DR. LONGLONG XIA (Orcid ID : 0000-0003-4026-4265)
MR. BENJAMIN WOLF (Orcid ID : 0000-0002-2636-1897)
DR. RALF KIESE (Orcid ID : 0000-0002-2814-4888)

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Title: Trade-offs between soil carbon sequestration and reactive nitrogen losses under straw return in global agroecosystems

Running head: Effects of straw return on Nr losses

Longlong Xia[a,b*], Shu Kee Lam[c], Benjamin Wolf[b], Ralf Kiese[b], Deli Chen[c], Klaus Butterbach-Bahl[b*]

[a.] State Key Laboratory of Soil and Sustainable Agriculture, Institute of Soil Science, Chinese Academy of Sciences, Nanjing 210008, China.

[b.] Institute for Meteorology and Climate Research (IMK-IFU), Karlsruhe Institute of Technology, Garmisch-Partenkirchen 82467, Germany.

[c.] School of Agriculture and Food, Faculty of Veterinary and Agricultural Sciences, the University of Melbourne, Victoria 3010, Australia.

[*]*Corresponding author*: Longlong Xia, Phone: +86 025 86881050, Fax: +86 025 86881000, Email: llxia@issas.ac.cn; Klaus Butterbach-Bahl, Phone: +49 8821183136, Fax: +49 8821183294, Email: klaus.butterbach-bahl@kit.edu

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Abstract: It is widely recommended that crop straw be returned to croplands to maintain or increase soil carbon (C) storage in arable soils. However, because C and nitrogen (N) biogeochemical cycles are closely coupled, straw return may also affect soil reactive N (Nr) losses, but these effects remain uncertain, especially in terms of the interactions between soil C sequestration and Nr losses under straw addition. Here, we conducted a global meta-analysis using 363 publications to assess the overall effects of straw return on soil Nr losses, C sequestration and crop productivity in agroecosystems. Our results show that on average, compared to mineral N fertilization, straw return with same amount of mineral N fertilizer significantly increased soil organic C (SOC) content (14.9%), crop yield (5.1%) and crop N uptake (10.9%). Moreover, Nr losses in the form of nitrous oxide (N_2O) emissions from rice paddies (17.3%), N leaching (8.7%) and runoff (25.6%) were significantly reduced, mainly due to enhanced microbial N immobilization. However, N_2O emissions from upland fields (21.5%) and ammonia (NH_3) emissions (17.0%) significantly increased following straw return, mainly due to the stimulation of nitrification/denitrification and soil urease activity. The increase in NH_3 and N_2O emissions was significantly and negatively correlated with straw C/N ratio and soil clay content. Regarding the interactions between C sequestration and Nr losses, the increase in SOC content following straw return was significantly and positively correlated with the decrease in N leaching and runoff. However, at a global scale, straw return increased net Nr losses from both rice and upland fields due to a greater stimulation of NH_3 emissions than reduction in N leaching and runoff. The trade-offs between increased net Nr losses and soil C sequestration highlight the importance of reasonably managing straw return to soils to limit NH_3 emissions without decreasing associated C sequestration potential.

Introduction

Globally, the annual production of crop straw reached approximately 4 billion metric tons at the beginning of the 21st century (Lal, 2005). Partial or full retention of crop straw is an effective and economically sound management practice to maintain or increase soil carbon (C) sequestration in arable soils (Powlson *et al.*, 2008; Liu *et al.*, 2014), although the effects depend on straw quality (e.g., C/N ratio) and quantity (Lal *et al.*, 2004; Smith *et al.*, 2008; Lugato *et al.*, 2018). Based on an extensive review of soil analyses, Zhao *et al.* (2018) estimated that the average soil organic C (SOC) stock in the topsoil (0−20 cm) of China's croplands increased from 1980 to 2011 at a rate of 140 kg C ha^{-1} yr^{-1}. Moreover, they concluded that straw return contributed to approximately 40% of the increment. A meta-analysis conducted by Liu *et al.* (2014) demonstrated that straw return significantly increased the average SOC content in global croplands by 12.8% (0−15 cm soil layer). Straw return may also improve soil fertility by supplying mineral elements and increasing water retention, thus improving crop yields (Majumder *et al.*, 2008; Singh *et al.*, 2008).

Because C and nitrogen (N) biogeochemical cycles are closely coupled (Luo *et al.*, 2006), straw return also affects soil N dynamics and associated biosphere-atmosphere-hydrosphere exchange processes of reactive N (Nr: all N species except dinitrogen). These effects depend on straw quality (e.g., C/N ratio) and soil properties (e.g., texture and clay content) (Miller *et al.*, 2008; Chen *et al.*, 2013). Generally, crop straw with a low C/N ratio (<30) is easily decomposed by soil microbes, and the mineralized N becomes available for nitrification, denitrification and hydrological losses (Frimpong & Baggs, 2010). Associated with these processes is increased production of nitrous oxide (N_2O) (Butterbach-Bahl *et al.*, 2013), dinitrogen and loss of N compounds such as nitrate and dissolved organic nitrogen via leaching (Hagedorn *et al.*, 1997). However, returning crop straw with a high C/N ratio (>30) might stimulate microbial immobilization of soil ammonium (NH_4^+) and nitrate (NO_3^-) (Aulakh *et al.*, 2001; Cheng *et al.*, 2017), thereby decreasing Nr loss through gaseous (N_2O and ammonia (NH_3) emissions) and hydrological pathways (N leaching and runoff) (Xia *et al.*, 2017).

The effects of straw return on soil Nr losses can also be regulated by soil properties. For example, soil texture and clay content determine pore size distribution, and thus soil aeration and oxygen availability for straw decomposition, which in turn controls the intensities of different soil N transformations and associated Nr losses (Skiba & Ball, 2002; Chen *et al.*, 2013). Moreover, management practices, such as lowland (e.g., rice) and upland arable cropping, mineral N fertilization rate and straw application method (surface application or incorporation), as well as climate conditions (temperature and precipitation), can also regulate the response of soil Nr losses to straw return by affecting straw decomposition and soil N transformations (Butterbach-Bahl *et al.*, 2013; Liu *et al.*, 2017). However, the intertwined response of various Nr losses (also C sequestration) to straw return in croplands under varying soil properties, straw quality and quantity, agricultural management and climate conditions, have not been comprehensively explored and documented.

Studies that have examined the effects of straw return on environmental Nr losses often focused on N_2O emissions (Miller *et al.*, 2008; Chen *et al.*, 2013; Shan & Yan, 2013) and not other Nr losses (e.g., NH_3 emissions, N leaching and runoff). However, the high complexity of cropland Nr dynamics highlights the importance of evaluating the net balance of Nr losses rather than focusing on a single Nr loss (Xia *et al.*, 2017), because the formation processes and regulatory conditions of Nr losses do vary substantially (e.g., biological process for N_2O production and physical-chemical processes for N leaching and runoff) (Hagedorn *et al.*, 1997; Davidson *et al.*, 2000).

Besides, to our knowledge, no comprehensive studies have explored the interactions between various Nr loss pathways and soil C sequestration following straw return, although a few studies have shown that Nr losses are closely coupled with C sequestration in agroecosystems (Li *et al.*, 2005; Liu *et al.*, 2014). For example, the accumulation of SOC following straw addition is largely derived from the greater formation of soil macroaggregates (Six *et al.*, 2002), which can in turn reduce the occurrence of soil erosion with rainfall and may consequently reduce Nr losses via hydrological pathways (Blanco-Canqui & Lal, 2009) but may on the other hand regulate soil N_2O emissions (Li *et al.*, 2005). This scenario highlights the necessity to consider the interactions between Nr losses and SOC sequestration to assess the overall environmental consequences of straw return to soils.

Here, we show the results of a global meta-analysis that was based on 363 peer-reviewed publications and aimed to (1) evaluate the overall effects of straw return on various Nr loss pathways (NH_3 emissions, N_2O emissions, N leaching and runoff), crop productivity and soil C sequestration in agroecosystems under different environmental and management conditions (straw quality and quantity, soil properties, agricultural management and climate conditions); (2) explore the interactions between various Nr losses and SOC sequestration following straw return; and (3) estimate how straw return has affected Nr losses from global croplands and the overall cropland Nr balance for the period 2005−2015.

