Dependency of nitrous oxide emission factors on nitrogen input rates: A meta-analysis

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Abstract
Rising atmospheric concentrations of nitrous oxide (N_2O) can cause global warming and associated climate change. It is typically assumed that there is a linear relationship between N_2O emission and nitrogen (N) input, and therefore, soil N_2O emission is often estimated as a proportion of N additions using emission factors (EF). However, a growing body of studies shows non-linear, exponential relationship between N_2O emission and N input. These studies commonly reported that N_2O emission abruptly increases at superoptimal level of N additions. Additionally, this rise in N_2O emission consistently causes EF to sharply increase and also to be directly dependent on N input rate. Meta-analysis revealed that increments in EF per additional unit of N input ranged from 0.0006 to 0.02, and these ΔEF/ΔN input values exhibited significant negative correlation with soil pH (r= -0.793, $P < 0.001$). Results support the importance of N management based on optimal rates particularly in intensive agricultural systems and low pH soils. It also indicates current IPCC default N_2O EF methodology could underestimate N_2O emission from these systems. Additional research efforts are needed to improve N_2O EF methodology and/or system modelling to capture the observed patterns of linear dependency of EF on N input rates.

Key Words
Greenhouse gas, ozone layer depletion, Intergovernmental Panel on Climate Change.

Introduction
Atmospheric N_2O contributes to both greenhouse effect (Wang *et al.* 1976) and ozone layer depletion (Crutzen 1970). A change in the N_2O mixing ratio from 270 ppb in 1750 to 319 ppb in 2005 caused an increased radiative forcing of 0.16 ± 0.02 W/m^2 in part because N_2O possesses a relatively high global warming potential (i.e., 298 and 25 times greater than carbon dioxide and methane, respectively; IPCC 2006). Of the entire anthropogenic N_2O emission (5.7 Tg N_2O-N/y), agricultural soils provide 3.5 Tg N_2O-N/y (IPCC 2006). Use of N fertilizers and animal manure is the main anthropogenic N_2O source and it is responsible for roughly 24% of total annual emissions (Bouwman 1996). Several early reports indicate a linear relationship between N input and N_2O emission in various agricultural systems (Bouwman 1996). This relationship is adapted for current IPCC Tier I EF methodology (IPCC, 2006) which estimates N_2O emission based on N additions in managed agricultural areas. However, there is a growing body of evidence indicating a nonlinear, exponential relationship between N input and N_2O emission (Grant *et al.* 2006; Zebarth *et al.* 2008). This nonlinear rise caused EF to increase with N additions, and therefore, N_2O EF values are not constant but dependent on N input rates (Grant *et al.* 2006). To date, these contradictory results pattern have not been clearly explained. Thus, the objectives of this study were to compile available data on the subject of direct N_2O EF in multiple agricultural systems with varying N input rates, to examine the dependency of N_2O EF on N input rates, and to establish hypotheses to potentially explain these relationships.

Materials and methods
Data were acquired by searching existing refereed literature as well as through personal communications with individual data owners. We compiled field data from 20 independent referred studies encompassing 40 experimental site-years worldwide. A complete list of the 20 assessed studies can be found within the caption for Figure 1. Based on this global dataset, direct N_2O EF were calculated following IPCC (2006) Tier I methodology, and subsequently, the changes in EF as a function of increments in N input (ΔEF/ΔN input with units/kg N ha) were also calculated. Statistics included Pearson correlation analyses.

Results and discussion
N_2O EF dependency on N input rates
This data compilation resulted in a very wide range of N_2O EF (i.e., 0.0005 to 0.16; Figure 1) revealing inconsistency amongst existing studies as well as when compared to the current IPCC single default value

for direct N_2O emissions (i.e., 0.01; IPCC 2006) as previously discussed by other researchers (Grant *et al.* 2006). Variations in N input sources across studies as well as dynamics soil-plant interactions in systems receiving superoptimal N addition rates could partly account for this ample EF variability. Extremely high EF values would likely occur when soil mineral N availability exceeds plant and soil N uptake capacities (McSwiney and Robertson, 2005). This collective evidence supports the critical need for enhancing existent N_2O estimation methodology for superior accuracy of both field-specific and global budget N_2O estimations. Furthermore, this meta-analysis also suggests that the pronounced variability in EF values across all assessed experimental site-years is directly associated with varying N input rates (Figure 1, Table 1). As N input (e.g., fertilizer or manure) rates increased, calculated N_2O EF consistently increased in all studied cases. Based on fitting linear regression models to the existing data, values of ΔEF/ΔN input (i.e., regression coefficient) ranged from 0.0006 to 0.02 and averaged 0.0074 (Table 1). In addition, the parameter ΔEF/ΔN input preliminary appears to behave linearly within several studies (Figure 1B). This numerical description of N_2O emission dynamics across multiple studies had not been previously documented in the existing literature.

