

1        **i.        Title**

2        A historical perspective on soil organic carbon in Mediterranean cropland (Spain, 1900-  
3        2008)

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25

26 **Abstract**

27 Soil organic carbon (SOC) management is key for soil fertility and for mitigation and  
28 adaptation to climate change, particularly in desertification-prone areas such as  
29 Mediterranean croplands. Industrialization and global change processes affect SOC  
30 dynamics in multiple, often opposing, ways. Here we present a detailed SOC balance in  
31 Spanish cropland from 1900 to 2008, as a model of a Mediterranean, industrialized  
32 agriculture. Net Primary Productivity (NPP) and soil C inputs were estimated based on yield  
33 and management data. Changes in SOC stocks were modeled using HSOC, a simple model  
34 with one inert and two active C pools, which combines RothC model parameters with  
35 humification coefficients. Crop yields increased by 227% during the studied period, but total  
36 C exported from the agroecosystem only increased by 73%, total NPP by 30%, and soil C  
37 inputs by 20%. There was a continued decline in SOC during the 20<sup>th</sup> century, and cropland  
38 SOC levels in 2008 were 17% below their 1933 peak. SOC trends were driven by historical  
39 changes in land uses, management practices and climate. Cropland expansion was the main  
40 driver of SOC loss until mid-20<sup>th</sup> century, followed by the decline in soil C inputs during the  
41 fast agricultural industrialization starting in the 1950s, which reduced harvest indices and  
42 weed biomass production, particularly in woody cropping systems. C inputs started  
43 recovering in the 1980s, mainly through increasing crop residue return. The upward trend in  
44 SOC mineralization rates was an increasingly important driver of SOC losses, triggered by  
45 irrigation expansion, soil cover loss and climate change-driven temperature rise.

46 **Keywords**

47 Climate change; Land use change; NPP; Irrigation; Roots; Woody crops

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50        **1. Introduction**

51        Carbon (C) dynamics in cropland soils is a hot topic in Mediterranean agriculture in the  
52        context of climate change. On the one hand, soil organic carbon (SOC) is highly sensitive to  
53        management practices in these environments (Aguilera et al., 2013), and soil C sequestration  
54        can offset a large share of the full life cycle greenhouse gas (GHG) emissions of crop  
55        production (e.g. Bosco et al., 2013, Aguilera et al., 2015a,b, Guardia et al., 2016). On the  
56        other hand, Mediterranean cropping systems are characterized by low SOC levels, resulting  
57        particularly vulnerable to climate change (Zalidis et al., 2002, Iglesias et al., 2011). This last  
58        condition stresses the need for increasing SOC levels to improve soil quality and fertility  
59        (Diacono and Montemurro, 2010).

60        Global cropland area expanded about 4-fold since 1700, up to ca. 20% of the vegetated area  
61        (Pongratz et al., 2008). In parallel, agroecosystems intensified to meet increases in population  
62        density (Ellis et al., 2013), culminating in a radical socio-metabolic transformation, with the  
63        transition from traditional organic, solar-based systems to industrial, fossil fuel based  
64        systems (Krausmann et al., 2008). The former usually operated at the local scale, relying on  
65        solar fluxes and internal biomass recycling as sources of energy and fertility. The latter are  
66        intensified through imports of fossil fuel-based industrial inputs and international or  
67        interregional trade to optimize the conditions for commodity production in a context of a  
68        global market economy (Guzmán and González de Molina, 2017, Gingrich et al., 2017).  
69        These structural characteristics shape the C cycle through effects on the type and quantity of  
70        soil C inputs and on the biotic and abiotic factors controlling C losses. Soil C inputs can  
71        potentially increase with industrialization due to a higher overall biomass production and a  
72        lower use of crop residues for animal feeding (e.g. Wiesmeier et al., 2014). Modern crops,

73 however, usually have higher harvest indices, which reduces the production of residue  
74 relative to the main product (Johnson et al., 2006). In addition, weed biomass is more  
75 effectively suppressed in modern cropping systems (Guzmán et al. 2014), and root growth in  
76 relation to aerial biomass is usually reduced (e.g. Chirinda et al., 2012). SOC mineralization  
77 is also affected by the changes in management practices such as tillage, irrigation and  
78 fertilization (Sainju et al., 2013, Shang et al., 2015).

79 Along with historical management changes, SOC dynamics are affected by shifts in  
80 environmental conditions associated to global change, particularly temperature increase,  
81 which would boost litter decay (Gregorich et al., 2017) and SOC mineralization (Davidson  
82 and Janssens, 2006), potentially representing a positive feedback to climate change. On the  
83 other hand, possible reductions in precipitation in Mediterranean areas (Giorgi and Lionello,  
84 2008) could increase water limitation of SOC mineralization.

85 SOC dynamics in modern conventional systems have been often compared to those of  
86 modern organic and/or low-input systems (Gattinger et al., 2012, Aguilera et al., 2013).  
87 However, specific studies on traditional organic cropping systems are very scarce,  
88 particularly at large spatial scales. Most large-scale assessments of SOC dynamics are based  
89 on crop-soil process-based simulations, validated with soil and yield data from databases  
90 such as EUROSTAT or FAOSTAT (e.g. Ciais et al., 2010, Bondeau et al., 2007). Most of  
91 these databases do not provide data before mid-20<sup>th</sup> century, or specific information on many  
92 management practices, a problem that can be overcome in studies covering smaller areas  
93 with better statistical information (e.g. Parton et al., 2015).

94 Spanish agriculture experienced vast technological and structural changes along the 20<sup>th</sup>  
95 century. During the second half of the century, there was a large increase in land and animal

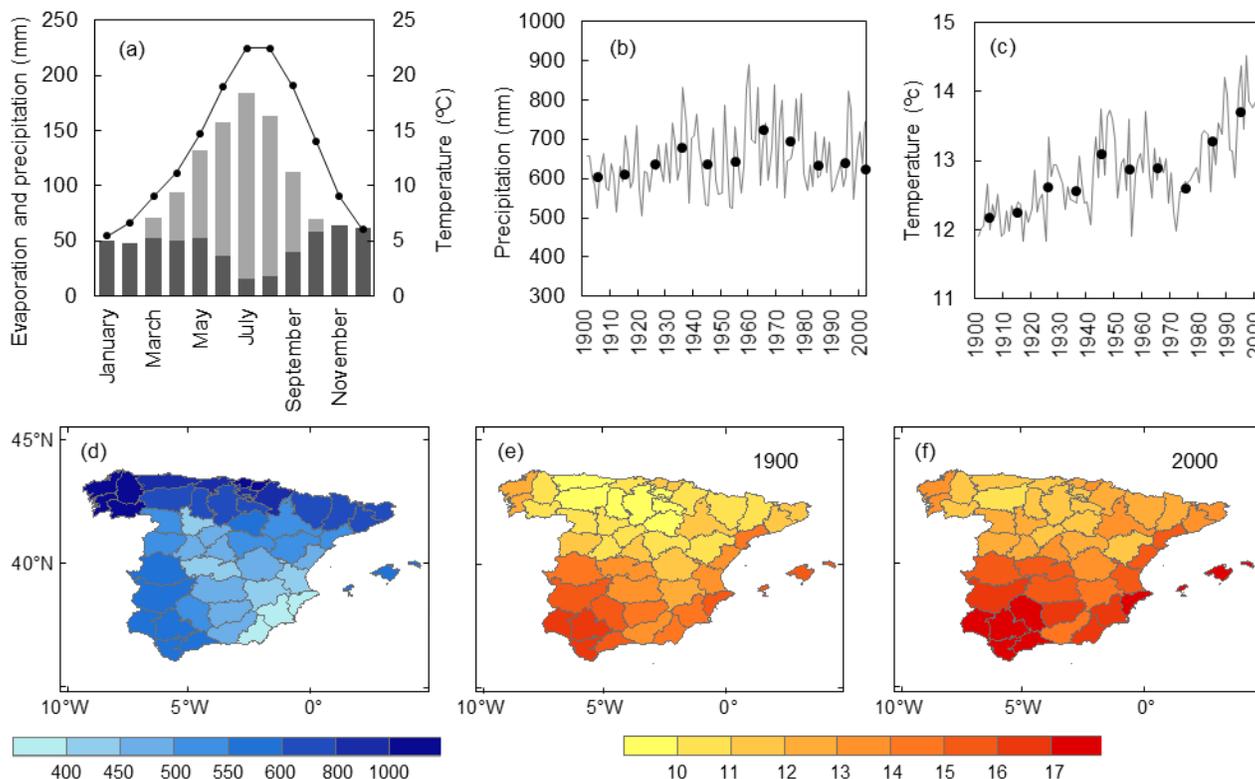
96 productivity, which was used to feed an increasing population, to increase the share of animal  
97 products in the diet, and to raise exports of high-value crop products (Lassaletta et al., 2014,  
98 Soto et al., 2016). Recent assessments have shown some of the biophysical costs of these  
99 productivity gains. The reliance on external and total energy consumed led to a significant  
100 decrease in the energy return on investment (EROI) (Guzmán et al., 2017), a growing  
101 dependence on feed biomass imports (Soto et al. 2016), a large nitrogen (N) surplus  
102 (Lassaletta et al., 2014) and a strong pressure on scarce water resources (Duarte et al., 2014).  
103 The main aims of this study were to analyze cropland SOC dynamics in Spain, used here as  
104 a model Mediterranean country, in the long-term (+100 years), and to identify the main  
105 drivers responsible for the observed trends. The specific objectives were: (i) to build and test  
106 a simplified SOC model for its use in historical studies; (ii) to reconstruct NPP and soil C  
107 inputs from 1900 to 2008; (iii) to simulate SOC stock changes from 1900 to 2008; and (iv)  
108 to test the sensitivity of the model outputs to changes in key model parameters.

109

## 110 **2. Methods**

### 111 *2.1 Study site characteristics*

112 Climate in Spain is mostly Mediterranean, with hot, dry summers and wet, mild autumns and  
113 winters. Severe water deficit during the summer (Fig. 1a) is one of the features controlling  
114 crops distribution and management. There is a strip of temperate climate in the northern  
115 coast, and a gradient of dryness towards the South-East (Fig. 1d).



116

117 **Fig. 1.** Agro-climatic features of Spain during the studied period. (a) 1901-2002 monthly average of potential evaporation  
118 (light bars), precipitation (dark bars) and temperature (dots and lines), (b) annual (lines) and decadal (dots) average  
119 precipitation (mm), (c) annual (lines) and decadal (dots) average air temperature, (d) provincial distribution of annual  
120 average precipitation in 1900-2002 (mm), (e) provincial distribution of annual average air temperature in 1900-1909 (°C),  
121 and (f) provincial distribution of annual average air temperature in 2000-2002. Data from Goerlich Gisbert (2012).

