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Stimulation of N_2O emission by manure application to agricultural soils may largely offset carbon benefits: a global meta-analysis

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Abstract

Animal manure application as organic fertilizer does not only sustain agricultural productivity and increase soil organic carbon (SOC) stocks, but also affects soil nitrogen cycling and nitrous oxide (N_2O) emissions. However, given that the sign and magnitude of manure effects on soil N_2O emissions is uncertain, the net climatic impact of manure application in arable land is unknown. Here, we performed a global meta-analysis using field experimental data published in peer-reviewed journals prior to December 2015. In this meta-analysis, we quantified the responses of N_2O emissions to manure application relative to synthetic N fertilizer application from individual studies and analyzed manure characteristics, experimental duration, climate, and soil properties as explanatory factors. Manure application significantly increased N_2O emissions by an average 32.7% (95% confidence interval: 5.1–58.2%) compared to application of synthetic N fertilizer alone. The significant stimulation of N_2O emissions occurred following cattle and poultry manure applications, subsurface manure application, and raw manure application. Furthermore, the significant stimulatory effects on N_2O emissions were also observed for warm temperate climate, acid soils ($pH < 6.5$), and soil texture classes of sandy loam and clay loam. Average direct N_2O emission factors (EFs) of 1.87% and 0.24% were estimated for upland soils and rice paddy soils receiving manure application, respectively. Although manure application increased SOC stocks, our study suggested that the benefit of increasing SOC stocks as GHG sinks could be largely offset by stimulation of soil N_2O emissions and aggravated by CH_4 emissions if, particularly for rice paddy soils, the stimulation of CH_4 emissions by manure application was taken into account.

Keywords: animal manure, emission factor, greenhouse gas balance, manure characteristics, meta-analysis, nitrous oxide, soil pH, soil texture

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Introduction

Agricultural production generates seven billion tons of animal manure per year globally (Thangarajan *et al.*, 2013), which is approximately two times greater than the global crop residue production (Lal, 2005). Application of these animal manures to arable land creates a great potential for sustaining crop productivity (Steiner *et al.*, 2007), improving soil fertility (Diacono & Montemurro, 2010), mitigating environmental N loss (Smith *et al.*, 2001; Bouwman *et al.*, 2010; Zhou *et al.*, 2016a), and enhancing soil C sequestration (Maillard & Angers, 2014). The enhanced agricultural soil C sequestration following manure application is urgently needed for mitigating climate change, as agricultural greenhouse gas (GHG) emissions contribute more than 13% to global anthropogenic GHG emissions (IPCC 2013). A recent global meta-analysis estimated that manure application could sequester an average of 5.6 Mg C ha^{-1} more organic C in the topsoil (0–22 cm) relative to synthetic N fertilizer over 18 years (Maillard & Angers, 2014). Therefore, substituting synthetic N fertilizer with animal manure has been suggested as a way to mitigate climate change while sustaining productivity in agricultural systems (Smith *et al.*, 2008).

Soil N_2O emissions can determine the overall GHG balance of agricultural production systems for a given agricultural practice, as N_2O is a potent GHG with a global warming potential (GWP) 265 times greater than

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CO_2 based on a 100-year time horizon (IPCC 2013). Both field experiments and process-based modeling have consistently indicated that agricultural practices that increase soil C sequestration (e.g., no tillage and/or reduced tillage, crop straw return) may induce additional N_2O emissions, which can offset the benefits of C sequestration (Six *et al*., 2004; Li *et al*., 2005; Zhou *et al*., 2014; Owen *et al*., 2015; Tian *et al*., 2016). For example, Li *et al.* (2005) estimated that soil N_2O emissions following reduced tillage and crop residue return practices offset 75% and 103% of the sequestered C, respectively. Therefore, it is necessary to determine the overall effect of manuring on N_2O emissions compared to synthetic N fertilizer if one aims to mitigate climate change in agricultural systems by substituting synthetic N fertilizers with animal manures.

Soil N_2O is mainly produced by microbial nitrification or denitrification, which are dependent on multiple factors, including the availability of carbon and nitrogen substrate as well as oxygen, soil properties (texture and pH), and environmental conditions (Firestone & Davidson, 1989; Groffman & Tiedje, 1991). Manure application by mediating the availability of soil inorganic N and bioavailable organic C as substrates for microbial N_2O production and consumption may affect N_2O emissions in agricultural soils (Baggs *et al*., 2000; Ball *et al*., 2004; Rochette *et al*., 2004; Aguilera *et al*., 2013; Thangarajan *et al*., 2013; Chen *et al*., 2014; Zhou *et al*., 2016a). In addition to direct effects, manure application could indirectly regulate soil N_2O emissions by changing soil aeration, specifically oxygen availability at microsites with the decomposition of organic matter (Xu *et al*., 2008). Furthermore, manure application cannot only increase soil pH (Whalen *et al*., 2000) but can also result in increased soil porosity, aggregation, and hydraulic conductivity (Haynes & Naidu, 1998), which can regulate various abiotic and biotic processes governing N_2O production in agricultural soils (Butterbach-Bahl *et al*., 2013; Heil *et al*., 2016, Zhou *et al*., 2016b).

Highly complex mechanisms associated with manuring regulate soil N_2O production and consumption. However, the available research results on the impact of manuring on N_2O emissions relative to synthetic N fertilizer have been contradictory (Akiyama & Tsuruta, 2003; Velthof *et al*., 2003; Ball *et al*., 2004; Rochette *et al*., 2004; Meijide *et al*., 2007; Ding *et al*., 2013; Zhou *et al*., 2014). For example, several previous studies have reported that manure application increased N_2O emissions in agricultural soils compared to synthetic N fertilizers (Baggs *et al*., 2000; Rochette *et al*., 2004; Zhou *et al*., 2014). In these studies, the enhancement of N_2O emissions following manure application was probably a result of an increased availability of labile organic C because soil labile organic C compounds often serve as an energy source for denitrifiers, thereby increasing N_2O production via denitrification (Ju *et al*., 2011). In contrast, some studies reported that manure application decreased N_2O emissions compared to synthetic N fertilizers (Ball *et al*., 2004; Meijide *et al*., 2007; Ding *et al*., 2013). The decrease in N_2O emission was likely a result of additions of organic C compounds that did not only enhance microbial inorganic N immobilization and competition for NH_4^+ for nitrification and NO_3^- for denitrification (Burger & Jackson, 2003; Zhou *et al*., 2016a), but also stimulated complete denitrification with further reduction of N_2O to N_2 (Ball *et al*., 2004; Meijide *et al*., 2007). Furthermore, some studies found no significant differences in N_2O emissions between manure and synthetic N fertilizer applications (Akiyama & Tsuruta, 2003; Vallejo *et al*., 2006). The sign and magnitude of the response of N_2O emissions to manure application relative to synthetic N fertilizer have been found dependent on manure characteristics, soil properties, and climate conditions (Velthof *et al*., 2003; Snyder *et al*., 2009; Chantigny *et al*., 2010). However, although many individual field studies have been conducted to investigate the effects of manuring on soil N_2O emissions, general conclusions have not been made due to the high variation of manuring effects across different experimental sites.

Detailed knowledge about how manure application affects soil N_2O emissions is critical to evaluating the potential of manure application to agricultural soils for climate change mitigation on a global scale. However, a quantitative synthesis of the overall response of N_2O emissions to manure application relative to synthetic N fertilizer is still lacking. Therefore, we conducted a meta-analysis to quantitatively estimate the overall sign and magnitude of manuring effects on N_2O emissions relative to synthetic N fertilizers in agricultural soils by integrating worldwide available measurements to evaluate the role of manure characteristics (e.g., animal species and manure management methods), climate, and soil properties as explanatory factors for N_2O emissions, as well as to evaluate the underlying mechanisms.

