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1 Nitrogen dynamics in cropping systems under Mediterranean climate: a systemic 2 analysis

3
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16 Abstract

17 Worldwide, Mediterranean cropping systems face the complex challenge of producing enough
18 high-quality food while preserving the quantity and quality of scarce water for people and
19 agriculture in the context of climate change. While good management of nitrogen (N) is
20 paramount to achieving this objective, the efficient strategies developed for temperate systems
21 are often not adapted to the specificities of Mediterranean systems. In this work we combined
22 original data with a thorough literature review to highlight the most relevant drivers of N
23 dynamics in these semiarid systems. To do so, we provide an analysis at nested scales combining
24 a bottom–up approach from the field scale with a top–down approach considering the agro-food
25 system where cropping systems are inserted. We analyze the structural changes in the agro-food
26 systems affecting total N entering the territory; the contrasting response of yields to N availability
27 under rainfed and irrigated conditions in a precipitation gradient; the interaction between N
28 management and climate change adaptation; the main drivers affecting the release of Nr
29 compounds (NO₃⁻, NH₃, NO, N₂O) as compared with temperate systems; and finally, the behavior
30 of N once exported to highly regulated river networks. We conclude that a sustainable N
31 management in Mediterranean cropping systems requires the specific adaptation of practices to
32 the particular local agroenvironmental characteristics with special emphasis on water availability
33 for rainfed and irrigated systems. This approach should also include a systemic analysis of N inputs
34 into the territory that are driven by the configuration of the agro-food system.

1. Background

The Mediterranean biome, characterized by its climate with warm, dry summers and mild, wet winters, is located between 30° and 45° latitude and is present in five areas of the world, namely, the Mediterranean Basin, California (USA), Chile, South Africa, and Australia (Fig. 1) (Aschmann, 1973). Mediterranean-type ecosystems are biodiversity hotspots concentrating a disproportionately high level of endemism (Myers *et al.*, 2000; Vogiatzakis *et al.*, 2006), making this biome a global conservation priority (Olson and Dinerstein, 2003). The total area covers 2.6 million km² and in 2010 had a population density of 95 inh/km² (i.e., 310 M inhabitants).

The land area of the Mediterranean Basin accounts for 66% of the world Mediterranean biome. The sustainability of this region for people and ecosystems is at stake due to: (1) scarcity of water (García-Ruiz *et al.*, 2011), (2) significant land-use changes, notably urbanization and abandonment of pastoralism (Malek *et al.*, 2018; Aguilera *et al.*, 2020), (3) increasing air and water pollution (Sanz-Cobena *et al.*, 2017), (4) biodiversity loss (Sala *et al.*, 2000), and (5) accelerating impacts of climate change, as well as other drivers of change (Cramer *et al.*, 2018; Zampieri *et al.*, 2020), making this region a “climate change hotspot” (Giorgi, 2006).