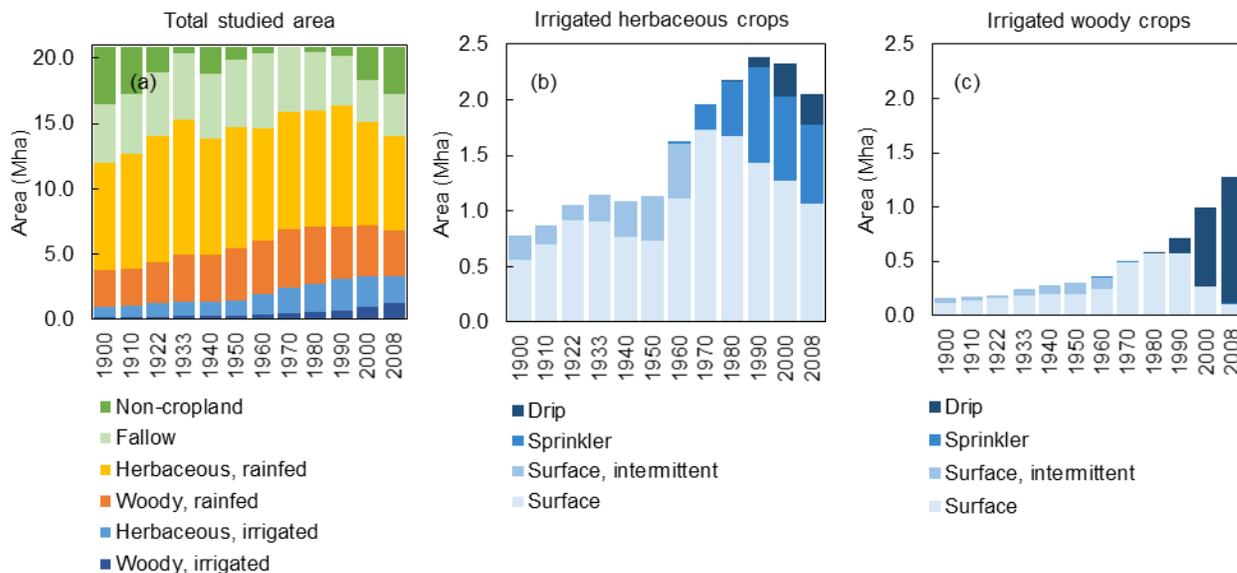
122

123 Annual mean precipitation during the 20<sup>th</sup> century in Spain ranged from 500 to 900 mm, with  
124 no clear trend (Fig. 1b). Mean temperature increased from 12.3°C in the 1900-1909 period to  
125 13.8°C in the 2000-2002 period (Fig. 1c,e,f), corresponding to an average increase rate of  
126 0.17°C per decade, which can be compared to the 0.1°C decadal average global land warming  
127 estimated for the 1901-2012 period (Hartman et al. 2013). These changes have resulted in  
128 increasing drought severity during the last 50 years (Vicente-Serrano et al., 2014).

129 The main soil orders in Spain are Entisols and Inceptisols, which account for more than three-  
130 quarters of the total national surface area (Gómez-Miguel and Badía-Villas, 2016). National

131 means and standard deviations (in parenthesis) for pH, soil organic matter, and sand, silt and  
132 clay proportions (%) are 7.47 (1.49), 2.53 (2.87), 51.77 (19.99), 26.50 (14.73) and 21.77  
133 (10.98), respectively (López Arias and Grau Corbí, 2005).

134 Area and production values for each crop type-management category were retrieved from the  
135 Agricultural Statistics Yearbooks, available online at MAPAMA (2017). In some cases, area  
136 and production values in rainfed and irrigated land had to be adjusted to match the total values  
137 provided in the source and the total irrigated area. Outliers in the data were also disregarded.  
138 The estimation of the total irrigated area and the segregation of the irrigated area by irrigation  
139 types was based on various official reports (MAICOP, 1904; MF, 1918; MAGRAMA, 2015)  
140 and secondary sources (Calatayud and Martínez-Carrión, 2005). Cropland area increased  
141 from 33% in 1900 to 41% in 1970, decreasing down to 34% by 2008 (Fig. 2a). Herbaceous  
142 crops represent the majority of cropland area (Fig. 2a), but the share of woody crops is also  
143 very significant (ranging from 18% in 1900 to 28% in 2008). Fallow land was highest in  
144 1960 (36% of herbaceous crops area), and lowest in 2000 (24%). Irrigated area increased  
145 from 6% to 19% of cropland from 1900 to 2008, with a growing share of sprinkler irrigation  
146 systems since 1970 and of drip irrigation systems since 1990 (Fig. 2b, c).



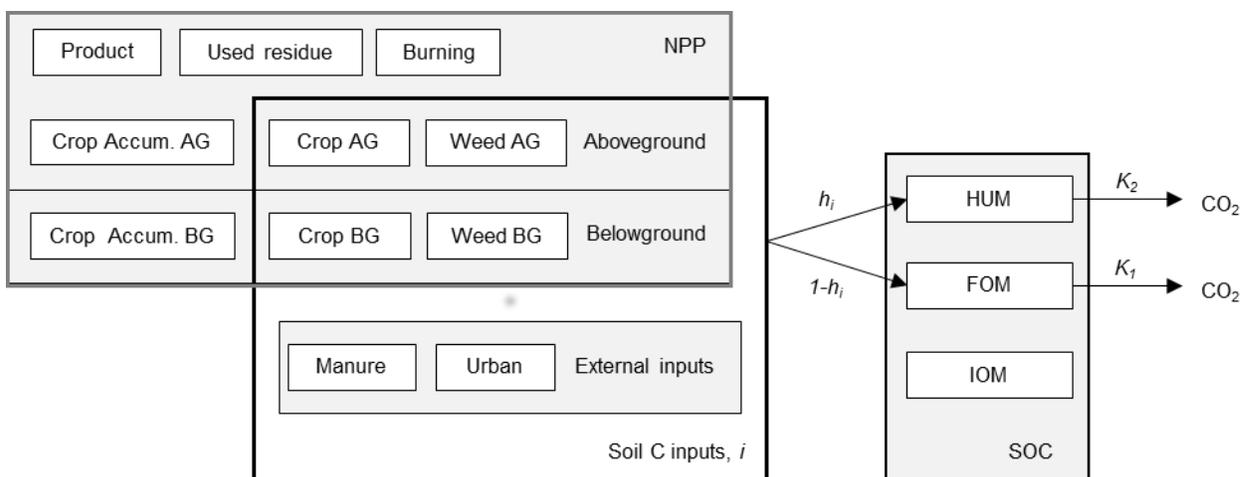
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148 **Fig. 2.** Land use distribution in the studied area of Spain, corresponding to the maximum cropland area over the study  
 149 period. (a) Area distribution by land use type and presence of irrigation, (b) irrigation types in herbaceous crops, and (c)  
 150 irrigation types in woody crops. Sources: MAPAMA (2017) for land uses, and MAICOP (1904), MF (1918), Calatayud and  
 151 Martínez-Carrión (2005) and MAGRAMA (2015) for irrigation types.

152

## 153 2.2 Soil organic carbon model description

154 Humified Soil Organic Carbon (HSOC) model is an adaptation of RothC model (Coleman  
 155 and Jenkinson, 1996), consisting in its simplification into two active SOC pools; fresh  
 156 organic matter, (FOM) and humus (HUM), and one inactive pool (IOM) (Fig. 3). In HSOC  
 157 model, the three labile C pools in RothC (resistant plant material, decomposable plant  
 158 material, and microbial biomass) are merged into a single pool (FOM). This allows for the  
 159 reduction of internal feedbacks and thus for an easier interpretation of the model functioning.  
 160 The simplification of the model also allows for a better integration of factors that have an  
 161 influence on SOC dynamics but are not considered in RothC, particularly the effect of C  
 162 input quality on humification rates.



163

164 **Fig. 3.** Basic structure of HSOC model, and components of net primary production (NPP)  
 165 components are categorized according to their source (crop or weed), their placement (aboveground, AG, and belowground,  
 166 BG), and their final use. The biomass of aboveground crop residues is divided into *Crop AG* (unharvested and applied to  
 167 the soil), *burning* (burnt on field), *used residues* (extracted from the field) and *accumulated* (incorporated into woody tissues  
 168 and remaining in the field).  $h_i$ : humification coefficient of each C input I, *HUM*: slow turnover soil organic carbon (SOC)  
 169 pool; *FOM*: fast turnover SOC pool; *IOM*: inert SOC pool;  $K_1$  and  $K_2$  stand for annual decomposition rates

170

171 Both active pools in HSOC model follow first-order kinetics, *FOM* with a fast turnover rate  
 172 and *HUM* a slow one. Decomposition rates of both pools are controlled by agro-climatic  
 173 factors. The inputs to *HUM* are calculated from annual soil C inputs using input-specific  
 174 humification coefficients ( $H_i$ ). Inputs to *FOM* are calculated as the total C inputs applied to  
 175 the soil minus those that are humified.

176 Starting from year 0, SOC content C in year t is given by:

$$177 \quad SOC_t = IOM + HUM_t + FOM_t \quad (1)$$

$$178 \quad HUM_t = HUM_0 \cdot e^{-K_2 \cdot t} + \sum_{i=0} \frac{I_i \cdot h_i}{K_2} \cdot (1 - e^{-K_2 \cdot t}) \quad (2)$$

$$179 \quad FOM_t = FOM_0 \cdot e^{-K_1 \cdot t} + \sum_{i=0} \frac{I_i \cdot (1-h_i)}{K_1} \cdot (1 - e^{-K_1 \cdot t}) \quad (3)$$

180 where  $K_1$  and  $K_2$  are the decomposition rates of *FOM* and *HUM* during the studied period,  
181 respectively,  $I$  is the amount of each C input  $i$  applied each year, and  $h_i$  is its corresponding  
182 humification coefficient. IOM is calculated following Falloon et al. (1998) equation, which  
183 was applied to the average SOC levels for Spanish cropland in 2008 (Rodríguez-Martín et  
184 al., 2016).

