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# A meta-analysis of fertilizer-induced soil NO and combined NO+N<sub>2</sub>O emissions

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## Abstract

Soils are among the important sources of atmospheric nitric oxide (NO) and nitrous oxide (N<sub>2</sub>O), acting as a critical role in atmospheric chemistry. Updated data derived from 114 peer-reviewed publications with 520 field measurements were synthesized using meta-analysis procedure to examine the N fertilizer-induced soil NO and the combined NO+N<sub>2</sub>O emissions across global soils. Besides factors identified in earlier reviews, additional factors responsible for NO fluxes were fertilizer type, soil C/N ratio, crop residue incorporation, tillage, atmospheric carbon dioxide concentration, drought and biomass burning. When averaged across all measurements, soil NO-N fluxes were estimated to be 4.06 kg ha<sup>-1</sup> yr<sup>-1</sup>, with the greatest (9.75 kg ha<sup>-1</sup> yr<sup>-1</sup>) in vegetable croplands and the lowest (0.11 kg ha<sup>-1</sup> yr<sup>-1</sup>) in rice paddies. Soil NO emissions were more enhanced by synthetic N fertilizer (+38%), relative to organic (+20%) or mixed N (+18%) sources. Compared with synthetic N fertilizer alone, synthetic N fertilizer combined with nitrification inhibitors substantially reduced soil NO emissions by 81%. The global mean direct emission factors of N fertilizer for NO (EF<sub>NO</sub>) and combined NO+N<sub>2</sub>O (EF<sub>c</sub>) were estimated to be 1.16% and 2.58%, with 95% confidence intervals of 0.71–1.61% and 1.81–3.35%, respectively. Forests had the greatest EF<sub>NO</sub> (2.39%). Within the croplands, the EF<sub>NO</sub> (1.71%) and EF<sub>c</sub> (4.13%) were the greatest in vegetable cropping fields. Among different chemical N fertilizer varieties, ammonium nitrate had the greatest EF<sub>NO</sub> (2.93%) and EF<sub>c</sub> (5.97%). Some options such as organic instead of synthetic N fertilizer, decreasing N fertilizer input rate, nitrification inhibitor and low irrigation frequency could be adopted to mitigate soil NO emissions. More field measurements over multiyears are highly needed to minimize the estimate uncertainties and mitigate soil NO emissions, particularly in forests and vegetable croplands.

**Keywords:** emission factor, fertilizer, meta-analysis, nitric oxide, nitrous oxide, trace gas

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## Introduction

Among the trace gases of great concern, nitric oxide (NO) and nitrous oxide (N<sub>2</sub>O) are involved in the production and consumption of atmospheric oxidants such as ozone (O<sub>3</sub>) and hydroxyl radical (OH) (Williams *et al.*, 1992). They are the potential precursors of photochemical formation of nitric acid (HNO<sub>3</sub>) that is the fast-growing component of acidic deposition, directly responsible for the acidification and eutrophication of terrestrial ecosystems (IPCC, 2013). Recently, the anthropogenic activities have greatly altered the background atmospheric NO and N<sub>2</sub>O concentration, indirectly or directly contributing to changes in concentration of atmospheric greenhouse gases and tropospheric chemistry (Bouwman *et al.*, 2002a).

Soils have been recognized as an important source of atmospheric NO (Davidson & Kingerlee, 1997;

Bouwman *et al.*, 2002a,b; Yan *et al.*, 2005; Stehfest & Bouwman, 2006; Pilegaard, 2013). Soil NO is mainly produced through the microbial processes of nitrification and denitrification. The nitrification predominates the pathways for soil NO emission, especially in tropical and subtropical climate regions (Godde & Conrad, 2000; Laville *et al.*, 2005; Stehfest & Bouwman, 2006). The controlling factors of soil NO emissions have been reviewed earlier, including N fertilizer application rate, soil N content, climate, land cover type, soil organic carbon content, pH and bulk density, drainage and length of the measurement period (Bouwman *et al.*, 2002a,b; Yan *et al.*, 2003a,b; Stehfest & Bouwman, 2006; Pilegaard, 2013).

In the past decades, a great many field measurements of soil NO fluxes have been taken in various ecosystems. Some earlier studies based on flux measurement data have developed statistic models to quantify global fertilizer-induced soil NO emissions (Bouwman *et al.*, 2002a; Yan *et al.*, 2003a; Stehfest & Bouwman, 2006). Parallel to these statistical models, it is useful to synthesize the flux measurement data with meta-analysis

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procedures to understand the processes and factors responsible for soil NO emissions. Meta-analysis approach has been developed for quantitative integration of available individual field measurements, which is increasingly used in studies with respect to ecological and biogeochemical issues (Knorr *et al.*, 2005; Akiyama *et al.*, 2010; Van Groenigen *et al.*, 2011; Shcherbak *et al.*, 2014). This approach can be used to update the mechanisms of soil NO emissions response to various factors (e.g., climate, soil properties, agricultural practice and experimental treatments) and partition N fertilizer-induced soil NO emissions from different ecosystems.

Although earlier statistical models have provided an insight into the fertilizer-induced soil NO emissions, their estimates had large uncertainties, partially due to limited available measurement data (Bouwman *et al.*, 2002a; Yan *et al.*, 2003a; Stehfest & Bouwman, 2006). For example, statistical models established by Yan *et al.* (2003a) were based only on a total of 53 data points from 16 individual reports. Stehfest & Bouwman (2006) updated the earlier dataset of Bouwman *et al.* (2002a), while only two measurements of NO from rice paddies were included in their studies. This limitation is challenged by the recent evidence that soil NO emissions were significantly different between rice paddies and upland croplands (Zhou *et al.*, 2010; Deng *et al.*, 2012). In particular, the earlier summary work did not consider field measurements taken from vegetable cropping fields, where high N fertilizer input rate and frequent irrigation often incur high NO fluxes (Mei *et al.*, 2009; Deng *et al.*, 2012; Yao *et al.*, 2015; Zhang *et al.*, 2016). Nevertheless, an increasing number of field measurements would allow us to use the meta-analysis to reexamine these limitations and quantify the direct emission factor of N fertilizer for NO among different ecosystems.

In addition, NO and N<sub>2</sub>O are generally interrelated in soil nitrogen cycling processes; an overall accounting of fertilizer-induced NO combined with N<sub>2</sub>O emissions (NO+N<sub>2</sub>O) across global soils would provide an insight into their integrative role in biogeochemistry (Bouwman *et al.*, 2002a; Van Lent *et al.*, 2015). Unfortunately, previous reviews have separately examined fertilizer-induced NO and N<sub>2</sub>O emissions and these two trace gases were not taken into consideration together (Bouwman *et al.*, 2002a; Yan *et al.*, 2003a; Stehfest & Bouwman, 2006). Indeed, simultaneous measurements of NO and N<sub>2</sub>O fluxes have been increasingly taken in recent field studies, and therefore, the fertilizer N ratio lost as (NO+N<sub>2</sub>O)-N emission, that is, fertilizer-induced direct emission factor of combined NO and N<sub>2</sub>O emissions, could be addressed using meta-analysis techniques.

Here, 520 field measurements derived from 114 peer-reviewed publications within an updated and comprehensive dataset were synthesized using meta-analysis procedures to identify the key factors influencing NO and the combined NO+N<sub>2</sub>O emissions across global soils. The objectives of this meta-analysis based on more extensive data were first to examine limitations of earlier reviews by extending the findings on the factors influencing soil NO emissions. Second, we aimed to highlight the global soil NO emission hot spots by comparing soil NO emissions from different ecosystems. Eventually, we used the updated data to assess the direct emission factors of N fertilizer for NO (EF<sub>NO</sub>) and a combination of NO and N<sub>2</sub>O emissions (EF<sub>c</sub>) partitioned by various ecosystems and fertilizer sources.

