



The main drivers of methane emissions differ in the growing and flooded fallow seasons in Mediterranean rice fields

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Received: 2 July 2020 / Accepted: 14 December 2020 / Published online: 9 January 2021
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Abstract

Purpose To assess 1) the cumulative greenhouse gas emissions –GHG- and global warming potential (methane – CH₄- and nitrous oxide) from rice fields in the growing and fallow seasons, and 2) the environmental and agronomic drivers of CH₄ emissions, and their relative capacity to explain CH₄ variation.

Methods A two-year multisite field experiment covering the agronomic and environmental variability of a rice growing area in NE Iberian Peninsula was conducted with monthly samplings of GHG and monitoring of both environmental and agronomic factors. Information-

theoretic framework analysis was used to assess the relative contribution of the environmental and agronomic variables on methane emissions.

Results Two thirds of the CH₄ is emitted in the fallow season. Edaphic factors exert more influence during the growing season whereas agronomic factors have a higher impact in the fallow. The implications of these findings on the design of improved mitigation options rice are discussed.

Conclusions Soils with higher soil sulphate concentration, bulk density and clay content emit less CH₄ in growing season. In the fallow season, the rates of both straw input and nitrogen fertilization stimulate CH₄ emissions.

Responsible editor: Zucong Cai

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Keywords Greenhouse gas emissions · Paddy rice · Post-harvest season · Seasonal methane emissions · Straw management · Soil sulfate content

Abbreviations

CH ₄	methane
DOC	dissolved organic carbon
GHG	greenhouse gas
GLMz	generalized linear models
GWP	global warming potential
N	nitrogen
NUE	nitrogen use efficiency
N ₂ O	nitrous oxide
SO ₄ ²⁻	sulphate
SOM	soil organic matter
SP	selection probability

Introduction

Anthropogenic greenhouse gas (GHG) emissions associated with agricultural production has increased 81% (from 3.1 Gt CO₂-eq yr⁻¹ to 5.8 Gt 17 CO₂-eq yr⁻¹) during the period 1961–2016 (IPCC 2019). The increase from agricultural activities is primarily due to methane (CH₄) emissions from paddy rice fields (Schaefer et al. 2016) which contribute 48% of cropland greenhouse gas emissions (Carlson et al. 2017). Because this flux is such a substantial portion of the global methane budget, it represents a large potential for GHG mitigations (IPCC 2019).

Methane emissions from paddy rice agroecosystems are a function of overall CH₄ production, oxidation, and release to the atmosphere (Cai et al. 2007). A suite of biologic, environmental and agronomic factors modulates the dynamics of CH₄ emissions. Soil microbiota contributes substantially to GHG emissions through concurrent CH₄ production and consumption processes in the soil. Anaerobic soils of paddy rice provide the proper environmental conditions for anaerobic bacteria and methanogenic archaea, thriving syntrophically on the degradation of soil native or plant-derived organic carbon (Conrad 2007). Nevertheless, a substantial fraction of CH₄ is oxidized by methanotrophic bacteria at the anoxic-oxic interfaces at the soil surface and in the rhizosphere (Watanabe et al. 1997), and also by anaerobic methane-oxidizing community (Nauhaus et al. 2005). The remaining CH₄ surplus is eventually released to the atmosphere, primarily through the aerenchyma of the rice plants (Cicerone and Shetter 1981; Laanbroek 2010). Consequently, the physiological traits of the rice plants and the associated crop management (e.g., flooding) are key factors in CH₄ emissions by providing a carbon source and anoxic environment to methanogens (Lu and Conrad 2005; Tokida et al. 2011) as well as an oxidative interface for the methanotrophs (Jiang et al. 2017).

Fertilization and crop residue management are, under permanent flooding conditions, major agronomic factors affecting CH₄ emissions in rice paddy fields (Linguist et al. 2018). Nitrogen fertilization has been widely studied but results on the net impact on CH₄ emissions have been mixed (Wassmann et al. 1993; Banik et al. 1996; Shang et al. 2011) while in the case of sulphate fertilizers, there is a general consensus that they can reduce CH₄ emissions (Rath et al. 1999; Denier van der Gon et al. 2001; Minamikawa et al. 2005). The

straw that remains in the field after the rice harvest is important for the overall CH₄ emissions and carbon budget because, depending on its management, it represents a large input of organic matter available for soil decomposing microorganisms. There are a variety straw management practices implemented worldwide, such as removal, mulching, composting, burning and incorporation into the soil; the selection is crop-context dependent. Finally, edaphic factors, mainly soil texture, are also relevant. There is wide consensus that coarser soil textures lead to higher emissions (Sass et al. 1994; Brye et al. 2013, 2016). However, soil texture is closely linked to rice productivity (Dou et al. 2016), fertilizer use efficiency of the crop (Alhaj Hamoud et al. 2019) and agricultural practices since farmers adapt their management to soil conditions. Therefore, the effect of soil texture favouring the gas diffusivity can be modulated by the agronomic management and the performance of the crop.

The interaction of these biological, environmental and, agronomic factors confers a complex system regulating CH₄ dynamics that yields variable cumulative emissions across and within the different rice growing areas. Addressing these processes individually may fail in both identifying the main processes responsible for net CH₄ emissions and provide realistic and precise estimations of the emissions. In spite of this, only a few studies have used a multivariate approach to assess GHG emissions in rice agroecosystem (e.g. Jia et al. 2002; Yan et al. 2005; Brye et al. 2016; Knox et al. 2016), but in most cases analyses were not controlled for collinearity among variables.

While N₂O emissions from continuously flooded rice fields are widely accepted to be negligible or small (Linguist et al. 2015; Wang et al. 2016) there are some managements, mainly related to nitrogen fertilization and water management that can induce N₂O pulses (Cai et al. 1997; Kritee et al. 2018; Lagomarsino et al. 2016; Pittelkow et al. 2013). Further, the warming potential of this gas is 298 times as high as that of CO₂ (Smith et al. 2014). Then, it is necessary to assess N₂O emissions to accurately estimate global warming potential of rice fields.

To address such complexity a two-year field study was conducted in a Mediterranean rice growing area in the Ebre Delta (Catalonia, Northeast Spain) based on a farm-to-farm approach covering the whole range of agronomic and environmental variability of the area. In a previous paper resulting from this study (Martínez-

Eixarch et al. 2018), we identified the key dynamic variables modulating CH₄ fluxes, including environmental (soil physic-chemical parameters such as temperature, pH, redox and conductivity) and agronomic (water level, plant cover) factors. In the present report, we focus on the relative effect of static factors (rather than dynamic) on cumulative (rather than fluxes) greenhouse gas emissions and the resulting global warming potential. Therefore, the main objectives set for the present report were to study the effect of static features (i.e. soil properties), agronomic management (i.e. fertilizer rates and straw incorporation), and productive traits (i.e. plant density, panicle density, and grain yield) on cumulative GHG emissions, including both CH₄ and N₂O. We applied a multivariate analysis and an information theoretic approach to find the best explanatory model. This approach allowed us to control for variables collinearity and calculate the relative importance of each independent variable in GHG emissions.

Material and methods

Study area

Rice production in Spain accounts for 28% and 5% of the European production and crop extension, respectively, while in the Ebre Delta (Catalonia, NE Spain), it covers 21,125 ha, representing ca. 19% of the total rice growing area in Spain (Spanish Ministry of Agriculture, Food and Environment).