185 Total SOC stock at equilibrium is given by:

$$186 \quad SOC_{eq} = IOM + \frac{\sum_{i=0} I_i \cdot h_i}{K_2} + \frac{\sum_{i=0} I_i \cdot (1-h_i)}{K_1} \quad (4)$$

187 Tillage is an important factor controlling SOC dynamics, but it was not included in the model  
188 due to the following reasons: 1) Tillage is not included in RothC model, which is the basis  
189 for HSOC model and has been validated without this factor. 2) Changes in tillage practices  
190 produce a vertical redistribution of SOC (e.g. Manley et al., 2005, Luo et al. 2010). This  
191 increases the uncertainty of the estimates of total SOC changes, that are strongly dependent  
192 on sampling depth; 3) Primary data regarding the historical changes in tillage practices in  
193 Spain is lacking, as far as we know.

194

### 195 *2.3 Net primary production*

196 Net Primary Production (NPP) in Spanish cropland was estimated as detailed in Soto et al.  
197 (2016) and Guzmán et al. (2017). In short, area and yield data of all crops cultivated in Spain  
198 from 1900 to 2008, collected from official statistics (MAPAMA, 2017), and expressed as 5-  
199 year averages in 12 selected time points. In this study, it was also distinguished between  
200 rainfed and irrigated crops. Total aboveground (AG) crop biomass production was estimated

201 by applying dry matter coefficients and harvest indices (Guzmán et al., 2014). Harvest  
202 indices of traditional and modern cereal varieties were distinguished.

203 Allometric functions (“Fixed root:shoot ratios”) are the most common approach for the  
204 estimation of root biomass production in GHG inventories and large-scale NPP or C  
205 assessments (Johnson et al., 2006, Taghizadeh-Toosi et al., 2016, Niedertscheider et al.,  
206 2016), and they have also been used in biomass and energy analyses of Spanish agriculture  
207 (Soto et al., 2016, Guzmán et al., 2017). There is increasing evidence, however, that  
208 root:shoot ratios are decreased by yield-promoting management changes, particularly by  
209 inputs of synthetic nutrients (Andrews et al. 2001, Chirinda et al., 2012, Poeplau, 2016,  
210 Poeplau et al., 2016, Grechi et al., 2007) and irrigation (Buwalda, 1993, Kozłowski and  
211 Pallardy 2002, Gan et al., 2009, Wilcox et al., 2016). Consequently, simulation studies  
212 suggest that using fixed root:shoot ratios overestimates the response of roots to shoot  
213 productivity changes (Poeplau, 2016), and that using a fixed amount of root mass (“Fixed  
214 root mass”) may lead to better fit with experimental data (Taghizadeh-Toosi et al., 2016).  
215 Other studies, however, suggest that root-shoot ratios of crops may have even increased along  
216 recent history (Weismeyer et al., 2014). Given this controversy, root biomass production was  
217 estimated as the average of the “fixed root:shoot ratio” and the “fixed root mass” approaches.  
218 The reference root mass value for the fixed root biomass approach was calculated by applying  
219 reference root:shoot ratios (Table A.1) to the aboveground production values of year 2000.  
220 In the case of traditional wheat varieties, the root:shoot ratio was set at 38% above that of  
221 modern varieties, based on our own data from a field experiment (Carranza Gallego et al.,  
222 Accepted), and in line with other published studies (Siddique et al., 1990, Bektas et al., 2016).

223 The effects of using fixed root:shoot ratios and fixed root mass approaches were assessed in  
224 a sensitivity analysis.

225 Above-ground and below-ground weed biomass production were taken into account in the  
226 estimation of cropland NPP (Soto et al., 2016), using data on modern organic agriculture for  
227 the 1900-1950 period, modern conventional agriculture for the 1980-2008 period (Guzmán  
228 et al., 2014, 2017), and linearly interpolating in the intermediate time points. A scenario  
229 without weeds was incorporated in the sensitivity analysis.

230 Carbon contents of crop products were taken from Wolf et al. (2015). Modeling studies  
231 usually assume a general value for the C content of inputs (e.g. Wolf et al., 2015, Nair, 2012).  
232 In this study, specific values were used for crop species or categories, depending on data  
233 availability (Table A.2).

#### 234 *2.4 Soil C inputs*

235 The annual plant C input to the soil was estimated by subtracting from total NPP the biomass  
236 extracted, burned and accumulated. Extracted biomass includes all crop production and the  
237 fraction of crop residues that is used, including harvested and grazed residues. Harvested  
238 crop and residue data were directly taken from statistics, while grazed biomass was estimated  
239 with a feed balance approach (Soto et al. 2016). The fraction of residues burned in the field  
240 was also taken from Soto et al. (2016), with modifications for cereals and industrial crops  
241 (Table A.3). Accumulated biomass refers to the fraction of the biomass of woody crops that  
242 is annually incorporated into permanent woody tissues.

243 External soil C inputs include manure and urban waste C inputs. Manure includes excretion  
244 of grazing animals and applications of managed manure. The amount of manure produced,

245 in terms of N, was estimated through a mass-balance approach, by subtracting gross N  
 246 production in slaughtered animals and livestock products (calculated using coefficients from  
 247 Bodirsky et al. 2012) from total N ingestion by livestock, estimated with biomass intake  
 248 values estimated in Soto et al. (2016) and N contents from García-Ruiz et al. (in prep). The  
 249 distribution of excreted N among animal species and fates is described in Appendix B. N  
 250 losses associated to manure management were estimated at 35.7% of excreted N (Pardo et  
 251 al. 2015). Manure N was converted to C using C:N ratios from the literature (Table 1).

252 The application of urban waste to cropland is reported by official statistics since 1990,  
 253 expressed as N (MAPAMA, 2017BNAE), which was converted to C using the data from  
 254 Table 1). For the previous period, it was assumed that the per capita agricultural use of urban  
 255 waste was the same as in 1990.

256 **Table 1.** Mean, standard deviation (SD), and number of studies (N) of C:N ratios of external  
 257 C inputs. See text for definitions of categories.

	Mean	SD	N	Sources
Manures "ready to apply"				
Farmyard manure	21.2	9.3	34	[1-11]
Pig manure	13.2	3.3	7	[3, 4, 8, 14]
Poultry litter	8.6	2.6	19	[3, 4, 8, 12-14]
Pig slurry	4.2	3.5	77	[15]
Bovine slurry	7.1	3.3	8	[3, 9, 16, 17]
Manures "as excreted"				
Bovine	19.1	4.4	18	[1, 10, 17-20]
Ovine/caprine	12.4	5.6	7	[19, 21, 22]
Pig	9.1	3.7	8	[8, 23-27]
Poultry	6.9	1.1	3	[8, 13, 26, 12]
Urban wastes				
Sewage sludge	7.3	1.8	20	[4, 6, 28, 29]
Municipal solid waste, composted	30.6	25.5	8	[4, 28, 29]

258 [1] Castellanos-Navarrete et al. (2015), [2] Chastain and Moore (2014), [3] Chadwik et al. (2000), [4]  
259 ECN (2017), [5] Ghosh et al. (2012), [6] Iakimenko et al. (1996), [7] Miller et al. (2003), [8] Mishima  
260 et al. (2012), [9] Pettygrove et al. (2009), [10] Tittonell et al. (2010), [11] Yamulki (2006), [12] Edwards  
261 and Daniels (1992), [13] Griffiths (2011), [14] Xu et al. (2017), [15] Antezana et al. (2016), [16] Amon  
262 et al. (2006), [17] Triberti et al. (2008), [18] Chen et al. (2003), [19] Jarvis et al. (1995), [20] Thomsen  
263 et al. (2013), [21] Ma et al. (2007), [22] Mafongoya et al. (2000), [23] Jacobs et al. (2011), [24] Jarret  
264 et al. (2012), [25] Jorgensen et al. (2013), [26] Kirchmann and Witter (1992), [27] Vu et al. (2009), [28]  
265 Mondini et al. (2017), [29] Plaza et al. (2016)

266

## 267 2.5 Humification coefficients

268 Humification coefficients ( $H_i$ ) indicate the fraction of soil C inputs entering the slow turnover  
269 *HUM* pool.  $H_i$  are specific for each C input type  $i$ , and they are calculated from basal  
270 humification coefficients,  $h_i$ , modified by a soil texture modifying factor,  $d$ , as follows:

$$271 H_i = h_i \cdot d \quad (5)$$

272 The literature was searched for input-specific humification coefficients ( $h_i$ ) derived from  
273 long-term (>4 years) experiments (Table 2). The definition and the calculation methodology  
274 varies among studies, which contributes to the variability found in the data. In many studies,  
275 a “carbon retention” coefficient is estimated by calculating the relative amount of cumulative  
276 C input retained in the soil after a certain period, either through  $^{13}\text{C}$  isotope analyses (e.g.  
277 Bolinder et al., 1999) or from the slope of the SOC increase versus the cumulative C input  
278 (e.g. Maillard et al., 2014). This approach is sensitive to SOC decomposition rate and study  
279 duration. In another group of studies, the C input and SOC dynamics data were fitted into  
280 first-order dynamics SOC models, usually with one active SOC compartment (e.g. Bayer et  
281 al. 2006), but also with two compartments (e.g. Andren and Kätterer, 1997). In spite of these  
282 conceptual differences, we did not find significant differences among the  $h$  values estimated

283 with the different approaches (data not shown), and the average study duration was similar  
 284 among the studied categories (Table 2).

285 **Table 2.** Mean, standard deviation (SD), number of studies (N), of the humification coefficients (*h*)  
 286 for C input categories (% of applied C). The mean duration of the experiments is also shown.

