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Elevated atmospheric CO_2 reduces yield-scaled N_2O fluxes from subtropical rice systems: Six site-years field experiments

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Abstract

Increasing levels of atmospheric CO_2 are expected to enhance crop yields and alter soil greenhouse gas fluxes from rice paddies. While elevated CO_2 (E_{CO_2}) effects on CH_4 emissions from rice paddies have been studied in some detail, little is known how E_{CO_2} might affect N_2O fluxes or yield-scaled emissions. Here, we report on a multi-site, multi-year in-situ FACE (free-air CO_2 enrichment) study, aiming to determine N_2O fluxes and crop yields from Chinese subtropical rice systems as affected by E_{CO_2}. In this study, we tested various N fertilization and residue addition treatments, with rice being grown under either E_{CO_2} (+200 μmol/mol) or ambient control. Across the six site-years, rice straw and grain yields under E_{CO_2} were increased by 9%–40% for treatments fertilized with ≥150 kg N/ha, while seasonal N_2O emissions were decreased by 23%–73%. Consequently, yield-scaled N_2O emissions were significantly lower under E_{CO_2}. For treatments receiving insufficient fertilization (≤125 kg N/ha), however, no significant E_{CO_2} effects on N_2O emissions were observed. The mitigating effect of E_{CO_2} upon N_2O emissions is closely associated with plant N uptake and a reduction of soil N availability. Nevertheless, increases in yield-scaled N_2O emissions with increasing N surplus suggests that N surplus is a useful indicator for assessing N_2O emissions from rice paddies. Our findings indicate that with rising atmospheric CO_2 soil N_2O emissions from rice paddies will decrease, given that the farmers' N fertilization is usually sufficient for crop growth. The expected decrease in N_2O emissions was calculated to compensate 24% of the simultaneously observed increase in CH_4 emissions under E_{CO_2}. This shows that for an agronomic and environmental assessment of E_{CO_2} effects on rice systems, not only CH_4 emissions, but also N_2O fluxes and yield-scaled emissions need to be considered for identifying most climate-friendly and economically viable options for future rice production.

KEYWORDS

climate change, elevated CO_2, FACE, greenhouse gas fluxes, N surplus, nitrous oxide, rice

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1 | INTRODUCTION

Human activities, specifically fossil fuel burning and land use change are rapidly increasing the level of carbon dioxide (CO_2) in the atmosphere (IPCC, 2013). The increase in atmospheric CO_2 concentration can directly and/or indirectly influence carbon (C) and nitrogen (N) biogeochemical cycles in soil-plant systems (e.g., Black et al., 2017; Hungate et al., 1999; Liu et al., 2018; Luo et al., 2004; Müeller et al., 2009; Rütting et al., 2010; Schaeffer et al., 2007). An important direct response to elevated CO_2 (E_{CO_2}) is enhanced plant photosynthesis and biomass production (e.g., Ellsworth et al., 2004; Wang et al., 2020), which has been found to possibly increase soil C input and soil C storage (e.g., Hungate et al., 1997; Yue et al., 2017). Along with the increase in soil C supply, soil microbial processes that drive N cycling and associated gaseous N losses, such as the emission of potent greenhouse gas (GHG) nitrous oxide (N_2O) from soils, might increase as well (e.g., Bhattacharyya et al., 2013; Billings et al., 2002; Kettunen et al., 2006). On the other hand, several studies indicate that under E_{CO_2} not only plant transpiration decreases, and thus soil water content increases, but that E_{CO_2} also results in increased plant N uptake. The latter might reduce the availability of N for soil microbial processes involved in N_2O production, namely nitrification and/or denitrification (Butterbach-Bahl et al., 2013; Hu et al., 2001; Stitt & Krapp, 1999). However, the interplay between plants, soil microorganisms and C and N cycling in response to E_{CO_2} varies widely both within and between terrestrial ecosystems (e.g., Baggs, Richter, Hartwig, et al., 2003; Dijkstra et al., 2012; Phillips et al., 2001).

Agricultural activities are the most important source for atmospheric N_2O, accounting for 60% of total global anthropogenic emissions (Ciais et al., 2013). The importance of agriculture as a driver of climate change has spurred substantial efforts to mitigate N_2O emissions from agricultural systems, particularly those where N fertilizers are used (Smith et al., 2007). Although N fertilizer-induced soil N_2O emissions are mostly associated with agricultural upland soils, rice paddies, especially those managed with intermittent irrigation (midseason drainage) or alternate wetting and drying have been found to contribute significantly to global N_2O fluxes (Akiyama et al., 2005; Zou et al., 2007, 2009). Rice is a staple food for >50% of the world's population and provides more calories than any other cereal crops for human consumption (Bouman et al., 2007). It is estimated that up to 155 million ha, or 11% of the planet's arable land, is cultivated with rice cropping systems (Bhattacharyya et al., 2013; Van Groenigen et al., 2013). These systems are being recognized as a large source of climate impact as a result of the enhancement of N_2O emissions (EPA, 2013; Kritee et al., 2018). Total direct N_2O emissions from China's rice systems are estimated to be 32–51 Gg N/year, or 8%–11% of total annual N_2O emissions from croplands in China (Liu et al., 2016; Zou et al., 2009).

To meet the needs of a growing world population, global cereal production is projected to increase at least 60% in 2050 relative to 2006 levels (Lv et al., 2020). The demanded increases in crop yields will require significant improvements (by 8–10 million Mg per year or 1.2%–1.5% of annual yield) in rice production (Seck et al., 2012). As agriculture is not only a source of GHGs but also is directly affected by climate change, the increase in production should be achieved at lower environmental costs, specifically with regard to GHG emissions. This challenge is well reflected when using the metric of yield-scaled N_2O emissions (Van Groenigen et al., 2010, 2013), though little is known if yield-scaled N_2O emissions of rice paddies might change under E_{CO_2}.

While much is known about the key role of increased CO_2 in driving global warming, its effect on soil N_2O emissions remains speculative. On the basis of a meta-analysis that included natural and agricultural soils (73 observations), Van Groenigen et al. (2011) concluded that E_{CO_2} is likely to stimulate soil N_2O emissions by about 18.8%. Similarly, Dijkstra et al. (2012) conducted a thorough literature review and showed that E_{CO_2} often increased N_2O emissions in fertilized ecosystems. In contrast, there were also meta-analysis results showing that E_{CO_2} caused insignificant increases in soil N_2O emissions (Liu et al., 2018). Moreover, all of these reviews and meta-analyses only included N_2O data from upland soils despite the potentially substantial effects of E_{CO_2} on N_2O emissions from rice paddies. So far, only a few studies have investigated N_2O fluxes from rice systems under E_{CO_2}, showing either a stimulation (Bhattacharyya et al., 2013), no significant response (Cheng et al., 2006; Pereira et al., 2013) or a reduction of soil N_2O emissions (Sun et al., 2018). This suggests that apart from changes induced by E_{CO_2}, other soil environmental factors (e.g., soil N availability) might also affect N_2O emissions. Moreover, so far, no field studies are available using the free-air CO_2 enrichment (FACE) technology, which allows to assess changes in soil N_2O fluxes from rice systems under more realistic environmental conditions as compared to studies relying on controlled-environment chambers (Zheng et al., 2006).