Materials and methods

Selection criteria and database

We used several databases such as Web of Science, Google Scholar, China National Knowledge Infrastructure database, China Wanfang Data, Current Contents Connect (ISI), Academic Search complete (EBSCO), Scopus and CAB Abstracts to search peer-reviewed publications (before August 2018) related to the effects of straw return on various Nr losses, crop productivity and soil C sequestration. The keywords used in the search included ‘crop straw or crop residue or crop stubble’, ‘Nr losses (NH_3 emissions, N_2O emissions, N leaching and runoff), SOC content, crop productivities (crop yield, crop N uptake and N use efficiency (NUE)) and/or other soil properties’. A study had to meet the following criteria to be included in this meta-analysis: a) the control (mineral N fertilization) and straw treatment (mineral N fertilization plus straw return) received equal mineral N fertilization rates, which indicated that the straw treatment provided additional straw N supply to soils; b) publications needed to report on at least one of the target variables and sample sizes for the control and treatment plots. Multiple observations that were conducted at the same experimental site over several sampling years were averaged; and c) the observation duration of the experiment must have covered the main discharge period of various Nr losses. Applying these criteria, a total of 363 peer-reviewed publications reporting results from global agroecosystems were selected for further analyses (Fig. S1).

The effects of straw return were evaluated under the following three categories: (1) Nr losses: NH_3 and N_2O emissions, N leaching and N runoff; (2) crop productivity: crop yield, crop N uptake and NUE; (3) SOC content and other soil properties (0−15 cm soil layer): soil total N content, soil microbial biomass N (MBN), soil microbial biomass C (MBC), dissolved organic carbon (DOC), soil labile carbon (LOC); soil NH_4^+, NO_3^- content and NH_4^+/NO_3^- ratio; soil available N, P, and K; cation exchange capacity, soil porosity, soil pH, soil urease activity, soil water content and crop water use efficiency. Crop N uptake refers to total aboveground N uptake. The NUE, i.e., fertilizer apparent N recovery, was calculated by crop N uptake of fertilized plots minus N uptake of nonfertilized plots and then divided by mineral fertilizer N rate (Congreves & Van Eerd, 2015). The effects of straw return were further categorized according to soil properties (soil clay content, texture, initial SOC and N contents, and soil pH), straw quality (straw C/N ratio) and quantity (straw input rate, straw N and C input rate), crop species, mineral N fertilizer rates, duration of straw return, straw application method and climate zones. Soil textures were classified based on the USDA soil texture classification system.

Meta-analysis

The effects of straw return on the variables (X) were quantified by the natural log of the response ratio (ln*RR*) using the following equation (Hedges *et al.*, 1999):

$$\ln RR = \ln (X_t/X_c) \qquad (1)$$

where X_t and X_c represent the mean of the treatment and control groups for variable X, respectively. The results are presented as the percentage of changes ((RR-1)×100) in the variables under straw return. Positive percentage changes denote an increase due to straw return whereas negative values indicate a decrease in the respective variables.

In previous meta-analyses, the effect sizes were generally weighted by the inverse of the pooled variance (Liu *et al.*, 2017) or replication (Lam *et al.*, 2012; Xia *et al.*, 2016), depending on the availability of standard deviations reported in the included studies. Most studies included in our database did not report the standard deviations of the mean values. In addition, there is a risk of generating extreme weights when weighting by the variance-based function, which is not the case for the replication-based method (van Groenigen *et al.*, 2011). Therefore, we adopted the replication-based weighting method in this meta-analysis using the following equation:

$$\text{weight} = (n_t \times n_c)/(n_t + n_c) \qquad (2)$$

where n_t and n_c denote the number of replicates of the treatment and control, respectively.

Mean effect sizes and the 95% confidence intervals (CIs) were generated by a bootstrapping procedure with 4999 iterations, using MetaWin 2.1 (Rosenberg *et al.*, 2000). Effects of straw return were considered significant if the 95% CIs did not overlap with zero. The means of the categorical variables were considered significantly different from each other if their 95% CIs did not overlap.

Net changes in Nr losses induced by straw return

To evaluate the effects of straw return on net changes in Nr losses, we attempted to quantify Nr losses from global croplands (rice and upland fields) for the period of 2005−2015. The amount of Nr losses (e.g., NH_3 emission, Gg N yr^{-1}) under straw return was calculated using the following empirical model:

$$NH_3\ \text{emission}_{\text{straw-induced}} = N_{\text{rate}} \times P \times F_{NH3} \times E \qquad (3)$$

where N_{rate} (Gg N yr^{-1}) denotes the rate of mineral N fertilizer applied to rice (14745.8 Gg N yr^{-1}) or upland fields (82265.8 Gg N yr^{-1}) during 2005−2015, which was derived from the FAO database; P denotes the proportion of the global harvested cropland area receiving straw return; F_{NH3} denotes the fraction of mineral N fertilizer that is lost to NH_3; and E denotes the effects of straw return on NH_3 emissions (Table 1). Because there are no data available for the proportion (P) of global cropland area receiving straw return, we calculated for different scenarios for P (S1, P=20%; S2, P=40%; S3, P=60%; and S4, P=80%). Straw-induced N_2O emissions, N leaching and runoff for the paddy field and uplands were calculated using their corresponding F and E values (Table 1). F-N_2O was derived from a recent global meta-analysis conducted by Liu *et al.* (2017). F-NH_3, F-$N_{leaching}$ and F-N_{runoff} were derived from a global literature synthesis (Table 1), which is described in detail in the Supporting Information (SI).

Results

Impacts of straw return on SOC content and other soil properties

Across all studies, straw return significantly increased SOC content by 14.9% (0−15 cm soil layer) (n=246) (Fig. 1), with an increase of 11.4% (n=86) for rice paddies and 17.0% (n=160) for upland fields (Table S2). The SOC content increased significantly with an increasing straw addition rate ($P<0.001$) and straw C input rate ($P<0.001$) (Table S1). For straw quality, the increase in SOC content is greater with a straw C/N ratio larger than 30 (e.g., cereal straws) (15.1%, n=219), compared to a smaller ratio (C/N ratio<30, e.g., legume straws) (10.3%, n=25) (Table S2). With regard to agricultural management, the effect of straw return on SOC content is similar for different mineral N fertilization rates and application methods (surface application versus incorporated), but a long-term (≥4 years) straw addition resulted in significantly higher C sequestration (27.7%, n=28) than a short-term addition (13.4%, n=213). For soil texture and clay content, straw return resulted in the highest C sequestration in silt loamy soils (21.0%, n=59) or soils with clay content between 20 and 40% (18.2%, n=127, Table S2). Climate conditions (temperature, precipitation and climate zones) had no significant impact on the responses of SOC content to straw return (Table S1).

Straw return also significantly increased the content of dissolved organic C (DOC, 22.4%, n=54), labile organic carbon (LOC, 21.2%, n=36), total N (9.8%, n=171), and other nutrients such as available N (14.1%, n=138), phosphorus (P, 10.4%, n=144) and potassium (K, 17.8%, n=148) (Fig. 1). The increase in nutrients availability was associated with an increase in microbial biomass C and N (MBC, 37.3%, n=101; MBN, 38.4%, n=80) (Fig. 1 and Table S3). In addition, straw return also increased soil aeration, as indicated by higher soil porosity (7.5%, n=28) and lower soil bulk density (6.1%, n=81). Besides, straw return significantly increased soil urease activity by 18.5% (n=133) (Fig. 1 and Table S4), which governs Nr loss through NH_3 emissions in agricultural soils. However, soil pH was not significantly affected by straw return (-0.6%, n=90), regardless of crop type (rice paddies, -0.9%, n=37; upland crops, -0.4%, n=53) (Table S5 and see SI for further details).