Materials and methods

Data collection

A comprehensive literature search was conducted of peer-reviewed articles that reported N_2O emissions following animal manure applications in agricultural soils in the Web of Science™ (Thomson Reuters, Philadelphia, PA, USA). 'Manure', 'nitrous oxide', 'N_2O', 'nitrogen', and 'N' were used as keywords to search for studies published prior to December of 2015. We only included studies that compared soil N_2O emissions between manure applications, and synthetic N fertilization and/or no fertilization treatments in the same agricultural system and at the same experimental site. Soil N_2O emissions from the unfertilized treatment were collected to estimate the direct N_2O emission factor with manure application. Studies included in the data pool had to meet the following criteria: a) replicated field experimental design; b) soil N_2O emissions measured under field conditions for at least a full crop season, with greenhouse and laboratory incubation experiments excluded; and c) field experiments that included at least one comparison of N_2O emissions between manure application and synthetic N fertilizer and/or no fertilization treatments. Furthermore, because of the positive response of N_2O emissions to N application rate, we only included comparisons of manure and synthetic N fertilizer applications if their differences in N application rate were <30 kg N ha^{-1}. Due to great temporal variations in N_2O emissions, only seasonal or annual cumulative N_2O emissions were considered, and one dataset of seasonal or annual cumulative N_2O emissions from one site was considered as one observation in this analysis. If a study was repeated for multiple growing seasons and/or years, the average value of the full experimental duration was considered as one observation. In total, 341 observations from 41 peer-reviewed articles were selected (Table 1; Data S1).

From each study, we extracted the cumulative N_2O emissions (kg N_2O-N ha^{-1}) and N application rate (kg N ha^{-1}) for manure and/or synthetic N fertilizer application treatment, with unit conversions performed where necessary. We also collected sample sizes and standard deviations for each treatment. If studies only reported standard errors, the corresponding standard deviations were converted from standard errors. GetData Graph Digitizer software (version 2.26: http://www.getdata-graph-digitizer.com/download) was used to extract data from graphs when data were presented with figures in the original publications. Other information from each of the selected studies that was compiled in the dataset included the following: geographic location, climatic conditions, soil properties (texture, total nitrogen [TN], SOC, C:N ratio, pH, and bulk density), synthetic N fertilizer type, manure characteristics, duration of fertilization treatment, crop yield, and N uptake. As some studies (30% of the selected studies) did not include soil texture and soil pH data, we obtained the missing data from the Harmonized World Soil Database v1.2 (FAO, 2012) in accordance with geographic locations (Data S1). As only 3% of experimental treatments were a combination of manure and synthetic N fertilizer application, the comparison of mixed application of manure and synthetic N fertilizer relative to pure synthetic N fertilizer was not considered in this study. Further, as there were only 25% and 20% of the selected studies presenting datasets of crop yield and N uptake, respectively, direct comparisons of crop yield and N uptake were not performed in the present analysis.

Seven categorical factors (three animal characteristics [animal species, manure preparation method, manure application method], duration of fertilization treatment, climate zone, soil texture, and soil pH), and six continuous factors (N rate, soil clay content, SOC, soil C:N ratio, soil pH, and manure C:N ratio) were retained for the analysis. Specifically, regarding manure characteristics, animal species were grouped in categories of cattle manure (n = 47 comparisons [same in the following in this section]), pig manure (n = 78), poultry manure (n = 9), and farmyard manure (FYM, n = 3); the manure preparation methods were grouped as raw (n = 80) and pretreated (composted or digested) manures (n = 57), and the manure application methods were grouped as surface (n = 59) and subsurface (n = 78) manure applications. The durations of fertilization treatment were grouped into short-term (<10 years, n = 120) and long-term (>10 years, n = 17) fertilization experiments. The precipitation and temperature at each experimental site were used to determine the climate zone from the world map of IPCC climate zones. In accordance with the generalized climate classification scheme of the IPCC climate zone (European Commission, 2012, Maillard & Angers, 2014), climate conditions at the experimental sites were grouped into three climate zones: cool temperate (n = 73), warm temperate (n = 61), and tropical (n = 3). Soil textures were grouped as sand (n = 4), sandy loam (n = 24), loam (n = 41), silt loam (n = 16), clay loam (n = 17), silt clay (n = 12), and clay (n = 23). The soil pH levels were grouped as <6.5 (acid, n = 84), 6.5–7.3 (neutral, n = 32), and >7.3 (alkaline, n = 21) based on the definitions recommended by the USDA (source: http://www.nrcs.usda.gov/).

Data analysis

We used the natural logarithm of the response ratio (ln RR) as the effect size of the comparisons of soil N_2O emissions between manure application and synthetic N fertilizer treatment in this meta-analysis (Hedges *et al.*, 1999).

$$\ln \mathrm{RR} = \ln\left(\frac{\overline{\mathrm{Xt}}}{\overline{\mathrm{Xc}}}\right) = \ln\left(\overline{\mathrm{Xt}}\right) - \ln\left(\overline{\mathrm{Xc}}\right) \tag{1}$$

where $\overline{\mathrm{Xt}}$ is the mean value of the manure application treatment and $\overline{\mathrm{Xc}}$ is the mean value of the synthetic N fertilizer treatment.

The variance of ln RR (v) for each study was estimated by the Eqn (2) (Hedges *et al.*, 1999).

$$\mathrm{V} = \left(s_t^2/n_t\overline{\mathrm{Xt}}^2\right) + \left(s_c^2/n_c\overline{\mathrm{Xc}}^2\right) \tag{2}$$

where s_t and s_c are the standard deviations for all comparisons in the treatment and control groups, respectively; n_t and n_c are the sample sizes for the treatment and control groups, respectively.

This meta-analysis was performed using a nonparametric weighting function, and the weighting factor w was calculated as the inverse of the pooled variance ($1/v$). When multiple observations were extracted from the same study, we adjusted the weights by the total number of observations (n) per site. The final weight (w') was calculated by the Eqn (3):

$$w' = w/n \tag{3}$$

The weighted effect size ln RR′ and mean effect size $\overline{\ln \mathrm{RR}'}$ for all observations were calculated with the Eqns (4) and (5):

Table 1 Geographic location, land use, soil texture, climate zone, animal species, N application rate, N_2O emission fluxes, and number of observation included in the present meta-analysis