In this study, we report on results of measurements of soil N_2O fluxes from two Rice-FACE sites in subtropical China, covering in total 6 years. The overall objective of this study was to determine the effects of E_{CO_2} on soil N_2O fluxes, as expressed on both an area- and yield-scaled bases, from rice paddies under various N fertilization treatments. We hypothesized that E_{CO_2} would increase soil N_2O fluxes, as root biomass and consequent C exudation by roots are increased under E_{CO_2} (Yang et al., 2008), which likely facilitates denitrification. Moreover, we hypothesized that yield-scaled N_2O emissions remain unchanged as not only soil N_2O emissions but also crop yields do increase under E_{CO_2} (Yang, Huang, et al., 2007; Yang et al., 2009). Furthermore, we assumed that soil N availability, here reflected by different N fertilization rates, will alter the response of soil N_2O fluxes to E_{CO_2} in rice systems.

2 | MATERIALS AND METHODS

2.1 | Experimental sites and field treatments

During the period from 2001 to 2006, a series of field measurements were carried out at the two Rice-FACE experimental sites of Wuxi (31°37′N, 120°28′E) and Jiangdu (32°35′N, 119°42′E). Study sites were located in the Chinese rice cropping region of the Yangtze River Delta, where rice is grown for hundreds of years. The typical cropping system consists of winter wheat (November to May) and paddy rice (June to October) throughout the year (Yao et al., 2010, 2013). While the winter wheat is not irrigated, the rice paddy is kept flooded from early June to early July, drained several times up to early August, before getting intermittently flooded until the final drainage prior to rice harvesting. The study region has a northern subtropical monsoon climate, with a mean annual precipitation of 924–1,079 mm, and mean annual air temperature of 15.6–15.9°C. Further details on climate, agricultural practices and soil properties are described by Yao et al. (2010).

For studying effects of E_{CO_2} on rice systems, the FACE technology was used at Wuxi from June 2001 onwards and at Jiangdu from June 2004 onwards. Details on the design, rationale, operation and performance of the FACE system are described in the work of Okada et al. (2001) and Zheng et al. (2006). Briefly, the system consists of three octagonal experimental rings (each with a diameter of 14 m), with atmospheric CO_2 concentration being elevated by 200 μmol/mol above the ambient concentration, and three additional control rings with ambient atmospheric CO_2 (A_{CO_2}). The area of each ring which could be used for rice growing was approximately 120 m^2. Each ring was separated into two plots and each plot was fertilized with synthetic N fertilizers (compound fertilizer and urea) at a rate of 125–150 or 250–350 kg N/ha, respectively. Application rates mirrored the upper and lower boundaries of recommended N fertilizer rates for rice cropping systems in subtropical China (Zhu & Chen, 2002). At the Jiangdu site, each plot was further separated into three subplots, the treatment of which differed with regard of crop residue (wheat straw) amendment. Table 1 summarizes and encodes the field treatments of N application and crop residue addition rates at both experimental sites.

At the Wuxi site, two N application rates were tested during the rice seasons of 2001–2003 (i.e., 150 and 250 kg N/ha, referred to as N_{150} and N_{250}, respectively), while the residue addition rate was the same for both N fertilization treatments, though differing across years. In 2001, the wheat straw was applied at a rate of approximately 2,000 kg C/ha, corresponding to all of the harvested residue yield from the previous season, hereinafter referred to as AR. In 2002 and 2003, the wheat straw from the previous season was applied at a rate of approximately 1,000 kg C/ha, equivalent to half the amount of harvested straw yield, hereinafter referred to as HR. Thus, across all study years four treatments were investigated: N_{150}-AR and N_{250}-AR in 2001; N_{150}-HR and N_{250}-HR in 2002 and 2003. These treatments were studied for E_{CO_2} and A_{CO_2} conditions, with each treatment having three replicated plots and each plot having an area of 60 m^2.

With regard to the Jiangdu site, two N application rates (i.e., 125 kg N/ha [referred to as N_{125}] and N_{250}, respectively) combined with three residue addition rates (i.e., no residue amendment [referred to as NR], HR and AR, respectively) were investigated during the rice seasons in 2004 and 2005. In 2006, the field plots received 350 kg N/ha (referred to as N_{350}) and no synthetic N fertilization (referred to as N_0), in combination with three residue addition rates (i.e., NR, HR and AR, respectively). Thus, in each rice season six fertilization treatments (i.e., N_{125}-AR, N_{125}-HR, N_{125}-NR, N_{250}-AR, N_{250}-HR and N_{250}-NR for the 2004 and 2005; N_{350}-AR, N_{350}-HR, N_{350}-NR, N_0-AR, N_0-HR and N_0-NR for the 2006) were tested under E_{CO_2} and A_{CO_2} conditions. Each treatment was examined on three replicated subplots (each subplot covering an area of 20 m^2).

For the N_{125} and N_{150} treatments, 60% of the synthetic N fertilizers were applied as basal fertilization and 40% as topdressing at the panicle initiation stage (Table 1). With respect to N_{250} and N_{350}, 36% of the N fertilizers were applied basally and 24% and 40% by topdressing at early tillering and at the panicle initiation stage, respectively. For the HR and AR treatments, the harvested wheat straw from the previous season was incorporated into the 0–15 cm soil depth before rice transplanting. In the NR treatment, neither harvested crop straw nor organic matter from other sources was used. Consistent with the local convention, all experimental treatments received 75 kg P_2O_5/ha and 75 kg K_2O/ha in the rice season to avoid P and K deficiency of the rice crop.