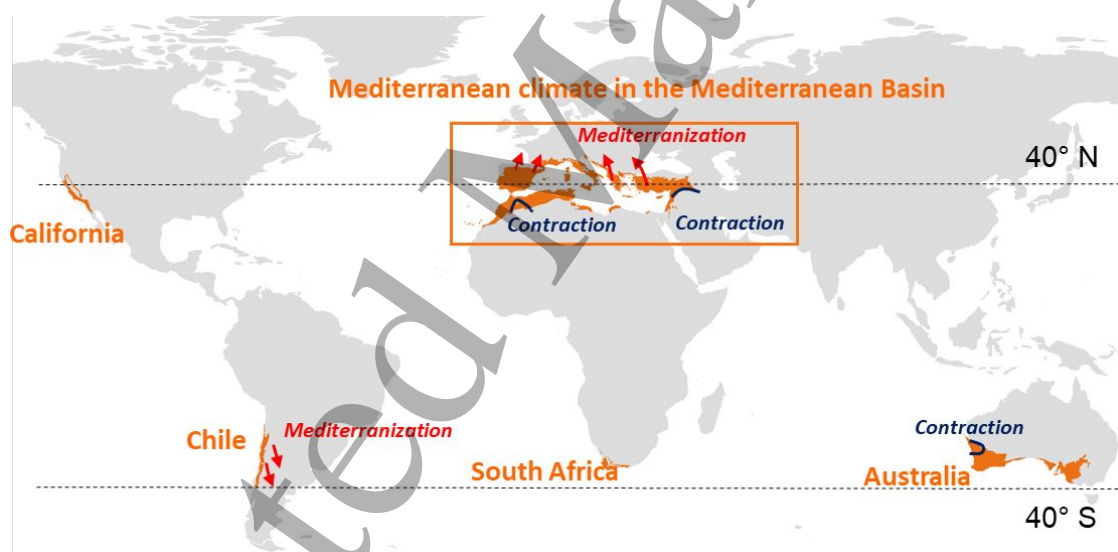


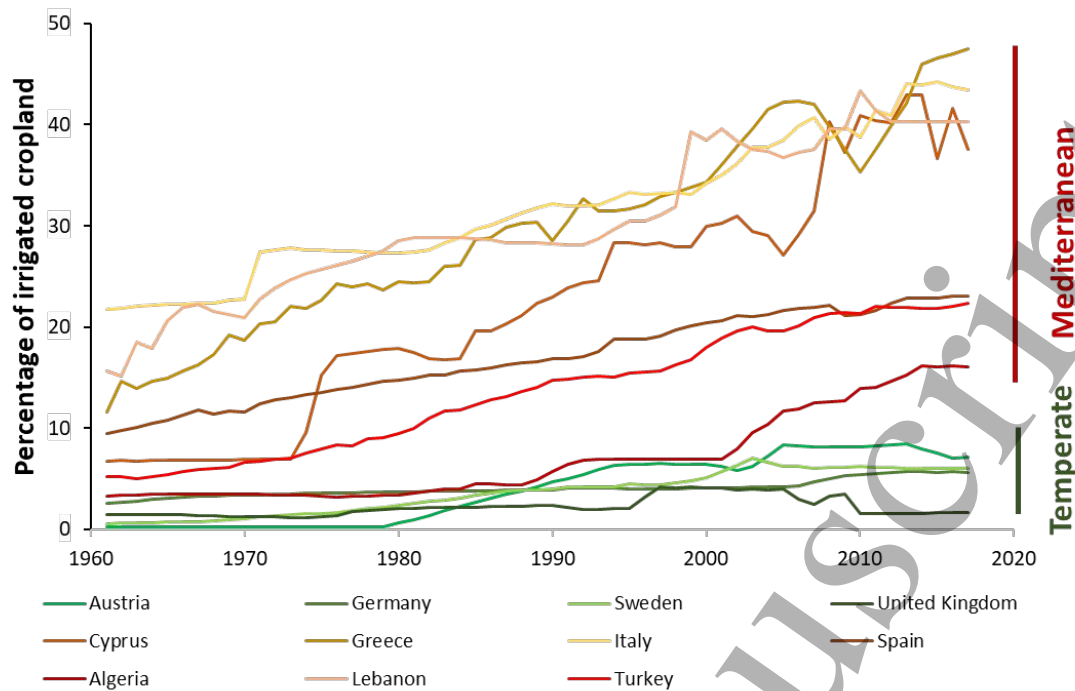
Figure 1. Global distribution of the Mediterranean biome (Olson *et al.* 2001) including potential areas of expansion (red arrows, “Mediterranization”) and contraction (blue lines). Adapted from Klausmeyer and Shaw (2009).

Due to global warming, some Mediterranean regions are expected to expand (“Mediterranization”) into currently temperate areas in South America and Europe, while contracting in others such as Australia and North Africa, giving way to a more arid climate (Klausmeyer and Shaw, 2009; Hernández, 2017). The key characteristic of the Mediterranean Climate is the asynchronicity between the maximum water availability (from autumn to spring) and the maximum irradiance and temperature, with important consequences for agricultural

1
2
3 60 production. It is also characterized by flash floods and droughts, which are projected to be
4
5 61 exacerbated (Michaelides *et al.*, 2018).
6

7 62 During thousands of years of evolution, agriculture in the Mediterranean Basin has been adapted
8
9 63 to environmental conditions of relative instability, rainfall variability, and summer water stress
10
11 64 through the selection of crops, varieties and rotations, as well as hydraulic infrastructures and
12
13 65 management practices, resulting in diverse and unique agricultural systems (Blondel, 2006). This
14
15 66 diversification permits the cultivation of rainfed crops such as cereals, legumes, oilseeds, and
16
17 67 some vegetables in winter, as well as of drought-resistant permanent crops such as almonds,
18
19 68 grapes, and olives. Efficient irrigation can increase crop yields from 10% to 90%, depending on the
20
21 69 annual water stress and the technology (Wriedt *et al.*, 2009). Irrigation also allows for the
22
23 70 cultivation and expansion of horticultural crops, maize, tobacco, and citrus fruits in these semiarid
24
25 71 areas. As a result, irrigation is a common practice in traditional Mediterranean cropping systems,
26
27 72 and it has expanded more than in temperate areas in the past 20 years (Fig. 2).

28
29 73 Climate change is affecting the entire planet, and Mediterranean climate zones are no exception.
30
31 74 As occurring in the entire planet (Klausmeyer and Shaw, 2009) climate change impacts
32
33 75 Mediterranean biomes (Famiglietti *et al.*, 2011), where it is expected to increase water scarcity
34
35 76 via sustained warming and decreasing precipitation, consequently leading to an increase in
36
37 77 evaporative demand and in the intensity, duration, and frequency of droughts (García-Ruiz *et al.*,
38
39 78 2011; Spinoni *et al.*, 2018). Without adaptation crop yields will likely be impacted (Iglesias *et al.*,
40
41 79 2011), (Zampieri *et al.*, 2020). Irrigation requirements may increase substantially with climate
42
43 80 change (Fader *et al.*, 2016), and therefore irrigation techniques will have to be adapted as water
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45 81 resources may turn out to be insufficient (Malek and Verburg, 2018).
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82
83 **Figure 2.** Evolution of cropland fraction equipped for irrigation in several Mediterranean countries (all of
84 them with than 50% of their agriculture under Mediterranean conditions) and a sample of temperate
85 countries in Europe (based on FAOSTAT, 2020).