Study number	References	Location	Land use	Soil texture	IPCC climate zone	Animal species	N rate (kg N ha^{-1})	N_2O emission fluxes (kg N ha^{-1})	Observations
1	Adviento-Borbe *et al.* (2010)	USA	Maize	Silt loam	Warm temperate	Cattle	90–225	0.10–0.89	8
2	Akiyama & Tsuruta (2003)	Japan	Vegetable	Loam	Warm temperate	Poultry and pig	150	0.49–1.84	3
3	Asgedom *et al.* (2014)	Canada	Rapeseed	Clay	Cool temperate	Cattle	0–137	0.39–3.05	10
4	Ball *et al.* (2004)	UK	Grassland	Loam	Warm temperate	Cattle	0–508	0.04–6.14	18
5	Bhatia *et al.* (2005)	India	Rice and Wheat	Clay loam	Warm temperate	FYM	0–120	0.32–0.86	6
6	Chantigny *et al.* (2013)	Canada	Maize	Clay soil and Silt clay	Cool temperate	Pig	0–150	1.5–10.7	14
7	Chantigny *et al.* (2010)	Canada	Maize	Clay soil and Loam soil	Cool temperate	Pig	0–130	0.68–18.50	42
8	Chen *et al.* (2014)	China	Maize	Clay loam	Cool temperate	Pig and Cattle	0–150	0.34–1.51	6
9	Collins *et al.* (2011)	USA	Maize	Silt loam	Cool temperate	Pig	0–336	0.12–0.48	6
10	Dalal *et al.* (2010)	Australia	Sorghum	Silt loam	Tropical	Cattle	0–373	3.29–5.52	4
11	Dambreville *et al.* (2008)	France	Maize	Loam	Warm temperate	Pig	0–180	0.34–2.17	6
12	Das & Adhya (2014)	India	Rice	Sandy clay loam	Tropical	Poultry	0–120	0.16–0.79	3
13	Ding *et al.* (2013)	China	Wheat and Maize	Silt loam	Warm temperate	Pig	0–150	0.10–1.11	8
14	Ellert & Janzen (2008)	Canada	Maize	Loam–clay soil	Cool temperate	Cattle	0–425	1.39–6.54	3
15	Guo *et al.* (2013)	China	Maize	Sandy loam	Cool temperate	Cattle	0–338	0.57–4.06	10
16	Hayakawa *et al.* (2009)	Japan	Vegetable	Loam	Warm temperate	Poultry	240	0.38–2.72	3
17	Hernandez-Ramirez *et al.* (2009)	USA	Maize	Silty clay loam	Warm temperate	Pig	157–255	3.29–8.17	3
18	Jarecki *et al.* (2008)	USA	Maize	Sandy soil and Clay soil	Warm temperate	Pig	0–200	0.30–7.46	6
19	Jin *et al.* (2010)	Japan	Grassland	Loam	Warm temperate	Cattle	0–331	0.6–4.9	9
20	Lampe *et al.* (2006)	Germany	Grassland	Sand	Warm temperate	Cattle	0–174	1.75–4.88	5
21	Li *et al.* (2013)	China	Maize	Sandy loam	Cool temperate	Pig	0–818	0.64–1.19	4
22	Li *et al.* (2002)	Japan	Vegetable	Loam	Warm temperate	Cattle	0–300	0.11–0.45	5
23	Lopez-Fernandez *et al.* (2007)	Spain	Maize	Loam	Warm temperate	Pig	0–170	2.91–5.89	5
24	Meijide *et al.* (2007)	Spain	Maize	Loam	Warm temperate	Pig	0–300	5.98–9.28	6
25	Meng *et al.* (2005)	China	Maize and Wheat	Sandy loam	Warm temperate	Pig	0–150	0.06–0.56	8
26	Mori & Hojito (2012)	Japan	Grassland	Loam	Warm temperate	Cattle	0–392	1.00–11.90	15
27	Mori *et al.* (2008)	Japan	Grassland	Loam	Warm temperate	Cattle	209–210	4.70–11.00	4

Table 1 (continued)

Study number	References	Location	Land use	Soil texture	IPCC climate zone	Animal species	N rate (kg N ha^{-1})	N_2O emission fluxes (kg N ha^{-1})	Observations
28	Nyamadzawo *et al.* (2014)	Zimbabwe	Rapeseed	Sandy loam	Tropical	Cattle	0–240	2.50–8.40	10
29	Pelster *et al.* (2012)	Canada	Maize	Silty clay and Sandy loam	Cool temperate	Poultry, Pig and Cattle	0–94.3	0.40–8.30	20
30	Perala *et al.* (2006)	Finland	Bayley	Sandy loam	Cool temperate	Pig	100–157	0.29–1.10	4
31	Rochette *et al.* (2004)	Canada	Maize	Loam	Cool temperate	Pig	150–219	1.74–5.99	3
32	Rochette *et al.* (2008)	Canada	Maize	Clay and loam	Cool temperate	Cattle	150	0.76–6.06	12
33	Schils *et al.* (2008)	The Netherlands	Grassland	Sandy loam	Warm temperate	Cattle	0–330	0.04–1.42	16
34	Sherlock *et al.* (2002)	New Zealand	Grassland	Silt clay	Warm temperate	Cattle	0–366	0.53–8.13	2
35	Singla *et al.* (2013)	Japan	Vegetable	Sand	Warm temperate	Cattle	0–120	0.03–0.54	6
36	Sistani *et al.* (2010)	USA	Maize	Silt loam	Warm temperate	Pig	0–200	0.47–8.20	10
37	Sosulski *et al.* (2014)	Poland	Rye	Loamy sand	Warm temperate	Cattle	90–128	0.64–0.73	3
38	Wang *et al.* (2013)	China	Rice	Silt clay	Warm temperate	Pig	0–180	0.18–0.24	3
39	Watanabe *et al.* (2014)	Japan	Vegetable	Loam	Warm temperate	Cattle	144	0.14–4.71	20
40	Zhai *et al.* (2011)	China	Maize and Wheat	Clay loam	Warm temperate	Pig	0–300	0.14–1.42	6
41	Zhou *et al.* (2014)	China	Maize and Wheat	Sandy loam	Warm temperate	Pig	0–150	0.03–1.03	6

$$\ln \mathrm{RR}'_{\mathrm{i}} = w'_i \times \ln \mathrm{RR}_i \tag{4}$$

$$\overline{\ln \mathrm{RR}}' = \sum_i \ln RR'_{\mathrm{i}} / \sum_i w'_i \tag{5}$$

where $\ln \mathrm{RR}'_{\mathrm{i}}$ and w'_i are $\ln \mathrm{RR}'$ and w_i of the ith observation, respectively.

In addition to the weighted mean effect sizes, confidence intervals (CIs, 95%) were generated using bootstrapping of 4999 iterations (Rosenberg *et al.*, 2000). The results were considered significant if the 95% confidence intervals did not overlap with zero and the randomization tests yielded P values <0.05. For improved explanatory power, the mean effect size was transformed back to the percentage change for manure application relative to the synthetic N fertilizer treatments. For the variables of N_2O emission, we conducted a categorical randomized-effects meta-analysis model to compare the effect sizes among the categorical groups through a framework similar to ANOVA. In other words, for each variable, the total heterogeneity (Q_T) in the categorical group was partitioned into within-group (Q_W) heterogeneity and between-group heterogeneity (Q_B) using chi-square distributions. The significance of Q_B represents mean effect sizes that are significantly different between various levels of the categorical group (Rosenberg, 2000). We also applied a continuous randomized-effects meta-analysis model to test the relationships between the effect sizes of N_2O emissions and the N application rate, soil clay content, SOC, soil C:N ratio, soil pH, and manure C:N ratio. The statistical results have been reported as the total heterogeneity (Q_T), the difference in the among-group cumulative sizes (Q_M), the residual error (Q_E), the slope, and the P values. The relationships were considered significant if $P < 0.05$. All of the meta-analysis procedures were conducted using MetaWin 2.1 software (Sinauer Associates, Inc., Sunderland, MA, USA).

We also calculated the direct N_2O emissions factors (EF, %) following the manure applications to estimate the net manuring effects on soil N_2O emissions if studies included a control (no fertilizer treatment):

$$\mathrm{EF}(\%) = (F_{\mathrm{N}} - F_{\mathrm{c}})/N * 100 \tag{6}$$

where F_{N} is the cumulative N_2O emissions (kg N_2O-N ha^{-1}) from each N addition treatment, F_{C} is the cumulative N_2O emissions (kg N_2O-N ha^{-1}) from the control, and N is the N application rate (kg N ha^{-1}).