2.2 | Measurement of N_2O flux

In-situ measurements of N_2O fluxes were performed at Wuxi in the rice seasons of 2001–2003 and at Jiangdu in the rice seasons of 2004–2006, using the static closed chamber method described by Yao et al. (2009, 2010). In brief, a square stainless-steel chamber frame, which covers an area of 0.25 m^2 and six hills of rice, was inserted into the soil in the center of each experimental plot or subplot. In total, there are 12 plots/frames for the Wuxi site and 36 subplots/frames for the

TABLE 1 Field treatments of nitrogen (N) fertilizer and crop residue additions for the investigated sites during the rice seasons 2001–2003 and 2004–2006, respectively

Code	Nitrogen application rate (kg N/ha) Basal fertilization	First topdressing	Second topdressing	Residue return (kg C/ha)	Transplanting/harvest date
Wuxi[a]					
N_{150}-AR	90	0	60	2,000	Jun. 13/Oct. 26, 2001; Jun. 13/Oct. 20, 2002; Jun. 13/Oct. 27, 2003
N_{150}-HR	90	0	60	1,000	
N_{250}-AR	90	60	100	2,000	
N_{250}-HR	90	60	100	1,000	
Jiangdu[a]					
N_0-AR	0	0	0	2,000	Jun. 15/Oct. 25, 2004; Jun. 14/Oct. 24, 2005; Jun. 14/Oct. 23, 2006
N_0-HR	0	0	0	1,000	
N_0-NR	0	0	0	0	
N_{125}-AR	75	0	50	2,000	
N_{125}-HR	75	0	50	1,000	
N_{125}-NR	75	0	50	0	
N_{250}-AR	90	60	100	2,000	
N_{250}-HR	90	60	100	1,000	
N_{250}-NR	90	60	100	0	
N_{350}-AR	126	84	140	2,000	
N_{350}-HR	126	84	140	1,000	
N_{350}-NR	126	84	140	0	

[a]Each treatment was applied under both elevated carbon dioxide (CO_2) and ambient CO_2 conditions; AR, the addition rate of crop residue was approximately equivalent to all of the harvested wheat straw yield; HR, the addition rate of crop residue was approximately equivalent to a half of the harvested wheat straw yield; ZR, no harvested wheat straw was incorporated into soils.

Jiangdu site in each experimental rice season. To minimize the mixing of paddy water, adjacent plots or subplots in each FACE and ambient ring were separated by a 30 cm PVC barrier pushed 15 cm depth into the soil. In dependence of crop height, a 50 or 100 cm high, gas-tight chamber was mounted on the base frame for flux measurements. Chambers were closed for approximately 30 min, and gas samples were collected from its headspace at regular intervals (usually at time 0 and after 8, 16, 24, and 32 min). Care was taken to minimize soil disturbance during the sampling period. Gas samples were analyzed for N_2O concentrations using an Agilent 4890D GC equipped with an electron capture detector (column and detector temperatures were 55°C and 330°C, respectively) within a 6 hr period at an on-site laboratory (Zheng et al., 2008). The N_2O fluxes were calculated on basis of linear or nonlinear curve fitting to observed temporal changes in headspace N_2O concentrations (Hutchinson & Livingston, 1993). To minimize the effect of diurnal temperature variation on fluxes, flux measurements were always carried out in the morning hours between 09:00 a.m. and 11:00 a.m. Gas flux measurements were performed twice per week, i.e., every 3–4 days.

2.3 | Auxiliary measurements

In addition to the flux measurements, daily precipitation, air temperature and barometric pressure were recorded by an automatic meteorological station established in direct vicinity to the experimental sites. Soil temperature in 5 cm depth was measured daily in the vicinity of the chamber frames with a thermocouple (JM624, Tianjin Jinming Instrument Co. Ltd.). Field floodwater depth during the experimental period was monitored daily using a stainless-steel ruler.

At the physiological maturity of rice, the aboveground biomass including straw and grain was harvested and oven-dried until constant weight. Straw and grain samples were ground and analyzed for carbon and nitrogen contents as described by Kim et al. (2003) and Inubushi et al. (2003).

2.4 | Data processing and statistical analysis

Total seasonal cumulative N_2O emissions were calculated by linear interpolation between sampling dates, assuming that the emissions followed a linear trend during the periods when no measurements were taken. The direct N_2O emission factor of applied N was determined as the difference between total cumulative emissions from the fertilized and unfertilized treatments divided by total N fertilization rate. To more effectively assess E_{CO_2} effects on soil N_2O emissions, seasonal cumulative emissions were expressed on both area- and yield-scaled bases. Yield-scaled N_2O emissions were computed by dividing seasonal cumulative emissions by total N in aboveground biomass of rice plants at harvest; for this the crop N content and the dry weight of aboveground biomass (i.e., straw + grain) was determined. Effects of E_{CO_2} on soil N_2O emissions (i.e., ΔN_2O) were calculated as the difference in seasonal N_2O emissions between E_{CO_2} and A_{CO_2}. Similarly, effects of E_{CO_2} on aboveground N uptake (i.e., ΔN uptake) were calculated as the difference in total aboveground N uptake between E_{CO_2} and A_{CO_2}. The N surplus was calculated as total N fertilization rate minus plant N uptake in aboveground biomass at harvest. The N use efficiency (NUE) was computed as the percentage of difference in total aboveground N uptake between the fertilized and unfertilized treatments in relation to the rate of N fertilizer application.

All data were analyzed using the SPSS 19.0 statistical package for Windows (SPSS China). Prior to analysis of variance, all data (or log-transformed data if necessary) were tested for normality using the non-parametric Kolmogorov–Smirnov test. Analysis of variance (ANOVA) was performed to test the significance of E_{CO_2} effects on N_2O emissions and crop yields under various fertilization treatments. Linear or nonlinear regression analysis was used to test the correlations between environmental and management-related variables and N_2O emissions. A significant level of $p < .05$ was utilized for all tests unless stated otherwise.

3 | RESULTS

3.1 | Environmental and agronomic variables

Across all study years, the seasonality of air and soil temperatures was comparable (Figures 1 and 2), with mean soil temperatures during the rice season ranging from 24.3°C to 24.5°C (Wuxi: 2001–2003) and from 23.1°C to 23.5°C (Jiangdu: 2004–2006). There was no significant difference in soil temperatures between E_{CO_2} and A_{CO_2}

