



# Linear and nonlinear dependency of direct nitrous oxide emissions on fertilizer nitrogen input: A meta-analysis

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## ABSTRACT

Rising atmospheric concentrations of nitrous oxide (N<sub>2</sub>O) contribute to global warming and associated climate change. It is often assumed that there is a linear relationship between nitrogen (N) input and direct N<sub>2</sub>O emission in managed ecosystems and, therefore, direct N<sub>2</sub>O emission for national greenhouse gas inventories use constant emission factors (EF). However, a growing body of studies shows that increases in direct N<sub>2</sub>O emission are related by a nonlinear relationship to increasing N input. We examined the dependency of direct N<sub>2</sub>O emission on N input using 26 published datasets where at least four different levels of N input had been applied. In 18 of these datasets the relationship of direct N<sub>2</sub>O emission to N input was nonlinear (exponential or hyperbolic) while the relationship was linear in four datasets. We also found that direct N<sub>2</sub>O EF remains constant or increases or decreases nonlinearly with changing N input. Studies show that direct N<sub>2</sub>O emissions increase abruptly at N input rates above plant uptake capacity. The remaining surplus N could serve as source of additional N<sub>2</sub>O production, and also indirectly promote N<sub>2</sub>O production by inhibiting biochemical N<sub>2</sub>O reduction. Accordingly, we propose a hypothetical relationship to conceptually describe in three steps the response of direct N<sub>2</sub>O emissions to increasing N input rates: (1) linear (N limited soil condition), (2) exponential, and (3) steady-state (carbon (C) limited soil condition). In this study, due to the limited availability of data, it was not possible to assess these hypothetical explanations fully. We recommend further comprehensive experimental examination and simulation using process-based models be conducted to address the issues reported in this review.

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## 1. Introduction

Atmospheric N<sub>2</sub>O contributes to both the greenhouse effect (Wang et al., 1976) and the ozone layer depletion (Crutzen, 1970). Nitrous oxide has a relatively high global warming potential (i.e., 298 times greater than carbon dioxide in a 100-yr time horizon; IPCC, 2006; Forster et al., 2007) and agricultural soils provide 3.5 Tg N<sub>2</sub>O-N yr<sup>-1</sup> of total anthropogenic N<sub>2</sub>O emissions (5.7 Tg N<sub>2</sub>O-N yr<sup>-1</sup>) (IPCC, 2006). Use of N fertilizers and animal manure is the main anthropogenic N<sub>2</sub>O source, and is responsible for roughly 24% of total annual emissions (Bouwman, 1996; Forster et al., 2007).

