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

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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.

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

Abbreviations
CH4 methane
DOC dissolved organic carbon
GHG greenhouse gas
GLMz generalized linear models
GWP global warming potential
N nitrogen
NUE nitrogen use efficiency
N2O nitrous oxide
SO4²− 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 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 (Linquist 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 (Linquist 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, Northeaster 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 (Martinez-
Eixarch et al. 2018), we identified the key dynamic variables modulating CH₄ fluxes, including environmental (soil physico-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.
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., Buckinghamsire, UK) and sent to laboratory. Methane (CH₄) and nitrous oxide (N₂O) concentration was determined using a THERMO TRACE 2000 (Thermo Finnigan Scientific, USA) gas chromatograph equipped with a flame ionization detector (GC-FID). Analysis of N₂O 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 CH₄ and N₂O standard in nitrogen provided by Carburos Metalicos S.A. (Spain).

The emission rates of the GHG, i.e. CH₄ and N₂O, 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$_2$O, 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 $\leq 5$, in order to avoid multicollinearity effects in regression models (Maggini et al. 2006). The second order Akaike information criterion (AICc), rescaled to obtain $\Delta$AICc values ($\Delta$AICc = AICc - 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 ± 1.1 °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 ± 0.8 °C) than during the flooded fallow (October to December: 13.9 ± 1.5 °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.
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\(^{-1}\)) where 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 ± 0.6 kg SO\(_4\)\(^{2-}\) ha\(^{-1}\)) than in 2015 (72.2 ± 9.4 kg SO\(_4\)\(^{2-}\) ha\(^{-1}\)).

Total GHG emissions and global warming potential

In total, rice fields in Ebre Delta emitted 262.6 ± 5.9 kg CH\(_4\) m\(^{-2}\) ha\(^{-1}\) (two-year mean) of which 36.9% was emitted during the growing season (96.6 ± 2.8 kg CH\(_4\) ha\(^{-1}\)) and 63.1% during the flooded fallow season (163.9 ± 9.8 kg CH\(_4\) ha\(^{-1}\)) mainly in October. October alone accounted for 45.4% of the annual emissions.

The monthly CH\(_4\) emissions rates followed a bimodal pattern (Fig. 3) with the first peak occurring in August (4.7 ± 0.3 mg CH\(_4\) m\(^{-2}\) h\(^{-1}\)) and the second, and the highest, in October (15.7 ± 2.0 mg CH\(_4\) 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 CH\(_4\) m\(^{-2}\) h\(^{-1}\)). By contrast, mean emission rate in the flooded fallow season was higher in 2015 (8.7 ± 0.6 mg CH\(_4\) m\(^{-2}\) h\(^{-1}\)) vs. 5.3 ± 0.4 mg CH\(_4\) 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 N\(_2\)O emissions were negative, −0.33 ± 0.02 kg N\(_2\)O ha\(^{-1}\), resulting in monthly emission rates that were negligible or negative (ranging from −0.003 mg N\(_2\)O m\(^{-2}\) h\(^{-1}\) to 0.028 mg N\(_2\)O m\(^{-2}\) h\(^{-1}\)) (Fig. S1).

The resulting global warming potential (GWP) of rice fields in the Ebre Delta, which integrates both CH\(_4\) and N\(_2\)O emissions, was 6466.7 ± 147.9 kg CO\(_2\)-eq ha\(^{-1}\). Only CH\(_4\) emission contributed to GWP.

Drivers of seasonal CH\(_4\) 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 CH\(_4\) emissions (Table 2) were applied for the growing and flooded fallow seasons.

We selected 39 models for the growing season to predict CH\(_4\) 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 CH\(_4\) emissions.