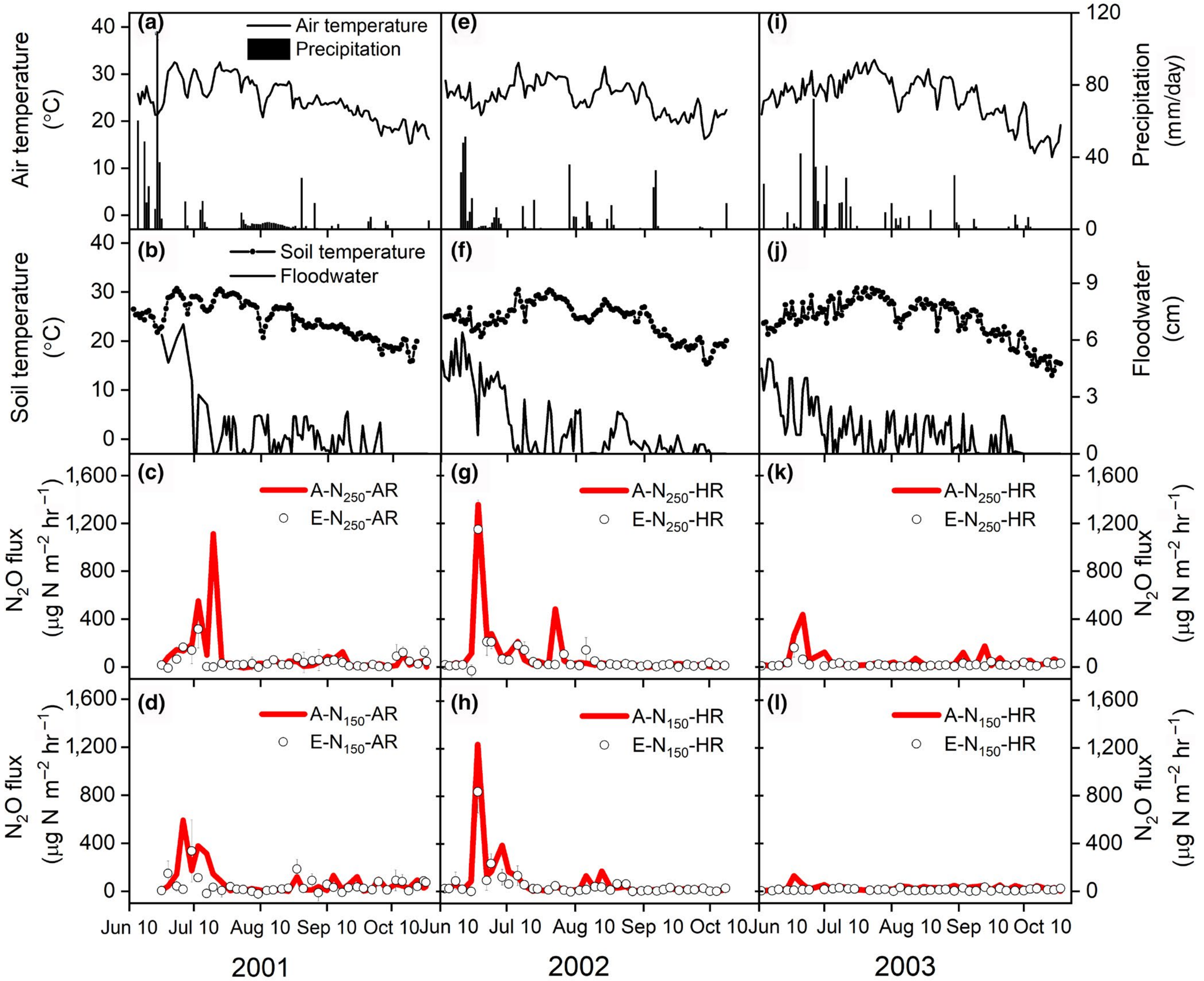

FIGURE 1 Seasonal variations in (a, e and i) air temperature and daily precipitation, (b, f and j) soil temperature and floodwater depth and (c, g, k, d, h and l) nitrous oxide (N_2O) fluxes for different fertilization treatments under elevated CO_2 (E) and ambient CO_2 (A) conditions during 2001–2003 rice seasons at the Wuxi site. Definitions of N_{150}, N_{250}, AR and HR are referred to in Table 1 and in the text [Colour figure can be viewed at wileyonlinelibrary.com]

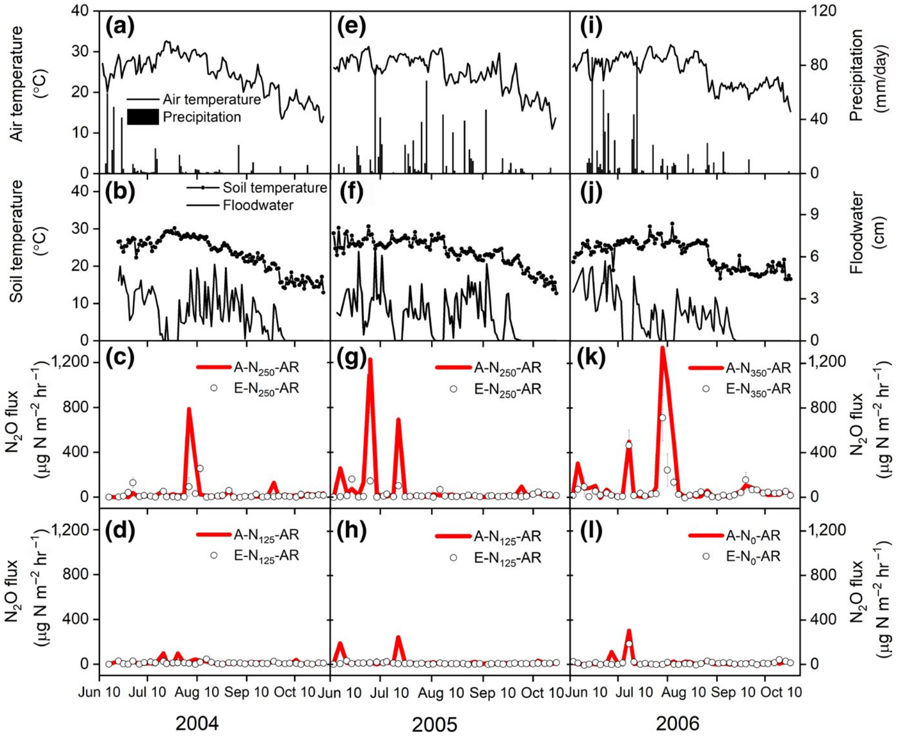

FIGURE 2 Seasonal variations in (a, e and i) air temperature and daily precipitation, (b, f and j) soil temperature and floodwater depth and (c, g, k, d, h and l) nitrous oxide (N_2O) fluxes for different fertilization treatments under elevated CO_2 (E) and ambient CO_2 (A) conditions during 2004–2006 rice seasons at the Jiangdu site. Definitions of N_0, N_{125}, N_{250}, N_{350} and AR are referred to in Table 1 and in the text [Colour figure can be viewed at wileyonlinelibrary.com]

plots. The seasonal amount and distribution of precipitation varied inter-annually, e.g., seasonal rainfall at the Wuxi site was 517, 413 and 455 mm, respectively, for the 2001, 2002 and 2003 rice seasons. At the Jiangdu site, the seasonal rainfall was lower in 2004 (322 mm) as compared to 2005 (658 mm) and 2006 (640 mm).