	Mean (%)	SD (%)	N	Mean duration (years)	References
Herbaceous residues	11.5	4.3	15	24	[1-9]
Herbaceous residue + roots	16.1	7.8	11	29	[10-20]
Pruning residues	32.5	3.5	2	Not available	[21, 22]
Roots	21.8	7.7	13	30	[2-4, 6, 7, 23-26]
Extra-root C <sup>a</sup>	8.0			Not available	Calculated from [26]
Manure	25.4	10.5	11	34	[1, 3-6, 9, 24, 27, 28]
Sewage sludge	38.6	13.7	5	35	[1, 4, 6, 27]

287 <sup>a</sup>Own calculation from root turnover and rhizodeposits data

288 References: [1] Andren and Kätterer (1997), [2] Barber (1979), [3] Berti et al. (2016), [4] Boiffin et al. (1986),  
 289 [5] Delas and Molot (1983), [6] Kätterer et al. (2011), [7] Plenet et al. (1993), [8] Thomsen and Christensen  
 290 (2010), [9] Thomsen and Christensen (2004), [10] Bayer et al. (2006), [11] Bolinder et al. (1999) (from Balesdent  
 291 et al. 1990), [12] Buyanovsky and Wagner (1998), [13] Campbell et al. (1991), [14] Kong et al. (2005), [15]  
 292 Lovato et al. (2004), [16] Gregorich et al. (1995), [17] Gregorich et al. (1996), [18] Poeplau et al. (2015), [19]  
 293 Saffih-Hdadi and Mary (2008), [20] Vieira et al. (2009), [21] Sofo et al. (2005), [22] Bosco et al. (2013), [23]  
 294 Angers et al. (1995), [24] Bertora et al. (2009), [25] Bolinder et al. (1999), [26] Brock et al. (2012), [27] Bhogal  
 295 et al. (2007), [28] Maillard et al. (2014)

296

297 Lower *h* values for herbaceous crop residues than for roots (Table 2) are in line with many  
 298 studies comparing both types of materials (e.g. Bolinder et al., 1999, Rasse et al., 2005,  
 299 Kätterer et al., 2011, Berti et al., 2016). In the case of pruning residues of woody crops, the  
 300 relatively high value is in line with studies in forest soils (Polglase et al., 2000, Zhou et al.,  
 301 2016). Values for external amendments are in agreement with the median C retention of 31%  
 302 (N=25) observed by Aguilera et al. (2013) for a wide range of external C inputs applied to  
 303 soils under Mediterranean conditions. In spite of this, Maillard et al. (2014) found a global  
 304 average C retention coefficient for manure of only 12%, which was used to build a scenario  
 305 in the sensitivity analysis.

306 Soil texture is usually assumed to influence the stabilization of SOC (e.g. Poeplau et al.  
 307 2015), although it may influence SOC dynamics through SOC mineralization (Saffih-Hdadi

308 and Mary, 2008). In HSOC model, humification coefficients are modulated by the texture  
309 modifying factor  $d$ , which is adapted from Coleman and Jenkinson (1996) equation for the  
310 partitioning of plant material between CO<sub>2</sub> and HUM and BIO pools in RothC.

$$311 \quad d = 3.51 / (1.67 \cdot (1.85 + 1.6 \cdot e^{-0.0786 \cdot CF})) \quad (6)$$

312 Where CF is the clay fraction of the soil (%). As in RothC, soil texture in HSOC model is  
313 only included as a modifying factor of humification, not on SOC mineralization. However,  
314 it is worth noting that texture indirectly affects the mineralization rates through its effect on  
315 soil moisture.

316

### 317 *2.6 Decomposition rates of SOC pools*

318 Decomposition rates  $K_1$  and  $K_2$  are based on RothC model approach and parameters.  
319 Decomposition rate constants  $k_1$  and  $k_2$  are modulated each month  $m$  by modifying factors,  
320 as follows:

$$321 \quad K = \sum_{m=12} (k \cdot a_m \cdot b_m \cdot c_m \cdot t) \quad (7)$$

322 Where  $a$  is the rate modifying factor for temperature,  $b$  for moisture,  $c$  for soil cover, and  $t$  is  
323 the time step, corresponding to 1/12.  $k_1$  was set at 48%, so that FOM matched the SOC  
324 fraction represented at equilibrium by the equivalent pools in RothC.  $k_2$  was set at 0.02, as in  
325 RothC. The equations for  $a$ ,  $b$ , and  $c$  modifying factors were taken from RothC (Coleman  
326 and Jenkinson, 2014).

327 Monthly average climate data for the 1901-2002 period in 46 out of 50 Spanish provinces  
328 (excluding Canary Islands, Ceuta and Melilla) was gathered from Goerlich Gisbert (2012),

329 who transformed the grid data from the CRU TS 2.1 database to the provincial level. Potential  
330 evapotranspiration was calculated with the Hargreaves equation (Hargreaves and Samani,  
331 1985). A sensitivity analysis scenario was also built using the average climate data (1901-  
332 2002) for the whole simulation period, in order to isolate the effect of changes in climate on  
333 SOC changes.

334 The crop area and production data was not spatially disaggregated for the whole study period,  
335 so the climate modifying factors had to be aggregated on a national level in order to run the  
336 model simulations. This aggregation was calculated as a weighted average of the provincial  
337 climate modifying factors. Weighting was based on the relative provincial distribution of  
338 crop-management categories in year 2000 (MAPAMA, 2017, Table A.4). Crop-management  
339 categories segregate production and area data based on main crop types (winter cereals,  
340 summer cereals, forage, other herbaceous, fruits, treenuts, olive, grapevine fallow and non-  
341 cropland) and two management types (rainfed and irrigated).

342 Annual irrigation water inputs were assumed to be 750, 650 and 500 mm for surface,  
343 sprinkler and drip irrigation systems, respectively (Corominas, 2010). Water inputs for  
344 intermittent irrigation were estimated as one third of those of surface irrigation systems.  
345 Water inputs were distributed monthly taking into account the crop-growing period and the  
346 water balance in each month.

347 Monthly soil cover for the selected crop-management categories was estimated according to  
348 our own field experience (Table A.5). The monthly average soil cover in woody crops was  
349 modeled by defining two extreme categories representing systems with and without cover  
350 crops (Table A.5), which were weighted according to an index (Table A.6) based on the  
351 amount of biomass produced by the cover crop, taken from Soto et al. (2016).

352

353 *2.7 Model application*

354 The model was first tested by comparing it to RothC and experimental data under  
355 Mediterranean conditions (Appendix B), including experiments with rainfed barley (Álvarez-  
356 Fuentes et al., 2012), rainfed cereal rotations (Sombbrero and Benito, 2010), and rainfed olives  
357 (Nieto et al., 2010). A good agreement between modelled and observed SOC values was  
358 observed, as well as between HSOC and RothC model outputs. In particular, we verified that  
359 the dynamics of the FOM pool were very similar to those of the sum of the three labile pools  
360 in RothC (DPM, RPM and BIO), indicating the validity of the unification of these pools into  
361 a single pool that was implemented in HSOC. After validation, the model was applied to  
362 reconstruct cropland SOC in Spain. The model was applied to the 0-30 cm layer of the soil.  
363 *Total studied area* corresponds to the maximum cropland area in Spain during the studied  
364 period, which was 20.9 Mha in 1970 (Fig. 2a). As cropland area changes along time, the  
365 remaining area (named “non-cropland”) was modelled as the weighted average of grassland  
366 and forestland in the country, with area and biomass production data from Soto et al. (2016).  
367 Woody belowground biomass values reported by Soto et al. (2016) were corrected to  
368 consider only the first 30 cm of the soil. This correction factor was set at 61%, which is the  
369 average of temperate coniferous forest, temperate deciduous forest, and sclerophyllous  
370 shrubs in Jackson et al. (1996).

371 *Total cropland area* represents the area actually cropped, including temporary fallow.  $SOC_{eq}$   
372 in the total cropland area is proportional to cropland SOC concentration, which can be  
373 considered a proxy of soil quality and adaptation potential (Lal et al., 2011, Aguilera et al.,

2013). A given configuration of land uses and practices resulting in high equilibrium SOC ( $SOC_{eq}$ ) in the total studied area but low  $SOC_{eq}$  in the total cropland area would be valuable for GHG mitigation (high total C stored) but not for long-term productivity and adaptability (low SOC in cropland soils).

It was assumed that SOC was at equilibrium at the starting point (1900). The model was first run up to the second time step (1910), and before initiating a new cycle, the land use changes that had taken place during the decade were applied by pooling together all land use types losing area, and correspondingly allocating their area to the land use types gaining area. This process was iterated in subsequent cycles.

383

## 2.8 Sensitivity analysis

The simulations were subjected to a sensitivity analysis to test the effect of changing key variable parameters on the results. The tested parameters were root biomass (two scenarios: Fixed root-shoot and Fixed root mass), weed biomass (one scenario: No weeds), manure (Manure h) and climate (Fixed climate). The scenarios are described in Table A.7.

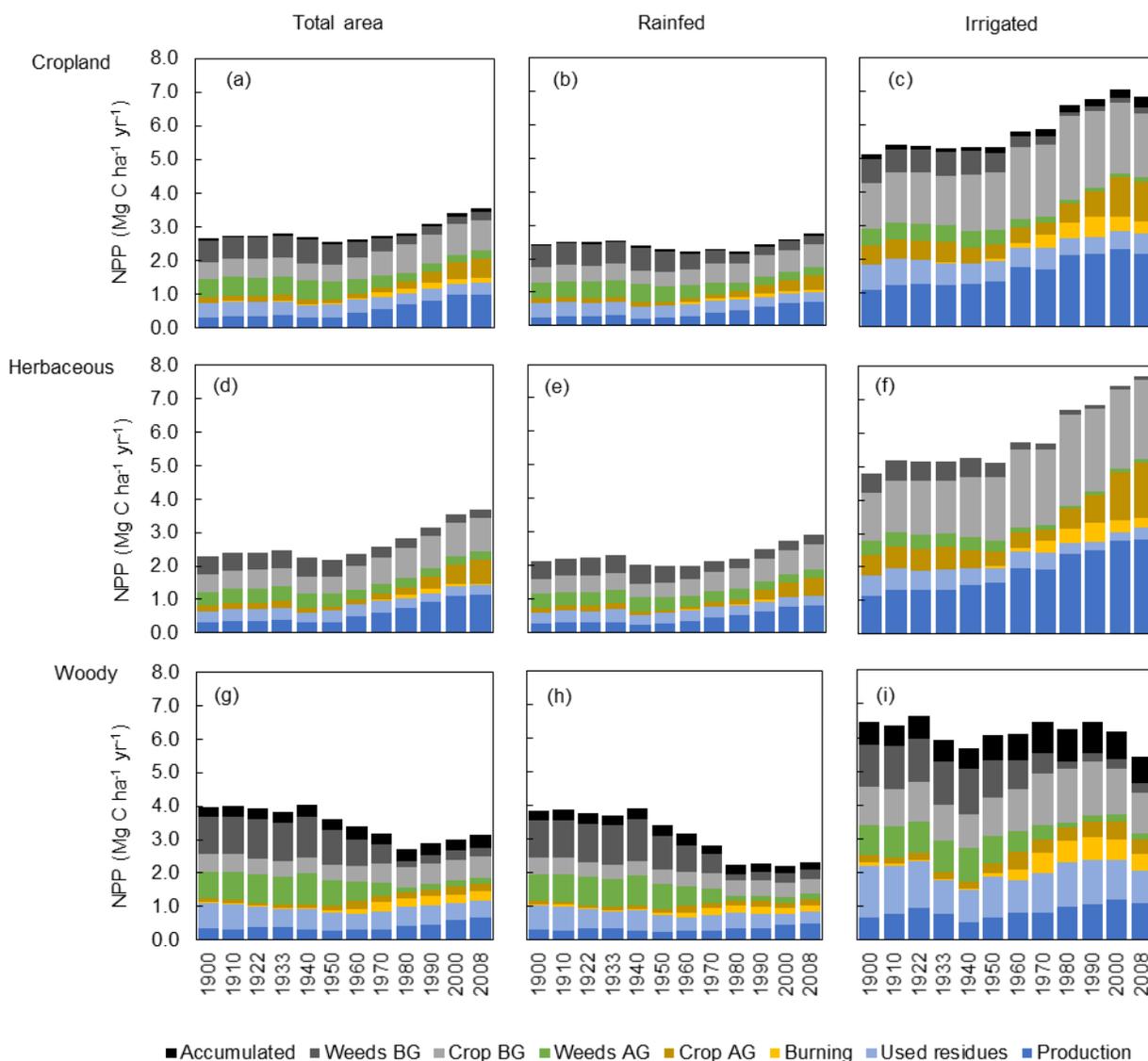
389

## 3. Results

### 3.1 Net primary production

Cropland NPP reached a first peak in 1933 and fell after the Spanish Civil War (1936-1939), reaching its minimum in 1950 at  $2.5 \text{ Mg C ha}^{-1} \text{ yr}^{-1}$ , when it started a steady growth, although it did not surpass the 1933 level until 1980 (Fig. 4a). Overall, cropland NPP, averaged across