## Materials and methods

### Data extraction and compilation

We conducted a detailed review of the literature published in peer-reviewed journals through the year 2016 (cutoff date on June 30, 2016). We extracted data from publications enclosing individual field measurements with only soil NO data or in the cases that both NO and N<sub>2</sub>O fluxes were simultaneously measured (Fig. 1, Table S1, Dataset S1). All published data were derived from the Web of Science and Google Scholar, and papers published in the China Knowledge Resource Integrated Database (CNKI) with English abstract, as well as gathering and re-evaluating the older literature cited in the prior review of Stehfest & Bouwman (2006). Different combinations of searching keywords ('nitric oxide' OR 'NO<sub>x</sub>' OR 'NO' AND 'soil') were used for data extraction. The final database consisted of 520 field measurements derived from 114 publications, of which 81 recent measurements taken from vegetable cropping fields and 17 measurements from rice paddies were not evaluated by Stehfest & Bouwman (2006). In addition, 43 studies that contain 173 simultaneous measurements of NO and N<sub>2</sub>O fluxes were available in the database (Table S1, Dataset S1).

We complied with the following criteria to avoid bias in selecting publications. Only field measurement data were included in this analysis, without considering laboratory or pot experimental data, vertical profile monitoring data over vegetation canopy or modeling results. For data from natural ecosystems, the occasional field NO flux measurements without covering the whole experimental period or the number of consecutive measurements less than three date-points were excluded. For data from croplands, the consecutive measurement period covers at least one whole cropping season. The raw data were either obtained directly from tables and texts or extracted by digitizing graphs using the GETDATA GRAPH DIGITIZER software (version 2.26, <http://www.getdatagraph-digitizer.com/download.php>). For each literature selected, the following original documented information was compiled: soil NO fluxes, total fluxes of soil NO and N<sub>2</sub>O, NO/N<sub>2</sub>O and/or NO/(NO+N<sub>2</sub>O) ratios, direct emission factors of N

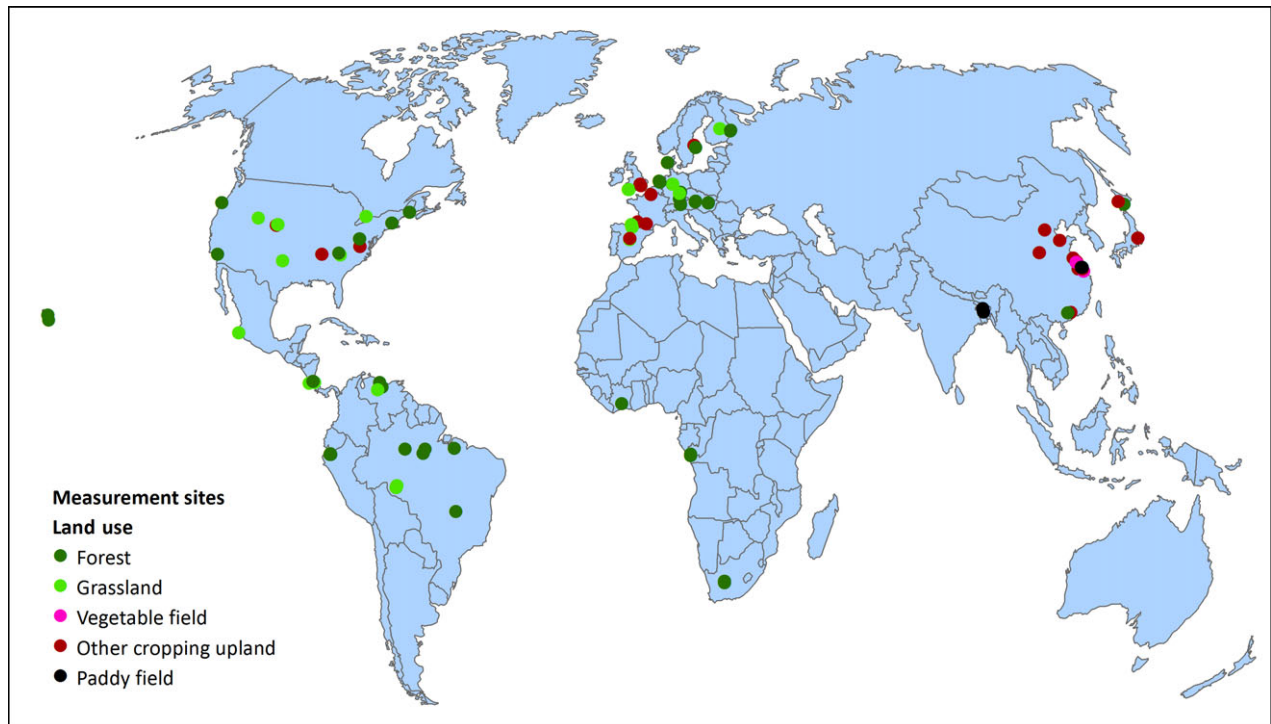


Fig. 1 World map of global soil NO fluxes measurement sites ( $n = 114$  locations).

fertilizer for NO and/or combined NO+N<sub>2</sub>O, location (longitude and latitude), general climate (tropical, subtropical, temperate and cool), experimental duration, land-use type, dominated vegetation, soil properties (texture, pH, mean temperature in 0–20 cm depth, WFPS, soil organic C (SOC) content, total N content, C/N ratio and mineral N), frequency of measurement, number of replicates, as well as fertilizer rate and source (Table S2). If only soil organic matter (SOM) was provided, we converted SOM to SOC using a Bemmelen index value of 0.58 to include as many data as possible.

Data were subjected to a standardization process to allow for comparisons. We calculated the balanced mean values of soil NO and N<sub>2</sub>O fluxes as well as their emission factors with the residual maximum likelihood (REML) procedure using GENSTAT release 4.2 to minimize the heterogeneity resulting from missing values and unequal number of observations among reviewed literature (Payne, 2000). Measurements from different sites or different fertilizer rates within a single study were considered as independent field measurements in data collection. In the cases that mean seasonal or annual soil NO and N<sub>2</sub>O emissions were not presented directly, hourly or daily fluxes were converted to kilograms of N per hectare for the experimental period. For grassland and forest soils, some nonconsecutive data were simultaneously obtained in both wet and dry seasons; in these cases, their grand mean was derived for the database. Otherwise, for measurements were only taken in a single season (dry or wet) and where temperature performed as the dominant factor driving soil NO and N<sub>2</sub>O fluxes with significant correlations, the flux data were further rectified by the annual mean temperature using the temperature-dependent statistical model that was established

in an earlier review of Yan *et al.* (2005). In addition, atmospheric N deposition was not considered in this analysis for most lack of information on actual N deposition rates under experimental field conditions.

In further data compiling prior to analysis, we categorized the soils into five land-use types as paddy field, vegetable field, other cropping upland (including wheat, corn, barley and cotton), grassland and forest based on the experimental land-use mode. Only two studies with three measurements were taken from deserts and three studies with four measurements were available in wetlands, which eventually led to the exclusion of them in our analysis. Besides fertilizer application, other five experimental factors including crop residue incorporation, experimental drought, tillage, biomass burning and atmospheric carbon dioxide concentration ([CO<sub>2</sub>]) enrichment were also taken into consideration in this analysis. Soil texture was grouped into three general classes (coarse, medium and fine) due to the inconsistent reporting of soil texture in the literature (e.g., general qualitative description, particle size distribution, soil taxonomical unit).

We quantified the direct emission factors of N fertilizer for NO (EF<sub>NO</sub>) and a combination of NO and N<sub>2</sub>O emissions (EF<sub>c</sub>) partitioned by various ecosystems and fertilizer sources. For studies without directly reporting the EF<sub>NO</sub> or EF<sub>c</sub> but with available data, we estimated them based on the gas flux data from both the control and fertilized treatments and total N input rate over the experimental period. Specifically, the EF<sub>NO</sub> is defined as NO emission from N fertilizer plots minus the emission from unfertilized control plots expressed as a percentage of N applied. Similarly, the EF<sub>c</sub> is calculated by the difference in the total of NO and N<sub>2</sub>O emissions between N

fertilizer treatments and controls expressed as a percentage of N input in this analysis.