At both sites and across all N fertilization schemes, rice straw and grain yields responded positively to E_{CO_2} (Table 2; Table S1). However, aboveground rice (straw + grain) yield responses to E_{CO_2} increased with increasing rates of N fertilization (Wuxi-N_{250}: +13%–22%; Wuxi-N_{150}: +9%–11%; Jiangdu-N_{250}: +13%–40%; Jiangdu-N_{125}: +4%–37%; Jiangdu-N_{350}: +19%–30%), while no significant E_{CO_2} effect was observed for non-fertilization (N_0) treatments.

Rice grown under E_{CO_2} showed lower total N concentrations in straw and grain, while total aboveground N uptake was significantly increased in treatments fertilized with ≥150 kg N/ha in 13 out of 15 cases (Table 2; Table S1). Such an effect was only observed in one out of nine cases for treatments receiving 125 kg N/ha or no N fertilization (i.e., N_{125} and N_0). As observed for dry matter yields, effects of E_{CO_2} on aboveground N uptake (i.e., ΔN uptake) increased with increasing N application rates (Figure 4). In contrast, the N surplus for all N fertilization treatments was lower for E_{CO_2} as compared to A_{CO_2}(Table S1). Consequently, the NUE was strongly enhanced under E_{CO_2} (NUE: 37%–39%) as compared to A_{CO_2} (NUE: 22%–30%) during the 2006 rice season.

3.2 | Nitrous oxide fluxes

As shown in Figures 1–3, seasonal patterns of N_2O fluxes across the six site-years were characterized by high pulse emissions, triggered by N fertilization (basal fertilization, top dressing) and water management (mid-season drainage) events, and relative low emissions (<30 μg N m^{-2} hr^{-1}) during other times. Peak emissions were variable and ranged from 437 μg N m^{-2} hr^{-1} (in 2003) to 1,356 μg N m^{-2} hr^{-1} (in 2002) at the Wuxi site, and from 787 μg N m^{-2} hr^{-1} (in 2004) to 1,340 μg N m^{-2} hr^{-1} (in 2006) at the Jiangdu site. Generally, peak N_2O emissions under E_{CO_2} were significantly lower than under A_{CO_2}, while for the rest of cropping periods N_2O fluxes between E_{CO_2} and A_{CO_2} were similar.

At the Wuxi site, area-scaled N_2O emissions during the 2001–2003 rice seasons were significantly lower (−23% to −61%) under E_{CO_2} as compared to A_{CO_2} (Table 3; Table S2). Comparable observations were also made for the Jiangdu site and for the 2004–2006 rice seasons. Here, area-scaled seasonal N_2O emissions under E_{CO_2} were reduced by 28%–73% for treatments receiving high rates of N fertilization (i.e., N_{250} and N_{350}). A comparable, though not statistically significant trend was also observed for treatments receiving low or no N fertilizer applications (i.e., N_{125} and N_0). Overall, integrating the observational results obtained at both sites and considering all N fertilization treatments during 2001–2006, the difference in seasonal cumulative N_2O emissions between E_{CO_2} and A_{CO_2} (i.e., ΔN_2O) in dependence of N fertilization rate could be described best by a negative linear relationship (Figure 4).

For the 2006 rice season we calculated direct emission factors of N_2O, as only for that year an unfertilized treatment was included in our measurements. The direct emission factors of N_2O ranged from 0.67% to 1.0% for the N_{350} treatments under A_{CO_2}, while they were reduced to 0.52% to 0.53% under E_{CO_2}.

When seasonal cumulative emissions were expressed relative to total N uptake of aboveground biomass, the yield-scaled N_2O emissions varied in the range of 2.47–12.5 g N/kg N uptake under

TABLE 2 Changes (in %) caused by the elevated CO_2 (E) relative to ambient CO_2 (A) in yields of rice straw and grain and the N uptake of aboveground biomass (i.e., straw + grain) for different fertilization treatments

			Changes ((E − A)/A × 100%)							
Site	Year	Code[a]	Straw	*p*	Grain	*p*	Straw + grain	*p*	N uptake	*p*
Wuxi	2001	N_{250}-AR	19	*	16	*	18	*	13	*
		N_{150}-AR	9	*	12	*	11	*	5	*
	2002	N_{250}-HR	12	*	15	*	13	*	10	*
		N_{150}-HR	11	*	7	n.s.	9	n.s.	4	n.s.
	2003	N_{250}-HR	32	*	11	*	22	*	11	*
		N_{150}-HR	11	*	12	*	11	*	7	*
Jiangdu	2004	N_{250}-AR	24	*	25	*	24	*	19	*
		N_{250}-HR	8	n.s.	19	*	13	*	8	*
		N_{250}-NR	10	*	44	*	25	*	11	*
		N_{125}-AR	0	n.s.	8	*	4	n.s.	2	n.s.
		N_{125}-HR	15	*	11	*	13	*	9	n.s.
		N_{125}-NR	22	*	14	*	18	*	6	n.s.
	2005	N_{250}-AR	11	n.s.	28	*	18	*	6	n.s.
		N_{250}-HR	21	*	27	*	24	*	27	*
		N_{250}-NR	34	*	50	*	40	*	37	*
		N_{125}-AR	−4	n.s.	31	*	11	n.s.	8	n.s.
		N_{125}-HR	17	*	21	*	19	*	9	n.s.
		N_{125}-NR	39	*	34	*	37	*	32	*
	2006	N_{350}-AR	24	*	13	*	19	*	20	*
		N_{350}-HR	28	*	10	*	20	*	12	*
		N_{350}-NR	45	*	13	*	30	*	24	*
		N_0-AR	8	n.s.	13	n.s.	10	n.s.	−2	n.s.
		N_0-HR	6	n.s.	1	n.s.	4	n.s.	−9	n.s.
		N_0-NR	6	n.s.	6	n.s.	6	n.s.	−13	n.s.

[a]Definitions of the treatment codes are referred to in Table 1 and in the text. The symbol * indicates the statistical significance at $p < .05$, while n.s. indicates no significance, i.e., $p > .05$.