86 Together with water, nitrogen (N) is the main limiting factor in agricultural production and must
87 be properly managed to sustain crop yields (Mueller *et al.*, 2012; Quemada *et al.*, 2020a).
88 Lassaletta *et al.* (2014a) showed that a robust hyperbolic relationship relates harvested yield,
89 expressed as N content and integrated over a whole crop rotation cycle, to average total annual
90 N inputs to soil, with a single parameter Y_{max} characterizing the pedo-climatic and technical and
91 agronomic context. This relationship implies that not all N applied to soil is recovered with the
92 harvested crop. Overall, N use efficiency (NUE) of the world cropping systems is less than 50%
93 (Bouwman *et al.*, 2013; Lassaletta *et al.*, 2014a; Bouwman *et al.*, 2017). Most of the N surplus, i.e.,
94 the amount of N input to soils not exported by harvested crops, is released to the environment),
95 where it undergoes a series of transformations, impacting different ecosystem compartments
96 through the so-called N cascade (Galloway *et al.*, 2003; Billen *et al.*, 2013). In agricultural systems,
97 reactive N (N_r , all nitrogen species except N_2) (Leach *et al.*, 2012) is preferentially emitted in the
98 form of: volatilized ammonia (NH_3), an aerosol precursor and soil-acidifying compound (Van
99 Damme *et al.*, 2018) that causes biodiversity losses when deposited (Ochoa-Hueso *et al.*, 2011);
100 nitrous oxide (N_2O), a potent greenhouse gas (Thompson *et al.*, 2019); and nitric oxide (NO), which
101 promotes the formation of tropospheric ozone (O_3) (Guardia *et al.*, 2017b). N can be also leached
102 as nitrate (NO_3^-), which is a threat to drinking water resources and freshwater biodiversity (van
103 Grinsven *et al.*, 2010) and promotes eutrophication of coastal waters (Garnier *et al.*, 2010). The
104 emission of these compounds imposes a high economic cost for society (Sutton *et al.*, 2013; Van

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3 105 Grinsven *et al.*, 2013) and their adequate management has been linked to nine of the United
4
5 106 Nations Sustainable Development Goals (Morsetto, 2019).

6
7 107 The total N inputs to the world's Mediterranean cropping systems in 2010 were 5.9 Tg N, most of
8
9 108 them in the form of synthetic fertilizers. 75% of this N is applied in the crops of the Mediterranean
10
11 109 Basin under Mediterranean climate (see the Supplements for a complete N budget of
12
13 110 Mediterranean cropping systems). Adequate crop management in the Mediterranean biome
14
15 111 involves the simultaneous control of water and N inputs (Sadras, 2005; Sadras *et al.*, 2016). Two
16
17 112 contrasting situations are experienced: rainfed systems subject to important spatiotemporal
18
19 113 variability, which complicates the adjustment of optimal fertilization doses (Ryan, 2008; Ryan *et al.*,
20
21 114 *et al.*, 2009; Quemada and Gabriel, 2016); and irrigated systems dealing in many cases with water
22
23 115 supply limitations and high risk of nitrate leaching (Causape *et al.*, 2006; Barros *et al.*, 2012; Malik
24
25 116 and Dechmi, 2019). Other intrinsic characteristics of the Mediterranean systems such as the high
26
27 117 temperatures and the low carbon content of soil (Aguilera *et al.*, 2013a; Rodríguez Martín *et al.*,
28
29 118 2016; Aguilera *et al.*, 2018) have a strong impact on the microbial activity that drives the form in
30
31 119 which N is emitted and the magnitude of the flow from croplands to other compartments (Sanz-
32
33 120 Cobena *et al.*, 2017). N₂O emission factors, for example, are lower than those in temperate
34
35 121 systems (Cayuela *et al.*, 2017).

36
37 122 Sustainable N management, adapted to the particularities of the Mediterranean biome, depends
38
39 123 on knowledge of specific processes at the plot and farm scale (bottom-up approach), but also on
40
41 124 the understanding of the agro-food system that controls the new N entering a territory (top-down
42
43 125 approach). Organic manure may be efficiently recirculated at the regional scale, reducing the
44
45 126 demand for new fertilizers, while different agricultural commodities can be locally produced or
46
47 127 imported, all of which configure the structure of the N cycle (Billen *et al.*, 2014; Billen *et al.*, 2015;
48
49 128 Le Noë *et al.*, 2017; Mueller and Lassaletta, 2020). For example, in Spain, the input of new N into
50
51 129 the national territory (NANI, net anthropogenic N input) (Howarth *et al.*, 1996) has increased
52
53 130 fourfold during the past 60 years, contributing to important yield improvements but also to
54
55 131 significantly increased N pollution. Once N reaches the waterbodies, the high regulation of
56
57 132 freshwater resources for crop irrigation and drinking water supply can affect N dynamics in
58
59 133 Mediterranean watersheds. The fraction of runoff exported to coastal waters is lower than in
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61 134 temperate watersheds, increasing the risk for pollution of aquifers, rivers, and lakes (Bartoli *et al.*,
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63 135 2012; Romero *et al.*, 2016).

