gas emissions.

Key Words
Nitrogen, nitrous oxide, fixation, ozone, carbon sequestration, greenhouse gases

Introduction

The severe anthropogenic perturbation of the global reactive nitrogen (N_r) cycle affects the Earth's climate through the manifold impacts of N on ecosystem C and N cycles and thus on emissions of greenhouse gases (GHGs), especially nitrous oxide (N_2O) and carbon dioxide (CO_2) (the impact on methane exchange is insignificant by comparison). Figure 1 summarizes three main pathways discussed in this paper through which N_r affects the climate. First, N fertilizer and manure use in agriculture lead to N_2O emissions from agricultural soils (denoted as direct N_2O emission); but also from terrestrial and aquatic systems, following volatilization or leaching of applied N and re-deposition and processing downwind or downstream of the agricultural regions (denoted as indirect N_2O emissions; Figure 1, top panel). Other sources of anthropogenic N_2O emissions include biomass burning, fossil fuel combustion, sewage, and industrial processes. Second, recent research has focussed on the impacts of N_r deposition on CO_2 emissions or uptake in terrestrial and aquatic ecosystems (Figure 1, middle panel). Since many (semi-) natural terrestrial and marine ecosystems are N-limited, increased N deposition (caused by agricultural and industrial NH_3 and NO_x emissions) usually increases productivity and thus C sequestration in those ecosystems. Third, emerging research focuses on the role of NO_x- (and hydrocarbon-) induced tropospheric ozone (O_3) formation. Ozone is damaging to plants and leads to a reduction of NPP, thus reducing C sequestration (Figure 1, bottom panel).

In this paper, we discuss and quantify the impacts of human N_r fixation on terrestrial and aquatic ecosystem N_2O and CO_2 exchange (emissions or uptake), in terms of net emissions expressed in CO_2-C equivalents (CO_2-C_{eq}), focusing on the year 2000. We present global-scale estimates of N_2O emissions, increased C uptake due to N deposition and decreased C uptake due to NO_x-induced O_3 exposure. This is done by multiplying N inputs with N_2O emission factors/functions, ecosystem C–N responses, and ecosystem C–O_3 responses, making use of results from experimental studies, field measurements and modelling approaches. We end with a summarizing overview of ranges in N_2O and CO_2 exchange in response to human N fixation.

1. Direct and indirect N_2O emissions

2. N-induced C sequestration in agricultural, terrestrial and marine systems

3. NO_x-induced O_3 formation reducing terrestrial C sequestration

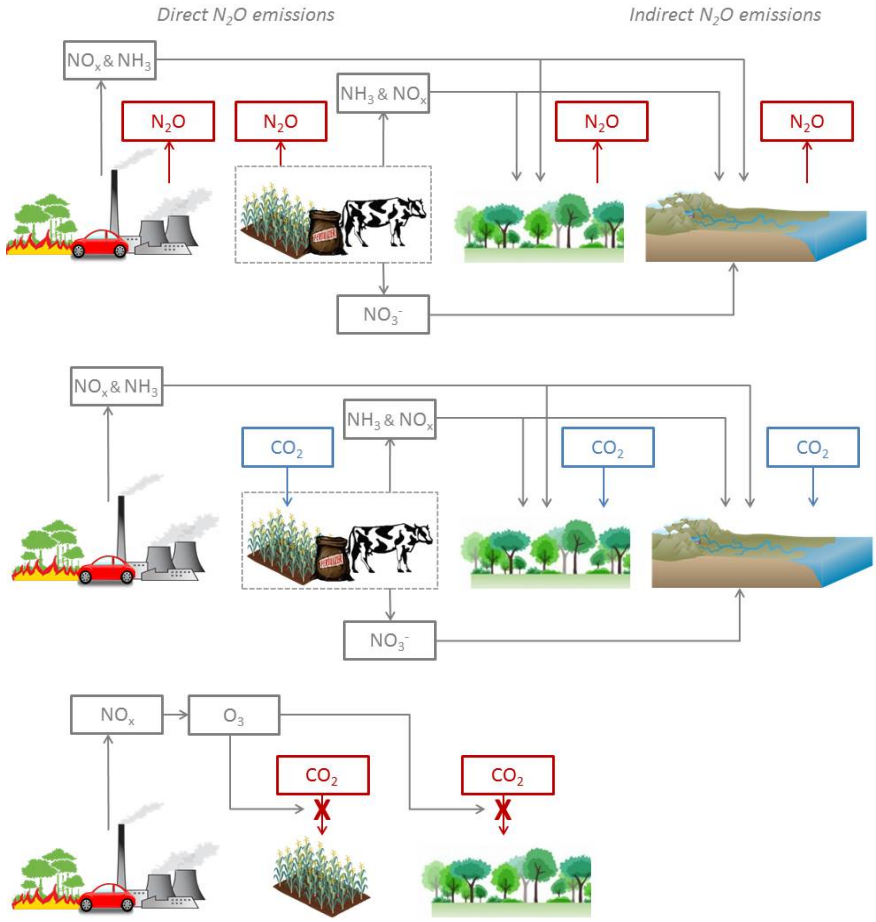

Figure 1: Linkages between anthropogenic N_r and climate discussed in this paper. Red boxes indicate a warming effect, while blue boxes indicate a cooling effect. Depicted processes are described in the text.