E_{CO_2} and 6.09–19.4 g N/kg N uptake under A_{CO_2} at the Wuxi site, and from 2.08 to 10.5 g N/kg N uptake and 3.46 to 22.3 g N/kg N uptake for E_{CO_2} and A_{CO_2}, respectively, at the Jiangdu site (Table S2). Similar to the area-scaled emissions, yield-scaled N_2O emissions were consistently lower (−30% to −65%) under E_{CO_2} as compared to A_{CO_2} at the Wuxi site (Table 3). Also, at the Jiangdu site yield-scaled N_2O emissions for high N fertilization rates (N_{250} and N_{350}) were significantly reduced under E_{CO_2} as compared to A_{CO_2}, while for low or no N fertilization (N_{125} and N_0) treatments this effect was not significant.

During the 2004–2006 rice seasons, different crop residue addition rates (i.e., NR, HR and AR) at a given N fertilization level did not significantly affect seasonal N_2O emissions for both E_{CO_2} and A_{CO_2}, irrespective of their expression on either an area- or yield-scaled bases (Table 3; Table S2).

Based on the data presented in Tables S1 and S2, we developed an exponential model which well described the positive relationship between N surplus and yield-scaled N_2O emissions across treatments (Figure 5). This model shows that variations in N surplus explain 53% and >90% of the variance in yield-scaled N_2O emissions at the Jiangdu and Wuxi site, respectively.

4 | DISCUSSION

Contrary to our hypothesis that soil N_2O fluxes would increase under conditions of E_{CO_2}, the current data clearly show that soil N_2O emissions from the fertilized rice systems were lower under E_{CO_2} as compared to those observed under A_{CO_2}. The decrease in seasonal emissions was due to a sharp reduction in N_2O fluxes under E_{CO_2} following fertilization and alternate wetting and drying events, but not during the rest of the season (Figures 1–3). However, total emissions during these peak flux periods generally contributed 50%–85% to seasonal cumulative emissions. Across the 6 site-years, seasonal N_2O emissions from rice systems under A_{CO_2} were estimated to be 0.43–4.11 kg N/ha, which falls within the range of estimates reported for

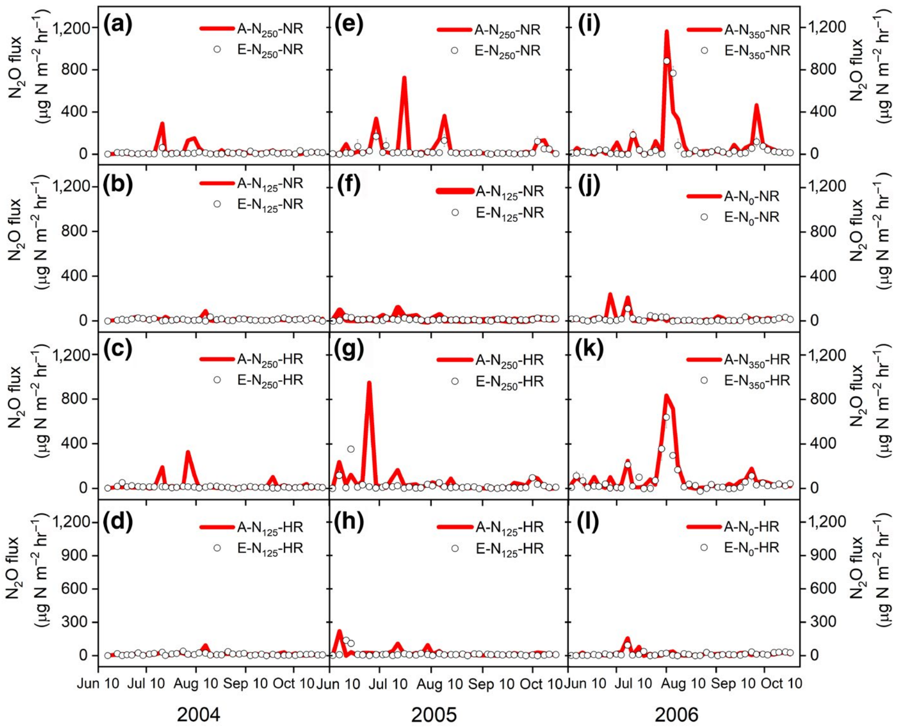

FIGURE 3 Seasonal variations in (a–l) nitrous oxide (N_2O) fluxes for different fertilization treatments under elevated CO_2 (E) and ambient CO_2 (A) conditions during 2004–2006 rice seasons at the Jiangdu site. Definitions of N_0, N_{125}, N_{250}, N_{350}, HR and NR are referred to in Table 1 and in the text [Colour figure can be viewed at wileyonlinelibrary.com]

intermittent waterlogged rice systems worldwide (0.026–4.42 kg N/ha, Akiyama et al., 2005). Our study is the first quantifying seasonal N_2O emissions under E_{CO_2} (+200 μmol/mol) using the FACE technology, with cumulative emissions ranging from 0.27 to 2.30 kg N/ha (Table S2). These values might be compared with results obtained by growing rice under varying climate and fertilization conditions in open-top chambers, with seasonal emissions ranging from 0.86 to 2.17 kg N/ha for a CO_2 enrichment of 155–175 μmol/mol (Bhattacharyya et al., 2013; Pereira et al., 2013).