Impacts of straw return on crop productivity

Overall, straw return significantly increased crop yield by 5.1% (n=636) in global agroecosystems (Fig. 2), with a similar increase of 5.3% (n=214) for rice paddies and 4.9% (n=422) for upland crops. The increase in crop yield was significantly and positively correlated with mineral N fertilization rate ($P<0.01$), straw N input rate ($P<0.01$) and mean annual temperature (MAT) ($P<0.001$) (Table 2). However, soil properties (SOC and N contents, clay content and pH) and straw C/N ratio had no significant effects on the response of crop yield to straw return (Table 2 and Table S6), except that significantly larger yield increases were observed at sites with a sandy rather than loamy and clay texture (Table S6). In addition, long-term practice of straw return (≥4 years) can also result in greater increases in yields (Fig. 2).

On average, crop N uptake and fertilizer NUE were significantly increased by 10.9% (n=157) and 15.0% (n=100) under straw return, respectively, with a similar increase for rice paddies and upland crops (Fig. 2). The increases in crop N uptake and fertilizer NUE were significantly and positively correlated with mean annual precipitation (MAP) (both $P<0.05$) and SOC content ($P<0.05$ for N uptake and $P<0.001$ for NUE) (Table 2). The increase in crop N uptake also significantly increased with straw N input rate ($P<0.001$) but significantly decreased with straw

C/N ratio (*P*<0.001) (Table 2). However, mineral N fertilization rate, soil total N and clay contents, soil pH and MAT were not significantly correlated with crop N uptake or NUE changes (Table 2). The application method and duration also had no significant impact on the response of N uptake or NUE to straw return (Fig. 2).

Impacts of straw return on gaseous Nr losses

On average, straw return significantly increased NH_3 emissions by 17.0% (n=116); the positive effect was smaller for rice paddies (11.4%, n=35) than for upland crops (20.3%, n=81) (Fig.3). Crop type significantly regulated the effects of straw return on soil N_2O emissions, which significantly decreased by 17.3% (n=82) for rice paddies but significantly increased by 21.5% (n=196) for upland crops (Fig. 3). The increases in NH_3 and N_2O emissions were both positively correlated with straw N input rate (both $P<0.001$) (Table 2) but negatively correlated with straw C/N ratio ($P<0.01$ for NH_3 and $P<0.001$ for N_2O) and soil clay content ($P<0.05$ for NH_3 and $P<0.01$ for N_2O) (Fig. 4 and Fig. 6). As shown in Figure 3, the largest increase in NH_3 and N_2O emissions occurred when returning straw with a C/N ratio<30 (e.g., legume straws) or to soils with a clay content<20%, whereas applying straw to soils with higher soil clay contents (>40%) or with a larger straw C/N ratio (>30) (e.g., cereal straws) did not stimulate NH_3 emissions, or even significantly decreased soil N_2O emissions.

In terms of soil texture, returning straw to sandy soils resulted in much higher increases in NH_3 (60.9%, n=11) and N_2O emissions (119.3%, n=4) compared to that for loamy (12.7% and n=51 for NH_3, and 3.9% and n=58 for N_2O) and clay soils (4.3% and n=16 for NH_3, and -10.4% and n=34 for N_2O) (Table 3). Neither mineral N fertilization rate nor other soil properties (total N and SOC contents and pH) were significantly correlated with NH_3 and N_2O emissions increases (Table 2). In addition, straw return stimulated NH_3 and N_2O emissions more at the sites with a warmer climate (e.g., warm temperate and subtropical) than those located in a cool temperate zone (Fig. 3).

Impacts of straw return on hydrological Nr losses

On average, straw return significantly decreased N leaching by 8.7% (n=60) and runoff by 25.6% (n=52), with no significant difference between rice paddies and upland crops (Fig. 5). The decreases in N leaching and runoff both positively correlated with straw C/N ratio and soil clay content, although these relationships were not significant (P>0.05) (Fig. 4 and Fig. 6). Other factors, such as the rate of mineral N fertilization, soil properties (total N and SOC contents and pH), or climate conditions (MAT and MAP) did not significantly impact the effects of straw return on hydrological N losses (Table 2). However, the decrease in N leaching was negatively correlated with straw addition rate and straw N input rate (both P<0.05). It is noteworthy that the negative effect of straw return on N leaching and runoff was significantly higher if straw was applied to the soil surface (N leaching: -26.2%, n=5; N runoff: -33.5%, n=28) and not incorporated into soils (N leaching: -6.8%, n=55; N runoff: -15.1%, n=24) (Fig. 5).

Relationships between SOC content and Nr losses changes under straw return

As shown in Figure 7, the decrease in the four Nr losses all showed a positive linear relationship with the increase in SOC content induced by straw return, i.e., higher soil SOC contents were accompanied by a greater reduction in Nr losses. However, this linear relationship was only significant for N leaching (R^2=0.37, P<0.05, n=14) and N runoff (R^2=0.52, P<0.05, n=9) but not for NH_3 (R^2=0.15, P>0.05, n=9) or N_2O emissions (R^2=0.09, P>0.05, n=29).

Straw return-induced net changes in Nr losses at a global scale

Assuming that 40% of global croplands receive straw return, global NH_3 emissions would increase by 132 Gg N yr^{-1} for rice paddies and 1010 Gg N yr^{-1} for upland fields compared that under a scenario that calculates NH_3 emissions based only on mineral N fertilizer application (Table 1). In the same scenario, N_2O emissions would decrease by 6 Gg N yr^{-1} for rice paddies but increase by 75 Gg N yr^{-1} for upland fields. N leaching would decrease by 32 Gg N yr^{-1} in rice paddies and

332 Gg N yr^{-1} in upland fields, and for rice paddies and upland fields, N runoff would decrease by 64 and 436 Gg N yr^{-1}, respectively. For all scenarios tested, straw return-induced increases in NH_3 emissions outweighed the reduction in other environmental Nr losses regardless of cropland type. Thus, total environmental Nr losses would increase by 181 Gg N yr^{-1} for the 20% scenario (S1) and up to 725 Gg N yr^{-1} for the 80% scenario (S4) (Table S8).

Discussion

Response of soil C sequestration and crop productivity to straw return

Straw return is widely considered to be one of the most sustainable and economically viable management practices for sequestering atmospheric CO_2 and improving global C storage in agricultural soils (Powlson *et al.*, 2008; Smith *et al.*, 2008). Our meta-analysis demonstrated that straw return significantly increased SOC content by 14.9% in global croplands (Fig. 8), which is comparable to the increase of 12.8% reported by Liu *et al.* (2014). Our results also showed a higher increase in SOC content in upland fields (17.0%) than in rice paddies (11.4%) (Table S2). This result can be explained by the higher initial SOC content in rice paddies (on average of 15.7 g C kg^{-1}, n=79) than in upland soils (10.6 g C kg^{-1}, n=143), as indicated by the negative correlation between the increase in SOC content and initial SOC content (Table S1). Soils with a lower initial C content have a greater saturation deficit, which may result in a higher initial soil C sequestration rate and a longer duration to reach a new C equilibrium (Powlson *et al.*, 2008).

Apart from cropland type, straw quality and soil texture may also alter the response of soil C sequestration to straw return. Our meta-analysis showed that the increase in SOC content is smaller for straw with a C/N ratio<30 (10.3%) than for straw with a larger C/N ratio>30 (15.1%) (Table S2). Generally, straw with a larger C/N ratio (>30) is rich in phenolic/lignin compounds that decompose slowly. These substances act as binding agents for the formation of soil aggregates, which promote SOC accumulation over longer time periods (Blanco-Canqui & Lal, 2009). Regarding soil texture, we found that straw return resulted in a higher increase in SOC content in loamy soils (14.5−21.0%) (e.g., silt loam, silty clay loam and loam) than in clay soils (11.5%) (Table S2). One possible explanation for this observation is the hampered degradation of straws in clay soils due to limited oxygen availability, which results in lower C transferring efficiency from straw C to SOC (Blanco-Canqui & Lal, 2009; Liu *et al.*, 2014).