395 all crops, grew 37% during the studied period, up to 3.5 Mg C ha<sup>-1</sup> yr<sup>-1</sup> in 2008. NPP grew  
 396 13% in rainfed systems and 34% in irrigated systems (Fig. 4b,c). NPP of irrigated systems  
 397 doubled that of rainfed systems in 1900, and tripled it in 1980. NPP growth in herbaceous  
 398 crops was less pronounced in rainfed (30%) than in irrigated systems (60%), while it was  
 399 negative both in rainfed (-40%) and irrigated (-16%) woody crops (Fig. 4e,f,h,i).



400

401 **Fig. 4.** Historical evolution of Net Primary Production (NPP) in Spanish total (a), rainfed (b) and irrigated (c) cropland, total  
 402 (d), rainfed (e) and irrigated (f) herbaceous crops, and total (g), rainfed (h) and irrigated (i) woody crops. NPP components  
 403 are categorized according to their source (crop or weed), their placement (aboveground, AG, and belowground, BG), and  
 404 their final use.

405

406 Crop production was the NPP component with the largest increase, ranging from 61% in  
407 rainfed and irrigated woody crops (Fig. 4h,i), to 220% and 146% in rainfed and irrigated  
408 herbaceous crops, respectively (Fig 4e,f). The growth in total NPP considering the total area  
409 of each crop type was higher, of 99% and 277% in woody and herbaceous crops, respectively,  
410 reflecting the effect of the expansion of irrigation (Fig. 2).

411 The share of crop residues in total cropland NPP grew from 23% in 1900 to 30% in 2008,  
412 despite the decline in harvest indices. Used residues represented more than two-thirds of the  
413 extracted biomass in 1900, falling to about one quarter in 2008. Meanwhile, the share of  
414 residues burnt in the field grew from 1% in 1900 to 21% in 1990, dropping to 11% in 2008.

415 Root:shoot ratios of most crop types declined (Table A.8), while root biomass production per  
416 hectare increased (Table A.9) during the studied period, which is related to shoot productivity  
417 increases.

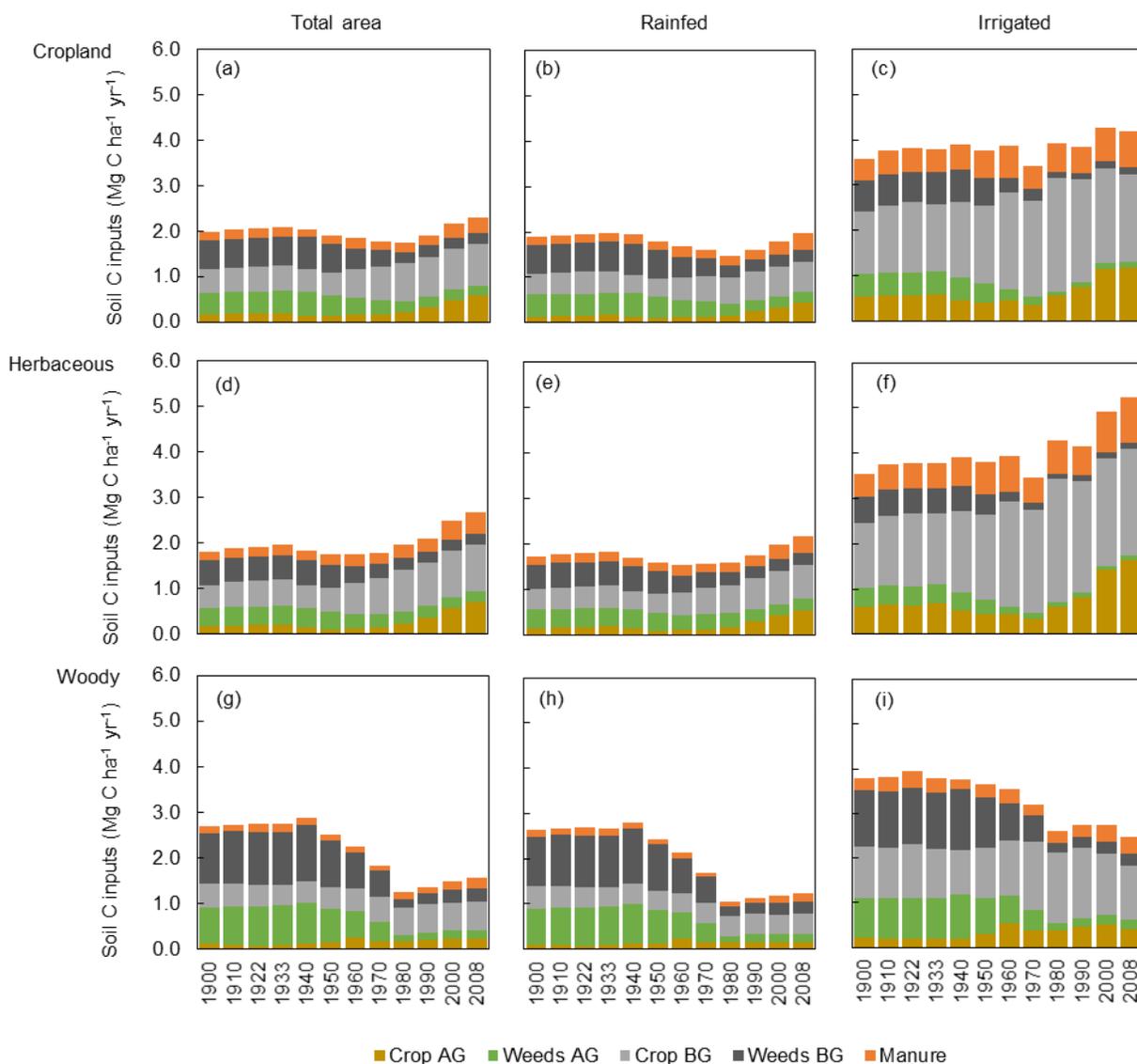
418 Weed biomass production averaged  $1.1 \text{ Mg C ha}^{-1} \text{ yr}^{-1}$  during the first half of the 20<sup>th</sup> century,  
419 falling to  $0.5 \text{ Mg C ha}^{-1} \text{ yr}^{-1}$  from 1980 onward (Fig. 4a). The drop was specially marked for  
420 woody crops (Fig. 4g), from  $1.9$  to  $0.5 \text{ Mg C ha}^{-1} \text{ yr}^{-1}$ , accounting for 48% and 15% of the  
421 NPP of woody cropping systems, respectively.

422

### 423 *3.2 Carbon inputs to the soil*

424 Average soil C inputs in the total cropland area grew from 2 to  $2.4 \text{ Mg C ha}^{-1} \text{ yr}^{-1}$  during the  
425 studied period, with a significant drop down to  $1.8 \text{ Mg C ha}^{-1} \text{ yr}^{-1}$  in the 1970s (Fig. 5a). This

426 decline was first associated to declining crop productivity and NPP during the 1933-1950  
 427 period, and from 1950 to 1980 due to the drop in annual weed biomass production (Fig. 4).  
 428 The subsequent increase from 1980 (Fig. 5a) can be largely explained by increasing crop  
 429 residue application due to a combination of an overall increase in crop productivity and a  
 430 decrease in the relative use and burning of crop residues (Fig. 4).



431

432 **Fig. 5.** Historical evolution of soil carbon (C) inputs in Spanish total (a), rainfed (b) and irrigated (c) cropland, total (d),  
 433 rainfed (e) and irrigated (f) herbaceous crops, and total (g), rainfed (h) and irrigated (i) woody crops. Soil C inputs  
 434 components are categorized according to their source (crop, weed or external), and their placement (aboveground, AG, and  
 435 belowground, BG). Manure category includes manure and urban waste.

436

437 Overall, soil C inputs in rainfed systems were similar in 1900 and 2008, at 1.9 Mg C ha<sup>-1</sup> yr<sup>-1</sup>  
438 <sup>1</sup> after having fallen 27% in 1980 from their 1933 peak at 2.0 Mg C ha<sup>-1</sup> yr<sup>-1</sup> (Fig. 5b). Soil C  
439 inputs in irrigated systems peaked in 2000 at 4.3 Mg C ha<sup>-1</sup> yr<sup>-1</sup>, up 20% from 1900 levels  
440 (Fig. 5c).

441 Crop residues applied to the soil reached a first peak (0.2 Mg C ha<sup>-1</sup> yr<sup>-1</sup>) in 1933, just before  
442 Spanish Civil War, and declined to 0.1 Mg C ha<sup>-1</sup> yr<sup>-1</sup> by 1950, but they started growing  
443 strongly after 1980, reaching 0.6 Mg C ha<sup>-1</sup> yr<sup>-1</sup> in 2008 (Fig. 5a). The contribution of crop  
444 residues to total soil C inputs was lowest for woody crops in older time steps, ranging 3-6%  
445 (Fig. 5h,i), and highest for herbaceous crops in modern time steps, ranging 21-31% (Fig.  
446 5e,f).