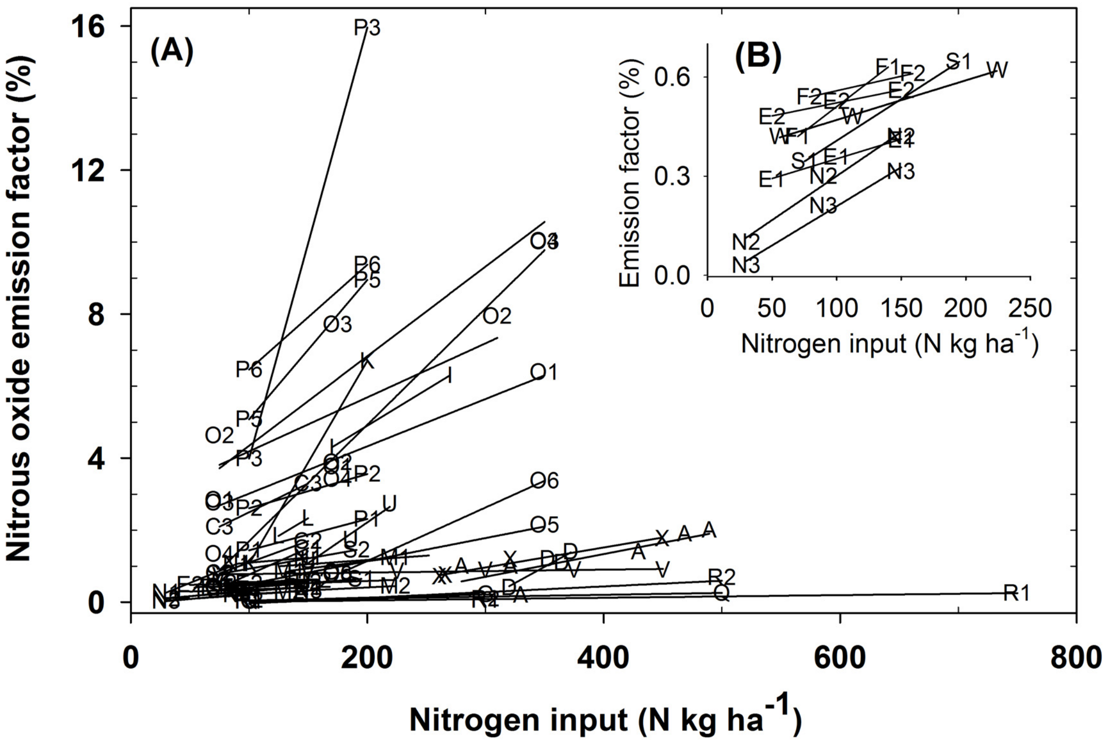

Figure 1. Direct nitrous oxide emission factors as a function of nitrogen (N) input rates across 20 studies. Ranges of N input from (A) 0 to 800 and (B) 0 to 250 kg N/ha. The 20 assessed studies are labelled as A (Hyde *et al.* 2006), C (Zebarth *et al.* 2008), D (Clayton *et al.* 1997), E (Zhang and Han 2008), F (Abdalla *et al.* 2010), G (Hua *et al.* 1997), I (Kilian *et al.* 1998), K (Allen *et al. in press*), L (Sehy *et al.* 2003), M (Mosier *et al.* 2006), N (Ma *et al.* 2009), O (Cardenas *et al. in press*), P (Engel *et al.* 2009), Q (Letica *et al.* 2009), R (Hoogendoorn *et al.* 2008), S (Jarecki *et al.* 2009), T (Rochette *et al.* 2000), U (Rochette *et al.* 2004), V (Breitenbeck and Bremner, 1986), and W (Mosier *et al.* 1982).