The climate of the region is Mediterranean with a mean annual precipitation of about 500 mm, mostly distributed during spring and autumn. The mean annual air temperature is 18 °C, ranging from 23 °C to 27 °C during the rice growing season (May to September) and from 6 °C to 13 °C during the post-harvest (October to December).

Rice cultivation in the Ebre Delta is divided in three periods: pre-sowing (January to early April), growing (late April to September) and post-harvest (October to December) seasons. The fields are left fallow over the post-harvest and pre-sowing seasons. This study covers the growing and post-harvest seasons.

Water management follows an annual pattern, with fields permanently flooded over the growing season to a depth ca. 5 to 15 cm deep. In the post-harvest season, fields are either flooded or left to progressively drain, according to the farmers' preferences, but even if the latter is the case, fields remain flooded and then soil-

saturated for most of this season, as straw addition is practiced in flooded conditions (puddling). Finally, fields are dry over the pre-sowing season because the irrigation supply from the canal network is cut off in December.

Soil operations are conducted in the dry pre-sowing season, i.e., harrowing and fertilizer application. The average mineral nitrogen fertilizer application rates range from 170 to 200 kg N ha⁻¹. Irrigation from the hydraulic network is provided from mid-April, then fields are flooded, and water seeded from late April to mid-May. The cultivars used in the area are Japonica-type with medium grain size and a growth cycle of ca. 120 and 140 days (from sowing to maturity). For this study, fields grown with two representative cultivars in the area, Gleva or JSendra, were selected. The harvest is conducted over the month of September, and it is followed by incorporation of the straw into the soil, mainly in October, which is the standard residue management.

Experimental layout

Experiments were conducted over the growing (June to September) and post-harvest seasons (October to December) in 2015 and 2016. Because CH₄ emissions were the priority of this study, samples were not collected during the pre-sowing season, when the fields are dry and emissions are minimal, in order to focus the sampling effort on the flooded or soil-saturated periods, when methane emission is expected. Furthermore, negligible CH₄ emission rates were found in May of 2015 (Martínez-Eixarch et al. 2018), so that, for the sake of optimizing resources, in the second year of the study gas sampling was started in June.

Hereafter, the post-harvest season will be referred as the flooded fallow season to stress both uncultivated and flooded conditions of the rice fields in this period. A total of 24 commercial rice fields (15 in 2015 and 9 in 2016), following standard agricultural management of the area, were selected for the study. Sites were widely distributed over the Ebre Delta (Fig. 1) to capture the environmental (soil properties and salinity, proximity to either river, sea, or lagoons) and agronomic (water management, phytosanitary applications and agronomic performance) variability of the area.

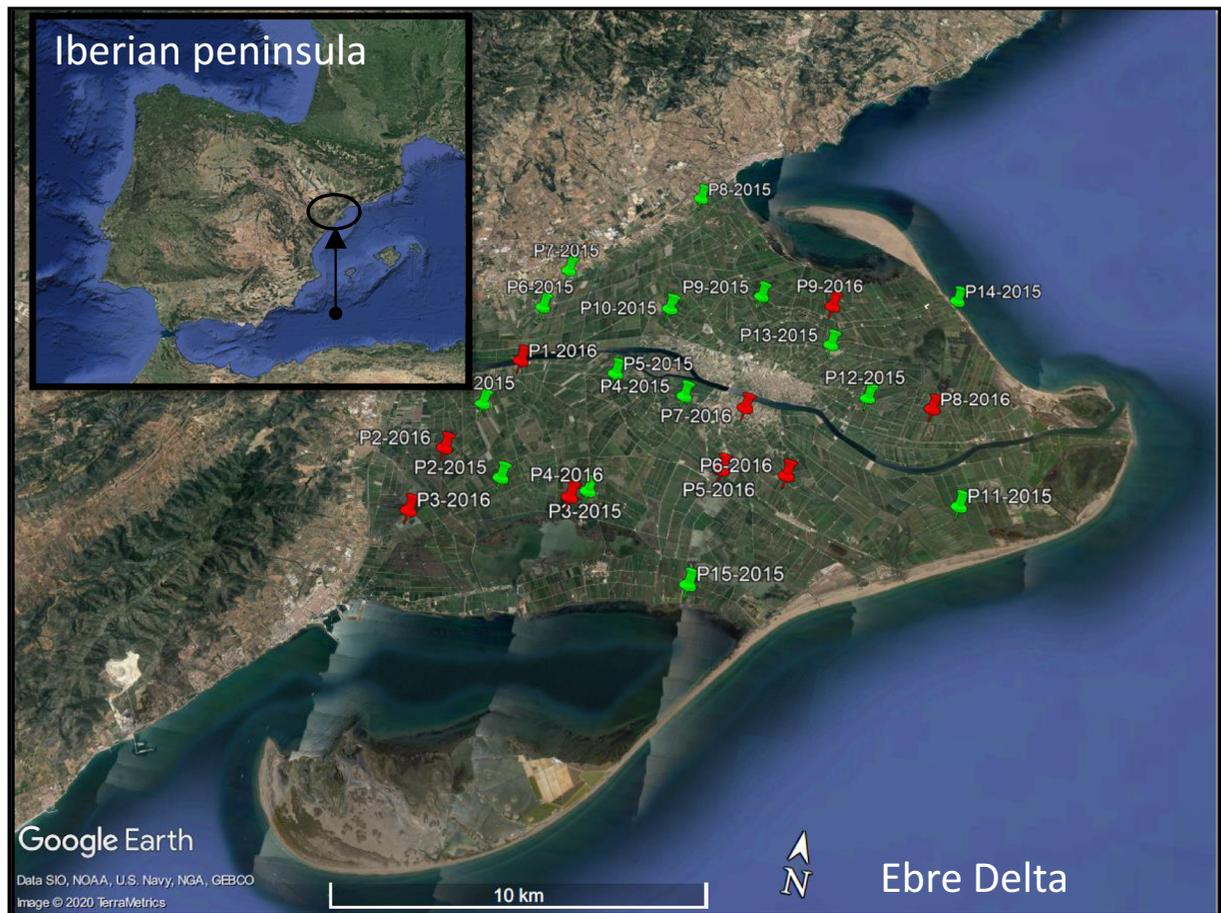


Fig. 1 Location of the 24 rice fields in Ebre Delta monitored in year 2015 (in green) and 2016 (in red)

Gas sampling and flux measurement

Gas sampling was conducted on a monthly basis, using non-steady state gas chambers (Altor and Mitsch 2008; Weishampel and Kolka 2008). The characteristics of the chambers as well as the procedure for chamber deployment and field sampling plan are detailed in Martínez-Eixarch et al. (2018). In brief, the chambers, were made of polyvinylchloride (PVC) structure covered by transparent plastic and they were equipped with a thermometer to monitor temperature within the chamber in each extraction. To avoid soil disturbance during gas sampling, blocks were installed in the field to support wooden boards to access the chamber. All the rice fields were sampled within the same day and consistently from 10:00 am to 3:00 pm to minimize variability derived from the daily emission variation (Hatala et al. 2012). During the sampling procedure, each gas sample was transferred overpressured to pre-evacuated 12.5 mL

vials (Labco Ltd., Buckinghamshire, UK) and sent to laboratory. Methane (CH_4) and nitrous oxide (N_2O) concentration was determined using a THERMO TRACE 2000 (Thermo Finnigan Scientific, USA) gas chromatograph equipped with a flame ionization detector (GC-FID). Analysis of N_2O were carried out using an Agilent 7820A GC System, (Agilent, USA) equipped with an electron capture detector (GC-ECD). The calibration of the gas chromatograph was carried out using a CH_4 and N_2O standard in nitrogen provided by Carbueros Metalicos S.A. (Spain).