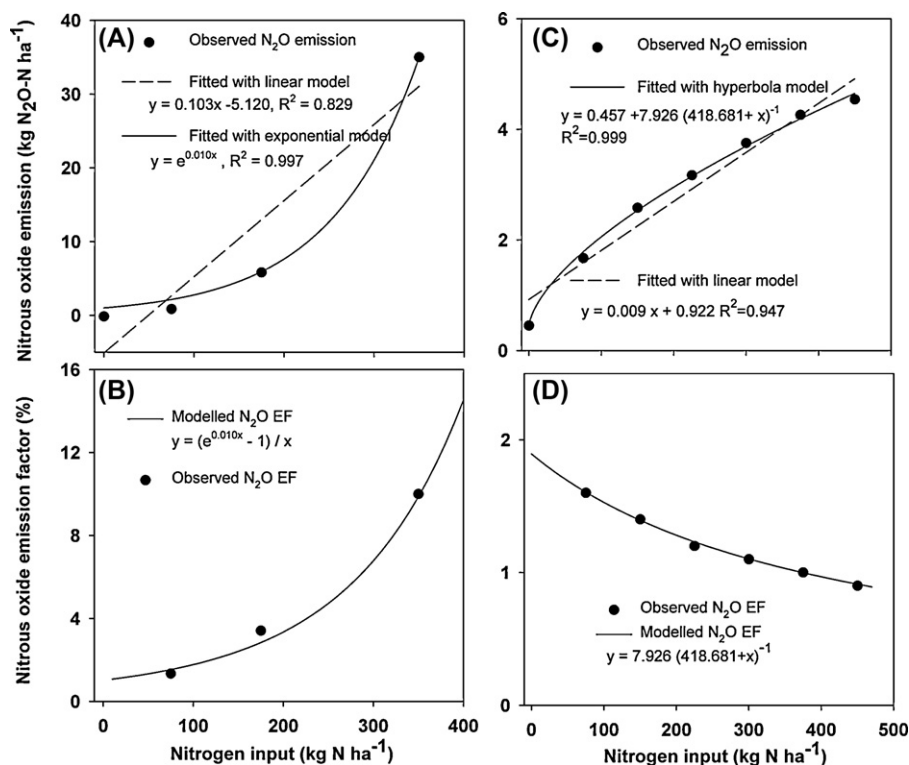
Nitrous oxide can be mainly produced from (1) aerobic autotrophic nitrification, the stepwise oxidation of ammonia (NH<sub>3</sub>) to nitrite (NO<sub>2</sub><sup>-</sup>) and to nitrate (NO<sub>3</sub><sup>-</sup>) (e.g., Kowalchuk and Stephen, 2001), (2) anaerobic heterotrophic denitrification, the stepwise reduction of NO<sub>3</sub><sup>-</sup> to NO<sub>2</sub><sup>-</sup>, nitric oxide (NO), N<sub>2</sub>O and

ultimately molecular nitrogen (N<sub>2</sub>), where facultative anaerobe bacteria use NO<sub>3</sub><sup>-</sup> as an electron acceptor in the respiration of organic material under low oxygen conditions (e.g., Knowles, 1982), and (3) nitrifier denitrification, which is carried out by autotrophic NH<sub>3</sub> oxidizing bacteria. This is the pathway whereby NH<sub>3</sub> is oxidized to NO<sub>2</sub><sup>-</sup>, followed by the reduction of NO<sub>2</sub><sup>-</sup> to NO, N<sub>2</sub>O and N<sub>2</sub> (e.g., Webster and Hopkins, 1996; Wrage et al., 2001).

Early reports suggested a linear relationship between increasing N input and increases in direct N<sub>2</sub>O emission (direct emissions of N<sub>2</sub>O from managed soils that occur through a direct pathway such as produced from synthetic N fertilizer; IPCC, 1996, 2006) in various agricultural systems (e.g., Bouwman, 1996; Dobbie et al., 1999). This relationship was adopted for the IPCC Tier I EF methodology (IPCC, 1996, 2006), which estimates direct N<sub>2</sub>O emission based on the amount of N added to agricultural soils. However, there is a growing body of evidence indicating a nonlinear, exponential response of direct N<sub>2</sub>O emission to N input (McSwiney and Robertson, 2005; Grant et al., 2006; Hellebrand et al., 2008; Zebarth et al., 2008; Jarecki et al., 2009; Cardenas et al., 2010; Kim et al., 2010; Hoben et al., 2011). This nonlinear increase in direct N<sub>2</sub>O emissions results in N<sub>2</sub>O EF values that are not constant but dependent on N input rates (Zheng et al., 2004; Grant et al., 2006;

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**Fig. 1.** Examples of better fitting with exponential or hyperbola increase rather than linear increase of nitrous oxide ( $\text{N}_2\text{O}$ ) emissions as a function of nitrogen (N) input, and non-linear increase of  $\text{N}_2\text{O}$  emission factor (EF) as a function of N input in cropland and grassland. Observed  $\text{N}_2\text{O}$  emission and fitted linear and exponential models (A) and observed  $\text{N}_2\text{O}$  EF and modeled  $\text{N}_2\text{O}$  EF by Eq. (3) (B) as a function of N input in cattle-grazed grassland, Aberystwyth, Wales (Cardenas et al., 2010). Observed  $\text{N}_2\text{O}$  emission and fitted linear and hyperbola models (C) and observed  $\text{N}_2\text{O}$  EF and modeled  $\text{N}_2\text{O}$  EF by Eq. (4) (D) as a function of N input in crop field, Iowa, USA (Breitenbeck and Bremner, 1986).