64
65 136 Worldwide, Mediterranean cropping systems face the complex challenge under the climate
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67 137 change context of producing enough high-quality food for an often increasing population,

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3 138 together with the need to preserve the quantity and quality of scarce water resources for people
4
5 139 and agriculture, while also reducing their impacts on air pollution. We present this review with
6
7 140 the overall aim of analyzing, from a systemic perspective, the particularities of N dynamics in
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9 141 Mediterranean cropping systems associated with the emission of Nr compounds. Our approach
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11 142 aims to extend from the plot or farm-scale to the agro-food systems that these cropping systems
12
13 143 are embedded in.

14 144 This study combines primary data with a literature review in order to provide an analysis at nested
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16 145 scales of (1) the evolution of the N cycle in agro-food systems at the regional scale including crop
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18 146 N budgets, (2) water and N interactions in contrasted irrigated and rainfed systems along a
19
20 147 precipitation gradient, (3) the implications of climate change adaptation for N management, (4)
21
22 148 the main drivers affecting the release of Nr compounds as compared with temperate systems, (5)
23
24 149 the behavior of N in Mediterranean watersheds, (6) the resulting policy recommendations for the
25
26 150 design of agro-environmental strategies tailored to Mediterranean conditions. The work mainly
27
28 151 focuses on the Mediterranean Basin, which forms a unique, conterminous and coherent territory,
29
30 152 suitable for developing a systemic analysis covering from the agro-food system to the river basin.
31
32 153 We postulate that the main lessons learnt for these semiarid systems managed under rainfed and
33
34 154 irrigated conditions are also relevant, not only for the rest of the world's Mediterranean areas but
35
36 155 also for the new areas undergoing "Mediterranization" due to climate change.

36 156 **2. Methods**

37
38 157 We analyzed the evolution of the N cycle in the agro-food system at the regional scale. Crop
39
40 158 production and pasture production are connected with livestock farming to supply food to the
41
42 159 local population, and also with the export or import of agricultural products to or from other
43
44 160 regions. Using N embedded in proteins or in N fertilizers as a marker, the structure of the agro-
45
46 161 food system is presented in a diagram showing N flows through the main transformation
47
48 162 compartments involved in the system, namely, crop and grass production, livestock breeding,
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50 163 food processing, international trade, and final human consumption (General Representation of
51
52 164 Agro-Food Systems, GRAFS) (Billen *et al.*, 2014; Le Noë *et al.*, 2017; Billen *et al.*, 2018).

53
54 165 To construct the GRAFS diagrams, and since some statistics were available at the country scale
55
56 166 only, we considered 11 out of the 22 countries in the Mediterranean Basin. In these countries,
57
58 167 more than 50% of the crop and grassland area is under Mediterranean conditions: Portugal, Spain,
59
60 168 Italy, Albania, Greece, Turkey, Cyprus, Israel, Tunisia, Algeria, and Morocco (Supplements Table
169 S1). Countries presenting a marked temperate-to-Mediterranean gradient (France) or with
170 incomplete observations (Palestine), or those with relevant data missing for other reasons (Syria

1
2
3 171 and Libya), were not included. In total, the selected countries comprise 91% of crops and
4 172 grasslands under Mediterranean climate in the Mediterranean Basin and 61% of crops and
5 173 grasslands under Mediterranean climate worldwide. The crop mix at country level was estimated
6 174 by clustering crop data from FAOSTAT into generic groups, namely, cereals, legumes, roots,
7 175 oilseed, horticulture, permanent crops, and others. N budgets in cropping systems of the
8 176 Mediterranean biome were calculated by combining spatially-explicit, crop-specific datasets on
9 177 crop and livestock distribution, yields and N inputs with country-level data from FAOSTAT (see a
10 178 full description of the procedure in the Supplements).

11 179 Water–N interactions were analyzed in Spain as a putatively representative Mediterranean
12 180 country with a strong precipitation gradient with cereal crops under rainfed and irrigated
13 181 conditions. Data on yearly crop-specific yield and N fertilization rates (synthetic and organic) were
14 182 provided by the Spanish Ministry of Agriculture for wheat cultivation, split into rainfed and
15 183 irrigated systems at the province level (40 Spanish provinces with Mediterranean climate) for 26
16 184 years (1990–2015). Yield instability during that particular period was calculated separately for
17 185 rainfed and irrigated conditions at the province level by means of the coefficient of variation (CV)
18 186 of crop yield in each province. Yearly precipitation data were obtained from the CRU global
19 187 dataset (Seager *et al.*, 2019). Irrigation water application was estimated for each crop in each
20 188 province at monthly time steps based on crop evapotranspiration, which was calculated as
21 189 described in (Vila-Traver *et al.*, 2021). Crop water requirements were calculated by subtracting
22 190 effective precipitation from crop evapotranspiration. Irrigation water application was calculated
23 191 by dividing crop water requirement by the water efficiency of each irrigation technology (Wriedt
24 192 *et al.*, 2009).