The emission rates of the GHG, i.e. CH_4 and N_2O , were obtained from the change of concentration of the respective gas in chambers over the 30-min sampling period in each chamber. The emission rate was estimated by the slope of the linear regression between gas concentration and sampling time. The increase of temperature in the headspace of the chamber was considered to correct GHG concentration of each sample

according to the ideal gas law. Only significant linear regressions ($P < 0.05$ and $R^2 > 0.80$) were accepted, and non-significant regressions were considered as zero emission rates.

Monthly mean emissions rates and cumulative emissions in each field were calculated assuming constant emission rates over the entire month. Seasonal (growing and fallow seasons) and total (from June to December) cumulative GHG were calculated by summing monthly cumulative emissions. The overall global warming potential (GWP), including CH_4 and N_2O , was estimated using the IPCC factors, i.e. 25 and 298, respectively, over the 100-year time scale (Smith et al. 2014).

Climatic, edaphic, and agronomic measurements

Environmental and edaphic traits

Climatic data was obtained from a meteorological station located in Ebre Delta (Municipality of Amposta; GPS coordinates, 40.708; 0.632) belonging to the Web of Agrometeorological Stations of Weather Services of Catalonia (Catalan Government).

In March, before flooding, a soil composite three sub-samples per field was extracted for soil characterization: texture (USDA characterization), soil granulometry (% of clay, sand and silt) and dry bulk density (by granulometry), content of organic matter, sulphates, and nitrogen (by Spectrometry).

Agronomic management and crop production traits

Agricultural practices, including fertilization (source, timing and rates), water (hydroperiod and water layer depth) and straw (rate and time of straw input) management, and agronomic traits (plant and panicle density, grain yield (of the current and preceding growing cycle) in each field was recorded according to farmer's communication. In 2015, the time of straw incorporation was broadly monitored by registering whether this practice was done or not the day of gas sampling. In 2016, given the relevance of post-harvest emissions observed in the preceding year, more detail was registered by surveying the farmers. Plant d and panicle density was assessed by IRTA staff by counting plants and panicles in 10 to 15 delimited subareas of 0.25 m^2 each, distributed along a diagonal transect of the field. Grain yield of the current and preceding season was used as a proxy of

the rate of straw input, assuming a mean harvest index of 0.5 (Matías et al. 2019).

Statistical analysis

Generalized Linear Models (GLMz) were used to study the association between cumulative GHG emissions and agroecological factors based on an information–theoretic approach to find the best approximating models (Tabachnick et al. 2007). GLMz were built including all possible combinations of independent variables, but excluding interactions, due to the large number of variables considered. The models meeting the following criteria were accepted as candidates: significant improved performance in relation to the null model and variance inflation factor of ≤ 5 , in order to avoid multicollinearity effects in regression models (Maggini et al. 2006). The second order Akaike information criterion (AICc), rescaled to obtain ΔAICc values ($\Delta\text{AICc} = \text{AICc}_i - \text{minimum AICc}$) was performed to evaluate the degree of support of each candidate model. Further details on the criteria for the selection of the models are provided in Aparicio et al. (2018). Prior to the analysis, quantitative variables were transformed to improve linearity and homoscedasticity. Analyses were performed with the R software version 3.4.

Results

Climatic data

Climatic data are presented in Fig. 2. Mean annual and seasonal temperatures followed similar values and temporal variation over the two years of the study. The mean annual temperature was $16.5 \pm 1.1 \text{ }^\circ\text{C}$ with highest mean maximum thresholds recorded in July and August. The mean temperature during the growing season (from May to September) was on average 8 degrees higher ($22.0 \pm 0.8 \text{ }^\circ\text{C}$) than during the flooded fallow (October to December: $13.9 \pm 1.5 \text{ }^\circ\text{C}$). Cumulative rainfall was higher in 2015 (445.0 mm) than in 2016 (376.0 mm) yet seasonal distribution differed in the two years with more precipitation in the growing season of 2015 but less in 2016 compared to the flooded fallow.

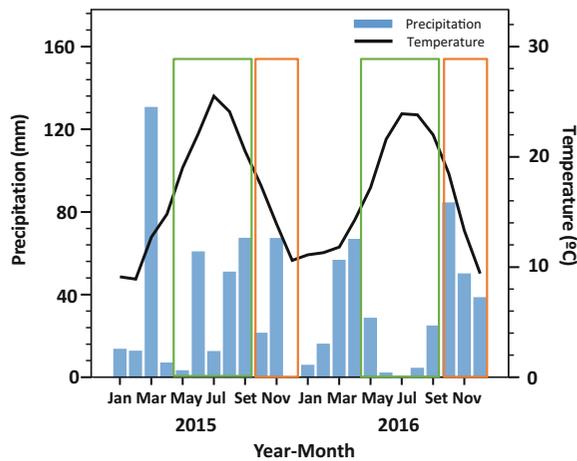


Fig. 2 Monthly mean temperatures and cumulative precipitation over the two years of the study (2015–2016). Rectangles represent the period of greenhouse sampling during the growing (green) and fallow (brown) seasons

Agronomic and soil characteristics of the rice fields

Agronomic and soil characteristics of the 24 monitored rice fields used in the study are detailed in Table 1.

Soil characteristics varied among rice fields as shown by the different soil textures, wide ranges of bulk density (809–1041 kg m⁻³), soil organic matter (1.89–7.70%), nitrogen (0.11–0.36%), sulphate (1928–6439 mg kg⁻¹), and the percent clay (11.3–36.7%) and sand (6.3–84.6%). Grain yield ranged from 5814 to 9897 kg paddy rice ha⁻¹. Rice fields yielded on average less in 2015 (7759.3 ± 255.4 kg paddy rice ha⁻¹) than in 2016 (8225.0 ± 698.5 kg paddy rice ha⁻¹) whereas the opposite pattern was found for panicle density (chronologically: 352.9 ± 12.2 vs. 303.0 ± 10.3 panicles m⁻²). Rates of N fertilization were similar in the two years (chronologically: 172.6 and 175.7 kg N ha⁻¹) but substantially lower sulphate fertilization rates were applied in 2016 (24.4 ± 5.6 kg S ha⁻¹) than in 2015 (72.2 ± 9.4 kg SO₄²⁻ ha⁻¹).

Total GHG emissions and global warming potential

In total, rice fields in Ebre Delta emitted 262.6 ± 5.9 kg CH₄ m⁻² ha⁻¹ (two-year mean) of which 36.9% was emitted during the growing season (96.6 ± 2.8 kg CH₄ ha⁻¹) and 63.1% during the flooded fallow season (163.9 ± 9.8 kg CH₄ ha⁻¹) mainly in October. October alone accounted for 45.4% of the annual emissions.