To estimate the climatic effects of N_2O emission changes by manure application, we calculated the GHG balance of soil N_2O emissions and soil C sequestration following manure application relative to synthetic N fertilizer. Here, the global mean N application rate of 115.7 kg N ha^{-1} yr^{-1} for arable land in 2014 that was estimated from the report of Food and Agriculture Organization of the United Nations (FAO, 2016) was applied in the estimation. The changes in soil C sequestration by manure application compared to synthetic N fertilizer were adopted from the newest global meta-analysis study, which estimated the average soil C sequestration of 5600 ± 2800 kg C ha^{-1} in the top 22-cm soil layer over 18 years (i.e., 311 kg C ha^{-1} yr^{-1}) following manure application relative to synthetic N fertilizer (Maillard & Angers, 2014). Soil N_2O emissions were converted into CO_2 equivalents by taking into account the specific radiative forcing potential of 265 relative to CO_2 on a 100-year time horizon (IPCC 2013).

Results

Effects of manure application on N_2O emissions compared to synthetic N fertilizer

On average, manure application significantly increased soil N_2O emissions by 32.7% (95% CI: 5.1–58.2%) compared to synthetic N fertilizers (Fig. 1). The continuous randomized-effects model analysis showed that the effect sizes of manure application on N_2O emissions were negatively correlated with soil clay content and soil pH ($P < 0.01$) but not with soil C:N ratio, SOC, N addition rate, or manure C:N ratio (Table 2). Regarding the duration of manure application treatment, relative to synthetic N fertilizer, soil N_2O emissions on average were increased by 22.9% (95% CI: 2.6–45.9%) and 70.7% (95% CI: −8.5 to 164.7%) from manure applications during short-term and long-term fertilization treatments, respectively, while the effect of long-term manure application treatment was not statistically significant (Fig. 1a).

The effect sizes were also dependent on the manure origin, that is, animal species, indicated by the marginally significant differences in effect size among animal species (Fig. 1b, P = 0.087). Compared to synthetic N fertilizer, cattle and poultry manure applications significantly increased soil N_2O emissions by an average 28.7% (95% CI: 5.2–67.2%) and 45.4% (95% CI: 7.8–159.2%), respectively. Furthermore, soil N_2O emissions increased by 41.6% (95% CI: −10.3 to 83.8%) for pig manure application and decreased by 21.4% (95% CI: –37.2–16.8%) for FYM application relative to synthetic N fertilizer while both effects were not statistically significant.

Compared with synthetic N fertilizer application, subsurface manure application significantly increased soil N_2O emissions by 74.8% (95% CI: 6.0–104.2%), whereas no significant increase was found for surface manure application (mean: 17.4%, 95% CI: −7.8 to 47.9%) (Fig. 2a). The categorical group analysis indicated that manure preparation methods also significantly affected the effect sizes of manure application on N_2O emissions relative to synthetic N fertilizer (Fig. 2b, $P = 0.0386$). The application of raw manure significantly increased soil N_2O emissions by an average of 46.9% (95% CI: 8.5–81.5%), whereas the pretreated manure application increased N_2O emissions, statistically

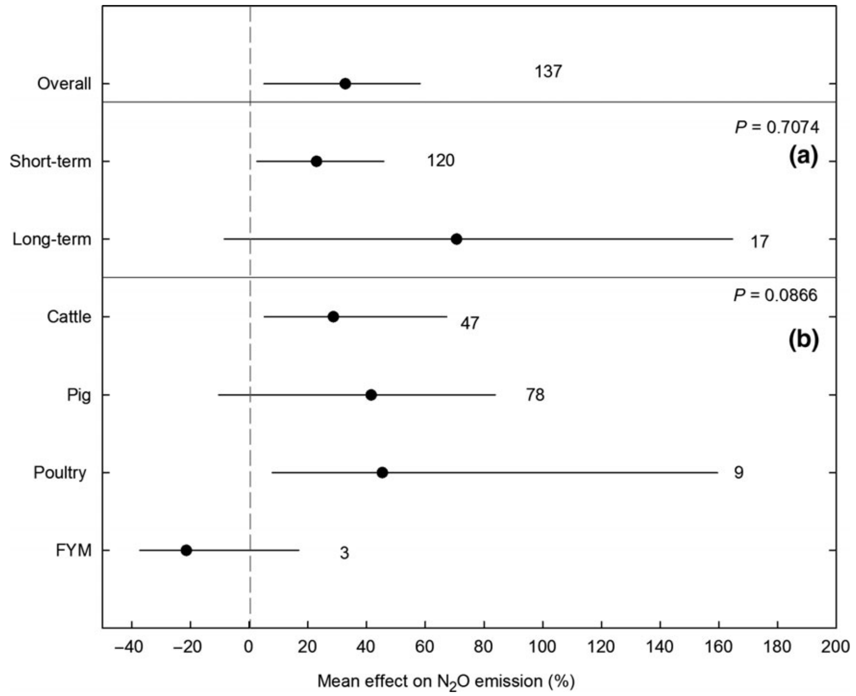

Fig. 1 Comparison of soil N_2O emission in manure vs. synthetic N fertilizer applications for the entire dataset (overall) and for subcategories of the duration of fertilization treatment (a, long-term or short-term fertilization treatment) and animal species (b). FYM represents farmyard manure. The number of observations included in each category is shown next to the error bars. Error bars represent 95% confidence intervals. The effect of manure application was considered significant if the 95% CI of the mean effect did not cover zero. The *P* value of the difference between subcategories is shown in the panel.

Table 2 Relationships between the effect sizes of manure application on soil N_2O emissions relative to synthetic N fertilizer application and N addition rate, soil clay content, soil organic carbon (SOC), soil C:N ratio, soil pH, and manure C:N ratio. Statistical results were reported as total heterogeneity in effect sizes among studies (Q_T), the difference among groups cumulative effect sizes (Q_M), and the residual error (Q_E) from continuous randomized-effects model meta-analysis. The relationship is significant if $P < 0.05$

	Q_T	Q_M	Q_E	Slope	P	n
N rate	201.1	2.1	199.0	0.002	0.1444	137
Clay content	139.5	34.3	105.2	−0.019	0.0000	90
SOC	146.3	2.3	144.0	−0.007	0.1259	101
Soil C:N ratio	138.9	0.1	138.8	0.008	0.7516	93
Soil pH	183.1	10.7	172.4	−0.243	0.0011	100
Manure C:N ratio	145.7	0.1	145.6	0.004	0.7427	113

not significantly, only by an average of 2.8% (95% CI: −18.5 to 30.3%).

Regarding climate zones, manure application significantly increased soil N_2O emissions relative to synthetic N fertilizer in the warm temperate climate zone (mean: 34.3%, 95% CI: 6.8–64.7%). By contrast, significant effects were not observed in cool temperate (mean: 32.2%, 95% CI: −8.7 to 33.8%) and tropical (mean: −4.8%, 95% CI: −43.3 to 3.9%) climate zones (Fig. 3).

Soil texture significantly affected manure application-induced N_2O emissions relative to synthetic N fertilizer-induced N_2O emissions (Fig. 4, $P < 0.0001$). On average, manure application significantly increased soil N_2O emissions by 40.0% (95% CI: 3.4–50.4%) in sandy loam soils, 63.8% (95% CI: 41.7–134.8%) in loam soils, and 17.4% (95% CI: 10.2–61.5%) in clay loam soils. However, manure application significantly decreased N_2O emissions by 15.9% (95% CI: −54.0 to −4.3%) in silt clay soils. However, the manuring effects were not statistically significant in the other three soil types, that is, clay, silt loam, and sandy soils (mean values: 13.6% [95% CI: −40.8 to 25.8%], −15.2% [95% CI: −51.0 to 28.8%], and 1.1% [95% CI: −35.9 to 6.6%], respectively).