25 193 Regional crop nutrient budgets at the Spanish province level include synthetic and organic N
26 194 fertilizers, and atmospheric deposition, as inputs (N_{in}), and N extracted in the main harvest as
27 195 outputs (NY). Nitrogen use efficiency (NUE) corresponds to NY/N_{in} and N surplus to $N_{in} - NY$
28 196 (Zhang *et al.*, 2020). Averaged NY vs. N_{in} response curves were estimated for each province and
29 197 fitted by the one-parameter hyperbolic curve :

$$30 \quad 198 \quad NY = Y_{max} \cdot N_{in} / (N_{in} + Y_{max})$$

31 199 Where the parameter $Y_{max} = (N_{in} * NY) / (N_{in} - NY)$ represents the maximum yield at N saturation
32 200 conditions. This approach is commonly used in regional and large-scale approaches (Lassaletta *et*
33 201 *al.*, 2016; Mueller *et al.*, 2017; Mogollón *et al.*, 2018).

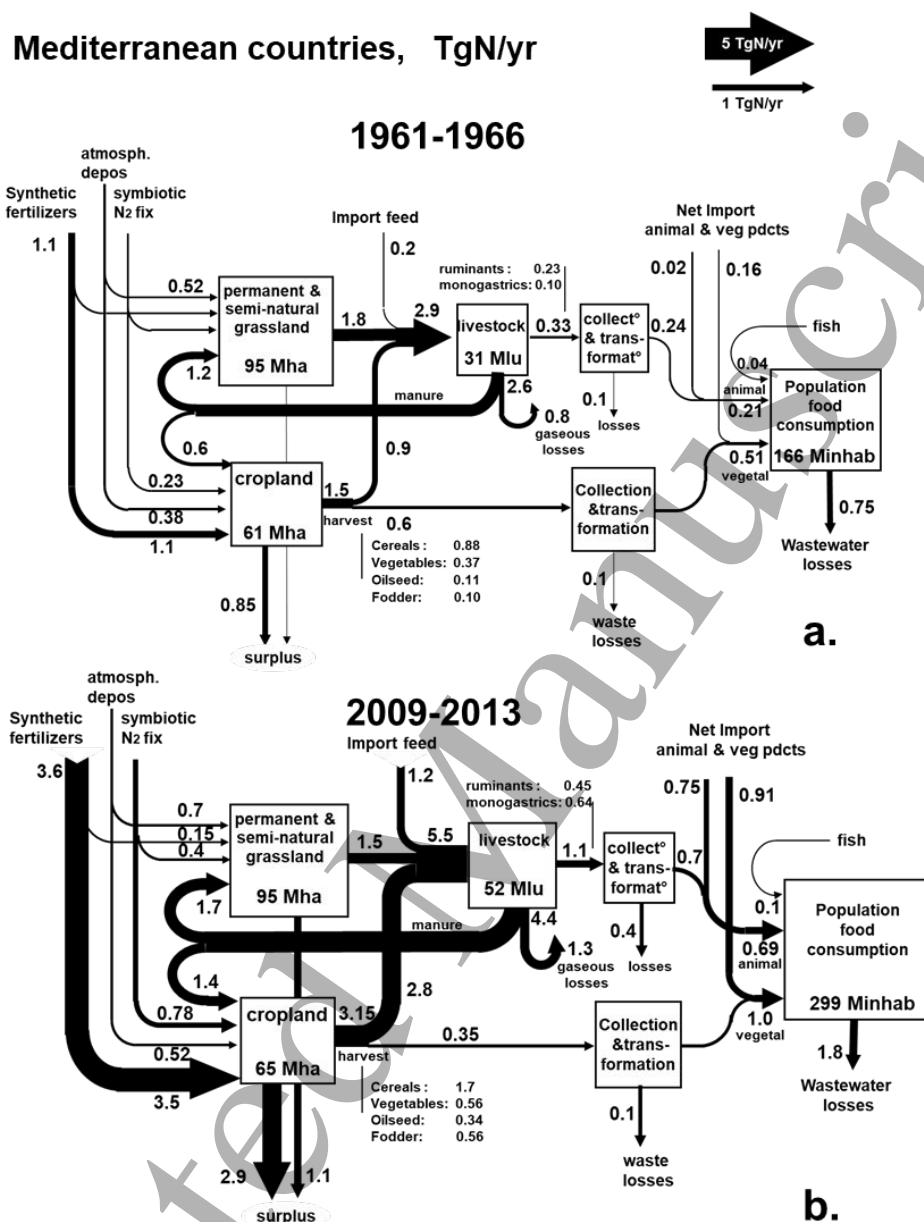
202 3. Trajectories of N use in the whole agro-food system in the Mediterranean Basin