447 Crop C allocated belowground represented a very significant fraction of soil C inputs. On  
448 average, their relative contribution rose from 26% in 1900 to 47% in 1980. Total  
449 belowground contribution to soil C inputs, including weeds, ranged 50-70% across all crop-  
450 management categories and periods (Fig. 5).

451 Weed biomass played a major role in the observed trends in soil C inputs. Until 1960, it  
452 represented about 50% and 75% of soil C inputs in herbaceous and woody crops, respectively  
453 (Fig. 5d,g), whereas by 2008 it had fallen to 18% and 30%, respectively. Weed C had a  
454 greater relevance in rainfed than in irrigated crops (Fig. 5e,f,h,i). This trend in weed biomass  
455 largely explains the decrease in soil C inputs during the 1950-1980 period in a context of  
456 strong growth in crop productivity (Fig. 4), as well as the diverging trends in soil C inputs in  
457 woody and herbaceous crops (Fig. 5d,g).

458 Manure C inputs, which include the deposition of animals grazing in cropland, rose from 0.2  
459 to 0.4 Mg C ha<sup>-1</sup> yr<sup>-1</sup> (Fig. 5a) along the studied period, but their contribution to total soil C  
460 inputs was always relatively low, ranging 8-16%. The role of urban waste in the C balance  
461 was always marginal, barely reaching 1% of soil C inputs in 2008.

462 Overall, the average changes in soil C inputs (Fig. 5a) can be largely explained by the net  
463 balance between the increase in C inputs due to the expansion of irrigation, and the decrease  
464 in C inputs due to the expansion of woody crops (Fig. 2), in which inputs decreased strongly  
465 from 1940 to 1980 due to the loss of soil cover (Fig. 5g). The analysis by crop types (Table  
466 A.9) also reveals strong differences in soil C inputs among the different crops, which  
467 contributed to the average changes observed. For example, highest soil C inputs were  
468 observed for alfalfa, and the expansion of this crop contributed to the growth in total cropland  
469 soil C inputs after 1950.

470

### 471 *3.3 SOC decomposition rates*

472 The changes in management and climate exerted strong effects on SOC decomposition rates,  
473 which varied depending on the geographical distribution and specific agronomic features of  
474 each crop-management category (Table 3). In rainfed herbaceous crops, with no management  
475 changes affecting decomposition rates, HUM decomposition rate increased by 21-34%, with  
476 lowest rates for winter cereal crops, cultivated mainly in the central provinces of the country  
477 (Fig. C.1), with a continental Mediterranean climate, and highest rates for summer cereals,  
478 cultivated mainly in the Northern provinces (Fig. C.2), with a temperate climate. The increase

479 in decomposition rates was higher for woody crops, in which the effect of soil cover loss was  
 480 added to the effect of the rise in mean annual temperature.

481 Decomposition rates under irrigation were higher than in rainfed systems (Table 3), but these  
 482 differences were much lower for winter cereals (56% in 1900), which are commonly irrigated  
 483 only in spring, than for summer cereals (108% in 1900), which are irrigated specially during  
 484 the period of high temperatures and with higher water doses. Differences between rainfed  
 485 and irrigated systems were even higher (above 200% in 1900) for other herbaceous crops and  
 486 treenuts, olive and vineyards, which are broadly cultivated in the coastal Mediterranean and  
 487 Southern provinces, with a warmer climate. The trends in decomposition rates estimated for  
 488 irrigated crops were very heterogeneous, increasing by 17-29% in cereals and forage crops,  
 489 mainly due to increasing temperatures, but declining by 7-19% in other herbaceous crops,  
 490 citrus, olive and grapevine. The observed decline took place during the last two decades, and  
 491 it was mainly related to the expansion of drip irrigation, which favors water deficit conditions  
 492 limiting SOC decomposition.

493 **Table 3.** Annual C mineralization rates,  $K_2$  (% SOC yr<sup>-1</sup>) of HUM (slow turnover SOC pool) in simulated crop and land use  
 494 categories in Spain along the studied period

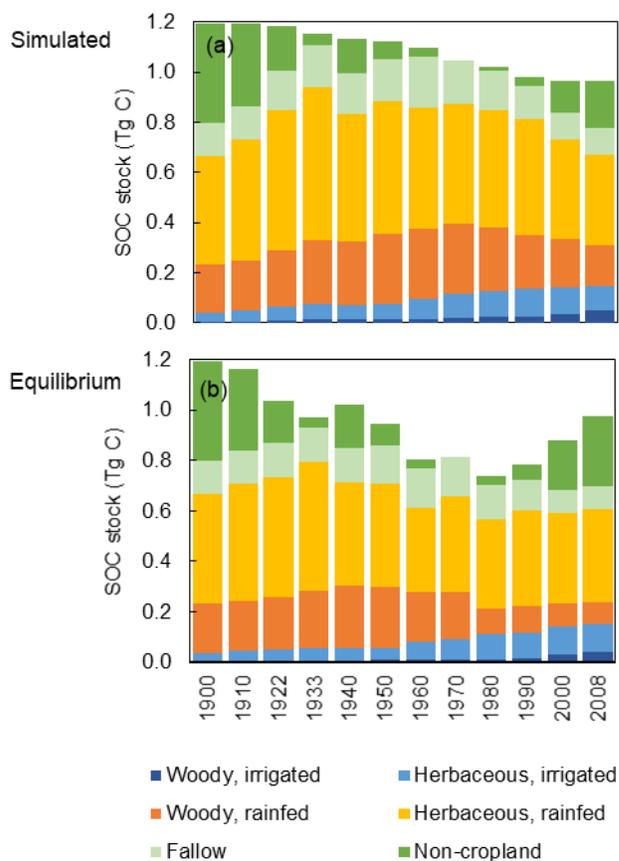
	1900- 1909	1910- 1921	1922- 1932	1933- 1939	1940- 1949	1950- 1959	1960- 1969	1970- 1979	1980- 1989	1990- 1999	2000- 2008
Rainfed											
Winter cereals	0.73	0.75	0.80	0.84	0.80	0.79	0.87	0.77	0.84	0.86	0.97
Summer cereals	1.14	1.20	1.33	1.31	1.39	1.30	1.43	1.30	1.36	1.60	1.49
Forage	0.89	0.92	1.00	0.99	1.02	0.98	1.05	0.96	1.00	1.10	1.08
Other herbaceous	0.87	0.88	0.95	0.96	0.95	0.92	0.99	0.88	0.97	0.96	1.09
Fruits	0.87	0.89	0.92	0.95	0.93	0.99	1.11	1.07	1.20	1.27	1.39
Treenuts	0.77	0.78	0.81	0.80	0.78	0.80	1.01	1.00	1.09	1.03	1.17
Olive	0.87	0.86	0.92	0.91	0.87	0.90	1.06	0.97	1.15	1.08	1.26

Grapevine	0.71	0.72	0.76	0.77	0.73	0.74	0.86	0.79	0.97	0.94	1.09
Fallow	0.89	0.89	0.95	0.97	0.93	0.92	1.01	0.91	0.98	0.97	1.08
Non-cropland	0.67	0.67	0.72	0.73	0.69	0.69	0.76	0.68	0.73	0.72	0.78
Irrigated											
Winter cereals	1.13	1.14	1.18	1.21	1.22	1.20	1.29	1.15	1.28	1.36	1.45
Summer cereals	1.97	1.99	2.04	2.04	2.14	2.13	2.13	2.04	2.12	2.11	2.32
Forage	1.95	1.95	1.98	1.99	2.07	2.09	2.09	2.01	2.17	2.24	2.29
Other herbaceous	2.68	2.67	2.78	2.76	2.92	2.84	2.84	2.70	2.77	2.58	2.26
Fruits	2.29	2.29	2.32	2.35	2.35	2.53	2.63	2.63	2.98	2.90	2.54
Citrus	2.67	2.65	2.71	2.69	2.80	2.88	3.06	3.06	3.49	3.24	2.49
Treenuts	2.59	2.58	2.65	2.62	2.70	2.80	3.00	3.19	3.38	3.22	2.54
Olive	2.69	2.69	2.80	2.77	2.90	2.91	3.12	3.11	3.49	3.21	2.19
Grapevine	2.38	2.38	2.43	2.42	2.51	2.55	2.62	2.59	3.07	2.89	2.22

495

#### 496 *3.4 SOC simulations*

497 The simulated maximum SOC level in the total studied area was 1.19 Tg C in 1900, declining  
 498 throughout the 20<sup>th</sup> century, with a minimum in 2000 at 0.96 Tg C (Fig. 6a). The estimated  
 499 equilibrium SOC reached a minimum at 0.74 Tg C in 1980, and in 2008 it was still 18%  
 500 lower than in 1900 (Fig. 6b).



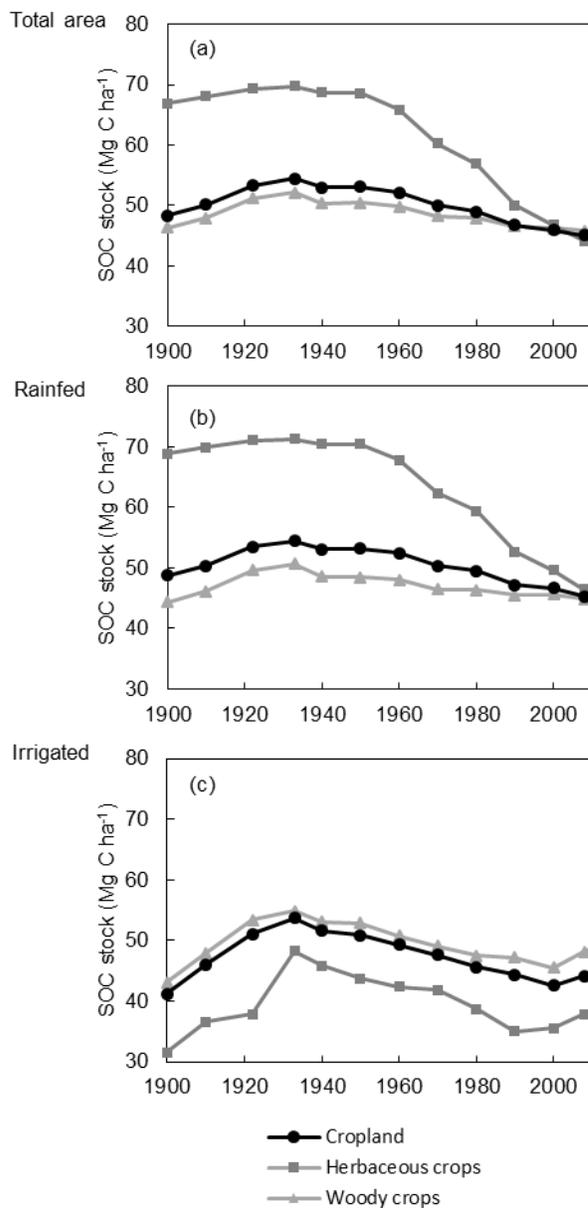
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502 **Fig. 6.** Simulated total SOC stock (a) and SOC stock at equilibrium (b) at 30 cm depth for the total studied area.