### Data analyses

In this study, data derived from fertilized soils (48 studies with 219 paired measurements) and from studies exposed to other available experimental factors (57 measurements from 23 studies) were separately analyzed using meta-analysis procedure to examine the general response of soil NO emissions to fertilizer and other typical experimental factors. The means of soil NO flux from treatment ( $X_t$ ) and control ( $X_c$ ) groups were used to calculate effect size in the form of natural log-transformed response ratio (RR). The standard deviations of both treatment and control were included as a measure of variance:

$$RR = \ln(X_t/X_c) = \ln(X_t) - \ln(X_c) \quad (1)$$

where  $X_t$  and  $X_c$  are means of NO fluxes for the treated and control groups, respectively. Its variance ( $v$ ) is estimated as:

$$v = \frac{s_t^2}{n_t x_t^2} + \frac{s_c^2}{n_c x_c^2} \quad (2)$$

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

We conducted a weighted meta-analysis using RRs, where mean effect size for each category was calculated using a categorical random effects model. Groups with less than two paired observations were excluded from this study to meet the criteria for meta-analysis. The overall mean effect size and 95% confidential interval (CI) of each grouping category generated by bootstrapping (9999 iterations) were calculated with METAWIN version 2.0 statistical software (Rosenberg *et al.*, 2000). Mean effect sizes of  $X_t$  were considered to be significantly different from those of  $X_c$  if the 95% CIs did not overlap with the line  $RR = 0$ , and significantly different from one another if their 95% CIs did not overlap.

In addition to the meta-analysis procedure, fitting of data to Gaussian distribution function was carried out using the SIGMAPLOT version 12.0 Software (Systat Software, Inc., San Jose, CA, USA), and frequency distribution of RR was plotted to reflect variability of among individual studies with the following Gaussian function (i.e., normal distribution) (Luo & Zhou, 2006):

$$y = a \exp \frac{(x - \mu)^2}{2\sigma^2} \quad (3)$$

where  $y$  is the frequency of RR values within an interval,  $x$  is the mean of RR for the given interval,  $\mu$  and  $\sigma^2$  are the mean and variance across all RR values, respectively, and  $a$  is a coefficient indicating the expected number of RR at  $x = \mu$ . One-way ANOVA was performed to evaluate the differences in soil NO emissions and emission factors among ecosystem types, fertilizer sources and other experimental factors. We conducted a Pearson correlation to identify key factors controlling soil NO and N<sub>2</sub>O emissions.

### Sensitivity analyses

We performed sensitivity analyses to test the robustness of our meta-analysis on response of soil NO emissions to fertilizer and other experimental factors. We removed the outlier studies to perform the same meta-analysis procedure and compared the results with those of the original meta-analysis. Furthermore, we conducted the same meta-analysis procedure by excluding datasets without variances reported, and then repeated the comparisons with the results of original meta-analysis.

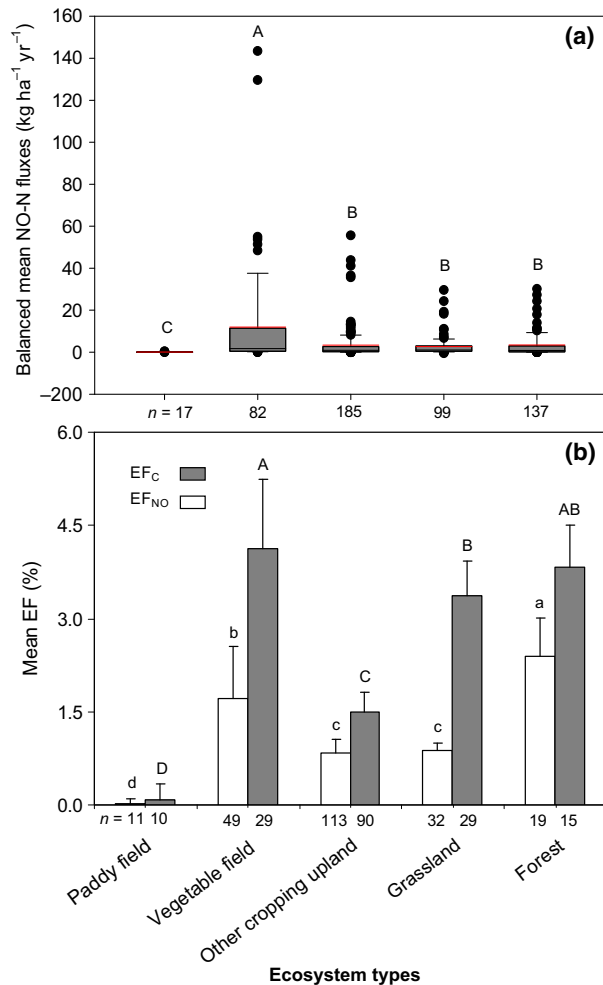
## Results

### Soil NO fluxes across ecosystems

When summarizing all field measurements, the global mean of soil NO-N fluxes was estimated to be 4.06 kg ha<sup>-1</sup> yr<sup>-1</sup>, and it significantly differed among ecosystem types (Fig. 2a). On average, soil NO-N fluxes were the greatest in vegetable croplands, with a grand balanced mean of 9.75 kg ha<sup>-1</sup> yr<sup>-1</sup>. The mean of soil NO-N fluxes was relatively comparable among other upland croplands (2.83 kg ha<sup>-1</sup> yr<sup>-1</sup>), grasslands (2.37 kg ha<sup>-1</sup> yr<sup>-1</sup>) and forests (2.74 kg ha<sup>-1</sup> yr<sup>-1</sup>), while paddy fields had the lowest NO-N fluxes (0.11 kg ha<sup>-1</sup> yr<sup>-1</sup>). For the controls without N fertilizer application, background emissions of soil NO-N averaged 0.05, 0.97, 1.02 and 1.24 kg ha<sup>-1</sup> yr<sup>-1</sup> for paddy field, vegetable field, other cropping upland and forest soils, respectively, while grasslands had the largest background emissions of soil NO-N (1.67 kg ha<sup>-1</sup> yr<sup>-1</sup>).

### Fertilizer effect on soil NO fluxes

The datasets were homogenous for fertilizer effect on soil NO fluxes with a normal distribution pattern of effect size in the meta-analysis (Fig. 3a). When averaged across all field measurements, N fertilization significantly increased NO emissions by 25%, with a 95% confidence interval (CI) of 20–32%, while NO response varied with N fertilizer types (Fig. 3b). When pooling data at different N fertilizer types, synthetic N fertilizer application induced the greatest increases in soil NO fluxes (SN, +38%), compared with organic N (ON, +20%) or mixed N fertilizer (Mixed, +18%). Relative to synthetic N fertilizer alone, synthetic N fertilizer combined with nitrification inhibitors substantially reduced soil NO emissions by 81% (SN+NI, Fig. 3c). Among chemical N fertilizer varieties, soil NO emissions had the largest response to ammonium nitrate (AN, +59%) and the smallest response to compound fertilizer (CF, +9%), relative to urea (U, +40%) or controlled-release urea (CRU: +36%).



**Fig. 2** Comparisons of balanced mean soil NO fluxes (a) and direct emission factors (b) of N fertilizer for NO ( $EF_{NO}$ ) and combined NO+N<sub>2</sub>O ( $EF_c$ ) among various ecosystem types. Different letters indicate significant difference in NO-N fluxes (uppercase letters in Fig. 2a),  $EF_{NO}$  (lowercase letters in Fig. 2b) and  $EF_c$  (uppercase letters in Fig. 2b) among ecosystem types at statistical probability level of 0.05 in ANOVA. In Fig. 2a, the red dashed and black solid lines, lower and upper edges and bars and black circles represent the mean and median values, 25th and 75th, 10th and 90th percentiles and outliers of all data, respectively. The mean direct emission factors with 1 standard deviation are shown in Fig. 2b. The number of measurements ( $n$ ) for each ecosystem type is shown next to the  $x$ -axis.