Our observation that the reduction in soil N_2O emissions under E_{CO_2} is greatly dependent on the rate of N fertilization generally supports the assumption that the response of soil N_2O fluxes to E_{CO_2} in rice systems will depend on soil N availability. While significant effects of E_{CO_2} on soil N_2O emissions (−23% to −73%) were observed for treatments with N fertilizer application rates ≥150 kg N/ha (i.e., N_{150}, N_{250} and N_{350}), effects remained mostly insignificant for treatments with N fertilizer application rates ≤125 kg N/ha (i.e., N_{125} and N_0; Table 3). This finding might help to understand the so far largely varying results regarding effects of E_{CO_2} on soil N_2O fluxes from terrestrial ecosystems, with positive (e.g., Kammann et al., 2008), negative (e.g., Xu et al., 2012) and no significant effects (e.g., Liu et al., 2018; Mosier et al., 2002; Pereira et al., 2013) being reported. It is well recognized that N_2O fluxes in terrestrial ecosystems are primarily regulated by environmental factors (e.g., soil water content) and nitrogen and carbon availability (e.g., Butterbach-Bahl et al., 2013; Conrad, 1996). However, all these factors are affected by E_{CO_2}. For example, E_{CO_2} may change soil inorganic N (ammonium and nitrate) contents as a result of progressive N accumulation in plant biomass (Luo et al., 2004), which depresses soil N availability, and thus N_2O emissions (Pleijel et al., 1998). Moreover, plants grown under E_{CO_2} have been shown to exhibit higher belowground plant-C allocation and higher root exudation (Milchunas et al., 2005), which stimulates microbial processes and subsequent N_2O emissions (Ineson et al., 1998). In the studied rice systems, a competition for soil N between plants and microbial N turnover is manifested by the observation that crop yields in terms of straw and grain were significantly stimulated by higher rates of N fertilization under E_{CO_2} (Table 2; Table S1). This generally agrees with findings by other researchers that under N-limiting conditions no significant E_{CO_2} effects are visible for upland plants (e.g., Kettunen et al., 2005; Stitt & Krapp, 1999). Also, our correlation analysis, which shows that the observed reduction in soil N_2O emissions (i.e., ΔN_2O) decreases linearly with increasing fertilization level, strongly indicates that the observed inhibitory effect of E_{CO_2} upon N_2O emissions from rice systems is due to a reduced substrate availability for microbial N turnover processes. This interpretation is further supported by the difference in plant N uptake between E_{CO_2} and A_{CO_2} (i.e., ΔN uptake), which is amplified with increasing N application rates (Figure 4). By using stable isotope techniques Baggs, Richter, Cadisch et al. (2003) showed that the ratio of N_2 to N_2O was often higher during the denitrification under E_{CO_2}, which might be as well explained by soil N availability as at low soil nitrate level denitrification tends to be complete and to predominantly end with N_2 as end product (Butterbach-Bahl et al., 2013). Unfortunately, in our study we did not measure soil ammonium (NH_4^+) and nitrate (NO_3^-) concentrations under E_{CO_2} and A_{CO_2} conditions due to logistical reasons. However, E_{CO_2}-induced changes in soil microbial community might have also affected the observed reduction in soil N_2O emissions in our rice systems. Sun et al. (2018) reported that the ratio of nitrite-reducing bacteria abundance (i.e., *nir*S + *nir*K) to N_2O-reducing bacteria abundance (i.e., *nos*Z) in rice systems was significantly lower under E_{CO_2} as compared to observations at ambient CO_2 concentrations. The work of Baggs,

TABLE 3 Changes (in %) caused by the elevated CO_2 (E) relative to ambient CO_2 (A) in seasonal cumulative nitrous oxide (N_2O) emissions, as expressed either on an area basis or relative to total N uptake of aboveground biomass for different fertilization treatments

Site	Year	Code[a]	Changes ((E − A)/A × 100%) Area-scaled N_2O flux	p	Yield-scaled N_2O flux	p
Wuxi	2001	N_{250}-AR	−48	*	−54	*
		N_{150}-AR	−51	*	−54	*
	2002	N_{250}-HR	−23	*	−30	*
		N_{150}-HR	−32	*	−35	*
	2003	N_{250}-HR	−61	*	−65	*
		N_{150}-HR	−57	*	−59	*
Jiangdu	2004	N_{250}-AR	−49	*	−57	*
		N_{250}-HR	−61	*	−64	*
		N_{250}-NR	−57	*	−61	*
		N_{125}-AR	−35	n.s.	−37	n.s.
		N_{125}-HR	−18	n.s.	−23	n.s.
		N_{125}-NR	−23	n.s.	−26	n.s.
	2005	N_{250}-AR	−73	*	−74	*
		N_{250}-HR	−60	*	−68	*
		N_{250}-NR	−58	*	−70	*
		N_{125}-AR	−62	*	−64	*
		N_{125}-HR	−43	n.s.	−48	n.s.
		N_{125}-NR	−38	n.s.	−52	n.s.
	2006	N_{350}-AR	−44	*	−54	*
		N_{350}-HR	−31	*	−38	*
		N_{350}-NR	−28	*	−42	*
		N_0-AR	−26	n.s.	−25	n.s.
		N_0-HR	−23	n.s.	−15	n.s.
		N_0-NR	−41	*	−32	n.s.

[a]Definitions of the treatment codes are referred to in Table 1 and in the text. The symbol * indicates the statistical significance at $p < .05$, while n.s. indicates no significance, i.e., $p > .05$.

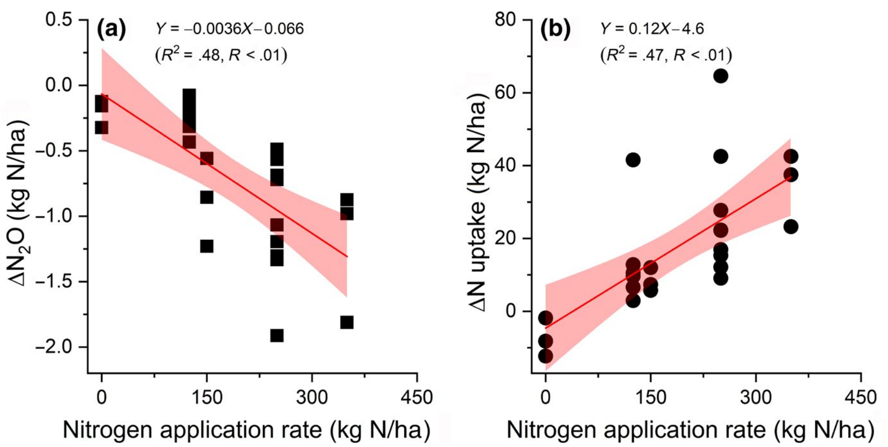

FIGURE 4 Effects of elevated CO_2 on differences in (a) the seasonal cumulative nitrous oxide (N_2O) emissions and (b) N uptake of aboveground biomass in dependence of the N fertilizer application rate. Effects of elevated CO_2 on N_2O emission (ΔN_2O) and N uptake (ΔN uptake) were calculated as the difference in mean values while comparing results from the elevated CO_2 treatments with those of the ambient control treatments. The shaded areas represent 95% confidence bands [Colour figure can be viewed at wileyonlinelibrary.com]

Richter, Cadisch et al. (2003) and Sun et al. (2018) suggests that E_{CO_2} generally facilitates N_2O consumption and favors N_2 production through denitrification, with an overall reducing effect on soil N_2O emissions.