Similarly, soil pH significantly affected N_2O emissions induced by manure application relative to synthetic N fertilizer application (Fig. 5, $P = 0.0025$). Manure application, on average, significantly increased N_2O emissions by 64.8% (95% CI: 14.0–70.8%) in acid soils. However, significant effects were not observed in pH neutral soils (mean: 7.3%, 95% CI: −28.6 to 25.4%) or alkaline soils (mean: −8.2%, 95% CI: −22.8 to 1.2%).

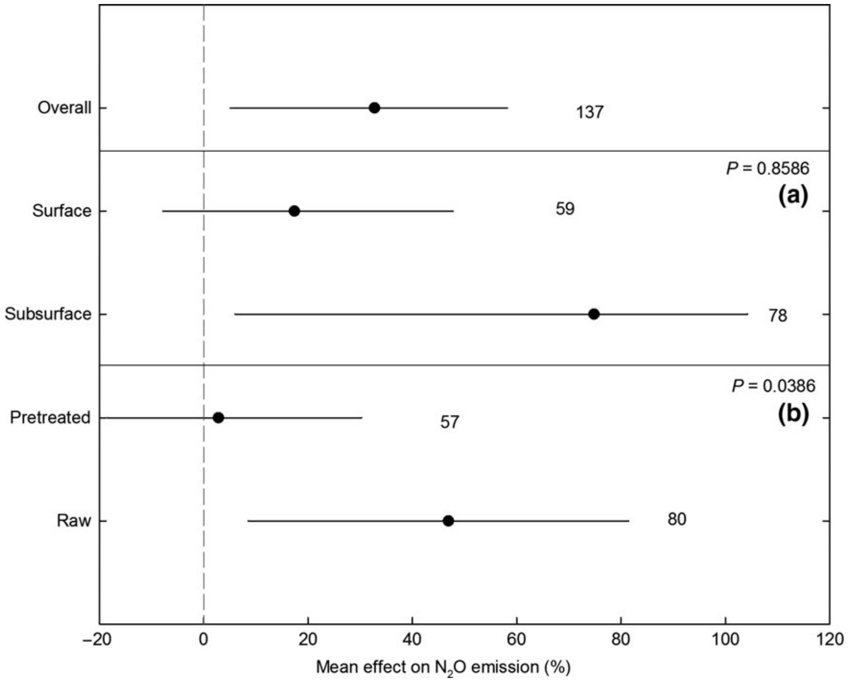

Fig. 2 Comparison of soil N_2O emission in manure vs. synthetic N fertilizer applications for application method (a) and manure preparation (b). The number of observations included in each category is shown next to the error bars. Error bars represent 95% confidence intervals. The effect of manure application was considered significant if the 95% CI of the mean effect did not cover zero. The *P* value of the difference between subcategories is shown in the panel.

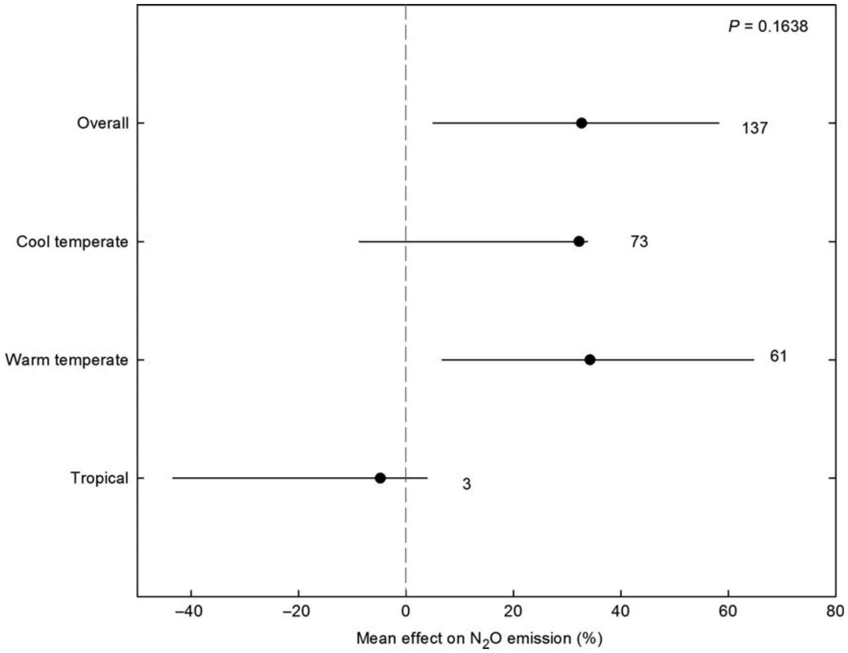

Fig. 3 Comparison of soil N_2O emission in manure vs. synthetic N fertilizer applications for subcategories of climate zone. The number of observations included in each category is shown next to the error bars. Error bars represent 95% confidence intervals. The effect of manure application was considered significant if the 95% CI of the mean effect did not cover zero. The *P* value of the difference between subcategories is shown in the panel.

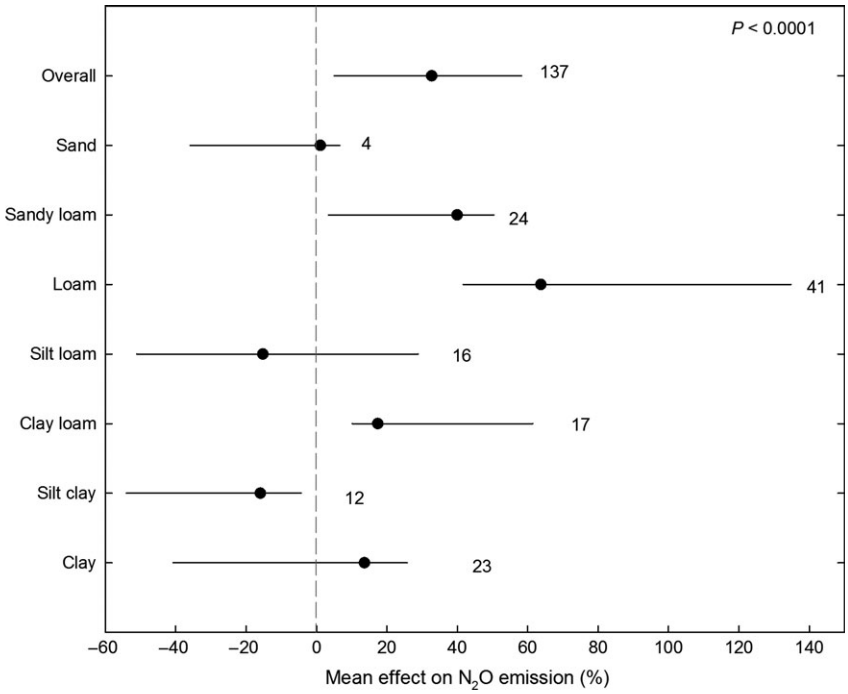

Fig. 4 Comparison of soil N_2O emission in manure vs. mineral nitrogen fertilizer applications for the entire dataset (overall) and for soil texture subcategories of sand, sandy loam, loam, silt loam, clay loam, silt clay, and clay soils. The number of observations included in each category is shown next to the error bars. Error bars represent 95% confidence intervals. The effect of manure application was considered significant if the 95% CI of the mean effect did not cover zero. P values represent significant differences between subcategories if $P < 0.05$.