The enhanced soil C sequestration following straw return may benefit crop yield, as evidenced by the positive correlation between SOC content and crop yield ($P<0.001$, Fig. S2). Crop growth can benefit directly from higher organic matter content (evidenced by higher SOC content) because its decomposition continuously provides nutrients, and SOC content is often a major factor of nutrient retention in agroecosystems (Lal, 2004; Singh *et al.*, 2008). In addition, crop straw is an important nutrient resource for crop growth (Majumder *et al.*, 2008); we found that the availability of soil nutrients (particularly N, P and K) was increased by 10.4−17.8% following straw return (Fig. 1). We further demonstrated that straw return improved soil physical properties (e.g., porosity and soil water retention capacity) and microbial biomass in soils (Fig. 1), which are known to support healthy crop development and contribute to higher crop water use efficiency (Ghuman & Sur, 2001).

The responses of crop yield to straw return are largely regulated by soil texture and climate conditions. For example, higher increases in yields were observed for sites with sandy and silt loamy texture or sites located in warmer climate zones (e.g., subtropical and tropical) (Table S6 and Fig. 2). These conditions favor straw decomposition and nutrient release due to better aeration conditions and/or higher temperatures (Singh *et al.*, 2008). In addition, the increase in yield increased with the straw returning period, likely due to the higher increase in SOC contents under continuous straw addition (Fig. 2 and Table S2).

Response of Nr losses to straw return

a) Nr loss through hydrological pathways

Apart from increasing soil C sequestration and crop yield, straw return also significantly decreased Nr losses via hydrological pathways (8.7–25.6%) (Fig. 5), which was mainly attributed to a stimulation of microbial N immobilization, as our meta-analysis showed that the soil N immobilization rate and MBN were significantly enhanced by 227% (n=28) and 38.4% (n=80), respectively (Fig. 1 and Fig. S3). This result is also in agreement with studies that show that increases in microbial N immobilization lead to a decrease in N runoff and leaching (Cheng *et al.*, 2017; Xia *et al.*, 2017).

Straw return also reduces N runoff by improving soil structure and consequently increasing the water infiltration rate (Blanco-Canqui *et al.*, 2006). The enhanced infiltration decreases surface runoff and the risk of soil erosion (Lindstrom, 1986), thereby reducing N runoff. Moreover, straw return can also decrease N leaching through diminishing leachate percolation. Blanco-Canqui *et al.* (2007) reported that soils subject to corn straw return can retain 20–50% more water for 0 to -6 kPa soil water potential. This reduces the frequency of leaching events and the amount of water transporting nutrients into the unsaturated zone and groundwater. Besides, higher SOC content after straw return increases the cation exchange capacity (CEC, 8.4%, n=33) that prevents NH_4^+ loss and increases the capacity to retain the very mobile anion NO_3^- due to deprotonated carboxyl groups (Blanco-Canqui & Lal, 2009).

A higher reduction in N leaching and runoff can be achieved by surface application of straw (26.2–33.5%) compared to soil incorporation (6.8–15.1%) (Fig. 5). Straw surface application would better protect the soil surface against the erosive impacts of rainfall and reduces the formation of surface cracks and crusts (Blanco-Canqui *et al.*, 2006), therefore leading to a higher reduction in hydrological N losses. Regarding the effects of soil texture, straw return to sandy soils significantly increased N leaching (19.7%), which was decreased in loamy and clay soils (Table 2).

Soils with a sandy texture generally have poor retention of water and nutrients due to their low SOC and clay contents (Six *et al.*, 2002). However, the increased water infiltration paired with additional N substrate from straw mineralization could aggravate N loss via leaching (Blanco-Canqui & Lal, 2009).

b) Nr loss through N_2O emissions

Soil N_2O is mainly produced through nitrification and denitrification, which depend on the availability of oxygen, soil N and C substrates (Davidson *et al.*, 2000; Butterbach-Bahl *et al.*, 2013). The remarkable decrease in N_2O emissions (17.3%) from rice paddies could be attributed to enhanced microbial N immobilization and complete denitrification (Aulakh *et al.*, 2001). Straw decomposition in rice paddies accelerates oxygen consumption in the soil aerobic layer and rhizosphere and increases DOC availability for denitrifiers (Fig. 1), which favors a further reduction of N_2O to N_2 (Firestone & Davidson, 1989).

However, straw return significantly increased N_2O emissions (21.5%) from upland soils (Fig. 3), also reported by Liu *et al.* (2014), mainly due to enhanced nitrification and denitrification. In upland soils, faster straw degradation provides additional N substrate for autotrophic nitrification and heterotrophic denitrification, which stimulate N_2O emissions (Davidson *et al.*, 2000; Chen *et al.*, 2013). We further found that this stimulation was significantly and positively correlated with straw N input rate (Table S1). Moreover, Zhao *et al.* (2018) reported that straw return also greatly increased the heterotrophic nitrification rate, possibly due to enhanced DOC availability. In addition, increased soil water content (14.0%, n=72, Fig. 1) together with decreased oxygen availability during straw decomposition would promote the formation of more anaerobic soil microsites, which can further accelerate N_2O emissions from denitrification process (Butterbach-Bahl *et al.*, 2013; Xia *et al.*, 2014).

The responses of N_2O emissions to straw return can also be regulated by soil properties and straw quality. Similar to Chen *et al.* (2013), we found the highest increase in N_2O emissions under straw return from loamy sandy soils, followed by loamy and clay soils (Table 3). Increasing clay content decreases soil aeration and oxygen availability, thereby decreasing straw decomposition and associated N release (Skiba & Ball, 2002). Moreover, soils with higher clay content (>40%) are generally characterized by low gas diffusivity, which may enhance the reduction of N_2O (produced in soil profiles) to N_2 through complete denitrification (Weitz *et al.*, 2001). This explains the negative relationship between N_2O emissions following straw return and soil clay content observed in our study (Fig. 6).

As for straw quality, the increase in N_2O emissions was significantly and negatively correlated with straw C/N ratio (Fig. 4), a result that was also reported by previous studies (Huang *et al.*, 2004; Chen *et al.*, 2013). Straw with a lower C/N ratio (<30) can be decomposed quickly, leading to higher N availability for nitrification and denitrification (Frimpong & Baggs, 2010). In contrast, a higher straw C/N ratio (>30) would increase microbial assimilation of soil N (Aulakh *et al.*, 2001), because low straw N contents may not satisfy the microbial N demand. The N depletion due to net N immobilization would decrease nitrification and denitrification rates, and consequently N_2O emissions (Liu *et al.*, 2017).

c) Nr loss through NH_3 emissions

Straw return significantly increased NH_3 emissions (17.0%), which was also observed by Pan *et al.* (2016), regardless of rice paddies (11.4%) and upland fields (20.3%, Fig. 3). Increases in NH_3 emissions following straw return can be first attributed to increased soil urease activity (overall: 18.5%, n=133; rice paddies: 8.8%, n=46; and upland fields: 24.6%, n=87) (Table S4). The presence of urease drives the hydrolysis of urea to NH_4^+ in paddy fields and upland soils and promotes NH_3 emissions (Pan *et al.*, 2016; Xu *et al.*, 2017). Besides, higher NH_4^+ availability from straw mineralization further stimulated NH_3 emissions, especially for straw with a C/N ratio<30 (Fig. 3). This result is further supported by the significant and positive correlation between the increase in NH_3 emissions and straw N input rate (Table 2). However, the increase in NH_3 emissions was lower with a straw C/N ratio>30 (Fig. 3 and Fig. 4), attributed to the enhanced microbial N immobilization (Aulakh *et al.*, 2001; Huang *et al.*, 2004).

In addition, straw return can also promote NH_3 emissions by stimulating ammonium-related soil N transformations (Wang *et al.*, 2015; Zhao *et al.*, 2018). For example, we found that straw return significantly increased the gross N mineralization rate by 82.4% (n=30) and dissimilatory NO_3^- reduction to NH_4^+ (DNRA) by 155% (n=9) (as supported by the increased soil NH_4^+/NO_3^- ratio by 14.2%) but decreased the NH_4^+ oxidation rate by 33.7% (n=18) (Fig. S3). These altered N transformations would provide more N substrates for NH_3 emissions, as further demonstrated by increased soil NH_4^+ content (5.7%, n=83) (Fig. 1).