Another interesting observation of this meta-analysis is that ΔEF/ΔN input exhibited a negative correlation with soil pH (r= -0.793, $P < 0.001$; Table 1). It is well established that N_2O reductase activity is inhibited at low pH (e.g., 4.5 to 6.5) (Knowles 1982), and consequently, N_2O:N_2 production ratio typically increases with decreasing pH. This pH effect can also partly account for variations in ΔEF/ΔN input across studies.

Hypothetical mechanisms for explaining observed patterns of EF and ΔEF/ΔN input

Several hypotheses can be postulated to account for the observed EF shifts depending on N input rates. As discussed above, this response appears to be primarily associated with excessive N supply (e.g., > 100 kg N ha^{-1}; Bouwman *et al.* 2002) and soil microbial mediation. This soil N surplus would concomitantly lead to lower plant N uptake efficiency, and therefore, the resulting soil residual N would likely served as substrate for additional N_2O production (Zebarth *et al.* 2008). Additionally, excess soil N under these conditions could also indirectly promote soil N_2O production as it is known increased NO_3^- can inhibit N_2O reduction to N_2 producing wider N_2O:N_2 ratios (Firestone *et al.* 1979). Alternatively, exogenous N additions to soils can cause priming effects by stimulating microbial mobilization of native N boned within pre-existing soil organic matter (Kuzyakov *et al.* 2000). This enhanced soil native N mobilization and accessibility can result in increased N_2O emissions derived from the soil N pool (Di and Cameron 2008). Finally, based on this meta-analysis (Table 1) and as discussed above, changes in soil pH can also determine the outcome of N_2O production by interfering with N_2O reductase activity. Therefore, it can also be hypothesized that soil pH decreases can drive abnormally greater soil N_2O emission as a result of increasing N additions in intensively managed agricultural systems using certain acidifying N sources [e.g., ammonium sulphate: $(NH_4)_2SO_4$].

Table 1. Estimated ΔEF/ΔN input parameter and soil properties for 18 selected assessed site-years.

Reference	ID (as shown in Figure 1)	ΔEF/ΔN input (kg^{-1} N ha)	pH	Sand (%)	Clay (%)	C (%)	N (%)	C/N
Hyde *et al.* 2006	A (2002)	0.0063	5.8	22	18	0.032	0.28	11.4
Zebarth *et al.* 2008	C (2003)	0.0147	6.6	59.2	10.6	1.54	0.1	15.4
Zebarth *et al.* 2008	C (2004)	0.012	6.6	36.7	13.4	2.07	0.086	24.1
Zebarth *et al.* 2008	C (2005)	0.016	6	38.2	13.8	1.5	0.096	15.6
Clayton *et al.* 1997	D (1993)	0.018	5.5	17	22	5.5		
Zhang *et al.* 2008	E (grassland)	0.0012	7.21			2.45	0.238	10.3
Zhang *et al.* 2008	E (cropland)	0.0008	7.07			1.89	0.187	10.1
Abdalla *et al.* 2010	F (2004)	0.003	7.4			1.94	0.19	10.2
Abdalla *et al.* 2010	F (2005)	0.0009	7.4			1.94	0.19	10.2
Hua *et al.* 1997	G (urea)	0.0006	7.99			1.85	0.116	15.9
Hua *et al.* 1997	G [$(NH_4)_2SO_4$]	0.0012	7.99			1.85	0.116	15.9
Kilian *et al.* 1998	I (compost-N)	0.02						
Sehy *et al.* 2003	L (synthetic-N)	0.0197	6.1	20	29	1.4	0.15	9.3
Mosier *et al.* 2006	M (no tillage)	0.0041	7.7	40.2	33.4	1.28	0.152	8.4
Mosier *et al.* 2006	M (with tillage)	0.0016	7.7	40.2	33.4	1.19	0.147	8.1
Ma *et al.* 2009	N (2006)	0.0081	6.1			2.38		
Ma *et al.* 2009	N (2007)	0.0027	7.7			2.18		
Ma *et al.* 2009	N (2007)	0.0024	6.9			2.55		

Conclusions

Results from this meta-analysis using global data for N_2O emission from agricultural fields underscore the need for additional hypothesis-driven studies to enhance the current understanding on how excess N input affect N_2O emissions in N managed ecosystems. In further detail, this study clearly evidences extremely inconsistent EF values across assessed site-years as well as a linear dependency for EF changes on N input (ΔEF/ΔN input). These preliminary findings could provide insights for improving N_2O EF methodology.

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