#### Direct emission factors of NO and combined NO+N<sub>2</sub>O

Soil NO and combined NO+N<sub>2</sub>O emissions were significantly and positively correlated with N fertilizer input rate when pooling all field flux measurements across fertilized soils (Table 1, NO-N:  $r = 0.78$ ,  $P < 0.001$ ; (NO+N<sub>2</sub>O)-N:  $r = 0.68$ ,  $P = 0.01$ ). When averaged all available data, the global mean direct emission factors of N fertilizer for NO ( $EF_{NO}$ ) and combined NO+N<sub>2</sub>O

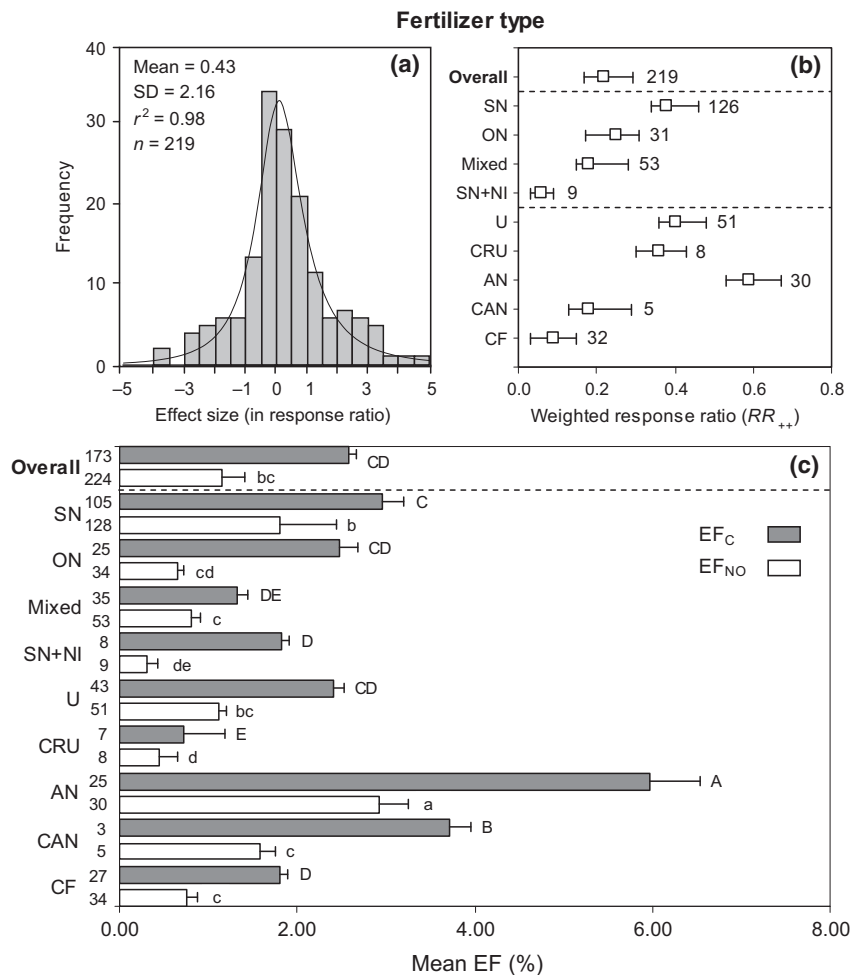
( $EF_c$ ) were estimated to be 1.16% and 2.58%, with CIs of 0.71–1.61% and 1.81–3.35%, respectively, while they were significantly different among various ecosystems and N fertilizer types (Table 2; Figs 2b and 3c). On average, the  $EF_{NO}$  was significantly greater in forests and vegetable fields relative to grasslands, but there was no significant difference in  $EF_c$  among them (Table 2). The  $EF_{NO}$  were significantly higher in forests (2.39%) than in grasslands (0.87%), while there was no significant difference in  $EF_c$  between them (3.82% vs. 3.36%). Within the croplands, vegetable fields showed the highest  $EF_{NO}$  (1.71%) and  $EF_c$  (4.13%), while the  $EF_{NO}$  (0.01%) and  $EF_c$  (0.08%) were the lowest in paddy fields.

The  $EF_{NO}$  was linearly and significantly correlated with  $EF_c$  (Fig. S1), suggesting that both NO and N<sub>2</sub>O emissions were simultaneously increased by N fertilizer application. However, both  $EF_{NO}$  and  $EF_c$  significantly differed with N fertilizer sources (Fig. 3c). Relative to organic N fertilizer, the  $EF_{NO}$  was significantly greater for synthetic N fertilizer, but there was no pronounced difference in  $EF_c$  between them. Relative to organic or synthetic N fertilizer application alone, the  $EF_c$  was significantly lower for organic N combined with synthetic N fertilizer (Mixed, Fig. 3c). Moreover, synthetic N fertilizer combined with nitrification inhibitors application greatly decreased  $EF_{NO}$  and  $EF_c$  by 82% and 39% as compared with synthetic N fertilizer being applied alone, respectively. Among chemical N fertilizer varieties, ammonium nitrate induced the highest  $EF_{NO}$  (2.93%) and  $EF_c$  (5.97%), while the  $EF_{NO}$  (0.45%) and  $EF_c$  (0.73%) were the lowest for controlled-release N fertilizer. On the other hand, the  $EF_{NO}$  was negatively correlated with soil pH (Fig. S2), suggesting that N fertilizer-induced NO emissions would be greater in soils with lower pH.

#### Soil NO and its link to N<sub>2</sub>O regulated by experimental and environmental factors

When averaged across all observations, soil NO emissions were significantly affected by experimental factors, including crop residue incorporation, drought, tillage, burning and atmospheric [CO<sub>2</sub>] enrichment (Fig. 4a). Soil NO emissions were significantly decreased by crop residue incorporation (−9%) and atmospheric [CO<sub>2</sub>] enrichment (−6%), in contrast to an increase in soil NO emissions due to biomass burning (+12%), experimental drought (+14%) or soil tillage (+30%).

Environmental factors such as climate and soil properties also played an important role in regulating soil NO emissions (Table 1; Fig. 4b). When pooling data at different climate zones, soil NO fluxes were the greatest in subtropical regions and the lowest in cool regions (Fig. 4b). Soil NO fluxes depended on soil properties,



**Fig. 3** Frequency distribution of fertilizer effect size among all observations (a) and soil NO emissions response to fertilizer sources (b), as well as direct emission factors of NO and NO+N<sub>2</sub>O partitioned by fertilizer sources (c). Symbols in Fig. 3b refer to the mean effect size with an interval of 95% confidence. The mean EF<sub>NO</sub> and EF<sub>C</sub> with 1 standard deviation are shown in Fig. 3c. Numerals following data points (Fig. 3b) or next to y-axis (Fig. 3c) indicate number of measurements. 'Overall' indicates the integrated response across fertilizer sources as compared to controls without fertilizer application. SN, synthetic N fertilizer; ON, organic N fertilizer; NI, nitrification inhibitor; U, urea; CRU, controlled-release urea; AN, ammonium nitrate; CAN, calcium ammonium nitrate; CF, compound fertilizer.

including soil pH, soil texture, soil organic carbon (SOC) content and soil C/N ratio. When experimental soils were divided into acid/neutral (pH  $\leq 7$ ) and alkaline soils (pH  $> 7$ ), soil NO emissions were significantly greater in acid or neutral soils than in alkaline soils (Fig. 4b). Soil NO fluxes depended on soil textures, decreasing from the coarse and medium soils to fine soils. Given that soil SOC content of 1.5% or soil C/N ratio of 10 was adopted as the threshold for further subgrouping, soil NO fluxes were greater in carbon-enriched soils (SOC content  $> 1.5\%$ ) relative to carbon-poor soils (SOC content  $< 1.5\%$ ), or in soils with lower C/N ratios ( $\leq 10$ ) relative to soils with higher C/N ratios ( $> 10$ ).