The responses of NH_3 emissions to straw return can also be affected by soil properties and climate conditions. A lower increase or even no effect on NH_3 emissions following straw return was observed at sites with clay content>40% or sites located in cool temperate zones (Fig. 3 and Table 3), where straw degradation was relatively hampered. Moreover, soils with higher clay content (>40%) generally have greater CEC (Parfitt *et al.*, 1995), which can increase NH_4^+ adsorption by clay particles and thus reduce NH_3 emissions (Xia *et al.*, 2017). This scenario explains the negative relationship between the increases in NH_3 emission following straw return with soil clay content observed in our study (Fig. 6).

Interactions between SOC content and Nr losses under straw return

A detailed investigation of the interactions between Nr losses and SOC sequestration provides a better understanding of the overall effects of straw return on soil N and C cycles. In this study, we found that the decreases in Nr losses, especially N leaching and runoff, were positively correlated with increases in SOC content under straw return (Fig. 7), which suggests that enhanced soil C sequestration may increase the reduction in Nr losses from croplands. This result can be attributed to the following reasons. First, the straw-induced increase in soil C sequestration is largely derived from the increase in soil macroaggregates (Tisdall & Oades, 1982; Six *et al.*, 2002; Liu *et al.*, 2014). The increase in macroaggregates would increase soil water and nutrient retention capacities and reduce the risk of soil erosion, consequently reducing N losses via hydrological pathways (Blanco-Canqui & Lal, 2009; Xia *et al.*, 2017).

Second, higher soil organic matter (SOM) content under straw return may facilitate a better synchronization between crop nutrient demand and soil nutrient supply (Singh *et al.*, 2008; Blanco-Canqui & Lal, 2009), which can promote nutrient uptake and crop growth. This scenario may explain the beneficial effect of straw return on crop N uptake and NUE (Fig. 2). The environmental Nr losses are closely linked to crop NUE (or crop N uptake), as shown in Cui *et al.* (2013) and Groenigen *et al.* (2010). Both studies indicated an exponential increase in Nr losses with increasing N surplus or decreasing NUE. In other words, increasing N uptake efficiency by crops associated with higher SOC content under straw return (P<0.001, Table 2) may reduce Nr losses to the environment.

Balance of global Nr losses under straw return to croplands

Overall, straw return significantly increased NH_3 emissions but decreased Nr losses through leaching and runoff (Fig. 8). As the stimulation of NH_3 loss was much higher than the overall reduction in the other Nr loss pathways, straw return increased net Nr losses from both rice paddies (30 Gg N yr^{-1}) and upland fields (316 Gg N yr^{-1}) (P=40%). Nevertheless, crop productivity under straw return still significantly increased (Fig. 8) because the total N content of the soils was significantly increased (9.8%) under straw return (Fig. 1), probably due to the straw N input rate exceeding the increased Nr losses from global croplands. Although returning straw with a higher C/N ratio would stimulate microbial N immobilization which is also known as the ‘N tie-up’ effect (Kirkegaard *et al.*, 2018), the immobilized N by microbes can be released across the growing season and benefit crop growth (Xia *et al.*, 2017), crop N uptake and consequently crop yield (Fig. 2).

It is still critical to minimize NH_3 emissions when straw returns are adopted to increase C sequestration and/or crop productivity in global agroecosystems. One possible management option is surface application of straw instead of incorporating it into soils, which may largely attenuate the increased NH_3 emissions (Fig. 3). This effect can be attributed to a weaker stimulatory effect on soil urease activity due to the incomplete mixing between straw and soils (Pan *et al.*, 2016), as demonstrated by the much lower increase in soil urease activity under surface application of straw (3.3%, n=25) than incorporation into soils (22.6%, n=101) (Table S4). Applying straw with a higher C/N ratio (>30) would also attenuate the increased NH_3 emissions (Fig. 3 and Fig. 6) and reduce other Nr losses. Application of urease inhibitor together with straw return can decrease urea hydrolysis and NH_4^+ concentration in soils and therefore decrease NH_3 emissions (Xia *et al.*, 2016). Reducing mineral N fertilization rates based on the amount of additional N input from straw into the soil may also decrease NH_3 emissions and other Nr losses (Wang *et al.*, 2015).

Implications and looking forward

Overall, our study shows that straw return is effective in increasing soil C storage and crop productivity (Fig. 8). Although the increase in SOC content is accompanied by a reduction in N leaching and runoff (Fig. 7), straw return increased net Nr losses from global croplands due to a greater stimulation of NH_3 emissions (Table 1). Since straw return is becoming more widely adopted (Lu *et al.*, 2009), our findings on the trade-offs between the increased net Nr losses to the environment and soil C sequestration provide a better understanding of N and C balances in global croplands. Our results also highlight that any initiative that aims to reduce the environmental footprint of agricultural production systems needs to consider that C and N cycles are closely coupled and that antagonistic effects, e.g., increased soil C sequestration and stimulation of NH_3 emissions, might occur simultaneously.

Our estimation of global Nr losses balance under straw return was based on an empirical model that only differentiated two cropland types (rice paddies and upland fields). This introduced some uncertainties in the upscaling of global Nr losses because other parameters, such as soil properties (soil texture), straw quality (straw C/N ratio) and quantity (straw N input rate), also greatly impact soil N transformations and Nr losses (Table 3). However, these parameters are not fitted into the model of this study due to a lack of data, particularly regarding the ‘mineral N fertilization rate (N_{rate})’ and ‘fraction of mineral N fertilizer lost to Nr (F)’ in equation (3) under different (parameter) categories. For example, there were deficient data on N_{rate} and F under different soil types (sand, clay and loam) which receive the application of straw with different C/N ratios. This underscores the importance of the inclusion of these parameters in future studies.

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Competing interests

The authors declare no competing financial interests.

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Table 1 Net changes in various reactive N (Nr) losses under straw return to global croplands (P=40%)

Nr losses	Paddy field	Upland field	Global croplands

	[a]F (%)	[b]E (%)	Nr change[c]	F (%)	E (%)	Nr change	F (%)	E (%)	Nr change
NH_3 emissions	19.7	11.4	132.2	15.1	20.3	1009.5	16.9	17.0	1114.5
N_2O emissions	0.62	-17.3	-6.3	1.1	21.5	74.5	0.95	5.9	21.5
N leaching	7.8	-7.0	-32.1	10.4	-9.8	-332.3	8.9	-8.7	-300.9
N runoff	4.5	-24.1	-63.9	5.0	-26.4	-436.0	4.8	-25.6	-472.7
Net Nr changes	--	--	29.9	--	--	315.8	--	--	362. 4

[a] F denotes the fraction of mineral N fertilizer that is lost as Nr.

[b] E denotes the effects of straw return on Nr emissions.

[c] The unit of (net) Nr changes is Gg N yr^{-1}. The calculation is based on the scenario of P=40%.

Table 2. Linear regression analysis between crop productivity and reactive N (Nr) losses with different climate conditions, soil properties, straw quality and quantity under straw return to global croplands

Parameters[a]	lnRR of variables						
	Yield	N uptake	NUE	NH_3 emissions	N_2O emissions	N leaching	N runoff
MAT	***[b]	ns	ns	ns	ns	ns	ns
MAP	ns	*	*	ns	***	ns	ns

Soil clay content	ns	ns	ns	*	**	ns	ns
SOC content	ns	*	***	ns	ns	ns	ns
Soil pH	ns	ns	ns	ns	ns	ns	ns
Total N content	ns	ns	ns	ns	ns	ns	ns
Mineral N rate	**	ns	ns	ns	ns	ns	ns
Straw input rate	ns	ns	ns	ns	ns	*	ns
Straw N input	**	***	ns	***	***	*	ns
Straw C input	ns	ns	ns	ns	ns	*	ns
Straw C/N ratio	ns	***	ns	**	***	ns	ns

[a] MAT, mean annual temperature; MAP, mean annual precipitation; soil properties refer to the initial soil properties prior to starting the experiment; mineral N rate means mineral N fertilization rate.

[b] * means 0.01<*P*<0.05, ** means 0.001<*P*<0.01, *** means *P*<0.001, 'ns' means the linear relationship is not significant, and stars with underline denote a negative linear relationship, while others represent a positive linear relationship.