Methods

Approaches to quantify large-scale impacts of nitrogen fixation on nitrous oxide emissions

N_2O emission estimates at the global scale are often based on the IPCC tier 1 emission factor approach. This approach assumes that a fixed percentage of N inputs is converted to N_2O (e.g. Syakila & Kroeze, 2011). Emission factors are applied both for direct N inputs (e.g. 1% for N fertilizer inputs or 2% for N excreted on grazing land) as well as indirect N inputs (e.g. 1% for NH_3 emissions or 0.75% for N leached from soils to groundwater and surface water). Davidson (2009) suggested emission factors of 2.8% for N_2O emissions from manure N and 2.2% for N_2O emissions from synthetic fertilizer N application. These values also integrate indirect N_2O emissions. Furthermore, N_2O emissions are estimated using empirical relationships between observed soil N_2O fluxes and environmental parameters such as (i) management (N application rate per fertilizer type, type of crop) and (ii) environmental factors (climate, soil organic C content, soil texture, drainage and soil pH), e.g. based on a global meta-analysis by Stehfest and Bouwman (2006). Finally, process-based models are increasingly used to assess N_2O emissions at regional to global scales.

Approaches to quantify large-scale carbon exchange in response to nitrogen inputs

In order to quantify the impact of global N inputs on CO_2 exchange in agricultural systems, forests, (semi-) natural vegetation and marine systems, we multiplied estimated global anthropogenic N inputs to these systems with ranges in C–N responses (defined as the additional mass unit of C sequestered per additional mass unit of N deposition). C–N responses for agricultural and semi-natural systems were obtained from Liu and Greaver (2009), while C–N response ratios for marine systems were based on Duce et al. (2008). Approaches used to assess C–N responses of forest ecosystems, which are the most important terrestrial C sink, included (i) N retention measurements combined with C:N ratios in forest ecosystem compartments, called stoichiometric scaling, (ii) experimental N addition studies assessing the impacts of N addition on C pools in biomass and soil, (iii) field-based monitoring studies on measured forest growth across N deposition gradients, and (iv) model simulations predicting carbon cycle response to environmental change. In this study, C–N response ratios for three main forest biomes (boreal, temperate and tropical) were obtained by

stoichiometric scaling using estimates for N retention fractions, N allocation fractions and C:N ratios obtained by a recent literature review by De Vries et al. (2014).

Approaches to quantify large-scale carbon exchange in response to ozone exposure
An assessment of the impacts of O_3 on global C sequestration can be derived by multiplying the spatial variation of O_3 exposure with the C response to O_3 exposure (C–O_3 response). There are three potential approaches to assess C–O_3 responses, i.e. (i) experimental O_3 addition studies, (ii) field based monitoring studies across O_3 gradients, and (iii) global carbon-nitrogen-ozone modelling approaches. In this study we used a combination of a meta-analysis of experimental O_3 addition studies with global-scale modelling to assess large-scale C exchange in response to O_3 exposure.

Results
An overview of global-scale N_2O emission estimates according to two different IPCC approaches as reported in Syakila & Kroeze (2011) and FAO (2013), the IMAGE model (Bouwman et al., 2013), the Edgar database (JRC-PBL, 2011) and a simple emission factor model by Davidson (2009) is given in Table 1. Considering the various sources, and the neglect of human waste and atmospheric N deposition on ocean in most of the estimates, total anthropogenic N_2O emissions are most likely between 7.0 and 9.0 Tg N_2O-N yr^{-1}.

Table 2 presents an overview of global-scale CO_2-C sequestration in response to human N inputs. The contribution of N deposition to the global forest C sink was estimated at 0.44 Pg C yr^{-1}, which is comparable to a relatively recent estimates from global-scale modelling focusing on N deposition impacts (0.46 ± 0.28 Pg C yr^{-1}; Fleischer et al., 2015). The magnitude of the combined C sequestration in agricultural (0.13 Pg C yr^{-1}) and marine systems (0.18 Pg C yr^{-1}) is comparable to forests, but the uncertainty is higher.