N_2O emission factors

Overall, the average N_2O emissions factor (EF) for manure applications was 1.83 ± 0.30% ($n = 146$) and significantly higher than zero across different experimental sites (Fig. 6). The mean EF following manure application was 1.87 ± 0.30% in upland soils ($n = 143$) and 0.24 ± 0.60% in rice paddy soils ($n = 3$). Pig manure applications had the highest mean EF (1.70 ± 0.21%), followed by cattle manure (1.35 ± 0.25%) and poultry manure (1.07 ± 0.39%) (Fig. 6). Applications of raw and pretreated manures were associated with mean EFs of 1.43 ± 0.21% and 1.66 ± 0.19%, respectively. On average, subsurface manure application induced a significantly higher EF (1.74 ± 0.22%) compared to surface manure applications (1.28 ± 0.20%). The manure application-induced EF in the warm temperate climate zone (0.79 ± 0.12%) was significantly lower than in cool temperate (1.95 ± 0.23%) and tropical (3.57 ± 1.04%) climate.

Global warming potentials

Our estimation showed that the average net change of GWP of soil organic C sequestration by manure application relative to synthetic N fertilizer was 1140.7 kg CO_2-eq ha^{-1} yr^{-1} (Table 3). However, due to changes of N_2O emissions, manure application induced a great increase in GWP of 419.2 kg CO_2-eq ha^{-1} yr^{-1} in upland soils and a slight decrease in GWP of 30.4 kg CO_2-eq ha^{-1} yr^{-1} in rice paddy soils. Overall, our calculations suggested that the increase in soil organic C sequestration as a CO_2 sink by manure application could be largely offset by at least 36.7% due to stimulation of N_2O emission in upland soils, but not in rice paddy soils.

Discussion

Effects of manure application on N_2O emissions

There has been an ongoing debate on whether manure application can increase N_2O emissions compared to synthetic N fertilizer (Petersen *et al.*, 1996; Van Groenigen *et al.*, 2005; Meijide *et al.*, 2007; Aguilera *et al.*, 2013; Zhou *et al.*, 2014). Our global meta-analysis showed that manure application significantly increased soil N_2O emissions an average of 32.7% (95% CI: 5.1–58.2%), relative to synthetic N fertilizer (Fig. 1). In this context, changes in quantity and quality of soil C and N

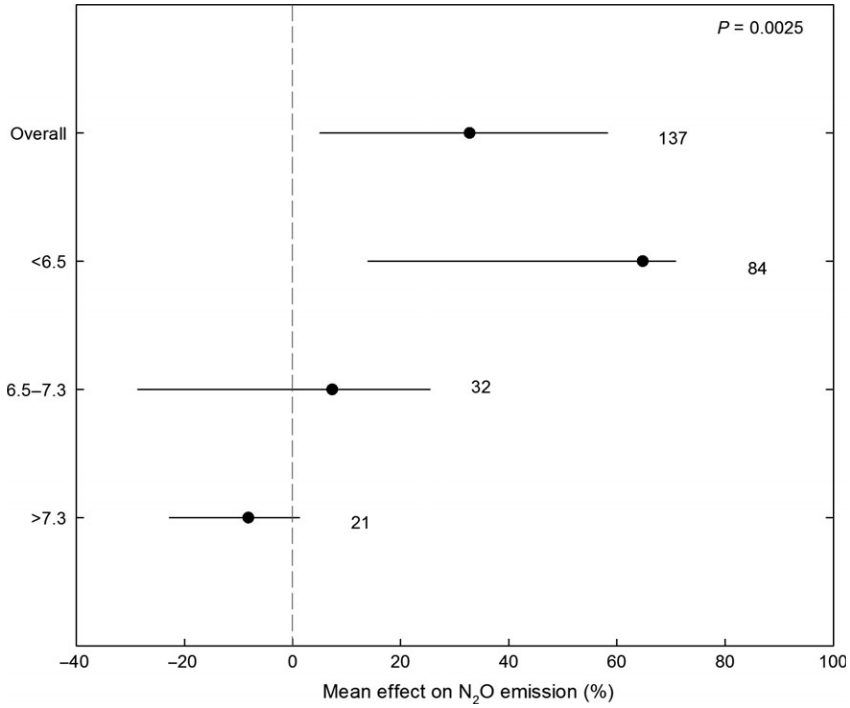

Fig. 5 Comparison of soil N_2O emission in manure vs. mineral nitrogen fertilizer applications for the entire dataset (overall) and for soil pH subcategories of <6.5 (acid), 6.5–7.3 (neutral) and >7.3 (alkaline). The number of observations included in each category is shown next to the error bars. Error bars represent 95% confidence intervals. The effect of manure application was considered significant if the 95% CI of the mean effect did not cover zero. P values represent significant differences between subcategories if $P < 0.05$.

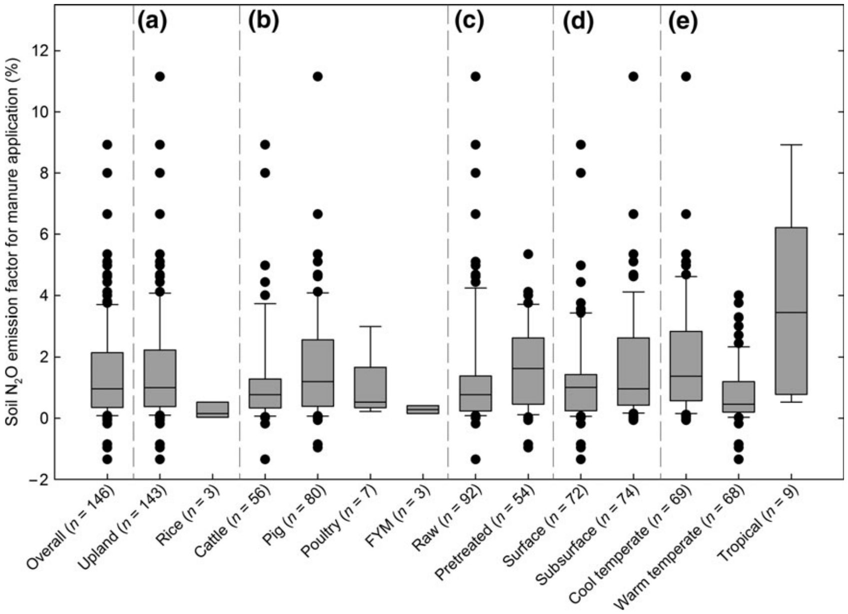

Fig. 6 Boxplots illustrating the soil nitrous oxide emission factors (EFs, %) for manure applications regarding different land use (a), animal species (b), manure preparations (c), manure application methods (d), and climate zones (e). Black circles represent outliers. Black solid lines in the boxes represent medium values.

substrate and environmental condition from manure applications may favor N_2O production and emission because soil N_2O emissions are moderated by multiple factors such as soil inorganic N and organic C availability, soil oxygen (O_2) availability, soil temperature, and soil moisture (Butterbach-Bahl *et al.*, 2013). First,

Table 3 Summary of manure application and synthetic N fertilizer application induced changes in SOC stocks (F_{SOC}, kg C kg N ha^{-1} yr^{-1} for synthetic N fertilizer and kg C ha^{-1} yr^{-1} for manure applications) and soil N_2O emission factor (F_{N2O}, kg N_2O-N ha^{-1} kg N^{-1})

	Upland soils	Rice paddy soils	References
N_2O emission factor (F_{N2O}, kg N_2O-N ha^{-1} kg N^{-1})			
Synthetic N fertilizer	0.01	0.003	IPCC (2006)
Animal manure	0.0187 ± 0.003	0.0024 ± 0.0064	This study
Changes in global warming potential (GWP, kg CO_2 eq ha^{-1} yr^{-1}) by manure application relative to synthetic N fertilizer			
N_2O emission	419.2	−30.4	This study
SOC	−1140.7	−1140.7	Maillard & Angers (2014)
Percentage of the increases in SOC stocks offset by N_2O emissions following manure application			
Animal manure	36.7%	−2.7%	

Estimated on basis of global average N application rate for agricultural soils in 2014: 115.7 kg N ha^{-1} (Food and Agriculture Organization of the United Nations (FAO), 2016).

relative to synthetic N fertilizer alone, manure application provides more labile organic C compounds as energy for microbial activity, thereby stimulating N_2O production of nitrifiers and denitrifiers (Firestone & Davidson, 1989). Second, the increase in availability of C and N substrates by manure application could enhance microbial activity and O_2 consumption, and hence create anoxic conditions in the soil due to O_2 depletion and facilitate denitrification, thereby increasing N_2O emissions (Petersen *et al.*, 1996; Van Groenigen *et al.*, 2005).