Table 3 Changes (%) in various reactive N (Nr) losses under straw return to global croplands in different soil textures with a 95% confidence interval (CI)

Soil texture	NH_3 emissions[a]		N_2O emissions		N leaching		N runoff	
	Mean	95% CI	Mean	95% CI	Mean	95% CI	Mean	95% CI
Sand	60.9	7.9–166.8	119.3	-34.2–7401	19.7	4.2–35.8	--[b]	--
Loam sand	78.5	21.5–125.6	483.5	210.9–1008	-28.2	-(32.7–23.4)	--	--
Sandy loam	--	--	-0.5	-15.4–18.2	-20.6	-(31.7–3.6)	--	--

Sandy clay loam	34.8	15.7–57.1	-8.2	-36.4–31.9	--	--	--	--
Silt loam	19.2	3.1–37.5	3.5	-11.7–22.2	9.4	-11.9–42.2	-39.3	-(48.9–28.7)
Silty clay loam	1.2	-32.5–38.2	7.1	-26.4–48.0	-22.5	-(26.9–17.0)	4.6	-8.9–30.6
Loam	12.7	1.5–26.2	3.9	-6.6–14.3	-23.3	-(39.1–6.3)	-21.5	-(29.5–13.1)
Clay	4.3	-7.6–19.2	-10.4	-29.8–17.0	-7.1	-26.6–16.2	-42.8	-(61.7–17.0)

[a] The number of experimental observations under different soil textures are 11, 3, 4, 18, 6, 51 and 16 for NH_3 emissions; 4, 13, 51, 16, 53, 20, 58 and 34 for N_2O emissions; 9, 2, 8, 8, 4, 11 and 14 for N leaching; and 13, 6, 24 and 5 for N runoff.

[b] No data are available.

Figure captions

Fig. 1. Changes in soil properties and crop water use efficiency under straw return to croplands. The number of experimental observations is in parentheses. MBN, microbial biomass nitrogen; SOC, soil organic carbon; MBC, microbial biomass carbon; DOC, dissolved organic carbon; LOC, labile organic carbon and CEC, cation exchange capacity.

Fig. 2. Changes in crop yield (a), crop N uptake (b) and N use efficiency (c) for croplands using straw return. The number of experimental observations is in parentheses. T<4 years denotes that crop straw has been continuously used for less than 4 years. N rate denotes the application rate of mineral N fertilizer (kg N ha^{-1}).

Fig. 3. Changes in NH_3 and N_2O emissions induced by straw return to global croplands. The number of experimental observations is in parentheses. Clay means soil clay content (%).

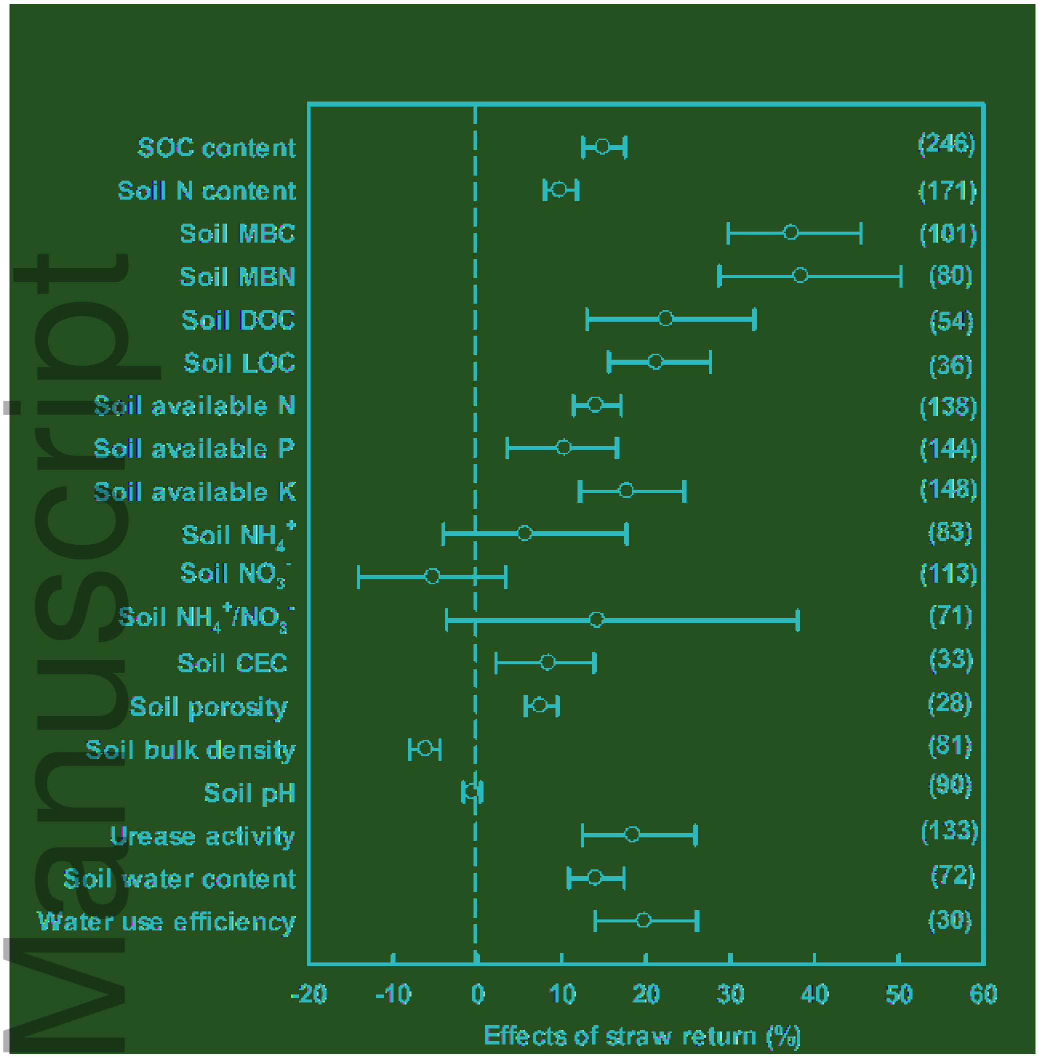
Fig. 4. Relationship between the changes in Nr losses (ln*RR*) under straw return and straw C/N ratio. Negative values of ln*RR* denote a reductive effect of straw return on Nr losses.

Fig. 5. Changes in N leaching and runoff induced by straw return to global croplands. The number of experimental observations is in parentheses. Clay means soil clay content (%).

Fig. 6. Relationship between the changes in Nr losses (ln*RR*) under straw return and soil clay content. Negative values of ln*RR* denote a reductive effect of straw return on Nr losses.

Fig. 7. Relationship between the changes in SOC content (ln*RR*) and Nr losses (ln*RR*) induced by straw return to global croplands. Negative values of ln*RR* denote a reductive effect of straw return on Nr losses.

Fig. 8. Overall effects of straw return on soil C dynamics, crop productivity and reactive N losses. SOC, soil organic carbon; DOC, dissolved organic carbon; MBC, microbial biomass carbon; MBN, microbial biomass nitrogen; DON, dissolved organic nitrogen. NUE, nitrogen use efficiency; WUE, water use efficiency. Data on the changes in CH_4 emissions from rice paddies and CO_2 emissions from upland (27.8%) and rice paddies (51.0%) under straw return were derived from Liu *et al.* (2014).