The monthly CH₄ emissions rates followed a bimodal pattern (Fig. 3) with the first peak occurring in August (4.7 ± 0.3 mg CH₄ m⁻² h⁻¹) and the second, and the highest, in October (15.7 ± 2.0 mg CH₄ m⁻² h⁻¹). The mean emission rate in the growing season was almost identical over the two years of the study (chronologically: 3.4 ± 0.1, 3.2 ± 0.2 mg CH₄ m⁻² h⁻¹). By contrast, mean emission rate in the flooded fallow season was higher in 2015 (8.7 ± 0.6 mg CH₄ m⁻² h⁻¹ vs. 5.3 ± 0.4 mg CH₄ m⁻² h⁻¹) because of the larger peak in October (16.8 ± 3.0 vs. 15.7 ± 2.0 mg m⁻² ha⁻¹) and the more progressive decline thereafter.

The overall cumulative annual N₂O emissions were negative, -0.33 ± 0.02 kg N₂O ha⁻¹, resulting in monthly emission rates that were negligible or negative (ranging from -0.003 mg N₂O m⁻² h⁻¹ to 0.028 mg N₂O m⁻² h⁻¹ (Fig. S1).

The resulting global warming potential (GWP) of rice fields in the Ebre Delta, which integrates both CH₄ and N₂O emissions, was 6466.7 ± 147.9 kg CO₂-eq ha⁻¹. Only CH₄ emission contributed to GWP.

Drivers of seasonal CH₄ emissions: growing and flooded fallow seasons

The information-theoretic framework analyses to provide predictive models of the effects of the agronomic and soil-treat variables on CH₄ emissions (Table 2) were applied for the growing and flooded fallow seasons.

We selected 39 models for the growing season to predict CH₄ emissions (Table 2). The good accuracy of the averaged model (Pearson's $r = 0.82$, Fig. 4) confirms the adequacy of the analyses to determine the best predictors of CH₄ emissions.

Soil sulphate content was the major driver of CH₄ emissions during the growing season displaying a selection probability (SP) close to 1 (Fig. 5a). It was followed, in order of importance, by soil-related traits, i.e., dry bulk density and clay content with similar prediction capacity (SP = 0.685, 0.506, respectively), and by the agronomic traits panicle density, rate of N fertilization and preceding grain yield (SP = 0.39, 0.22, 0.15, respectively). Marginal factors were the sand content, soil organic matter, current grain yield, soil nitrogen, rate of sulphate-fertilization and plat density (SP < 0.09). Soil sulphate and panicle density ($\beta = -0.023$ and -0.179 , respectively) negatively affected the growing season CH₄ emissions (Fig. 5a, d) as opposed to the stimulatory effect of the N fertilization rate ($\beta =$

Table 1 Soil and agronomic traits of the 24 monitored rice fields in Ebre Delta. ND: data no available. Note: the exact date of straw addition in 2015 was not available

Year	#	Rice field	Bulk density (kg m ⁻³)	Soil organic matter (%)	Clay (%)	Sand (%)	Soil sulphates (mg kg ⁻¹)	Soil Nitrogen (%)	Grain yield_preceding growing season (kg rice ha ⁻¹)	Grain yield_current growing season (kg rice ha ⁻¹)	Plant density (Plants m ⁻²)	Panicle density (Panicles m ⁻²)	Nitrogen fertilization (kg N ha ⁻¹)	Sulphate fertilization (kg SO ₄ ²⁻ ha ⁻¹)	Time of straw addition	
2015	P01	969	3.42	24.1	9.6	2424	0.222	6566	6840	228.0	330.4	167.0	0	0	November	
	P02	903	3.74	14.3	58.3	1928	0.223	6258	6019	206.0	236.8	159.3	76.6	0	November	
	P03	688	4.94	34.6	6.3	5329	0.282	7489	8310	114.8	359.2	183.5	0	0	October	
	P04	869	3.45	23.1	9.3	4417	0.202	8208	8550	249.2	338.0	183.3	21.9	0	October	
	P05	812	3.38	29.3	13.6	3688	0.207	8311	8707	249.2	432.0	197.0	98.5	0	October	
	P06	884	3.38	29.6	7.6	1983	0.207	9302	7456	145.2	366.4	167.6	40.2	0	October	
	P07	1014	4.1	28.5	28.0	1357	0.252	7866	9302	247.6	329.2	152.3	0	0	November	
	P08	1016	2.95	14.0	60.8	1366	0.171	7695	7934	210.0	357.2	170.0	0	0	October	
	P09	878	4.12	36.7	4.5	3724	0.251	8139	8721	212.8	420.8	173.3	0	0	October	
	P10	879	3.05	23.4	5.5	1669	0.190	7421	8174	204.4	380.4	180.1	0	0	October	
	P11	975	2.21	4.9	84.6	1266	0.109	ND	6313	ND	ND	153.8	86.6	0	December	
	P12	896	4.17	25.1	16.8	4394	0.243	7524	7182	73.2	257.2	175.0	0	0	October	
	P13	809	4.57	35.3	6.2	4433	0.265	9644	8652	203.2	470.8	190.0	0	0	October	
	P14	1104	1.89	21.3	48.2	2244	0.110	6498	7524	175.6	383.6	170.0	0	0	November	
	P15	1013	3.54	9.8	75.6	4395	0.182	8208	6703	165.2	278.4	164.2	109.4	0	December	
2016	P01	1041	2.73	30.6	7.0	2141	0.176	6019	9149	ND	ND	170	0	0	26/10/2016	
	P02	806	7.7	20	9.8	8565	0.359	5814	5850	113.5	251.5	195	25	0	27/10/2016	
	P03	879	3.5	11.3	61.3	3128	0.195	6840	6156	169.0	288.5	170.7	0	0	18/10/2016	
	P04	906	2.76	13.7	61.3	2721	0.157	6840	7095	164.5	290.5	180.0	8.0	0	21/10/2016	
	P05	1066	4.56	23.4	10.1	6439	0.275	ND	ND	ND	ND	ND	ND	ND	ND	
	P06	ND	ND	ND	ND	ND	ND	9878	11643	ND	ND	ND	150.0	ND	0	20/10/2016
	P07	900	3.16	22.6	17.5	2982	0.185	7011	7353	132.0	334.5	180.0	46.0	0	13/10/2016	
	P08	ND	4.94	36.2	6.4	5959	0.229	8550	8721	116.0	298.5	180.0	46.0	0	11/10/2016	
	P09	1011	2.78	13.7	50.5	4768	0.149	7319	9832	202.5	358.0	180.0	46.0	0	24/10/2016	

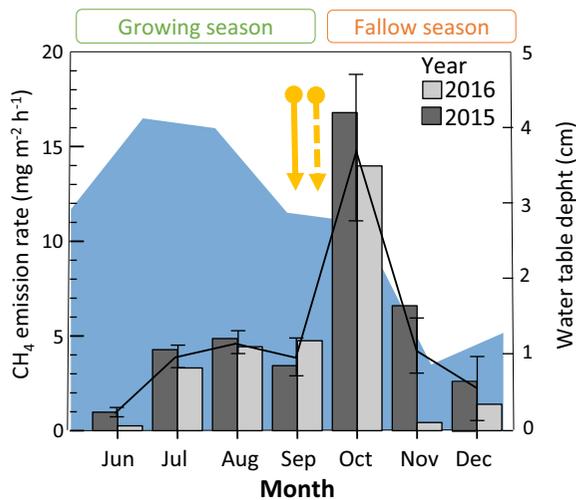


Fig. 3 Mean monthly CH₄ emissions rates in rice fields in Ebre Delta over the growing (June to September) and flooded fallow (October to December) seasons in years 2015 and 2016. Columns indicate monthly mean emissions within each year and line represent the two-year monthly emission rates. Blue shaded area represents water level (monthly means across rice fields and years). Solid arrow represent harvest (September) and dashed arrow straw incorporation (in early October). Fields are sown from late April to early May (not represented in the Figure)

0.610). Soils with higher dry soil bulk density and clay content emitted less CH₄ ($\beta = -0.231, -1.940$, respectively; Fig. 5b, c).