However, it should be noted that the magnitude of soil N_2O emission is related to the ratio of gaseous end products of denitrification (i.e., N_2O / (N_2 + N_2O)). This is one mechanism to explain the decrease in N_2O emissions by manure application compared to synthetic N fertilizer (Meijide *et al.*, 2007; Aguilera *et al.*, 2013). For example, Meijide *et al.* (2007) observed that manure applications decreased N_2O emissions from a maize system in Mediterranean climate by increasing the tendency to complete denitrification with reduction of N_2O to N_2, in particular after irrigation and rainfall events (Meijide *et al.*, 2007). Owing to the presence of a number of studies reporting the inhibitory effects of manure application on N_2O emissions relative to synthetic N fertilizer (Meijide *et al.*, 2007; Zhou *et al.*, 2014), the present analysis revealed the overall sign and magnitude of manuring effects on N_2O emissions relative to synthetic N fertilizers (Fig. 1), which were dependent on manure characteristics (Figs 1 and 2), climate (Fig. 3), soil texture (Fig. 4), and soil pH (Fig. 5).

Manure characteristics. Compared to synthetic N fertilizer, N_2O emissions increased due to applications of cattle (mean: 28.7%, 95% CI: 5.2–67.2%) and poultry (mean: 45.4%, 95% CI: 7.8–159.2%) manures (Fig. 1b). These variations in the effect of animal-specific manure on N_2O emissions were likely attributable to the different manure compositions (Chadwick *et al.*, 2000; Bell *et al.*, 2016). For example, effect sizes for poultry (mean 45.4%, 95% CI: 7.8–159.2%) manure were greater than those for cattle manure (mean: 28.7%, 95% CI: 5.2–67.2%) (Fig. 1b), although cattle manure contained a relatively larger inorganic N pool than poultry manure per unit (Chen *et al.*, 2014). The significantly greater N_2O emissions by application of poultry manure were likely due to their higher content of easily decomposable organic C (e.g., DOC and volatile fatty acids) relative to cattle manure (Kirchmann, 1991; Kirchmann & Lundvall, 1993; Chadwick *et al.*, 2000; Velthof *et al.*, 2003). First, application of manure with a larger content of easily degradable organic C substrates tended to alleviate efficiently the inhibition of denitrification by limited organic C substrate supply and to generate higher N_2O emissions than those with more resistant organic C (e.g., Velthof *et al.*, 2003; Mori & Hojito, 2012). Second, application of manure with more labile organic C could create a more anoxic soil environment that favors denitrification and N_2O emissions (Cayuela *et al.*, 2010; Aguilera *et al.*, 2013).

Besides animal species, manure effects on soil N_2O emissions could be also dependent on manure application methods (Webb *et al.*, 2010). The present study found that subsurface manure application significantly increased N_2O emissions by an average of 74.8% (95% CI: 6.0–104.2%), relative to synthetic N fertilizers, while there were no significant effects for surface manure application (17.4%, 95% CI: −7.8 to 47.9%, Fig. 2a). Some previous studies also observed greater soil N_2O emissions following subsurface manure application compared to surface manure application (Wulf *et al.*, 2002; Velthof *et al.*, 2003). First, subsurface manure application could enhance the contact between soil and added C and N compounds, which may intensify microbial O_2 consumption and induce strong O_2 depletion that facilitates denitrification and N_2O emission (Zhu *et al.*, 2015). Second, subsurface manure application could decrease NH_3 volatilization and retain more N in soil, thereby increasing the supply of N substrate for N_2O production (Webb *et al.*, 2010). However, previous studies have demonstrated that subsurface manure application could also decrease or have no effect on N_2O emissions compared to surface manure application (Velthof *et al.*, 2003; Mkhabela *et al.*, 2008). This phenomenon was likely because subsurface manure application might increase the length of N_2O diffusion path from the microsites of denitrification into the atmosphere and hence increase the potential for reduction of N_2O to N_2. It should be noted that subsurface manure application is widely recommended to mitigate NH_3 volatilization in agricultural soils (Webb *et al.*, 2010). The trade-offs between N_2O emission and NH_3 volatilization by manure application have not been well considered to date. In this context, the overall effect of subsurface manure application on N_2O emission may change if both indirect N_2O emission due to NH_3 volatilization (IPCC 2006) and direct N_2O emission were taken into account.

Our analysis found that N_2O emissions significantly increased by 46.9% (95% CI: 8.5–81.5%) by application of raw manure but not for pretreated manure (mean: 2.8%, 95% CI: −18.5 to 30.3%) relative to synthetic N fertilizer (Fig. 2b). The results indicate that application of pretreated manure may be effective to mitigate N_2O emissions in manure-amended agricultural systems. In general, compared to pretreated manure raw manure has a higher availability of easily degradable organic C and inorganic N compounds that may enhance the microbial nitrification–denitrification processes and N_2O production in soils (Zhou *et al.*, 2016a). By contrast, pretreated manure has a greater C:N ratio than raw manure (Bernal *et al.*, 2009). In addition, several studies reported that pretreated manure with a greater C:N ratio (>15) could enhance microbial N immobilization (Widmer *et al.*, 2002; Velthof *et al.*, 2003; Mooshammer *et al.*, 2014), which leads to a decrease in availability of inorganic N substrate for nitrification and/or denitrification and inhibition of N_2O emissions. Nevertheless, one caveat is that N_2O emission during manure pretreatment (e.g., composting) may substantially offset the benefit of pretreated manure application in agricultural soils (Hou *et al.*, 2015).

Climate. Climate, through regulating soil moisture and temperature regimes, may affect the microbial nitrification–denitrification process and N_2O emissions (Barnard *et al.*, 2006; Xu *et al.*, 2012). Relative to synthetic N fertilizer, manure application significantly increased N_2O emission by an average of 34.3% (95% CI: 6.8–64.7%) in warm temperate climate but not in cool temperate climate (Fig. 3). First, the warmer climate may directly enhance microbial nitrogen turnover rate coefficients and increase availability of C and N substrate in soils; second, the warmer climate could stimulate microbial decomposition of organic matter and increase soil respiration which favors the development of soil anoxic conditions. Thus, the warmer climate may favor microbial nitrification–denitrification associated with N_2O production, and hence increase soil N_2O emissions (Barnard *et al.*, 2005; Xu *et al.*, 2012). It is noteworthy that soil N_2O emissions may not consistently increase with increasing temperature as the increase in denitrification could stimulate the production of N_2, the gaseous end product of denitrification, thereby outweighing the effect on N_2O production (Smith, 1997). For example, previous studies have demonstrated that addition of organic C substrate by manure application may lead to complete denitrification, with higher production of the gaseous end product N_2 than N_2O, and hence a decrease in N_2O emission under warm and wet environmental conditions (Dalal *et al.*, 2010; Das & Adhya, 2014). It still remains an open question how climatic condition (i.e., temperature and precipitation regime) affects N_2O emissions in agricultural soils and further studies are therefore highly recommended in particular for tropical region.