The NO-N/N<sub>2</sub>O-N or NO-N/(NO+N<sub>2</sub>O)-N ratio was negatively correlated with soil water content (WFPS, %)

(Table 1, Fig. 5a), suggesting that nitrification instead of denitrification might dominate soil NO emissions irrespective of soil or ecosystem type. In addition, soil NO fluxes were linearly increased with soil temperature (Table 1, Fig. 5b). The combined NO+N<sub>2</sub>O emissions were positively correlated with soil mineral N content (Table 1, Fig. 5c). However, no significant differences were found between cropping and noncropping soils in terms of above correlations. Across all observations, soil NO fluxes tended to increase with the extension of experimental duration (Fig. 5d), suggesting that soil NO emissions might have been underestimated in short-term field studies. For a systematic review, an overall schematic response of soil NO to its controlling factors is presented in Fig. 6.

**Table 1** Correlations of soil nitric oxide (NO) emission, combined NO and nitrous oxide (NO+N<sub>2</sub>O) emission and NO-N/(NO+N<sub>2</sub>O)-N ratio against soil and environmental variables

Variables	NO-N emission			(NO+N <sub>2</sub> O)-N emission			NO-N/(NO+N <sub>2</sub> O)-N ratio		
	<i>n</i>	<i>Corr. C.</i>	<i>P</i>	<i>n</i>	<i>Corr. C.</i>	<i>P</i>	<i>n</i>	<i>Corr. C.</i>	<i>P</i>
Soil temperature (°C)	488	0.69	<b>0.02</b>	353	0.13	0.12	353	0.01	0.87
Soil WFPS (%)	440	-0.09	0.15	334	0.04	0.52	334	-0.67	<b>0.01</b>
Soil pH	482	-0.64	<b>0.03</b>	350	-0.06	0.42	350	-0.78	<b>&lt;0.01</b>
Soil organic C (g kg <sup>-1</sup> )	395	0.01	0.61	292	0.12	0.21	292	0.01	0.93
Soil total N (g kg <sup>-1</sup> )	358	-0.02	0.43	260	0.09	0.32	260	0.12	0.19
Soil C/N ratio	340	-0.75	<b>0.01</b>	250	-0.71	<b>&lt;0.01</b>	250	0.07	0.50
Soil mineral N (mg kg <sup>-1</sup> )	373	0.89	<b>&lt;0.001</b>	312	0.82	<b>&lt;0.01</b>	312	0.03	0.64
N input rate (kg N ha <sup>-1</sup> )	358	0.78	<b>&lt;0.001</b>	283	0.68	<b>0.01</b>	283	0.04	0.41
Experimental duration (days)	520	0.62	<b>0.04</b>	382	0.06	0.56	382	-0.06	0.38

*n*, number of datasets included in the correlation analysis; *Corr. C.*, Pearson's correlation coefficients; *P*, *P*-value of correlation analysis and the values in bold indicate statistical significance at *P* < 0.05 probability level.

**Table 2** Mean direct emission factors of NO (EF<sub>NO</sub>), N<sub>2</sub>O (EF<sub>N<sub>2</sub>O</sub>) and combined NO+N<sub>2</sub>O (EF<sub>C</sub>) and their 95% confidence intervals (CIs)

Grouping category	NO			N <sub>2</sub> O			NO+N <sub>2</sub> O		
	<i>n</i>	EF <sub>NO</sub>	CI	<i>n</i>	EF <sub>N<sub>2</sub>O</sub>	CI	<i>n</i>	EF <sub>C</sub>	CI
Grand mean	224	1.16	0.71–1.61	173	1.42b	1.04–1.76	173	2.58b	1.81–3.35
Ecosystem type									
Forest	19	2.39a	0.12–5.56	15	1.43b	0.03–1.91	15	3.82a	0.42–7.22
Grassland	32	0.87c	0.47–1.27	29	2.49a	2.20–5.18	29	3.36ab	2.58–6.14
Other cropping upland	113	0.84c	0.31–1.37	90	0.66c	0.57–1.05	90	1.50c	0.72–2.28
Vegetable field	49	1.71b	0.48–2.94	29	2.42a	0.69–2.69	29	4.13a	1.20–7.06
Paddy field	11	0.01d	0–0.02	10	0.07d	0.04–1.00	10	0.08d	0.05–0.11
Fertilizer type									
SN	128	1.81a	1.23–2.39	105	1.15b	0.73–1.57	105	2.96a	2.10–3.82
ON	34	0.66bc	0–1.57	25	1.81a	0.58–2.86	25	2.47ab	0.79–4.15
Mixed	53	0.80b	0.52–1.08	35	0.52c	0.33–0.71	35	1.32c	0.98–1.66
SN+NI	9	0.31d	0.02–0.60	8	1.51ab	0–3.48	8	1.82b	0–3.86
Chemical N varieties									
U	51	1.12bc	0.06–2.18	43	1.29c	0.84–1.74	43	2.41cd	0.92–3.90
CRU	8	0.45d	0.05–0.85	7	0.28e	0–0.76	7	0.73e	0–1.49
AN	30	2.93a	0.87–4.55	25	3.04a	1.53–4.17	25	5.97a	3.57–8.21
CAN	5	1.58b	0.03–1.71	3	2.14b	0–10.41	3	3.72b	0–10.93
CF	34	0.75c	0.29–0.79	27	1.05cd	0.45–1.49	27	1.80d	0.79–2.43

*n*, number of observations; abbreviations for N fertilizer sources see Fig. 3; different lowercase letters indicate statistical significance at *P* < 0.05 probability level among subgroups within each category.

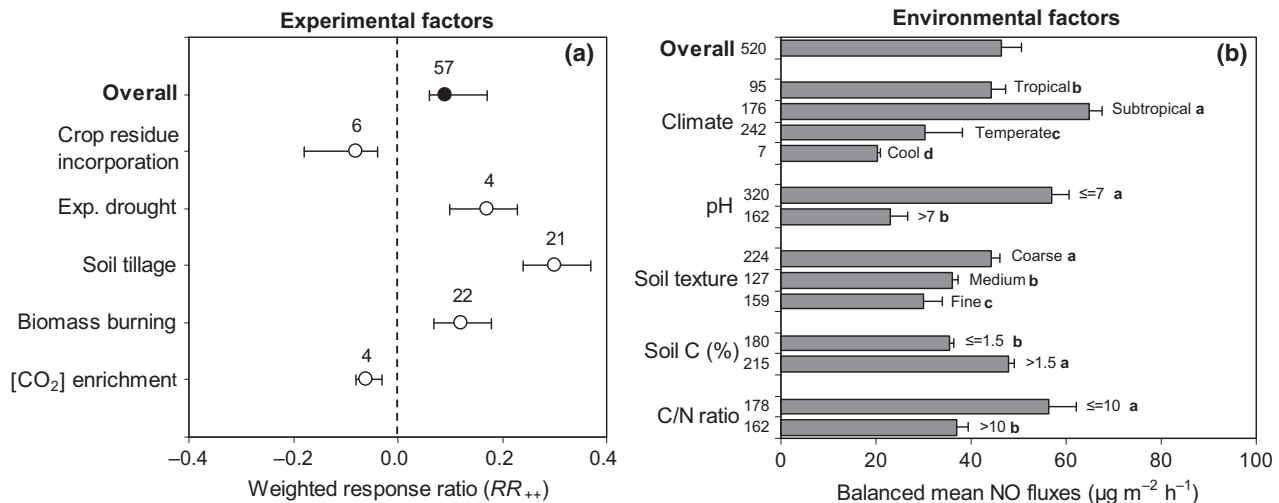
## Discussion

### Soil NO fluxes varying with land-use type

In this meta-analysis, the vegetable fields were highlighted from upland croplands category due to its substantially high NO emission rates. On average, soil NO fluxes were the greatest in vegetable fields and the lowest in paddy fields (Fig. 2a). The higher NO emissions from vegetable croplands have been recently

documented in field studies (Akiyama & Tsuruta, 2003; Mei *et al.*, 2009; Pang *et al.*, 2009; Deng *et al.*, 2012; Yao *et al.*, 2015). Typically, intensified vegetable production is characterized by excessive N fertilizer input rate, especially in greenhouse vegetable cropping systems to maintain high yield. It is generally believed that soil NO emissions were strongly correlated with N applied rates (Smith *et al.*, 1997; Bouwman *et al.*, 2002a), and thus, high N input rate would incur substantial soil NO emissions in vegetable croplands (Fang & Mu, 2006;