In the fallow season, 35 models were selected to predict CH₄ emissions (Table 2). The good accuracy of the averaged model (Pearson's $r = 0.88$) confirms the adequacy of the analyses to determine the best predictors of CH₄ emissions (Fig. 6). The main explanatory variables ($SP > 0.99$) were the grain yield of the preceding crop season (hereafter, preceding grain yield) and the rate of N fertilization, which both showed positive effects ($\beta = 0.110, 7.7672$, respectively; Fig. 7). They were followed by panicle density and current grain yield ($SP = 0.37$), which negatively influenced emissions ($\beta = -0.368, -0.017$ respectively). Soil nitrogen content, rate of sulphate fertilization, plant density, soil texture, organic matter, and dry bulk density were variables of minor importance.

Drivers of total CH₄ emissions

The good accuracy (Pearson's $r = 0.94$, Fig. S3) of the averaged model confirms the adequacy of the analyses to determine the best predictors of CH₄ emissions. Eighteen plausible models were selected to explain annual

CH₄ emissions for the growing and flooded fallow seasons combined. The most influencing variables coincided with those provided by the flooded fallow season analyses (i.e. preceding grain yield and rate of N fertilization), given the large contribution of CH₄ during this season in relation to the total emissions (Table 2). Therefore, the results on the main drivers will be discussed for each season separately, i.e. growing and flooded fallow season, as the total emissions are explained by those in the fallow.

Discussion

On average, rice fields in the Ebre Delta emitted 262.6 ± 5.9 kg CH₄ ha⁻¹ during the growing and flooded fallow season, of which almost two-thirds occurred in the flooded fallow period. The bi-modal temporal distribution of CH₄ emission rates and the more cumulative emissions in the fallow than in the growing season were consistent across the years. Substantial fallow CH₄ emissions are reported in other Mediterranean monocrop rice systems with winter flooded fallow and rice straw addition, contributing from 10 to 63% to the total emitted annually (Fitzgerald et al. 2000; Knox et al. 2016; Xu and Hosen 2010). The ca. 60% herein reported falls within the upper limit of this range, and it is particularly close to some specific years with similar straw and water management reported within both Fitzgerald's and Knox's works. Lesser fallow CH₄ emissions are reported in winter flooded fields with no straw input (Fitzgerald et al. 2000; Reba et al. 2019). The overall GWP, measured as kg CO₂-eq ha⁻¹, was 6466.7 ± 147.9 kg CO₂-eq ha⁻¹, with CH₄ as the main contributor whereas N₂O emissions were almost negligible. Similar results were found by Wang et al. (2016); Wu et al. (2018) and Wang et al. (2019) who reported N₂O emissions either several orders of magnitude lower than CH₄ emissions or even negative, thus rice fields becoming small sink of N₂O. Given the minor role of N₂O emissions on the GWP, the following discussion is focused on the main drivers of CH₄ emissions.

Main drivers of CH₄ emissions during the growing season

Several soil features were associated with CH₄ emissions during the growing season. Overall, the information theoretic analysis of the candidate set of GLMs

Table 2 Results from the information-theoretic framework analysis to assess the relative contribution of agronomic and environmental factors on annual, growing season and fallow CH₄ emissions in the Ebre Delta rice field area. Model-averaged regression coefficients (β) are parameter coefficients averaged by model weight across all candidate models ($\Delta AICc < 7$) in which the given

parameter occurs; selection probability (SP) indicates the importance of an independent variable, and parameter bias is the difference between the averaged estimates (β) and the full model coefficients. The number (N) of candidate models ($\Delta AICc < 7$) is also shown. Parameters included in the best model, in each case, are shaded in grey colour

Model term	Growing season			Fallow season			Annual		
	<i>N</i> = 39	<i>N</i> = 35	<i>N</i> = 18	<i>N</i> = 39	<i>N</i> = 35	<i>N</i> = 18	<i>N</i> = 39	<i>N</i> = 35	<i>N</i> = 18
	SP	β	Bias	SP	β	Bias	SP	β	Bias
(Intercept)	1.00	417.40	-0.76	1.00	-1745.0	-0.53	1.00	-2042.7	0.05
Clay soil fraction	0.51	-1.94	1.62	0.06	-0.06	20.62	0.03	0.05	-46.72
Dry bulk soil density	0.69	-0.23	-0.96	0.06	0.00	115.69	0.03	0.00	-4.79
Grain yield_current crop season	0.05	-0.0001	56.11	0.37	-0.02	-1.88	0.19	-0.01	-4.03
Grain yield_preceding crop season	0.15	-0.002	-1.26	1.00	0.11	-0.27	1.00	0.12	-0.11
Nitrogen content in the soil	0.07	-16.91	-100.9	0.07	13.81	-112.5	0.04	10.18	16.36
Panicle density	0.39	-0.18	-2.42	0.37	-0.37	-2.53	0.86	-1.41	-0.35
Plant density	0.03	0.00	338.68	0.09	0.03	-17.65	0.12	0.08	-10.59
Rate of nitrogen fertilization	0.22	0.61	-0.23	0.99	7.67	-0.52	1.00	11.74	-0.06
Rate of sulphate fertilization	0.04	-0.01	-23.65	0.09	0.05	-3.43	0.03	0.01	-3.38
Sand soil fraction	0.09	0.04	-4.75	0.07	-0.04	-25.69	0.04	-0.03	-28.13
Soil organic matter	0.07	-0.97	69.23	0.06	0.36	227.63	0.05	-0.10	-143.8
Sulphate content in the soil	0.98	-0.02	-0.34	0.07	0.00	-25.68	0.73	-0.03	0.16

selected 38 plausible models (i.e. $\Delta AICc < 7$) to explain the variability in CH₄ emissions. The best explanatory variables (SP values in Table 2) were soil sulphate, bulk

density, and clay concentration, all show negative relation to CH₄ emissions.