203 The structure of the agro-food system for the periods 1961–1965 and 2009–2013 in the selected
204 Mediterranean countries is represented in diagrams showing N fluxes through the main
205 compartments. The components involved are crop and grass production, livestock breeding, food
206 transformation, international trade, and human consumption (Fig. 3). In the early 1960s,
207 Mediterranean countries as a whole (as defined in the Methods section) were close to self-
208 sufficiency for feeding their population of 166 million inhabitants, with a diet composed of 3.1
209 kg N/cap/yr of proteins from vegetal origin, and 1.5 kg N/cap/yr of animal proteins, including 0.2
210 kg N/cap/yr from fish. Livestock manure, together with symbiotic fixation of atmospheric N₂ and
211 atmospheric deposition of Nr, was the dominant form of N fertilizing input to cropland, even if
212 synthetic fertilizers already contributed significantly at that time. Livestock, dominated by
213 ruminants, thus played a major role by conveying N from permanent and seminatural grassland
214 to cropland soils. Imported feed was of limited significance, and approximately 60% of crop
215 production was dedicated to animal feeding. The remaining crop production was used for local
216 human nutrition together with animal production, such as meat and milk. Net imports of animal
217 and vegetable food products were minimal.

218 During the past 50 years, the population of the Mediterranean Basin countries increased by 80%
219 and changed its diet by increasing total protein ingestion by 30% (from 4.5 to 6.1 kg N/cap/yr),
220 while the share of animal proteins in this total rose from 32 to 43%. This transition has been more
221 evident in European Mediterranean countries and Israel than in the rest of the basin. On the one
222 hand, these changes made it possible that the protein consumption per capita did not fall below
223 WHO recommendations at any time. But these changes also indicate the gradual abandonment
224 of the so-called Mediterranean diet that was replaced by a “Western-type” diet that totally alters
225 the recommended relative proportions of each food group and negatively affects human health
226 (Blas *et al.*, 2019). This dietary change has profoundly impacted the structure of the entire agro-
227 food system, with a considerable intensification of animal breeding: livestock production
228 increased 3.5-fold, mostly in terms of monogastric animals feeding on grains.

229 Despite a concomitant intensification of cropping systems, increasing its use of synthetic N
230 fertilizers by a factor of 3, the production of forage and cereal was not enough to feed the
231 livestock. The rising demand for feed occurred in parallel to a change in the crop mix of the region.
232 The cultivation of cereals was reduced by 16% of the region’s total cropland area during the past
233 few decades. Only Morocco significantly increased cereal cropping areas. Legume crop surfaces
234 decreased in European countries but not in south and east Mediterranean countries. On the other

235 hand, cultivation of oilseed with potential allocation to animal consumption has almost doubled,
 236 although today its share is still too low to provide a sustained source of protein (5.2% of cropland).



237

238 **Figure 3.** Generalized representation of the Agro-Food System (GRAFS) of countries in the Mediterranean
 239 Basin with more than 50% of their agricultural land under Mediterranean climate (11 selected countries,
 240 see Methods Section), in 1961–1965 (a) and 2009–2013 (b)

241 Massive imports of feed, namely, soybean and maize from the Americas, were required for
 242 livestock production (Lassaletta *et al.*, 2014b; Leip *et al.*, 2015; Billen *et al.*, 2019). Also,
 243 considerable net imports of animal food and cereals were needed to feed the human population.

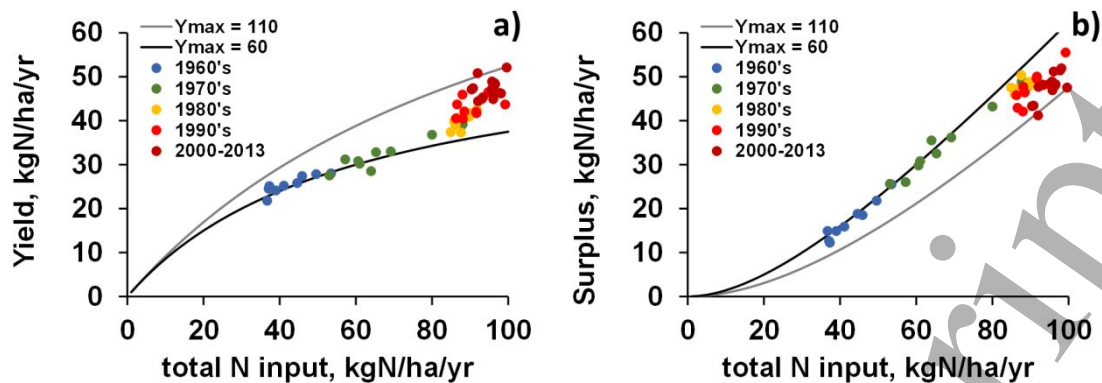
244 As a result, the external dependency of the agro-food system of Mediterranean countries, defined

245 as the ratio of net import of food and feed proteins to the local animal and human ingestion, rose

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3 246 from 6% in 1960 to 30% in 2013. This trend contributes to surpassing the carrying capacity of the
4
5 247 region by a factor of 2.5 (Galli *et al.*, 2015).
6