**Fig. 4** The effect of experimental (a) and environmental (b) factors on soil NO fluxes. The symbols in Fig. 4a refer to the weighted response ratio to experimental factors with an interval of 95% confidence. Numerals above symbols (Fig. 4a) or next to y-axis (Fig. 4b) indicate the number of measurements. 'Overall' refers to the integrated effect of all experimental factors across soils as compared with controls without relevant experimental manipulations. Different lowercase letters in Fig. 4b indicate significant pairwise differences in soil NO emissions among subgroups within a given category.

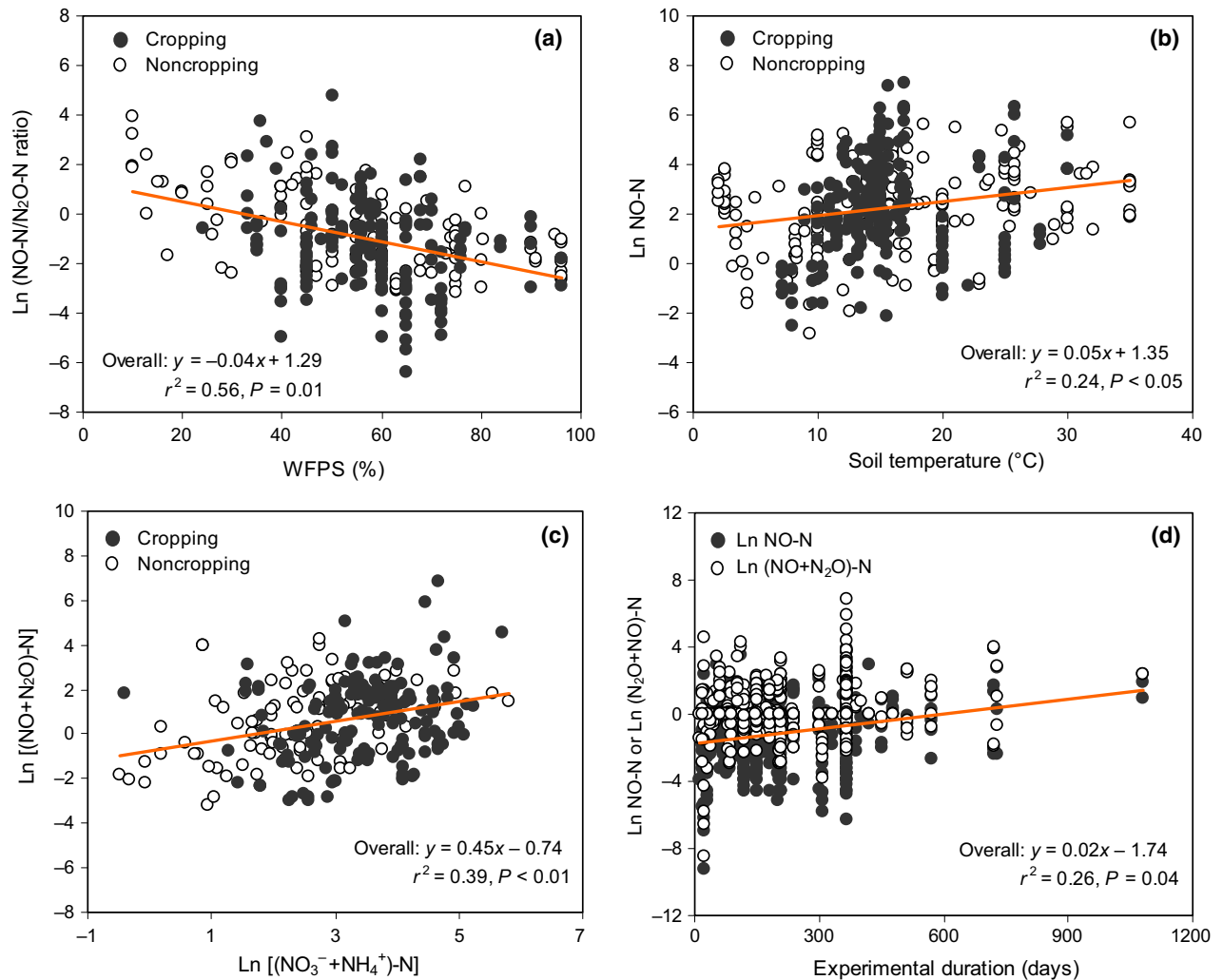
Yao *et al.*, 2015). In addition, higher soil NO fluxes might be also due to frequent irrigation in vegetable cropping fields because soil NO fluxes have been found to be greatest when dry soils are rewetted (Homyak *et al.*, 2016). Relative to vegetable croplands, soil NO fluxes were relatively lower in other upland croplands, mainly due to lower N fertilizer input rate and less irrigation in terms of per unit area. Soil NO fluxes were comparable between grasslands and the other upland croplands. Although synthetic N fertilizer is seldom applied in grasslands and forests, organic N input by animal waste could be occasionally used in such seminatural or natural ecosystems. Indeed, the 'mining' of soil N following land-use change from natural forest or grassland to cultivated agricultural land has been of great concern to its important role in global N budget (Davidson, 2009; Smith *et al.*, 2012).

Among all ecosystem types, paddy fields were shown to have the lowest NO source strength. The lowest NO fluxes in paddy fields might be closely related to the waterlogged soil conditions (Zhou *et al.*, 2010; Gaihre *et al.*, 2015; Zhao *et al.*, 2015). It is generally believed that soil NO is predominantly produced through microbial nitrification rather than denitrification (Russow *et al.*, 2008). The produced NO could be further reduced toward the formation of N<sub>2</sub> under poor aeration conditions due to waterlogging in paddy fields (Russow *et al.*, 2009).

#### Fertilizer effects on soil NO fluxes

When averaged across all fertilizer sources, fertilizer application significantly increased soil NO emissions by 25% (Fig. 4b). As reported in previous individual studies, linear or nonlinear positive responses of soil NO emissions to N fertilizer input have been recently documented (e.g., Shcherbak *et al.*, 2014; Zhao *et al.*, 2015). Synthetic N fertilizer relative to organic or mixed N fertilizer application was more effective at stimulating NO emissions in this meta-analysis, generally in agreement with previous field studies (Hayakawa *et al.*, 2009). However, Deng *et al.* (2012) reported that soil NO emissions were greater for manure relative to synthetic N fertilizer treatments in a vegetable cropping system. Nevertheless, optimizing fertilizer types, apart from cutting down N input amounts, would be an effective option for mitigating soil NO emissions, especially in agro-nomic cropping soils.

Synthetic N fertilizer combined with nitrification inhibitor or controlled-release N fertilizer application decreased soil NO emissions as compared with common synthetic N fertilizer application alone in this meta-analysis, although it is limited by few available data (Akiyama *et al.*, 2000; Cheng *et al.*, 2002; Hou & Tsuruta, 2003; Sanz-Cobena *et al.*, 2012). As proposed by Sánchez-Martín *et al.* (2010), the lowered pool of NH<sub>4</sub><sup>+</sup> in soils as a consequence of controlled-release N fertilizer can weaken the nitrification potential driving NO release.