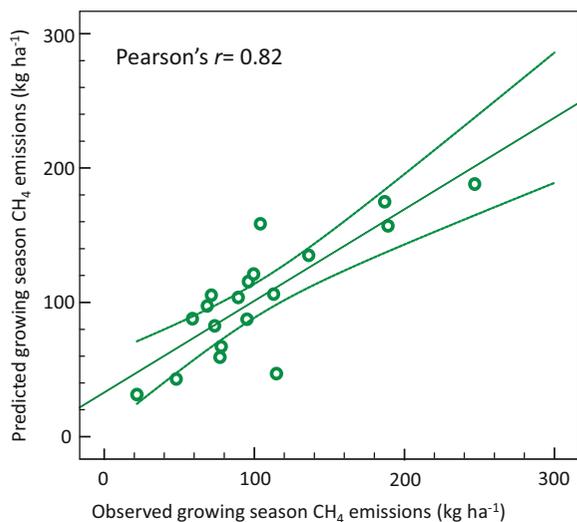


Fig. 4 Relationship between the observed and the predicted growing season CH₄ emissions estimated by the GLM averaged through an information theoretic approach

Soil sulphate content is the major driver inhibition of CH₄ emissions during the growing season. The observed inhibitory effect results from sulphate outcompeting methanogens as terminal electron acceptors (Ponnamperuma 1972). Despite the dominant influence of the soil sulphate content, the effect of sulphate-containing fertilizers on CH₄ emissions was minor. A number of studies demonstrate the mitigation effect of sulphate-based fertilization (Cai et al. 1997; Denier van der Gon et al. 2001; Rath et al. 2002; Minamikawa et al. 2005; Ro et al. 2011). The information- approach used here, in contraposition to the univariate analyses, allows the estimation of the relative contribution of each studied factor and suggests that, despite the demonstrated inhibitory effect of sulphates on CH₄ emissions in this study and elsewhere, the mitigation potential of sulphate-based fertilizers is minor in comparison to that derived from the indigenous sulphate concentration in soils.

Both dry bulk density and soil clay content showed negative effects on CH₄ emissions. Soils rich in clay emitted less CH₄, which is aligned with Brye et al.

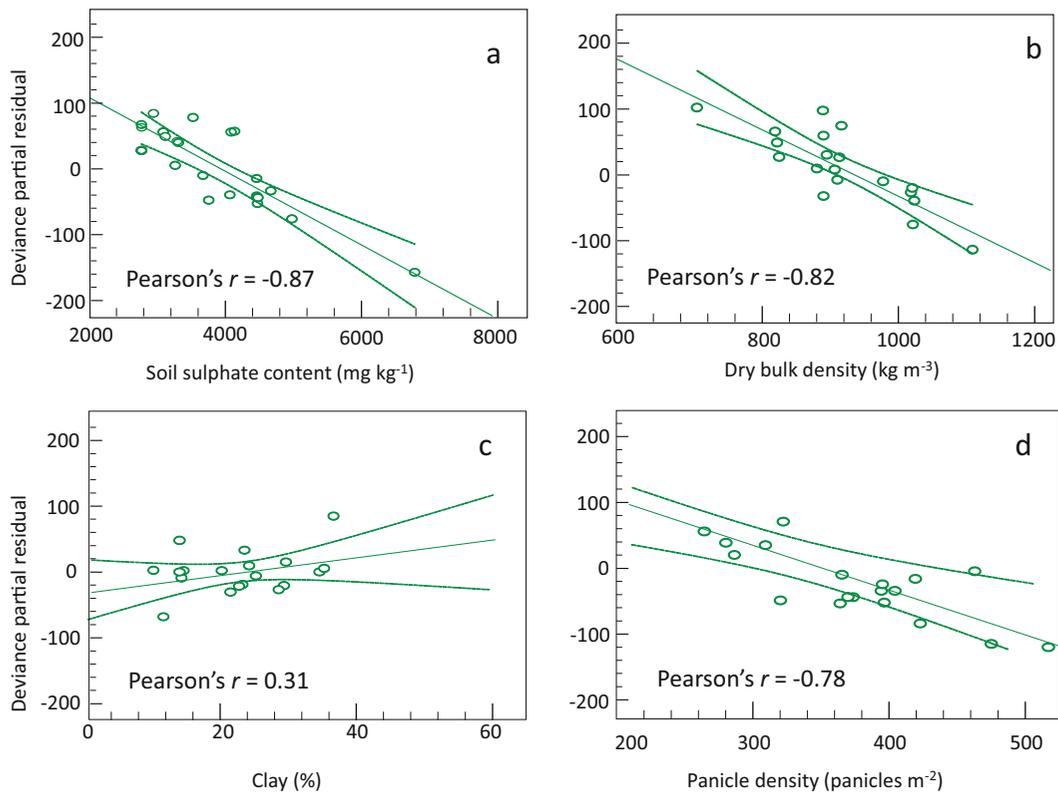


Fig. 5 Partial residual plots for the most influencing variables in growing season CH₄ emission rates. Partial residuals show the effect of a given independent variable on the response after taking into account all other explanatory variables. Solid line shows the

(2013); Zhang et al. (2012); Brye et al. (2016) and Sass et al. (1994). Coarser soil texture favours the diffusivity of CH₄ from the soil to the atmosphere (Brye et al. 2013)

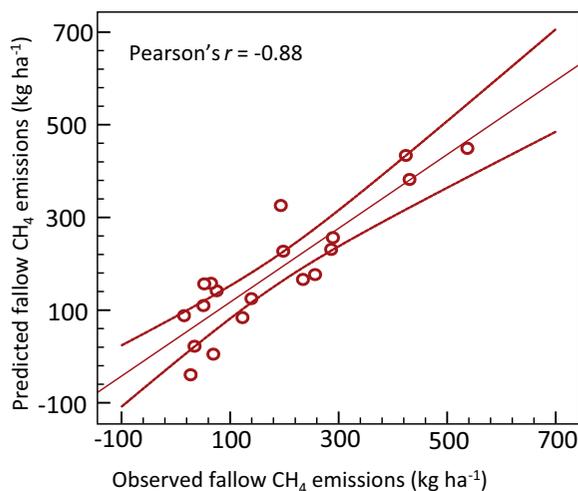


Fig. 6 Relationship between the observed and the predicted fallow CH₄ emissions estimated by the GLM averaged through an information theoretic approach

linear relationship between partial residuals and the given explanatory variable, and dashed lines are the 95% confidence interval. Pearson's correlation coefficient is shown

thus reducing residence time of the molecule in the soil (Sass and Fisher 1997) and so limiting its availability for methanotrophs (Brye et al. 2016; Smartt et al. 2016). In addition, clay particles impede the accessibility of organic matter to microbial decomposers by the formation of microaggregates (Six et al. 2002) and increase the soil ferric iron (Fe³⁺) concentration (Sass et al. 1994), thus facilitating the ability of iron-reducing bacteria to out-compete methanogens (Van Bodegom et al. 2001). Dry bulk density is a pedo-transfer function of several soil properties (Aimrun and Amin 2009; Caldwell et al. 2007) influencing gas and water fluxes, biochemical reactions and processes and, subsequently, CH₄ dynamics. There are different mechanisms through which high bulk density can reduce CH₄ emissions, such as prolonging either the direct or plant-mediated transport of CH₄ from the soil to the atmosphere (Smith et al. 2001), reducing the fraction of large pores and subsequently the availability of organic matter (Ahmad et al. 2009) or reducing the concentration of the dissolved organic carbon (DOC), the precursor of CH₄ (Liu et al. 2011).