7 248 The trends in agro-food system components from both the demand side and the supply side
8
9 249 indicate massive temporal change (Fig. S2 Suppl.). The gradual increase of animal and human
10
11 250 consumption cannot be met by the increase of cropland yield and the stagnation of grassland
12
13 251 production, requiring recourse to increased importation. Thus, the level of external dependency
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15 252 paralleled this gradual increment (Fig. S3 Suppl.).
16

17 253 Yield of arable land increased by a factor of two over the past 50 years. This is largely the result
18
19 254 of increased N fertilization. The trajectory of N yield versus soil N inputs follows the same distinct
20
21 255 hyperbolic relationship with a Y_{max} of approximately 60 kg N/ha/yr until the end of the 1970s (Fig.
22
23 256 4). From this time onward, improvements in yield were obtained with much lower increases in
24
25 257 fertilization than in the first decades. Consequently, the surplus, i.e., the excess of total N soil
26
27 258 inputs over N in harvested products, increased until the early 1980s and then levelled off until the
28
29 259 present period. Crop NUE decreased from 60% in the early 1960s to 44% in the 1980s, and then
30
31 260 increased again up to 50%. There are several reasons to explain the changes observed in the
32
33 261 aggregated N crop yield and NUE. On the one hand, the expansion of irrigated land (see Fig. 2)
34
35 262 and other agronomic improvements such as better fertilization and irrigation techniques, as well
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37 263 as the development of improved crop varieties through breeding and selection, had a significant
38
39 264 effect on the observed yield increases. For example, wheat and tomato yields grew between two-
40
41 265 and threefold, and two- and fourfold, respectively. On the other hand, over-fertilization may have
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43 266 moved the system to the inefficient part of the yield vs. N input response curve. In addition to
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45 267 these agronomic factors, the aforementioned changes in the crop mix may have had an important
46
47 268 effect on the NUE of the system (Zhang *et al.*, 2015). High N input/N output systems such as
48
49 269 cereals were replaced by low N input/low N output systems such as permanent crops, systems
50
51 270 that have doubled in presence. This is linked to the specialization of Mediterranean agriculture in
52
53 271 vegetables, fruits, and olive oil for export to temperate European countries. Counterintuitively,
54
55 272 the cultivated area with high N input/low N output systems such as horticulture has been reduced
56
57 273 in European Mediterranean countries while their total production has increased, indicating high
58
59 274 intensification practices (e.g., greenhouses). By contrast, both the agricultural area and the
60
275 productivity of horticulture substantially increased in south and east Mediterranean countries.



276

277 **Figure 4.** Intensification and improvement of cropping practices of countries in the Mediterranean basin
 278 over the period 1961–2013: relationship between yield (in terms of protein content of harvested products
 279 per ha) and total N soil inputs (a) and corresponding surplus (N soil input in excess over harvested N).

280 Intensification of animal production, accompanied by an increase in the share of monogastric
 281 animals (representing 10% of the livestock units in 1961 and 27% in 2013), resulted in a limited
 282 improvement of aggregated protein conversion efficiency from 8% in the 1960s to 13% (at the
 283 herd level and considering edible protein only) in recent years. These NUE values aggregate many
 284 contrasting types of livestock systems. The most intensified systems have increased N efficiency
 285 significantly, gradually reducing the total demand for feed (expressed as dry matter) to produce
 286 1 kg of meat during the past decades and subsequently reducing the N excretion/kg meat during
 287 recent years (Lassaletta *et al.*, 2019; MAPA, 2020a, b). These systems strongly depend on
 288 imported feed, causing the externalization of N impacts (Bai *et al.*, 2014; Quemada *et al.*, 2020a)
 289 and increasing the input of new N embedded in the feed via international trade.

290 Alongside the intensification of both livestock and crop farming, environmental N losses increased
 291 considerably, as shown by the rise in Net Anthropogenic Nitrogen Input (NANI) from 620 to 1900
 292 kg N/km²/yr (Fig S3. Suppl.). While the input of synthetic N fertilizers was stabilized from 1985,
 293 the net import of food and feed steadily increased and today accounts for 2 Tg N/yr, i.e., 50% of
 294 synthetic fertilizers. This massive and increasing amount of feed largely transformed into manure
 295 represents a huge challenge for cropping and livestock systems. As a result of these changes,
 296 agricultural surplus and gaseous N emission from livestock breeding have also increased. Clearly,
 297 the trend toward losing self-sufficiency of the agro-food system was accompanied by the opening
 298 of nutrient cycles. The period from 1960 to the 1980s experienced the fastest increase in
 299 environmental losses, mostly from cropland. A stabilization of these losses was observed in the
 300 latest period in parallel to NUE increases.