Nevertheless, E_{CO_2} often increases belowground plant-C allocation with the enhancement in rice growth. Though belowground C development was not measured in this study, Yang et al. (2008) and Hu et al. (2020) found significant increases in root biomass under E_{CO_2} in supplementary studies on other seasons/years at the Wuxi and Jiangdu sites. Such higher root growth may provide C supply source for heterotrophic processes such as denitrification or fungal co-denitrification and associated N_2O fluxes (Kammann et al., 2008),

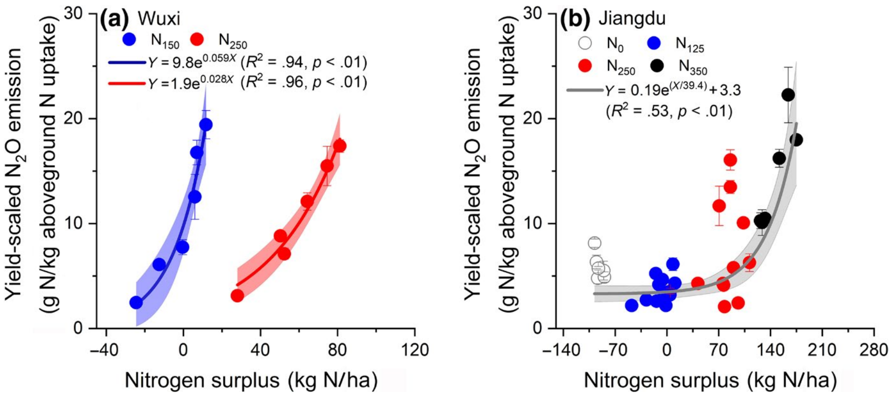

FIGURE 5 Relationships between the nitrogen surplus and yield-scaled nitrous oxide (N_2O) emissions at the (a) Wuxi and (b) Jiangdu sites. The shaded areas represent 95% confidence bands [Colour figure can be viewed at wileyonlinelibrary.com]

while on the other hand increased belowground C may increase microbial demand for inorganic N, and thus, resulting in a temporary immobilization of soil N (Hungate et al., 1999) and a reduction of N_2O emissions. Based on our experiments we only can speculate that both effects are canceling each other out, i.e., that the effect of soil N immobilization due to increased C rhizo-deposition is likely in the same magnitude as the stimulating effect due to the provisioning of additional C substrate. This was partially supported by our observation that different rates of crop residue addition (i.e., NR, HR and AR) did not significantly affect soil N_2O emissions under both E_{CO_2} and A_{CO_2} conditions (Figures 2 and 3).

The metric of yield-scaled emissions has been proposed as an effective tool to value the environmental cost of crop production (Van Groenigen et al., 2010, 2013). In our study, the estimated yield-scaled N_2O emissions for rice systems were in the range of 3.46–22.3 g N/kg N uptake under A_{CO_2} (Table S2). Based on a meta-analysis of yield-scaled N_2O emissions from non-leguminous annual crops, Van Groenigen et al. (2010) estimated that yield-scaled N_2O emissions were in the range of 8.4 to 26.8 g N/kg N uptake, i.e., comparable to our estimates. Moreover, our study now provides first estimates of yield-scaled N_2O emissions under E_{CO_2}, which were 2.08–12.5 g N/kg N uptake for rice systems (Table S2). In contrast to our hypothesis, E_{CO_2} reduced yield-scaled N_2O emissions as compared to A_{CO_2} due to two effects: (a) reductions in soil N_2O emissions and (b) increased crop growth and plant N uptake. Overall, our data show a positive exponential response of yield-scaled N_2O emissions to increasing values of N surplus (Figure 5), thus, corroborating the finding by Van Groenigen et al. (2010). This suggests that apart from the NUE (EUNEP, 2015; SDSN, 2015; Yao, Yan, et al., 2017), the N surplus is a valuable indicator for assessing agricultural N management and controlling environmental costs in terms of N_2O emissions of crop production. The general importance of N surplus for assessing potential losses of N to the environment from agricultural soils has been acknowledged as well by various countries and organizations (Oenema et al., 2005; Zhang et al., 2015, 2019).

It has to be mentioned that although E_{CO_2} increases crop NUE and subsequent production of rice grain, it goes as well along with a significant decline in integrated rice quality, including proteins, micronutrients and vitamins (Yang, Wang, et al., 2007b; Zhu et al., 2018). Besides, Van Groenigen et al. (2013) conducted a meta-analysis that indicated that E_{CO_2} increases soil CH_4 emissions by on average 42.2% or yield-scaled CH_4 emissions by on average 31.4% across rice systems worldwide. At the Wuxi Rice-FACE site, Zheng et al. (2006) measured soil CH_4 emissions from the same plots and at the same time along with N_2O fluxes reported in this study. They found that E_{CO_2} stimulated CH_4 emissions by on average 41.7 kg C/ha across the studied 2001–2003 rice seasons. This increase in CH_4 emissions under E_{CO_2} is equivalent to 1,890 kg CO_2-eq/ha (based on a global warming potential of 34 for CH_4 if compared to CO_2, IPCC, 2013). In our study, for the Wuxi site E_{CO_2} reduced N_2O emissions by on average 0.95 kg N/ha, equivalent to 445 kg CO_2-eq/ha (based on a global warming potential of 298 for N_2O if compared to CO_2, IPCC, 2013). Thus, integrating the findings of E_{CO_2} effects on N_2O (this study) and CH_4 (Zheng et al., 2006) shows that total non-CO_2 GHG emissions from rice systems will rise with increasing atmospheric CO_2, but that the E_{CO_2}-induced reductions in soil N_2O emissions negate as much as 24% of the increases in soil CH_4 emissions. However, this increase in non-CO_2 GHG emissions from rice systems under E_{CO_2} can be significantly reduced by improving water regimes and using alternative fertilization practices such as alternate wetting and drying irrigation schemes, ground cover rice production system and urea deep placement (Beach et al., 2008; Linquist et al., 2015; Yao, Zheng, Liu, et al., 2017; Yao, Zheng, Zhang, et al., 2017). Also, adopting rice cultivars with potentially strong responses to E_{CO_2} may eventually not only improve crop yield but also enhance soil C sequestration (Hu et al., 2020), thereby curbing the future growth of greenhouse effect contributed by rice systems.

5 | CONCLUSIONS

On the basis of the Rice-FACE experiments in subtropical China, our study shows that under common agricultural practice increasing atmospheric CO_2 will result in decreasing soil N_2O emissions and increasing rice yields in terms of straw and grain. Our results further show that the reduction of N_2O emissions is strongly linked to increasing plant N uptake of the crop under elevated CO_2. The positive exponential curve describing the increase in yield-scaled N_2O emissions with increasing N surplus shows that N surplus is a strong indicator for assessing environmental costs of crop production. While under elevated CO_2 soil N_2O emissions from rice systems can be expected to decrease, other studies show the opposite is true for soil CH_4 emissions. Our findings emphasize the need for exploring alternative irrigation and fertilization schemes for rice to mitigate CH_4 emissions, while stabilizing or even increasing soil C stocks. Overall, under future atmospheric CO_2 concentrations soil N_2O emissions from rice systems seem to play a rather low role for the greenhouse gas balance of rice production.

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DATA AVAILABILITY STATEMENT

All data used for this study are freely available from the corresponding author upon the reasonable request.

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section.

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