Soil sulphate content was the major driver of CH\(_4\) 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 CH\(_4\) emissions (Fig. 5a, d) as opposed to the stimulatory effect of the N fertilization rate (\(\beta = \))
| 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                          | November                  |
|      | P02        | 903                           | 3.74                    | 14.3     | 58.3     | 1928                          | 0.223            | 6258                            | 6019                            | 206.0                       | 236.8                         | 159.3                       | 76.6                       | November                  |
|      | P03        | 688                           | 4.94                    | 34.6     | 6.3      | 5329                          | 0.282            | 7489                            | 8310                            | 114.8                       | 359.2                         | 183.5                       | 0                          | October                   |
|      | P04        | 869                           | 3.45                    | 23.1     | 9.3      | 4417                          | 0.202            | 8208                            | 8550                            | 249.2                       | 338.0                         | 183.3                       | 21.9                       | October                   |
|      | P05        | 812                           | 3.38                    | 29.3     | 13.6     | 3688                          | 0.207            | 8311                            | 8707                            | 249.2                       | 432.0                         | 197.0                       | 98.5                       | October                   |
|      | P06        | 884                           | 3.38                    | 29.6     | 7.6      | 1983                          | 0.207            | 9302                            | 7456                            | 145.2                       | 366.4                         | 167.6                       | 40.2                       | October                   |
|      | P07        | 1014                          | 4.1                     | 28.5     | 28.0     | 1357                          | 0.252            | 7866                            | 9302                            | 247.6                       | 329.2                         | 152.3                       | 0                          | November                  |
|      | P08        | 1016                          | 2.95                    | 14.0     | 60.8     | 1366                          | 0.171            | 7695                            | 7934                            | 210.0                       | 357.2                         | 170.0                       | 0                          | October                   |
|      | P09        | 878                           | 4.12                    | 36.7     | 4.5      | 3724                          | 0.251            | 8139                            | 8721                            | 212.8                       | 420.8                         | 173.3                       | 0                          | October                   |
|      | P10        | 879                           | 3.05                    | 23.4     | 5.5      | 1669                          | 0.190            | 7421                            | 8174                            | 204.4                       | 380.4                         | 180.1                       | 0                          | October                   |
|      | P11        | 975                           | 2.22                    | 4.9      | 84.6     | 1266                          | 0.109            | ND                              | 6313                            | ND                          | ND                            | 153.8                       | 86.6                       | December                  |
|      | P12        | 896                           | 4.17                    | 25.1     | 16.8     | 4394                          | 0.243            | 7524                            | 7182                            | 73.2                        | 257.2                         | 175.0                       | 0                          | October                   |
|      | P13        | 809                           | 4.57                    | 35.3     | 6.2      | 4433                          | 0.265            | 9644                            | 8652                            | 203.2                       | 470.8                         | 190.0                       | 0                          | October                   |
|      | P14        | 1104                          | 1.89                    | 21.3     | 48.2     | 2244                          | 0.110            | 6498                            | 7524                            | 175.6                       | 383.6                         | 170.0                       | 0                          | November                  |
|      | P15        | 1013                          | 3.54                    | 9.8      | 75.6     | 4395                          | 0.182            | 8208                            | 6703                            | 165.2                       | 278.4                         | 164.2                       | 109.4                      | December                  |
| 2016 | P01        | 1041                          | 2.73                    | 30.6     | 7.0      | 2141                          | 0.176            | 6019                            | 9149                            | ND                          | ND                            | 170                         | 0                          | 26/10/2016                |
|      | P02        | 806                           | 7.7                     | 20       | 9.8      | 8565                          | 0.359            | 5814                            | 5850                            | 113.5                       | 251.5                         | 195                         | 25                         | 27/10/2016                |
|      | P03        | 879                           | 3.5                     | 11.3     | 61.3     | 3128                          | 0.195            | 6840                            | 6156                            | 169.0                       | 288.5                         | 170.7                       | 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                        | 21/10/2016                |
|      | P05        | 1066                          | 4.56                    | 23.4     | 10.1     | 6439                          | 0.275            | ND                              | ND                              | ND                          | ND                            | ND                          | ND                         | ND                        |
|      | P06        | ND                            | ND                      | ND       | ND       | ND                            | ND               | 9878                            | 11643                           | ND                          | ND                            | 150.0                       | ND                         | 20/10/2016                |
|      | P07        | 900                           | 3.16                    | 22.6     | 17.5     | 2982                          | 0.185            | 7011                            | 7353                            | 132.0                       | 334.5                         | 180.0                       | 46.0                       | 13/10/2016                |
|      | P08        | ND                            | 4.94                    | 36.2     | 6.4      | 5959                          | 0.229            | 8550                            | 8721                            | 116.0                       | 298.5                         | 180.0                       | 46.0                       | 11/10/2016                |
|      | P09        | 1011                          | 2.78                    | 13.7     | 50.5     | 4768                          | 0.149            | 7319                            | 9832                            | 202.5                       | 358.0                         | 180.0                       | 46.0                       | 24/10/2016                |
0.610). Soils with higher dry soil bulk density and clay content emitted less CH$_4$ ($\beta = -0.231$, -1.940, respectively; Fig. 5b, c).

In the fallow season, 35 models were selected to predict CH$_4$ emissions (Table 2). The good accuracy of the averaged model ($r = 0.88$) confirms the adequacy of the analyses to determine the best predictors of CH$_4$ 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 CH$_4$ emissions

The good accuracy ($r = 0.94$, Fig. S3) of the averaged model confirms the adequacy of the analyses to determine the best predictors of CH$_4$ emissions. Eighteen plausible models were selected to explain annual CH$_4$ 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 CH$_4$ 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 CH$_4$ 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 CH$_4$ emission rates and the more cumulative emissions in the fallow than in the growing season were consistent across the years. Substantial fallow CH$_4$ 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 CH$_4$ 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 CO$_2$-eq ha$^{-1}$, was 6466.7 ± 147.9 kg CO$_2$-eq ha$^{-1}$, with CH$_4$ as the main contributor whereas N$_2$O emissions were almost negligible. Similar results were found by Wang et al. (2016); Wu et al. (2018) and Wang et al. (2019) who reported N$_2$O emissions either several orders of magnitude lower than CH$_4$ emissions or even negative, thus rice fields becoming small sink of N$_2$O. Given the minor role of N$_2$O emissions on the GWP, the following discussion is focused on the main drivers of CH$_4$ emissions.