**Fig. 5** Correlations of soil NO emissions, total NO and N<sub>2</sub>O emissions as well as NO-N/N<sub>2</sub>O-N ratio with soil moisture in water-filled pore space (WFPS) (a), temperature (b) and mineral nitrogen substrate (c) of the 0–20 cm soil depth and experimental duration (d).

Menéndez *et al.* (2009) found a significant effect of urease inhibitor on reducing NO emissions but greatly depended on soil water status. Among different chemical N fertilizer varieties, ammonium nitrate was most effective at increasing soil NO emissions. Presumably, direct mineral N substrate enrichment greatly facilitates soil NO release relative to other indirect N chemical sources, but more field verifications are needed.

#### *Direct emission factors of NO and combined NO+N<sub>2</sub>O*

The direct emission factor of fertilizer N for NO ( $EF_{NO}$ ) has been reported to show large variabilities, highly dependent on climate, soil conditions and site-specific management practices (Bouwman *et al.*, 2002a; Dobbie & Smith, 2003). By summarizing available data, the  $EF_{NO}$  was the greatest in forests among different ecosystem types (Fig. 2b), suggesting that N input has

a high risk of being lost as NO in forests (Venterea *et al.*, 2003). The soil pH might have played an important role in regulating  $EF_{NO}$  in different ecosystems. It is generally believed that nitrite is stable at soil pH >5.5 and most NO can be resulted from chemical decomposition (chemo-denitrification) of  $NO_2^-$  in acidic soils (Kesik *et al.*, 2006; Heil *et al.*, 2016). By examining the database of this study, soil pH was significantly lower in tropical/subtropical forests than in grasslands (4.6 vs. 6.8), and negative correlation of  $EF_{NO}$  with soil pH suggested that lower pH would benefit for higher N fertilizer-induced soil NO emissions from forests (Table 1; Figs 2 and S2). Overall, the forest soils subjected to long-term atmospheric N deposition would result in high ratio of anthropogenic N loss as NO (Venterea *et al.*, 2003).

The global mean  $EF_{NO}$  was estimated to be 1.16%, with a CI of 0.71–1.61%, in this study. This estimate

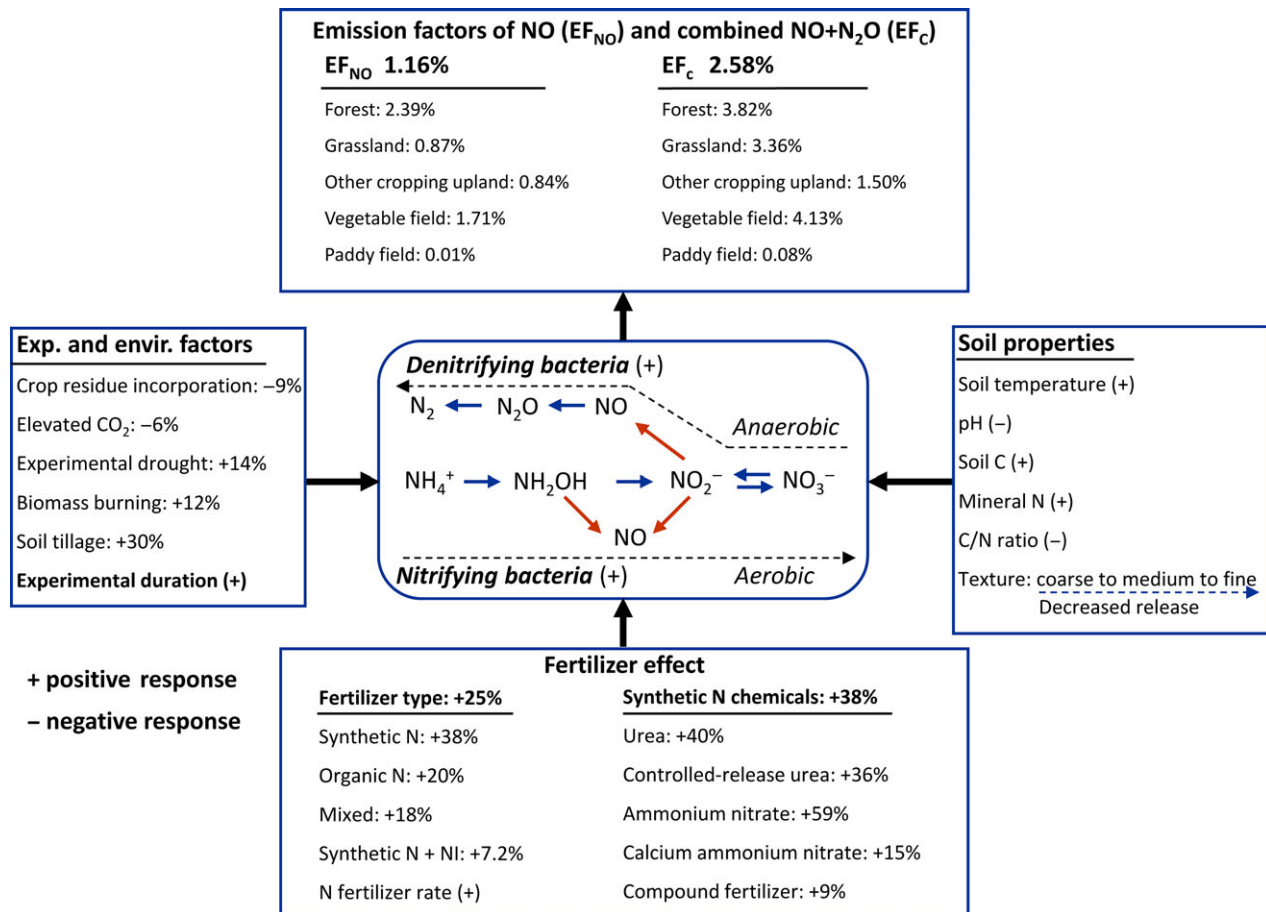


Fig. 6 A summary panel of soil NO responses to environmental and experimental factors and the direct emission factors of N fertilizer for NO and combined NO+N<sub>2</sub>O.

based on updated data samples, particularly including new 81 recent measurements taken from vegetable cropping fields, is obviously greater than the previous estimates of 0.55% or 0.71% for global agriculture that were based on limited available data (Bouwman *et al.*, 2002b; Yan *et al.*, 2003b, 2005; Stehfest & Bouwman, 2006). When averaged across studies within croplands, vegetable fields had the greatest  $EF_{NO}$  (1.71%) and  $EF_c$  (4.13%), which would be largely associated with drying–rewetting episodes created by frequent irrigation in vegetable cropping systems. The  $EF_{NO}$  was significantly greater in vegetable fields than in grasslands, but  $EF_c$  did not differ between them, suggesting that relatively higher NO-N/N<sub>2</sub>O-N ratio would be obtained in vegetable fields with larger N input rate (Zhang *et al.*, 2016). In addition, the  $EF_{NO}$  was negligible (mean: 0.01%) in paddy fields, mainly ascribed to negligible NO emissions from paddy soils under waterlogging conditions (Zhou *et al.*, 2010).