Soil texture. Soil texture significantly affected effect sizes of manure application on N_2O emissions ($P < 0.0001$, Fig. 4). In coarser-textured sandy soils, nitrification is the dominant process of N_2O emissions (Mctaggart *et al.*, 2002; Zhou *et al.*, 2013). The air-filled porosity of sandy soils frequently results in low denitrification activity (Groffman & Tiedje, 1991); in this context, manure application may not appear to increase nitrification and N_2O emissions compared to synthetic N fertilizer (Fig. 4), which is consistent with the general understanding of soil texture serving as an important control on soil N_2O emissions (Mctaggart *et al.*, 2002; Skiba & Ball, 2002; Gu *et al.*, 2013). Soil texture regulates soil N_2O emissions through moderating soil O_2 availability (Corre *et al.*, 1999), as soil texture has a strong impact on the size and distribution of soil pores. By contrast, manure applications significantly increased soil N_2O emissions in the sandy loam, loam, and clay loam soils (Fig. 4). The fine-textured soils can frequently develop anoxic microsites and favor denitrification, perhaps due to clay particles that hold water tightly in the soil aggregates (Gu *et al.*, 2013). Thus, manure application with a sufficient supply of organic C substrate could stimulate denitrification and hence increase N_2O emissions. However, relative to synthetic N fertilizer, manure application significantly decreased N_2O emissions in silt clay soil (Fig. 4) that contains over 40% clay particles. First, the silt clay soil with high clay content (>40%) generally has low gas diffusivity, which could promote reduction of N_2O produced in the soil profile to N_2 through complete denitrification before being emitted to the atmosphere (Weitz *et al.*, 2001), particularly after manure amendment with sufficient easily degradable organic C substrates. Second, the silt clay soil with high clay content (>40%) has great cation-exchange capacity (CEC) and can increase NH_4^+ adsorption by clay particles, which in turn decreases soil NH_4^+ availability and constrains nitrification and denitrification, thereby decreasing N_2O emissions (Jarecki *et al.*, 2008).

Soil pH. Soil pH has been identified as another key regulator of soil N_2O emissions (e.g., reviewed by Butterbach-Bahl *et al.*, 2013). The present analysis indicated that, compared with synthetic N fertilizer, manure application significantly increased N_2O emissions in acid soils (pH < 6.5, Fig. 5) but not in neutral and alkaline soils. As low soil pH generally prevents the assembly of functional N_2O reductase (N_2OR) and inhibits the reduction of N_2O to N_2 by this enzyme during denitrification (e.g., Bakken *et al.*, 2012), the mole fraction of $N_2O/(N_2O + N_2)$ during denitrification was greater in acid soils (Stevens *et al.*, 1998). Therefore, in acid soils manure application could promote greater production of N_2O than N_2 by denitrification and subsequently increase soil N_2O emissions. It is noteworthy that there may be potential uncertainties in effect sizes of manure application on N_2O emissions as a function of soil pH. Because the missing soil pH values in the selected publications were extracted from the Harmonized World Soil Database v1.2 of the FAO in accordance with the geographic locations, these extracted data may not accurately represent the true values of the experimental sites thereby inducing uncertainty. Moreover, as reviewed by Butterbach-Bahl *et al.* (2013), there is still a lack of knowledge on how soil pH regulates soil N_2O emission. In addition, manure application tends to neutralize soil acidity and raise soil pH (Thangarajan *et al.*, 2013), which may increase the complexity of the mechanism of manure application effects on soil N_2O emissions across various soil pH levels.

N_2O emissions factors of manure application

The present analysis, including various land uses, manure characteristics, climate, and soil characteristics, estimated an average EF of 1.83% for manure application (Fig. 6), which was higher than the current IPCC default value of 1% (IPCC 2006). The average EFs of manure application in upland soils (mean: 1.87%) were greatly higher than for rice paddy soils (mean: 0.24%, Fig. 6a), which was even lower than the IPCC default factor of 0.30% for rice paddy soils (IPCC 2006). Because rice paddy soils are often submerged, this can cause a large proportion of the produced N_2O to be further reduced to N_2, thereby leading to lower N_2O emissions, in particular in rice paddies receiving manure (Firestone & Davidson, 1989; Zhou *et al.*, 2015). Nevertheless, the present estimation provided the latest EFs of manure application, which reduced the knowledge gap on how much of the N input by manure application can be lost as N_2O emission in agricultural soils at the global scale.

Implications and perspectives

Globally, application of animal manure to arable land as organic fertilizer enhances SOC stocks compared to synthetic N fertilizer alone (e.g., Maillard & Angers, 2014). However, the potential of manure application for climate change mitigation by increase in SOC stocks can be attenuated by enhanced N_2O emissions as indicated in the present analysis (Fig. 1). The estimation of the present study suggests that increases in N_2O emissions offset the GHG sink strength of manure application-induced C sequestration by roughly 37% in upland soils (Table 3), if the annual N rate in agricultural soils was equal to the global average value of 115.7 kg N ha^{-1} in 2014 (Food and Agriculture Organization of the United Nations (FAO), 2016). In addition, Owen *et al.* (2015) found that the stimulation of N_2O emissions following long-term manure application offsets the GHG sink strength of soil C sequestration by 75–100% in the rangelands of California, USA. By contrast, there was no stimulatory effect of manure application on N_2O emission in rice paddy soils (Table 3). As CH_4 emission dominantly contributes to GHG balance in rice paddy soils (e.g., Shang *et al.*, 2011), the GHG sink strength of increasing SOC stocks could be also largely offset by CH_4 emissions, if the stimulation of CH_4 emissions by manure application for rice paddy soils was taken into account. Overall, our analysis highlights an important concept that the benefits of increasing the soil C sink may be largely offset by increased N_2O emission (and increased CH_4 emission, which requires additional quantitative analysis) by manure application to agricultural soils.

It is noteworthy that the primary goal of manure application to arable land is to increase and/or at least sustain crop productivity. This suggests that further evaluation is necessary to reconcile the concerns of GHG emissions with food security regarding manure application, even though manure application has been shown to be able to sustain crop yield (e.g., Steiner *et al.*, 2007). Second, we recommend that further research should consider a potential saturation of the SOC level following long-term manure application. As some studies have proposed that soil has an upper limit of organic C storage capacity (Six *et al.*, 2002; Liu *et al.*, 2014), C addition by manure application may result in a limitation of soil C sequestration if the C saturation point has been reached in a long-term perspective (Steiner *et al.*, 2007). Third, we emphasize that particular attention should be paid to the dynamic interactions between the C and the N cycle after manure application in order to maximize the climate change mitigation potential of substituting synthetic N fertilizer application with animal manure in agricultural soils. It would appear that the benefit of SOC stocks from manure application may be further offset by increased N_2O emissions as the significant positive relationship between SOC content and N_2O emissions existed in most agricultural soils (Li *et al.*, 2005). This emphasizes that further studies should contribute to improving dynamic biogeochemical models and conduct measurements in long-term experiments for the accurate assessment of the effect of manure application on the net GHG balance of agroecosystems in the long-term perspective.

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Supporting Information

Additional Supporting Information may be found in the online version of this article:

Data S1. Basic information of the selected publications extracted for this meta-analysis.