Main drivers of CH$_4$ emissions during the growing season

Several soil features were associated with CH$_4$ emissions during the growing season. Overall, the information theoretic analysis of the candidate set of GLMs
selected 38 plausible models (i.e. \( \Delta \text{AIC}_c < 7 \)) to explain the variability in CH\(_4\) emissions. The best explanatory variables (SP values in Table 2) were soil sulphate, bulk density, and clay concentration, all show negative relation to CH\(_4\) emissions.

Soil sulphate content is the major driver inhibition of CH\(_4\) 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 CH\(_4\) 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 CH\(_4\) 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 CH\(_4\) emissions. Soils rich in clay emitted less CH\(_4\), which is aligned with Brye et al.
Coarser soil texture favours the diffusivity of CH$_4$ from the soil to the atmosphere (Brye et al. 2013) 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 (Fe$^{3+}$) 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, CH$_4$ dynamics. There are different mechanisms through which high bulk density can reduce CH$_4$ emissions, such as prolonging either the direct or plant-mediated transport of CH$_4$ 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 CH$_4$ (Liu et al. 2011).
To a lesser extent, panicle density, rate of N fertilization
and grain yield from the preceding growing season (here-
after, preceding grain yield) also affected CH4 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 CH4 emis-
sions. A fraction of the produced CH4 is consumed by
methanotrophs associated with the oxidized layers of the
rhizosphere (Kruger et al. 2002), which consume 80% of
the total CH4 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 corre-
lation between the root-oxygenated area and the number of
methanotrophs. According to this, we hypothesize that the
observed reduction in CH4 emissions in the more produc-
tive rice fields (Fig. 5d) result from enhanced CH4 oxida-
tive rates.

Main drivers of CH4 emissions during the flooded
fallow season

The information theoretic analysis of the candidate set
of GLMs selected 38 plausible models (i.e. ΔAICc < 7)
to explain the variability in CH4 emissions during the
flooded fallow season. Two variables were clearly se-
lected as the best explanatory variables (SP values in
Table 2), namely, preceding grain yield and nitrogen
fertilization rate, both showing a positive effect on CH4
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 CH4 emissions in the flooded fallow. Instead,
straw incorporated in the ongoing season (hereafter, re-
cently 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

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**Fig. 7** Partial residual plots for the most influencing variables in
fallow CH4 emission rates. Partial residuals show the effect of a
given independent variable on the response after taking into ac-
count 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.
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 (Taraafdar et al. 2001). The contrasting results with our study can be explained by differences in physical and biochemical availability for the biodeterioration 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 (millimetre) 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 labouring 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 Martinez-Eixarch et al. 2018) for the initiation of CH₄ production (Wang et al. 1993), and the crop becomes more physiologically active (Martinez-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 persist 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; Martinez-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 CH$_4$ 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 aforementioned environmental benefits.

The second agronomic implication is that enhanced rice yield may reduce CH$_4$ emissions. Our study demonstrates that rice production and rate of nitrogen fertilization are the main agronomic controlling factors of CH$_4$ emissions in both growing and flooded fallow season. Rice production shows an ambivalent effect on CH$_4$ emissions with an inhibitory effect during the growing season but stimulatory in the flooded fallow season, whereas N rate stimulates both growing and fallow CH$_4$ emissions. The effect of rice production on CH$_4$ 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 CH$_4$ during the flooded fallow season. It is then derived that reduction of CH$_4$ 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 CH$_4$ emissions by ca. 4% in permanently flooded cultivated rice.

In the growing season, nitrogen stimulates the production of root exudates fuelling CH$_4$ 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 CH$_4$ emissions point out the improvement of nitrogen use efficiency (NUE) as a promising strategy to mitigate CH$_4$ 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 CH$_4$ emissions. There are few studies testing the hypothesis that enhanced NUE can reduce CH$_4$ 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 CH$_4$ 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 CH$_4$ emissions are different in the growing than in the fallow seasons. Edaphic factors exert more influence on CH$_4$ 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 CH$_4$ emissions surpass that obtained by sulphate-based fertilizers, which resulted in only minor decreases. Soils with higher bulk density and clay content emit less CH$_4$. 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 CH$_4$ 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 CH$_4$ emissions, so that effective mitigation strategies should include them.

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