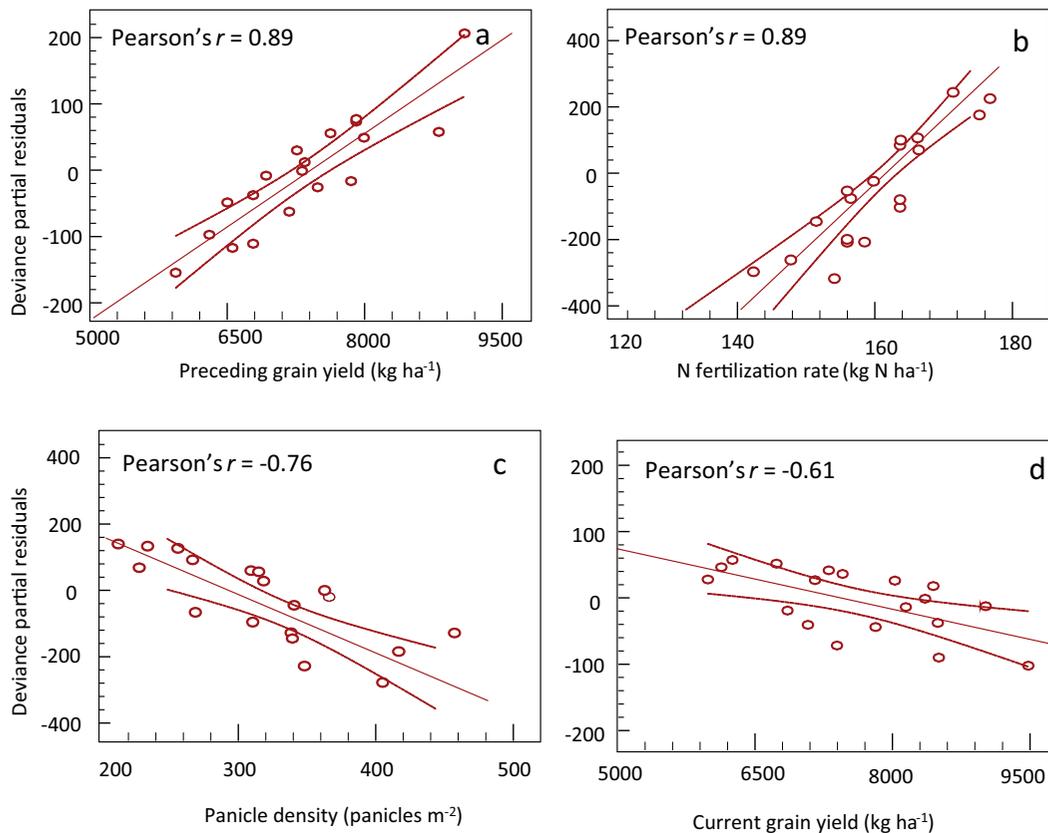


Fig. 7 Partial residual plots for the most influencing variables in fallow CH_4 emission rates. Partial residuals show the effect of a given independent variable on the response after taking into account all other explanatory variables. Solid line shows the linear

relationship between partial residuals and the given explanatory variable, and dashed lines are the 95% confidence interval. Pearson's correlation coefficient is also shown

To a lesser extent, panicle density, rate of N fertilization and grain yield from the preceding growing season (hereafter, preceding grain yield) also affected CH_4 emissions during the growing season. The negative effect of both panicle density and preceding grain yield, combined with the residual effect of current grain yield and plant density suggest that favourable crop growth mitigates CH_4 emissions. A fraction of the produced CH_4 is consumed by methanotrophs associated with the oxidized layers of the rhizosphere (Kruger et al. 2002), which consume 80% of the total CH_4 produced (Hanson and Hanson 1996). In addition, crop productivity is correlated to root biomass and thus to root oxygenated area (reviewed by Yang et al. 2012) while Gutierrez et al. (2014) found a positive correlation between the root-oxygenated area and the number of methanotrophs. According to this, we hypothesize that the observed reduction in CH_4 emissions in the more productive rice fields (Fig. 5d) result from enhanced CH_4 oxidative rates.

Main drivers of CH_4 emissions during the flooded fallow season

The information theoretic analysis of the candidate set of GLMs selected 38 plausible models (i.e. $\Delta\text{AICc} < 7$) to explain the variability in CH_4 emissions during the flooded fallow season. Two variables were clearly selected as the best explanatory variables (SP values in Table 2), namely, preceding grain yield and nitrogen fertilization rate, both showing a positive effect on CH_4 emissions. Preceding grain yield, which was used as a proxy of straw incorporated into the soil in the preceding fallow season (hereafter, preceding straw), was the main driver of CH_4 emissions in the flooded fallow. Instead, straw incorporated in the ongoing season (hereafter, recently added straw) has a minor yet slight negative effect (Table 2). Several studies have described the persistence of the added straw residues (30–50%) in the soil for one year or even longer (Neue and

Scharpenseel 1987; Cai et al. 2018; Yan et al. 2019). Thus, one year after adding the straw into the soil, the intermediate and recalcitrant carbon-derived straw pools may persist as components of the soil organic matter (SOM) matrix (Cai et al. 2018), and the intermediate carbon pool might then account for the largest and most available carbon source for methanogenesis, representing the dominant mechanism of CH₄ production. However, some previous works (Ye et al. 2014; Ye and Horwath 2017) have proposed a contrasting pattern, with more than 50% of CH₄ emissions related to the recently added straw while the remaining emissions were claimed to arise from SOM. In these studies, straw was chopped or powdered into small fragments (< 1 mm) and homogeneously distributed over the soil depth, thus favouring accessibility of the recently incorporated straw easily accessible to methanogens (Tarafdar et al. 2001). The contrasting results with our study can be explained by differences in physical and biochemical availability for the biodegradation of the recently added straw. In the Ebre Delta, resulting from wet tillage combined with straw incorporation, a set of different sizes of straw fragments (see Supplementary Fig. S2), from fine (millimetres) to coarse particles (> 5 cm long), are deposited from the soil surface down to approximately 10 cm deep, and so are partially exposed to air due to O₂ penetration by diffusion. Thus, the shallow allocation of the straw might limit its degradation by methanogenic archaea, which are strictly anaerobic, either because it is not fully integrated into the soil (air-exposed) or because anaerobic conditions are not met. The dry soil operations (soil labouing and basal fertilization operations) during the pre-sowing season (February to April) allow the full integration of straw residues into SOM matrix. In spite of this, CH₄ emissions remained low over the early growing season, i.e., until July (Fig. 2) when the soil reached critical reductive conditions (redox between -150 mV to -160 mV; see Martínez-Eixarch et al. 2018) for the initiation of CH₄ production (Wang et al. 1993), and the crop becomes more physiologically active (Martínez-Eixarch et al. 2018). This delay in CH₄ emissions up to several weeks after flooding was also reported by Brye et al. (2013).

To summarize, this pattern suggests: firstly, the predominance of growing season CH₄ emissions derived from the organic matter produced from the rice plants and less from the straw-derived SOM route (Ström et al. 2003; Yuan et al. 2012) and, secondly; that straw is not

decomposed and evolved to CH₄ until the subsequent fallow season.