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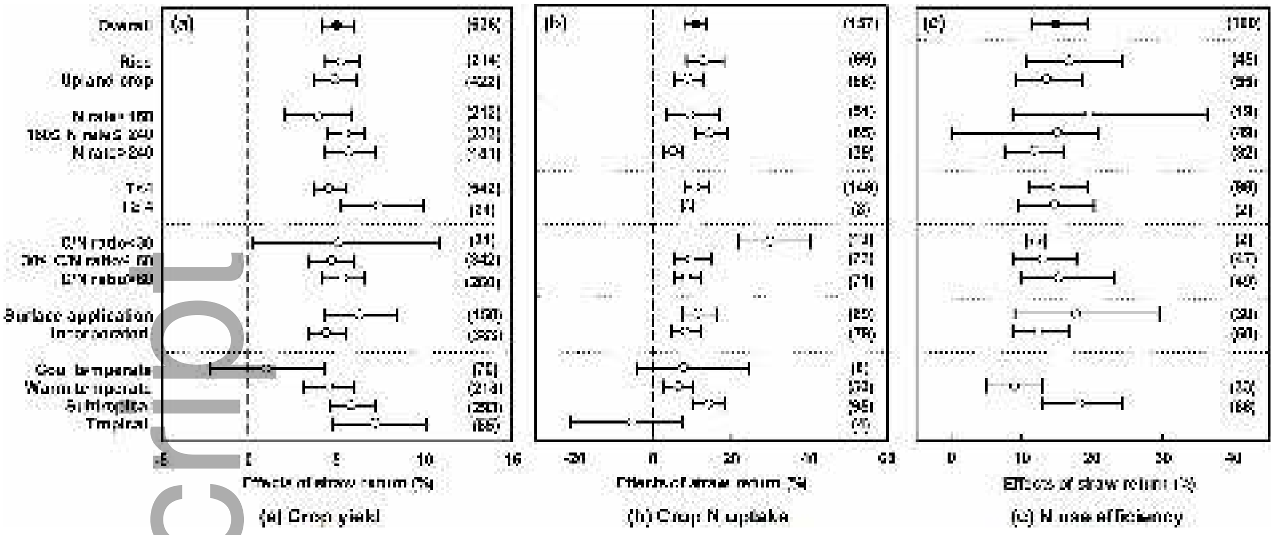

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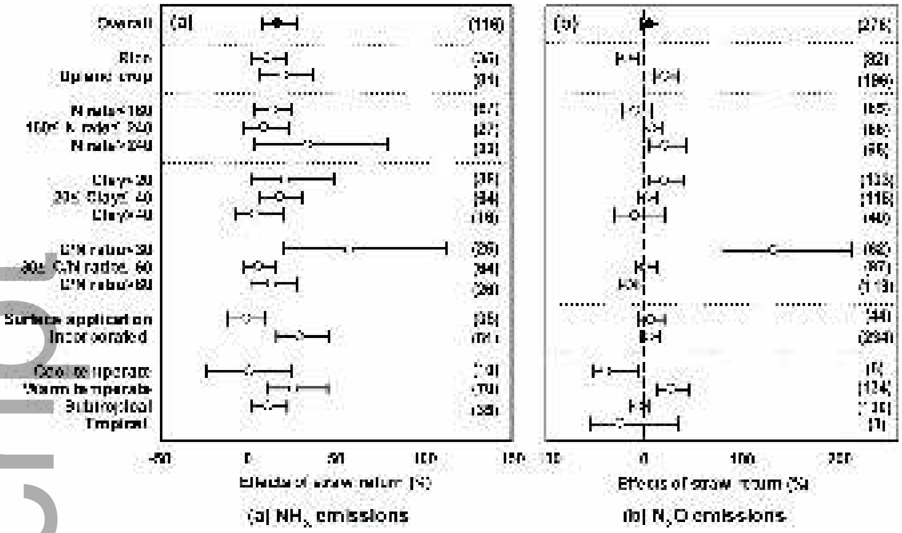

(a) NH_3 emissions (b) N_2O emissions

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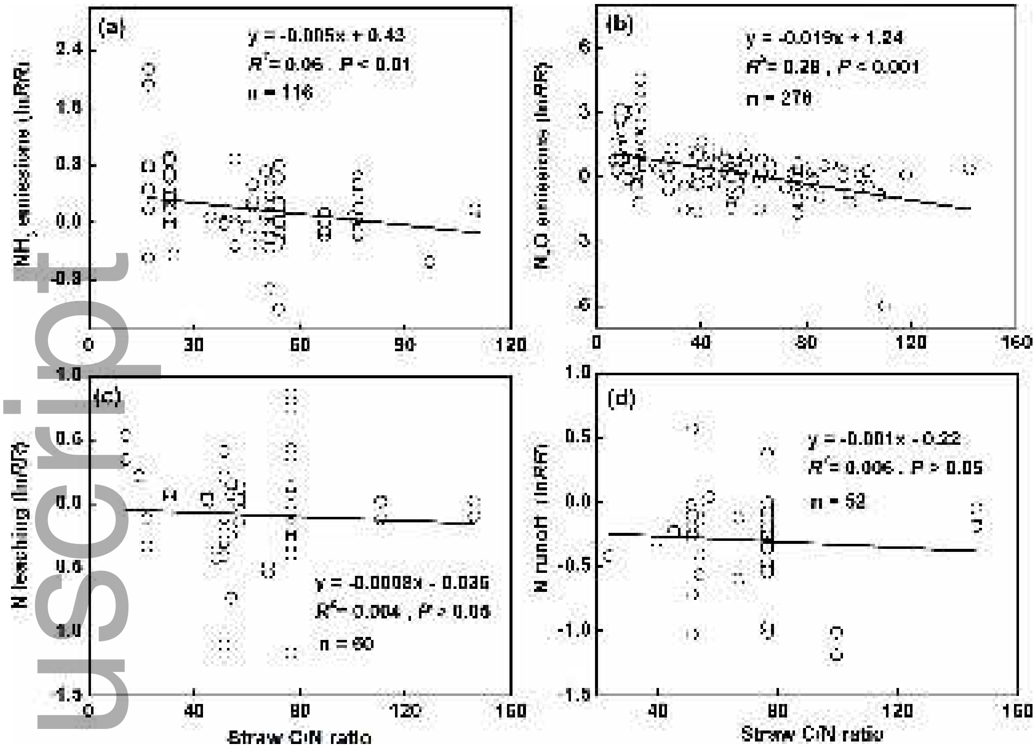

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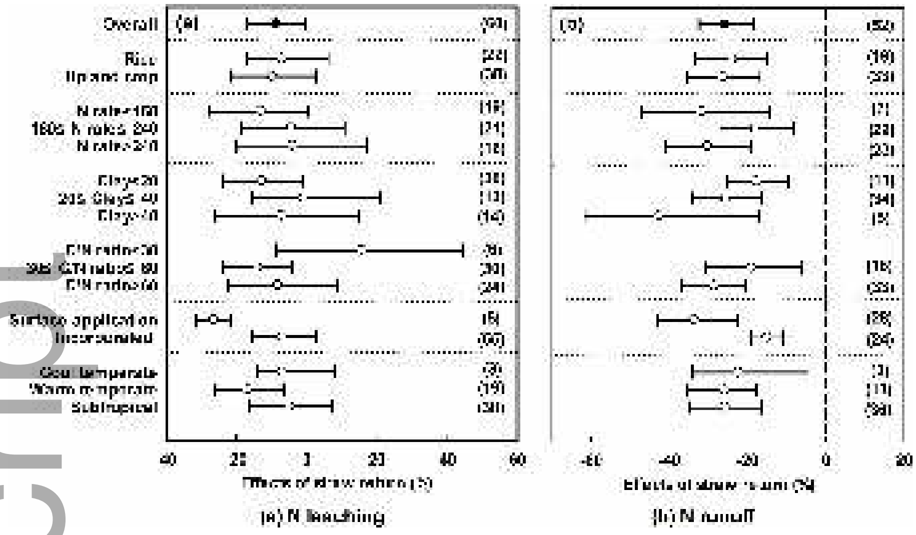