Table 1. Global-scale anthropogenic N_2O emission estimates (Tg N_2O-N yr^{-1}) according to various approaches (details are given in De Vries et al., 2016). '-'= Not estimated, EF = emission factor, NA = Not available (included in estimates)

Source:	Syakila and Kroeze (2011)	Syakila and Kroeze (2011)	FAO (2013)	Bouwman et al. (2013)	Edgar 4.2 (JRC/PBL, 2011)	Davidson (2009)
Approach:	EFs based on IPCC 1997 Tier 1	EFs based on IPCC 2006 Tier 1	EFs based on IPCC 2006 Tier 1	Empirical relationships (IMAGE)	EFs from various sources	EFs and top-down approach
Year:	2006	2006	2000	2000	2000	2000
Agriculture						
Direct N_2O emissions from agricultural soils	2.2	1.8	1.6	4.8	2.4	2.2
Animal production	2.3	2.3	1.3	1.6	0.2	2.8
Indirect N_2O emissions	3.6	2.3	0.8	1.6	0.5	NA
Total agriculture	**8.1**	**6.3**	**3.7**	**8.0**	**3.2**	**5.0**
Biomass burning	**0.5**	**0.7**	**0.3**	**0.3**	**0.7**	**0.5**
Energy and transport		**1.2**	**-**	**0.6**	**1.0**	**0.8**
Human waste	**-**	**-**	**-**	**-**	**0.2**	**-**
Total emissions	**-**	**8.2**	**-**	**8.9**	**5.6**	**6.3**

Table 2. Estimated impacts of anthropogenic N inputs in agricultural land, grassland/woodland, forests, other land and marine systems for the year 2000 on global-scale carbon sequestration by multiplying inputs with average C–N responses (details for all estimates are given in De Vries et al., 2016).

Ecosystem	N inputs (Tg N yr^{-1}) (1)	C-N response (kg CO_2-C kg N^{-1}) (2)	CO_2-C sequestration (Tg CO_2-C_{eq} yr^{-1}) (1) × (2)
Agricultural land	248	0.53 (0.33–0.73)	131 (82–181)
Pastoral grassland/ Woodland	18.9	0[3]	0
Forests	22.9		
Boreal Forest	1.3	39.8 (31.7–49.6)	52 (41–64)
Temperate Forest	6.9	31.2 (24.8–39.3)	215 (171–271)
Tropical Forest	14.7	11.5 (8.5–15.2)	169 (125–223)
Other land	5.1	0	0

Total terrestrial	29		567 (419–739)
Marine	46	4 (3–5)	184 (138–230)
All systems	341		751 (557–969)

Based on global-scale modelling results by Sitch et al. (2007) and a meta-analysis by Wittig et al. (2009), we assumed that the global-scale impact of ozone exposure reduces forest growth by 10% (5–15%). The total C sink in global established forests has been estimated at 2.30 ± 0.49 Pg C yr^{-1} from an inventory-based global assessment (Pan et al., 2011). A 10% (5–15%) reduction thus implies a C sink reduction of 0.23 (0.115–0.345) Pg C yr^{-1}. Assuming that the O_3 increase since 1900 is determined for 60% by an increase in NO_x (Wang and Jacob, 1998) implies a CO_2 release due to N-induced O_3 exposure of 0.14 (0.07–0.21) Pg C yr^{-1}.

Conclusion

Ranges in impacts of human N fixation on N_2O and CO_2 exchange in response to N inputs and O_3 exposure, expressed in Pg CO_2-C_{eq} yr^{-1}, are given in Table 3 using a time scale of 100 years. This overview indicates that the overall impact of human N fixation is most likely a net increase in GHG emissions. Note, however, that the negative radiative impact (cooling effect) of N aerosol formation (mainly NH_4NO_3 and $(NH_4)_2SO_4$) nor the O_3 effects on atmospheric OH radical concentrations and thus atmospheric lifetime of atmospheric CH_4 have been included. Considering those impacts the effect of human N fixation might be cooling (Erisman et al., 2011).

Table 3. Ranges in N_2O emissions and CO_2 emission estimates in response to N inputs and O_3 exposure and total effect on the emission in Pg CO_2-C_{eq} yr^{-1}.

Effects of human N fixation	Effect of human N fixation on GHG in Pg CO_2-C_{eq} yr^{-1}
N_2O emissions due to N production	1.02 (0.89–1.15)
CO_2 uptake due to N deposition	-0.75 (-0.97 to -0.56)
CO_2 release due to N-induced O_3 exposure	0.14 (0.07–0.21)
Overall effect	**0.41 (-0.01–0.80)**

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