When averaged 173 field simultaneous measurements of NO and N<sub>2</sub>O fluxes, the direct emission factor

of combined NO+N<sub>2</sub>O ( $EF_c$ ) was estimated to be 2.58%, with a CI of 1.81–3.35%. This value of  $EF_c$  is greater than the sum of global mean direct emission factor of NO (1.16%) estimated in this meta-analysis and the global IPCC default emission factor of 1.0% for N<sub>2</sub>O (IPCC, 2006). In this meta-analysis, the direct emission factor of fertilizer N for soil N<sub>2</sub>O ( $EF_{N_2O}$ ) was estimated to be 1.42%, with a CI of 1.04–1.76%, higher than the global IPCC default value of 1.0%, derived from over 1000 field N<sub>2</sub>O flux measurements prior to the year of 2006 (IPCC, 2006). The higher  $EF_{N_2O}$  in this meta-analysis was largely contributed by recent flux measurements showing much greater fertilizer-induced N<sub>2</sub>O emissions from grassland and vegetable soils (Dataset S1, Table 2). Our mean  $EF_{N_2O}$  estimates in fertilized grasslands (2.49%) and vegetable cropping systems (2.42%) were highly close to the global top-down mean  $EF_{N_2O}$  estimates of 2.5% over the period of 1860–2005 (Davidson, 2009). Indeed, the global IPCC bottom-up  $EF_{N_2O}$  of 1% has been shown to be much lower than some recent regional/global top-down estimates of 3–5% (Crutzen

*et al.*, 2008; Davidson, 2009; Smith *et al.*, 2012; Griffis *et al.*, 2013) or bottom-up estimates of 1.75–1.8% (Grace *et al.*, 2011; Griffis *et al.*, 2013; Shcherbak *et al.*, 2014).

This meta-analysis primarily focused on fertilizer-induced NO and combined NO and N<sub>2</sub>O emissions; the estimation of EF<sub>N<sub>2</sub>O</sub> was only based on much smaller sample size (173 field simultaneous measurements of NO and N<sub>2</sub>O fluxes). Some EF<sub>NO</sub> and EF<sub>C</sub> estimates were also limited by few available data in this study and deserve to be further examined with more field measurements (Fig. 2b). Nevertheless, this study first attempted to estimate the bottom-up EFs in forest and grassland given that land-use change from natural forest or grassland to cultivated agricultural land could constitute an important source of global N<sub>2</sub>O (Davidson, 2009). In particular, the fertilizer N-induced total emissions of NO and N<sub>2</sub>O should be of more concern in future studies in terms of their interrelation in soil N cycling.

#### *Dependence of soil NO emission on experimental and environmental factors*

We also examined the response of soil NO emissions to other experimental factors apart from fertilizer application (Fig. 6a). When averaged across all observations, available experimental factors had significant effects on NO emissions, but the size of effect substantially differed. Of which, crop residue incorporation and atmospheric [CO<sub>2</sub>] enrichment significantly decreased soil NO emissions, in contrast to significant positive responses to experimental drought, soil tillage and aboveground biomass burning. The negative response of soil NO emissions to crop residue incorporation may be largely attributed to the declined soil oxygen availability for nitrification following crop residue amendment, dependent on specific crop residue type and its C/N ratio (Yao *et al.*, 2009; Zhang *et al.*, 2011). The negative response of soil NO fluxes to elevated [CO<sub>2</sub>] was associated with the highly enhanced N uptake by plants grown under elevated [CO<sub>2</sub>], which would decrease mineral N availability to soil microbes (Hungate *et al.*, 1997; Hu *et al.*, 2001). Moreover, the decreased surface soil water content under elevated [CO<sub>2</sub>] could also constitute a key limiting factor for soil NO emissions (Mosier *et al.*, 2002). However, the current data on soil NO emissions in response to elevated [CO<sub>2</sub>] are only limited in grassland soils, and more individual evidence in other extensive ecosystem types is anticipated.

Experimental drought significantly increased soil NO emissions in this meta-analysis. As suggested by Davidson *et al.* (2008), reduced precipitation may have important feedback effects on climate change by altering soil-atmospheric N oxide gas emissions. It has also

been found that dry soil conditions tended to facilitate soil NO emissions over N<sub>2</sub>O emissions (Firestone & Davidson, 1989; Davidson *et al.*, 2000). In addition, soil N availability to nitrifying bacteria may be improved in drought-exposed soils, subjected to site-specific soil characteristics (Goldberg & Gebauer, 2009).

Soil tillage relative to no-tillage practice significantly increased soil NO emissions, and our datasets generally presented uniform positive responses in this analysis. The improved soil aeration following tillage events would greatly increase oxygen availability favoring nitrification that dominates soil NO production (Sanhueza *et al.*, 1994; Yamulki & Jarvis, 2002; Yao *et al.*, 2009). Nevertheless, soil tillage effects on soil N oxide emissions were dependent on soil textures, where coarse- and medium- vs. fine-textured soils may weaken nitrification potential in tilled soils (Mummey *et al.*, 1998).

Biomass burning, as a typical management practice in chaparral and tropical savannas, was found to have a significant positive effect on soil NO emissions in this analysis. The burning-induced postfire pulse emissions of NO have been often documented (e.g., Williams *et al.*, 1992; Poth *et al.*, 1995). As suggested by Levine *et al.* (1996), for example, the increased ammonium substrate for nitrification may account for the stimulated NO emissions following burning of combustible matter. However, the extent of stimulation substantially varied with original soil moisture conditions and vegetation cover types (Anderson & Poth, 1998).

Soil NO emissions also depended on the environmental parameters (Fig. 4b). The subtropical soils presented to most facilitate NO emissions, relative to the lowest NO release rate in cool regions. Our meta-analysis results are well in line with the modeling estimates by Yan *et al.* (2005), showing that the highest NO emissions occurred in subtropical regions with relatively dry climates favoring soil nitrification. Among soils with different pH values, the acid or neutral relative to alkaline soil conditions significantly increased soil NO emissions. Low soil pH could inhibit NO reduction to N<sub>2</sub>O (Simek & Cooper, 2002), or low soil pH may promote chemical decomposition of nitrite, yielding NO as the primary product (McKenney *et al.*, 1990). As reviewed by Bouwman *et al.* (2002b), aerobic soil conditions may be more easily reached and maintained for longer periods within aggregates in coarse- than in fine-textured soils, which would in turn facilitate soil NO release with improved oxygen availability and gas diffusion. Besides, soils with relatively higher soil C contents but lower C/N ratios were observed favorable for NO emissions. Soils rich in SOC but with low C/N ratios tend to create more available C and N substrate for nitrifiers or denitrifiers driving NO production.

*Future studies on soil NO emission*

This meta-analysis provided an insight into fertilizer-induced soil NO emissions and the link to N<sub>2</sub>O emissions across various ecosystems, soil types and climatic regions (Fig. 6). However, most field studies were limited within seasonal or annual timescale, and there is an urgent need for continuous long-term (multiyear) measurements to elucidate the temporal variation. In particular, the continuous measurements are extremely limited in forest and grassland ecosystems and few measurements of NO fluxes from croplands are available in South America, India and South-East Asia, directly leading to large uncertainties existed for the current available bottom-up estimates of NO emissions from related soils. On the other hand, although few studies have examined indirect NO emissions or overall accounting of both indirect NO and N<sub>2</sub>O emissions from N leaching and runoff in agro-ecosystems, more field measurements at expanded geographic range should be strengthened in future work due to much anthropogenic N loss through the above two major pathways.

Besides field measurements, more condition-controlled experiments are still needed to further explore the mechanisms controlling soil NO emissions, such as closely linking it to the microbial analysis of abundance dynamics and composition of functional microbes involved in NO production from soils. In addition, more credible statistic or process-oriented models should be developed based on updated data sources to minimize the current large uncertainties in global or regional soil NO estimates. Some options such as mixed N fertilizer, synthetic N fertilizer combined with nitrification inhibitors instead of synthetic N fertilizer alone, decreasing N fertilizer application rate and low irrigation frequency could be adopted to mitigate soil NO emissions.

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## Supporting Information

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

**Table S1** Locations of field studies included in this systematic analysis.

**Table S2** Variables involved in this study.

**Figure S1** Correlation of soil NO emission factors (EF<sub>NO</sub>) with those of N<sub>2</sub>O (EF<sub>N<sub>2</sub>O</sub>).

**Figure S2** Dependence of soil NO emission factors (EF<sub>NO</sub>) on soil pH.

**Dataset S1** Dataset of 520 field NO flux measurements compiled from 114 publications.