Nitrogen fertilization, along with the rate of preceding added straw, is the major driver of the fallow CH₄ emissions. Despite the narrow rate of N fertilization (150 kg N ha⁻¹ to 200 kg N ha⁻¹), our study provides evidence of the persist and stimulatory effect of N fertilization on the flooded fallow CH₄ emissions. Such a persistent effect beyond the growing season have been demonstrated elsewhere (Kim et al. 2019; Datta et al. 2013) but it is in contrast with Pittelkow et al. (2013). The effect of N fertilization on CH₄ emissions depends on the trade-off between its influence on CH₄ production and oxidation (Dan et al. 2001; Xu et al. 2004) resulting from complex interactions with environmental, soil microbiota, and agronomic site-specific conditions (Banger et al. 2012). The stimulatory effect of N rates on both total and seasonal (growing and flooded fallow) CH₄ emissions (Table 2) is in agreement with the overall trend found in two different meta-analyses conducted by Banger et al. (2012) and by Liu and Greaver (2009).

Agricultural-based options to mitigate CH₄ emissions in rice fields

Understanding the key drivers of CH₄ emissions provides valuable knowledge to either design more effective mitigation agronomic strategies or adapt the existing ones. The assessment of the relative contribution of agronomic factors on CH₄ emissions allows for inferring their relative mitigation capacity thereby pointing out the factors that should be prioritized when designing agronomic strategies to reduce CH₄ emissions. Two main implications in regard of CH₄ mitigation options in the Mediterranean rice agroecosystem are derived from our study. The first one is that in Mediterranean rice growing areas such as Spain, France, and California, where winter flooding and straw return are practiced, measures to mitigate CH₄ emissions in the flooded fallow season have a larger overall impact than those designed to reduce emissions in the growing season. The rice straw management and winter flooding have multiple implications on GHG emissions, crop production and ecologic functions in the rice agroecosystems. This poses the interesting challenge to find compatible solutions for mitigating CH₄ emissions while preserving the agronomic (Van Groenigen et al. 2003; Pathak et al. 2006; Wang et al. 2015; Martínez-Eixarch et al. 2016) and environmental benefits that

straw addition and winter flooding offer, such as carbon sequestration (Chaudhary et al. 2017) and the preservation of biodiversity (Elphick and Oring 2003; Pernollet et al. 2015). For example, alternating wetting and drying cycles (Linquist et al. 2015; Runkle et al. 2018) or implementing a single mid-season drainage (Wang et al. 2020) during the fallow season to reduce CH₄ emissions while maintaining the favourable aquatic ambient for aquatic birds could be considered. In addition, limiting straw incorporation to cooler periods (Martínez-Eixarch et al. 2018) when methanogenic activity is reduced has the potential to substantially mitigate emissions and maintains the abovementioned environmental benefits.

The second agronomic implication is that enhanced rice yield may reduce CH₄ emissions. Our study demonstrates that rice production and rate of nitrogen fertilization are the main agronomic controlling factors of CH₄ emissions in both growing and flooded fallow season. Rice production shows an ambivalent effect on CH₄ emissions with an inhibitory effect during the growing season but stimulatory in the flooded fallow season, whereas N rate stimulates both growing and fallow CH₄ emissions. The effect of rice production on CH₄ emissions is driven by two contrasting processes: more methanotrophic activity proportionally to root development and increased organic matter readily available for methanogenic communities in the soil eventually evolving to CH₄ during the flooded fallow season. It is then derived that reduction of CH₄ emissions via grain yield optimization could be driven by boosting root development simultaneously to favour yield partitioning to grain, i.e., increasing harvest index. Jiang et al. (2019) found that increasing harvest index (i.e., ratio of harvested grain to total shoot dry matter) could reduce CH₄ emissions by ca. 4% in permanently flooded cultivated rice.

In the growing season, nitrogen stimulates the production of root exudates fuelling CH₄ production whereas in the fallow season, the fraction of the remaining N fertilizer available in the soil can inhibit methanotrophic activity (Bodelier and Laanbroek 2004). The narrow and bidirectional associations found in this study between yield, nitrogen rates and CH₄ emissions point out the improvement of nitrogen use efficiency (NUE) as a promising strategy to mitigate CH₄ emissions by increasing rice productivity. NUE is determined by two components, the N uptake efficiency of the crop and the efficiency of N assimilation and remobilization of the plant

to produce grain. The enhancement of these two components results in limiting soil N availability (Che et al. 2015; Guo et al. 2017) and favouring harvest index (Huang et al. 2018), which, according to our study, are two necessary features to reduce CH₄ emissions. There are few studies testing the hypothesis that enhanced NUE can reduce CH₄ emissions in permanently flooded rice fields (Liu et al. 2015; Adviento-Borbe and Linquist 2016; Zhu et al. 2016) and yet they provide contrasting results because either the tested practices are different (i.e., plant density, N placement, N rates) or the rice cropping systems also differ. Taking all this together, we think that NUE enhancement is an unexplored promising win-win strategy to both mitigate CH₄ emissions and improve rice production that should be further explored on a site-specific crop context to address the high complexity of the rice agroecosystems.

Before concluding, some considerations on the sampling strategy. This is a multi-site study conceived to assess both the temporal pattern of GHG emissions and their main drivers so that covering the spatial environmental and agronomic variability of the whole area of rice cultivation was crucial for the pursue of this objective. However, multi-site sampling was at the expense of intense sampling rate, which was conducted monthly, thus less frequently than the recommended weekly or biweekly samplings (Minamikawa et al. 2012). Nevertheless, the consistent temporal pattern, the narrow range of variation of the emission rates provides confidence in the reliability of the results and the derived conclusions.

Conclusions

This study argued that the main drivers of cumulative CH₄ emissions are different in the growing than in the fallow seasons. Edaphic factors exert more influence on CH₄ emissions during the growing season whereas agronomic factors have a higher impact in the fallow season. In the growing season, soil sulphate content, followed by bulk density and clay content are the main emission drivers. The effect of the inherent soil sulphate in lowering CH₄ emissions surpass that obtained by sulphate-based fertilizers, which resulted in only minor decreases. Soils with higher bulk density and clay content emit less CH₄. In the fallow season, the rate of both straw incorporated in the preceding crop (rather than that added in the ongoing season) and nitrogen fertilizer

are the major controlling factors. The stimulating effect of Nitrogen fertilization on CH₄ emissions is larger in the fallow season in relation to the growing season. Of all the agronomic factors analysed in this study, rice productivity and nitrogen fertilization rates are those with the most influence on CH₄ emissions, so that effective mitigation strategies should include them.

Supplementary Information The online version contains supplementary material available at <https://doi.org/10.1007/s11104-020-04809-5>.

Acknowledgements This work was supported by the European Union through the Life + Environmental Policy and Governance, project entitled Pilot project for adaptation and mitigation measures for Climate Change in Ebre Delta (Life13 ENV/ES/001182); and by the Ministry of Economy and Competitiveness through the National Institute for Agricultural and Food Research and Technology (INIA) Spanish, project entitled Mitigation and adaptation to climate change in coastal rice fields (RTA2014-00058-C03-03). The support of the CERCA Programme / Generalitat de Catalunya is also acknowledged. The funders had no role in study design, data collection and analysis, decision to publish, or preparation of the manuscript. We also acknowledge the logistic support provided by the Kellogg's Origins Program and the farmers within this program for authorizing us to sample in their fields.

The authors would like to thank Lluís Jornet, Pep Cabanes and David Mateu (IRTA-Marine and continental waters), Andrea Bertomeu (IRTA- Extensive crops) and, Bea Grau, Inma Funes and Eulàlia Serra (IRTA- Fruticulture) for their technical support and field assistance.

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