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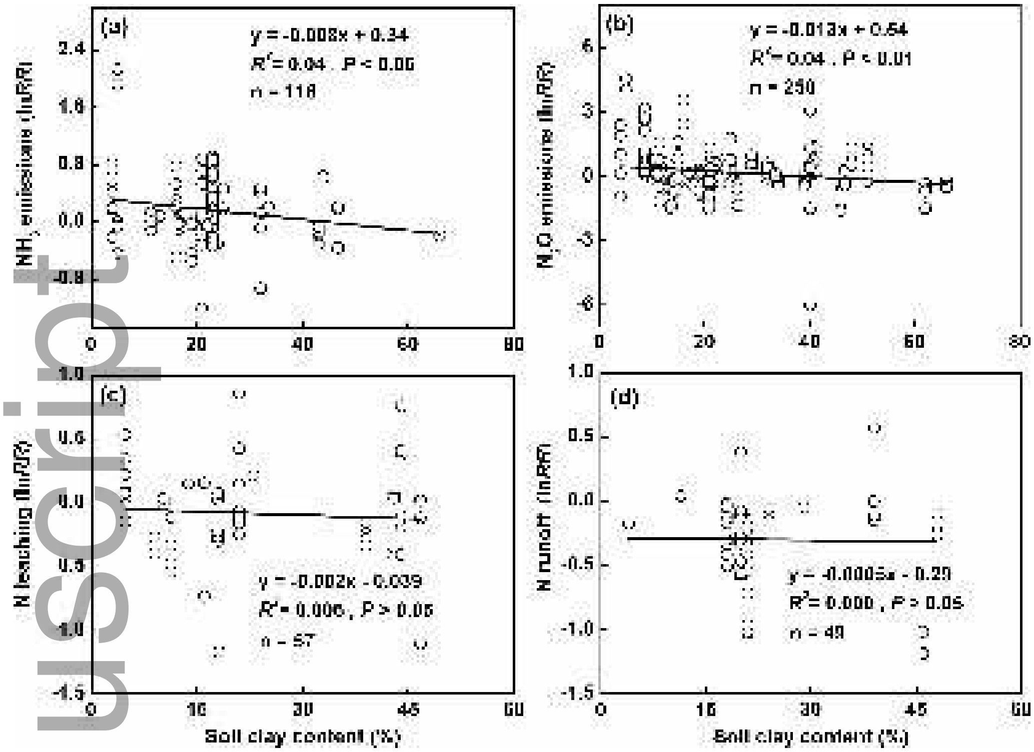

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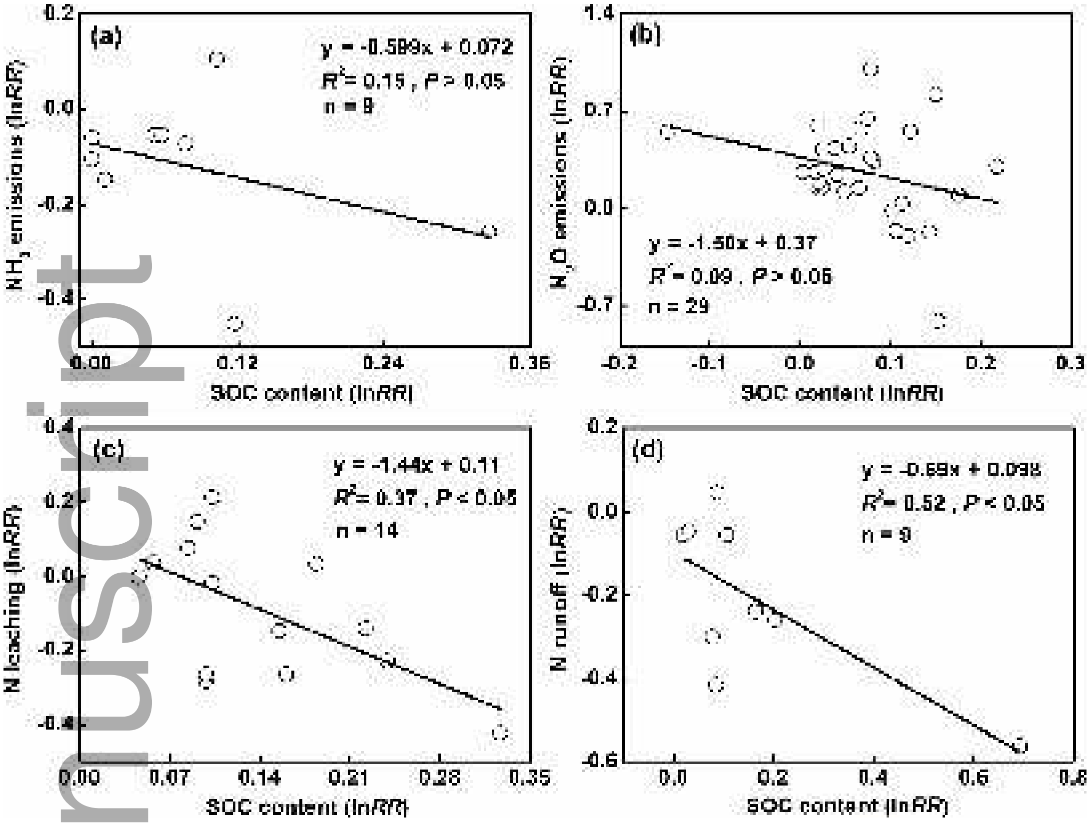

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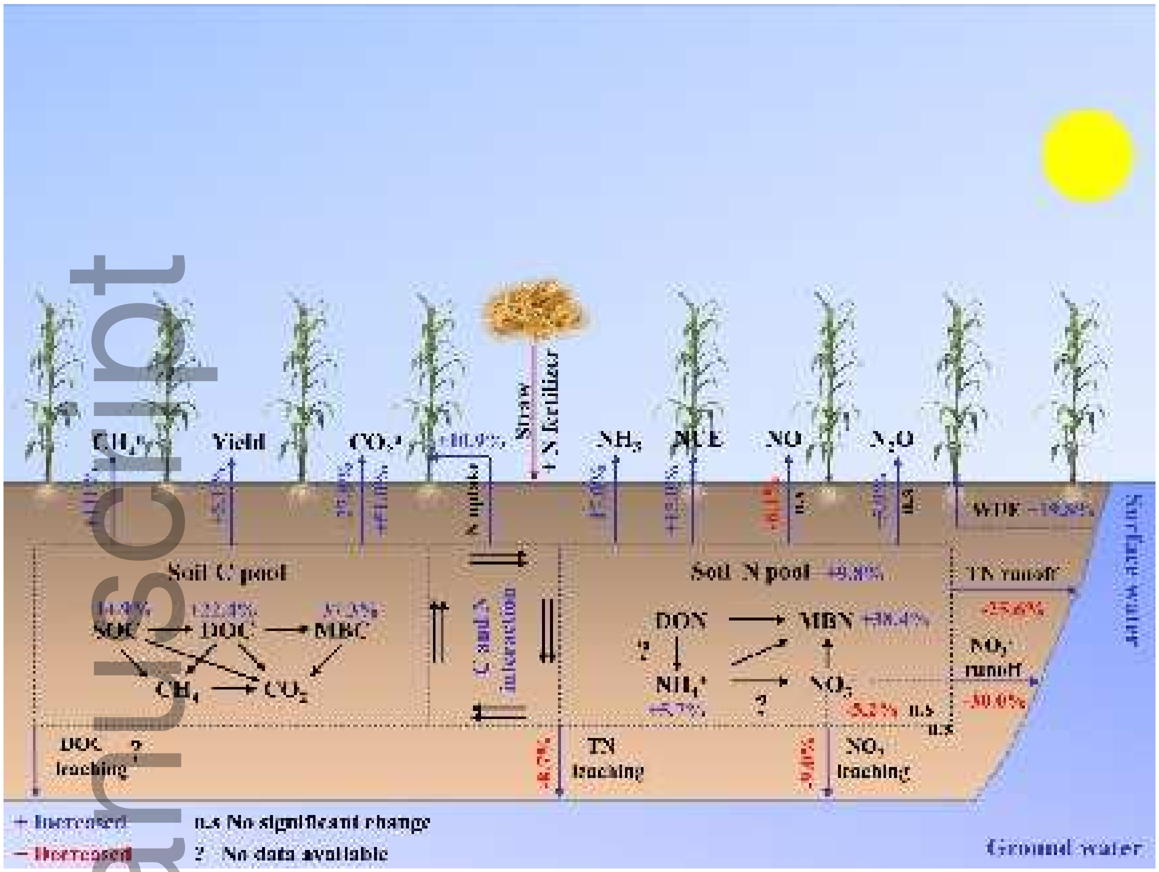

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Author/s:
Xia, L;Lam, SK;Wolf, B;Kiese, R;Chen, D;Butterbach-Bahl, K

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