503

504 In the general analysis of the cultivated cropland area of Spain (Fig. 7a), there was a first  
505 period of growth in SOC from 1900 to 1933, partially associated to the increase in soil C  
506 inputs, but mainly to the expansion of cropland over non-cropland. This is, part of the  
507 increase was only apparent, as the new cropland area included previously non-cropped land  
508 with a higher SOC content. Cropland SOC fell steadily after this peak, reaching its minimum  
509 at 45.1 Mg C ha<sup>-1</sup> by 2008. Fig. 7a also reveals the contrasting trends between herbaceous  
510 and woody crops: while SOC stocks declined 12% from 1933 to 2008 in herbaceous crops,  
511 the decrease was 37% for woody crops. The observed SOC patterns for rainfed crops (Fig.  
512 7b) were similar to the general trends in cropland, but they were markedly different for

513 irrigated crops (Fig. 7c). Under irrigation, SOC levels started recovering by 2000, and they  
514 were lower for woody crops than for herbaceous crops.



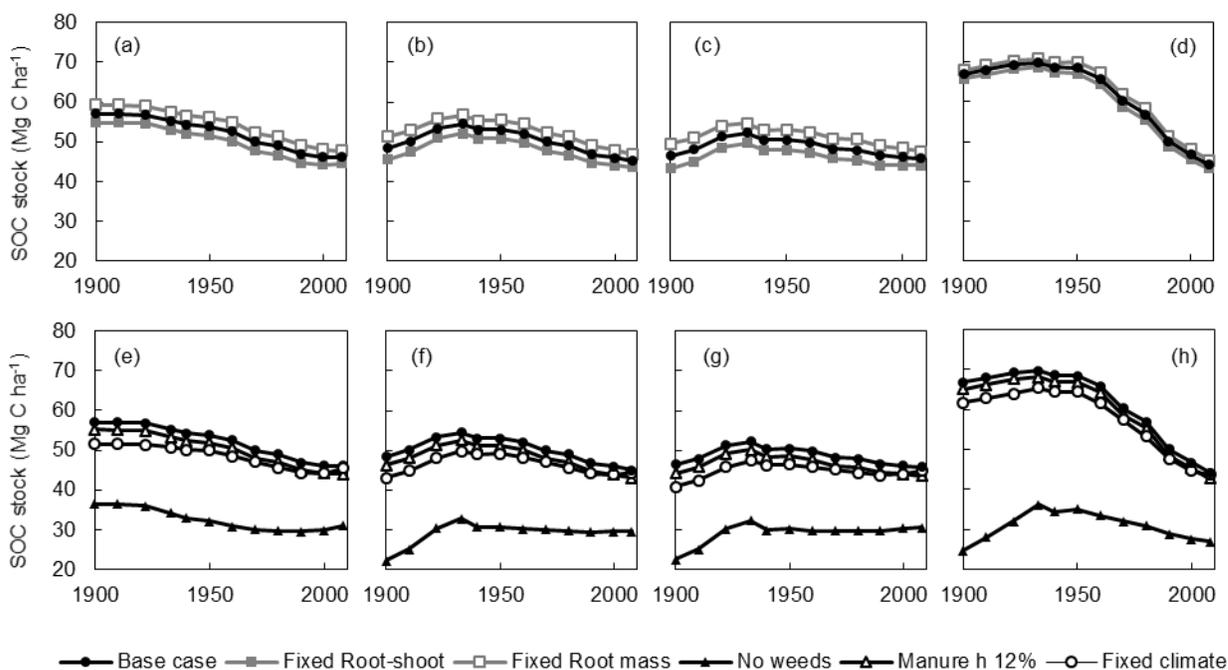
515

516 **Fig. 7.** Simulated SOC stocks (at 30 cm depth) per hectare for total (a), rainfed (b), and irrigated (c) cropland in Spain.

517

518 *3.5 Sensitivity analysis*

519 Most of the scenarios in the sensitivity analysis showed SOC values similar to those of the  
 520 base scenario. The root biomass calculation method (Fig. 8a-d) had a very limited influence  
 521 on SOC values, with slightly lower estimates for the fixed root-shoot approach, and higher  
 522 for the fixed root mass approach. The observed decline in SOC from 1900 to 2008 in the total  
 523 studied area was similar for the fixed root:shoot approach and for the fixed root mass  
 524 approach (Fig. 8a). The differences between the crop root estimation approaches were higher  
 525 for herbaceous crops (Fig. 8c) than for woody crops (Fig. 8d), reflecting the lower share of  
 526 crop roots in the NPP of woody crops.



527

528 **Fig. 8.** Sensitivity analysis of some key methodological choices affecting SOC in the total studied area of Spain (a, e),  
 529 total cropland (b, f), herbaceous crops (c, g) and woody crops (d, h). Studied scenarios include the base case (the  
 530 simulation performed in the rest of the paper), Fixed root-shoot (fixed crop-specific root:shoot ratios for the calculation of  
 531 root biomass), Fixed root mass (Fixed crop-specific root biomass values), No weeds (weeds are excluded from the model),  
 532 Manure h (manure humification coefficient is changed), and Fixed climate (climate parameters are maintained constant  
 533 along the simulation period)

534

535 Applying a manure humification coefficient of 12% hardly affected SOC stocks, leading to  
536 just 2-8% lower SOC estimates than in the Base scenario (Fig. 8e-h), with an estimated total  
537 SOC decline 5% higher than in the Base scenario.

538 Fixed climate parameters instead of decadal averages led to 10% lower total SOC stock at  
539 the beginning of the 20<sup>th</sup> century, a difference that was progressively reduced up to 2% in  
540 2008 (Fig. 8e). This implies that temperature increase was responsible for the loss of ca. 4.8  
541 Mg C ha<sup>-1</sup> in the total studied area during the 1900-2008 period. For the whole period, the  
542 average rate of decline due to changes in climate was 0.04 Mg CO<sub>2</sub>e ha<sup>-1</sup> yr<sup>-1</sup>, but it  
543 progressively increased from 0 in the 1900-1910 period to 0.13 Mg CO<sub>2</sub>e ha<sup>-1</sup> yr<sup>-1</sup> in the  
544 2000-2008 period.

545 Weed biomass was the factor with the highest influence on SOC estimates (Fig. 8e-h).  
546 Despite a reduction in SOC in the total area is also observed when weeds were excluded (Fig.  
547 8e), the total amount of SOC stored was less than half of that of the Base scenario at the  
548 beginning of the study period. This is mainly due to the major contribution of weeds to total  
549 C inputs of traditional cropping systems (Fig. 5). SOC remained stable for cropland in the  
550 scenario without weeds in the 1940-2008 period, a markedly different pattern compared with  
551 all the other scenarios.

552

#### 553 **4. Discussion**

554 This study depicts the evolution of SOC stock in Spanish cropland as a complex story  
555 involving shifting trends driven by changes in land uses, management and climate. In turn,  
556 these changes are determined by historical events in Spain and the world. The results shed

557 light on the implications of the transition from an organic to an industrial socio-metabolic  
558 regime on SOC dynamics, and on the potential and limitations of traditional organic  
559 management practices to increase SOC storage in modern Mediterranean agroecosystems.

#### 560 *4.1 Historical periods associated to the observed trends*

561 Four distinct historical periods help explaining the observed trends in SOC stocks. In the first  
562 period (1900-1936), both cropland area expansion and intensification took place within the  
563 traditional organic agriculture model. The result was a decline in SOC stocks considering the  
564 total studied area (Fig. 6a), as the expansion of cropland dominated over the increase in  
565 cropland soil C inputs (Fig. 5) in a context of stable SOC decomposition rates (Table 3).

566 The second period (1936-1955), was characterized by low yields (Fig. 4) and cropland  
567 abandonment (Fig. 2, Soto et al., 2016) due to the Spanish Civil War and Franco's dictatorship  
568 autarky period. In this period, SOC stocks further declined because of a decrease in cropland  
569 NPP (Fig. 4) and soil C inputs (Fig. 5), which prevailed over cropland abandonment (Fig. 6).

570 During the third period (1955-1986), Franco's regime transitioned from autarky to liberal  
571 economic policies, combined with investment in irrigation infrastructure, promoting a rapid  
572 adoption of industrial inputs. This policy was further developed after the onset of democracy  
573 in 1978. This was the most intensive stage of the industrialization process, in which external  
574 energy inputs increased 11-fold (Guzmán et al., 2017). This period shows the highest SOC  
575 decrease rate (Fig. 6), which was mainly related to declining soil C inputs, particularly from  
576 weeds (Fig. 4), combined with increasing SOC decomposition rates (Table 3) due to  
577 irrigation expansion and soil weed-cover loss in woody crops. The decline in cropland SOC  
578 observed after the mid-20<sup>th</sup> century is in line with field observations in England and Wales

579 from 1978 to 2003 (Bellamy et al., 2005) or Belgium from 1955 to 2005 (Goidts et al., 2009),  
580 and with modeling studies in Europe from 1951 to 1965 (Ciais et al., 2011). In Belgium,  
581 changes in the agricultural practices and in the precipitation regime were identified as the  
582 major causes of cropland SOC loss (Goidts et al., 2009). In the European simulation, the  
583 decrease was mainly linked to the shifting from organic to mineral fertilizers (Ciais et al.,  
584 2011). In the US Great Plains, the soil shifted from being a CO<sub>2</sub> source to a C sink during  
585 this period (Evrendilek and Wali, 2001, Parton et al. 2015), mainly due to the increase in  
586 residue production and soil application.