Halvorson et al., 2008; Hoogendoorn et al., 2008; Cardenas et al., 2010; Kim et al., 2010; Velthof and Mosquera, 2011). In Irish grassland, annual  $\text{N}_2\text{O}$  emissions increased sharply when N fertilization rates were above optimal levels, and direct  $\text{N}_2\text{O}$  emission showed an exponential relationship with N input (Kim et al., 2010). In spring barley (*Hordeum vulgare* L.) fields in eastern Canada,  $\text{N}_2\text{O}$  EF increased two-fold (i.e., 1.1–2.1%) when the applied fertilizer N rate was increased two-fold (Zebarth et al., 2008). In maize (*Zea mays* L.) fields in southwest Michigan USA, direct  $\text{N}_2\text{O}$  emissions increased sharply at N fertilizer rates above  $134 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ , and  $\text{N}_2\text{O}$  EF was up to 7% of the N-fertilizer input (McSwiney and Robertson, 2005).

Linear models of responses of direct  $\text{N}_2\text{O}$  emissions to N input are not always adequate and the underlying causes of these non-linear behaviors have not yet been clearly elucidated. Thus, the objectives of this study were to examine the dependency of both direct  $\text{N}_2\text{O}$  emission and  $\text{N}_2\text{O}$  EF on N input through meta-analysis of available, worldwide data and to establish preliminary hypotheses to mechanistically explain observed relationships.

## 2. Materials and methods

### 2.1. Data collection

Data were acquired by searching existing peer-reviewed literature (1980–2011), as well as through personal communications with individual data owners. We compiled field-measured direct  $\text{N}_2\text{O}$  emission data from 11 independent experimental studies encompassing 27 datasets worldwide (Tables 1–5). We selected studies where at least four different levels of N input were applied and the criteria were necessary to examine the best model fit (linear or nonlinear response) and to derive the corresponding model

parameters for direct  $\text{N}_2\text{O}$  emissions as a function of the respective N input levels (see 2.2 in details). Our compilation of global experimental data was also supplemented with three published modeling studies for direct  $\text{N}_2\text{O}$  emissions estimated using an agricultural production systems simulator (APSIM), *Ecosys*, and New Zealand Denitrification-Decomposition (NZ-DNDC) models. Where relevant and available, biomass productivity data were also used along with the corresponding measurements of direct  $\text{N}_2\text{O}$  emission and N input. If the N input, cumulative  $\text{N}_2\text{O}$  emissions and biomass productivity were only presented in graphical form without directly reporting in the literature we quantified the values using the software ADOBE® ACROBAT® 8 PROFESSIONAL ver. 8.2 (Adobe Systems, Inc. Burlington, NJ, USA). Through cross-checking the values quantified by graphical form with the corresponding numeric values reported in the existing literature, we estimated that quantification via graphical method introduced a 1–5% reading error, depending on the resolution of the graphical form provided.

Experimental  $\text{N}_2\text{O}$  emission datasets containing both control treatments with no N fertilizer additions (i.e., for background  $\text{N}_2\text{O}$  emissions) and different levels of N input were used to calculate direct  $\text{N}_2\text{O}$  EF following IPCC (2006) Tier I methodology as follows:

$$\text{N}_2\text{O EF}(\%) = \frac{\text{N}_2\text{O emission}_{\text{N treatment}} - \text{N}_2\text{O emission}_{\text{control}}}{\text{N input}} \times 100 \quad (1)$$

It should be noted that our data compilation includes a wide variety of studies that were conducted under diverse biophysical conditions using a range of methodologies (e.g., sampling protocol, chamber design, and emission rate calculation).

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