587 The last period (1986-2008) started with the entrance of Spain in the European Union,  
588 strengthening the market orientation of its agricultural sector but also incorporating  
589 environmental policies, such as restriction of residue burning. This period was also  
590 characterized by the relative abandonment of cereals straw use for animal feeding and  
591 bedding, supported by imported biomass (Soto et al., 2016). SOC stocks at equilibrium, and  
592 ultimately simulated SOC, progressively increased (Fig. 6), mainly driven by: (i) growing  
593 crop residue soil C inputs (Fig. 5), due to a combination of increasing residue production,  
594 decreasing residue field burning, and a stabilization of residue use, and (ii) diverging  
595 trajectories in SOC decomposition rates (Table 3), which grew in some areas due to rising  
596 temperatures, but decreased in other due to the expansion of drip irrigation and cropland  
597 abandonment. This land sparing process improved equilibrium SOC levels compared to those  
598 of the 1970s, but not to those of the early 20<sup>th</sup> century. The observed non-linear trend in SOC  
599 stocks suggest that it is not intensification per se which drives SOC changes, but the way in  
600 which this intensification is implemented. In Spain, as in other areas (e.g. Firbank et al. 2013),

601 agricultural intensification is currently getting more sustainable, but there is still a very large  
602 potential for improvement (González de Molina and Guzmán, 2017).

#### 603 *4.2 Comparison of simulated SOC with field studies*

604 We have found no historical data to validate our simulations of SOC trends in Spanish  
605 cropland. However, there are some published estimations of SOC contents in the last decades  
606 that can be used for comparison. Rodríguez-Murillo (2001) estimated SOC contents in  
607 Spanish cropland based on soil surveys and found slightly higher SOC contents in irrigated  
608 than in rainfed areas, similar to our simulation around 1980. However, the variability in the  
609 data was very high, probably due to the large range of soil depths (up to 1 m) and time points  
610 (1960-1995) included, hindering the comparability with our results. In another field  
611 assessment of Spanish soils, Romanya and Rovira (2011) found higher SOC values for  
612 irrigated cereals than for rainfed cereals in Mediterranean alkaline soils, a similar pattern than  
613 the one observed in this study for the whole cropland area in 2008. In the case of olives and  
614 nuts, Romanya and Rovira (2011) observed similar SOC contents for both categories,  
615 whereas we found lower SOC values under irrigation than in rainfed areas. This may indicate  
616 an under-estimation of SOC in irrigated woody crops in our assessment, probably related to  
617 an over-estimation of water inputs in these systems.

618 The most appropriate comparison of our results with measured values can be done using the  
619 comprehensive assessment of Rodríguez-Martín et al. (2016). Simulated SOC levels in year  
620 2008 were 0.3 Mg C ha<sup>-1</sup> lower for herbaceous crops and 5.3 Mg ha<sup>-1</sup> higher for woody crops  
621 than the average values estimated from field measurements. Both simulated data, however,  
622 were within the confidence intervals of the data in Rodríguez-Martín et al. (2016).

623 Equilibrium SOC values modeled for non-cropland, ranging 69-90 Mg C ha<sup>-1</sup> (data not  
624 shown), were within the range of values reported by Rodríguez-Martín et al. (2016) and  
625 Doblas-Miranda et al. (2013), based on field measurements. Because of these relatively high  
626 SOC values, there was a strong response of SOC to abandonment, which has also been  
627 observed in experimental studies in Spain (Segura et al., 2016).

#### 628 *4.3 Factors affecting the observed changes in NPP, soil C inputs and SOC*

##### 629 *General considerations*

630 The limited growth in NPP can be partially attributed to climate constraints, but it has also  
631 been linked to the degradation of the agroecosystem functions caused by industrialization  
632 (Guzmán et al., 2017), which may include the observed decline in SOC and the effect of  
633 anthropogenic climate change. Ciais et al. (2010) found that rainfall changes consisting on  
634 dryer springs and wetter autumns contributed to C losses in western Mediterranean regions.  
635 These authors also estimated that atmospheric CO<sub>2</sub> increase was responsible for a 10% NPP  
636 increase in European arable lands, indicating that a large part of the NPP increase that we  
637 observed could be attributed to CO<sub>2</sub> enrichment. Another important factor in the NPP and  
638 SOC trends was cover crop loss in woody cropping systems. The situation observed in Spain  
639 contrasts with that of Italy, where cover crops in woody crops have not declined. Farina et  
640 al. (2017) found higher SOC values for woody crops than for herbaceous crops in Southern  
641 Italy, which was associated to the high contribution of cover crops to soil C inputs, as we  
642 observed for the first half of the 20<sup>th</sup> century in Spain.

643 The small disagreement with measured data from Rodríguez-Martín et al. (2016) for woody  
644 crops could be due to a number of factors not accounted for in the model, such as previous

645 management history, soil drying and wetting cycles, erosion, increases in atmospheric CO<sub>2</sub>,  
646 or management practices such as tillage. Drying and wetting cycles are an important factor  
647 controlling SOC dynamics in Mediterranean environments (Jarvis et al. 2007), but they may  
648 have a limited effect on annual SOC changes (Borken and Matzner, 2009). Soil C loss by  
649 erosion in Mediterranean woody cropping systems was enhanced by a factor of 2-40 by the  
650 absence of soil cover (Gomez et al., 2011), which can result in a loss of 1-12 Mg C ha<sup>-1</sup> over  
651 a 50-year period. Vanwallegen et al. (2011) found that soil loss increased at least 3-fold from  
652 1900-1970 to 1970-2000 in olive groves in Southern Spain. The strong increase in  
653 mechanical traction in the 1950-1980 period (Guzmán et al., 2017) was probably related to  
654 an increase in tillage intensity and depth. After 1990, conservation tillage practices expanded  
655 up to more than 3 Mha in 2010, possibly promoting SOC storage in these areas (González-  
656 Sánchez et al., 2012). However, the net effects of these changes on SOC is highly uncertain,  
657 as they could be offset by changes in soil layers below the 30 cm depth limit of our model.

### 658 *Irrigation*

659 The results stress the key role of irrigation in the productivity of Mediterranean systems  
660 (Wriedt et al., 2009), following the global trend (Ozdogan 2011), but also in soil C inputs  
661 and SOC decomposition rates, both of which roughly doubled with irrigation (Fig. 5, Table  
662 3). These opposing effects resulted in mixed effects on SOC, in line with studies reporting  
663 either a decrease (Condrón et al., 2014, Nunes et al., 2007) or an increase (e.g. Montanaro et  
664 al., 2009) in SOC associated to irrigation. Additionally, the results suggest that drip irrigation  
665 was responsible for a substantial drop in C mineralization rates, due to water limitation. Drip  
666 irrigation, however, could also reduce the root:shoot ratio when compared to furrow  
667 irrigation (Araujo et al., 1995). Therefore, there is a need for experimental studies specifically

668 addressing the net effect of drip irrigation on SOC, especially given its interaction with other  
669 GHG (Cayuela et al., 2017, Sanz-Cobena et al., 2017). Irrigation should also be assessed  
670 considering its broader impacts on the water cycle, as its expansion has severely affected  
671 freshwater ecosystems in most Mediterranean and semiarid basins (Romero et al., 2016,  
672 Jaramillo et al., 2015) and may lead to changes in rainfall regimes (Lo and Famiglietti, 2013).

### 673 *Weeds*

674 The outcomes of the simulations and the sensitivity analysis reveal the prominent role of  
675 weed biomass in the changes in cropland NPP and SOC related to agricultural  
676 industrialization. The reduction in weeds due to herbicide expansion was offset by crop  
677 biomass growth in the case of herbaceous crops, but not of woody crops. Cover crops in  
678 woody cropping systems have been widely acknowledged as an effective soil C-building  
679 practice under Mediterranean conditions (Gonzalez-Sanchez et al., 2012, Aguilera et al.,  
680 2013, Vicente-Vicente et al., 2016). Furthermore, decreases in weed biomass production  
681 have been associated to lower biodiversity of weeds (Alignier et al., 2017) and wild  
682 heterotrophs (Rundlöf et al., 2008, Gabriel et al., 2013).

### 683 *Crop residues*

684 The increase in harvest indices was another important factor of the initial reduction in  
685 cropland soil C inputs during Spanish agriculture industrialization. Sinclair (1998) argued  
686 that low harvest indices in traditional systems could allow for higher biomass production  
687 under low N availability, being functional in a context in which straw had a high value.  
688 Currently, the search for renewable energy sources and soil C sequestration have made straw  
689 production desirable again (e.g. Lorenz et al., 2010). High yields can indirectly reduce GHG

690 emissions and other environmental impacts (e.g. Bennetzen et al., 2016), so breeding efforts  
691 should focus on dual-purposes varieties characterized by a high residue production without  
692 decreasing crop product yield (Lorenz et al., 2010). The focus on residue production may  
693 make even more sense in a context of stagnating yields in Europe (Wiesmeier et al., 2015).  
694 In Mediterranean organic farming systems, traditional crop varieties could be able to perform  
695 as well as modern varieties in terms of grain yield (Carranza et al., 2017), although this has  
696 not been observed under temperate climate (Konvalina et al., 2014).

#### 697 *Roots*

698 The estimated root biomass production values and root:shoot ratios of the major herbaceous  
699 crops are within the ranges of the global revision by Mathew et al. (2017). Roots represented  
700 the majority of C inputs across most of the crop-management categories and time-periods,  
701 emphasizing the need to include roots in cropland C assessments. In a global change context,  
702 and particularly in low-productivity environments such as many Mediterranean systems,  
703 promoting root growth may help simultaneously increasing yields and SOC storage (Paustian  
704 et al., 2016), while improving water use (Bodner et al. 2015).

705 The contribution of manure to soil C inputs was very limited even in a situation of strong  
706 growth of the livestock herd (Soto et al. 2016), which by 2007 consumed half of the feed  
707 protein from abroad (Lassaletta et al., 2014). This is related to the decrease in manure C:N  
708 ratios in modern livestock farming, even if changes due to animal diet composition, which  
709 could be significant (Yamulki, 2006) were not considered.

#### 710 *Climate*

711 Temperature rise gradually became the main driver explaining the observed decrease in SOC,  
712 representing a positive feedback to climate change in this Mediterranean area. The negative  
713 impact of climate change on SOC levels is particularly worrisome given the low SOC content  
714 of Mediterranean soils. Modeling studies suggest that yields could also be negatively affected  
715 by climate change in Mediterranean areas (Bindi and Olesen, 2011), and accelerate  
716 phenology driven by warming could decrease NPP (Ventrella et al., 2012), which would  
717 further reduce C availability for soil application. These trends warn us about the need for  
718 substantial changes in management practices in order to maintain soil quality in a global  
719 warming context.

720

## 721 **5. Acknowledgements**

722 This work was supported by the Social Sciences and Humanities Research Council of Canada  
723 [SSHRC 895-2011-1020], and the Ministerio de Economía y Competitividad of Spain  
724 [HAR2015-69620-C2-1-P]. The authors want to thank Dr. Luis Lassaletta for useful  
725 comments on previous versions of this manuscript.

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