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4. The relevance of the interaction between N and water exemplified in Spanish regions

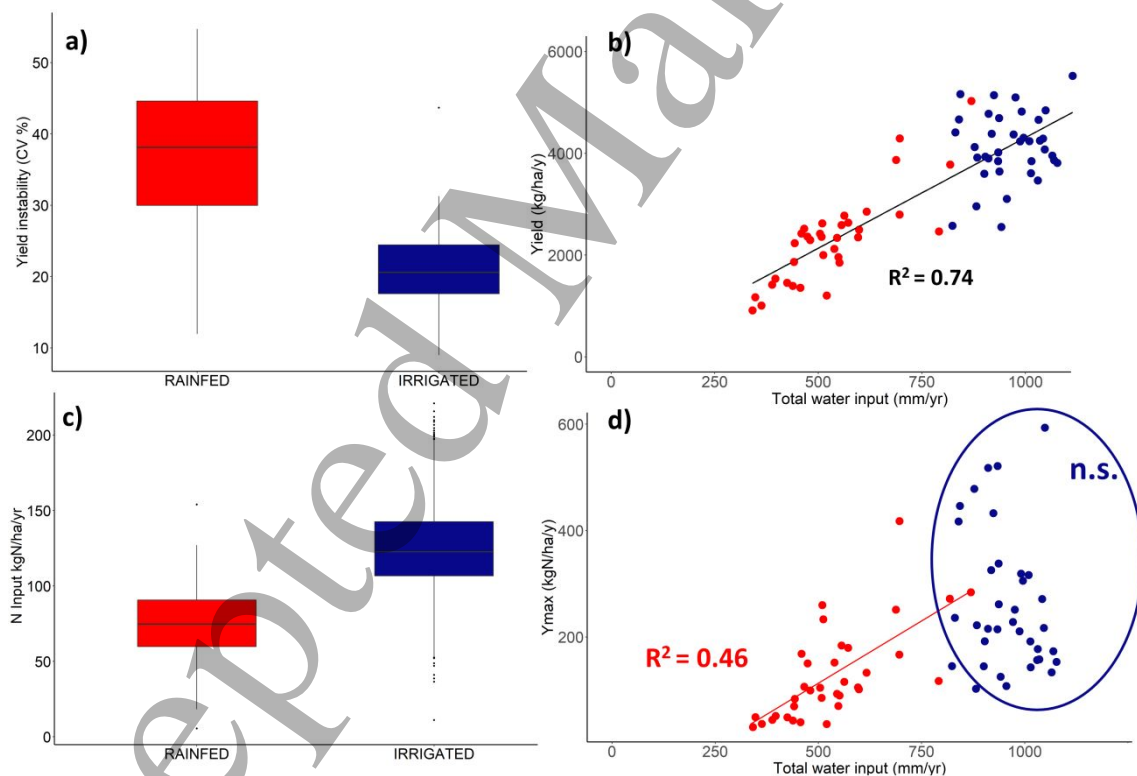
The crop yield response to water and N availability was stated as a paradigm of co-limitation, the simultaneous limitation of plant productivity by multiple resources. This co-limitation has been well studied in cereal crops of Mediterranean-type environments in Australia and Europe (Cossani and Sadras, 2018). Prolonged water stress leads to yield reduction and decreased N uptake as well as NUE, whereas insufficient N may limit water response and water use efficiency (Savin *et al.*, 2019). The implication of co-limitation is that strategies aiming to maximize crop response to both resources should ensure that water and N are equally available (Quemada and Gabriel, 2016; Hochman and Horan, 2018). Also, water is one of the main drivers of N losses at a cropping systems level, as excessive water input leads to leaching losses and favors denitrification, thereby enhancing N₂O emissions (Section 6). As a consequence, proper management of the interaction between NUE and water use efficiency (WUE) is required in many cropping systems in order to reach a compromise between productivity and environmental sustainability.

Water availability is thus a limiting factor in Mediterranean areas that determines the pattern of land use (López-Bellido, 1992). High solar radiation and extended frost-free periods make these regions capable of attaining high crop yields. When water is not available, agricultural systems are either adaptations of woody crops to xeric conditions (i.e., olive orchards, vineyards, oak *dehesas*) or low-productivity rainfed arable crops (i.e., barley, wheat, sunflower, leguminous grain) subject to high yield variability. N application in these rainfed systems tends to be opportunistic depending on the weather forecast, and the NUE attained is determined by the adjustment of fertilization to the actual crop growth limited by rainfall (Sadras *et al.*, 2003). Water application to compensate for the frequent spring and summer droughts contributes not only to stabilized productivity but also to crop diversification, providing the opportunity for more complex rotations including highly profitable crops (i.e., horticultural) usually characterized by a demand for large amounts of inputs including N. However, this increase in productivity is not always associated with an enhancement of NUE because crops are frequently over-fertilized or over-watered in order to achieve high yields (Quemada *et al.*, 2013; Salazar *et al.*, 2019), thereby generating a high risk of N leaching (Section 6). Therefore, increasing WUE and NUE in the Mediterranean is challenging and requires the development of specific practices adapted to rainfed and irrigated systems.

The analysis of the N budgets for wheat cultivation under rainfed and irrigated conditions at the province scale in Spain clearly illustrates the agro-environmental implications of N–water co-limitation. This analysis includes information on N input and wheat yields for 40 Spanish provinces (administrative units ranging 5000–20,000 km²) over 26 years (1990–2015). These provinces have

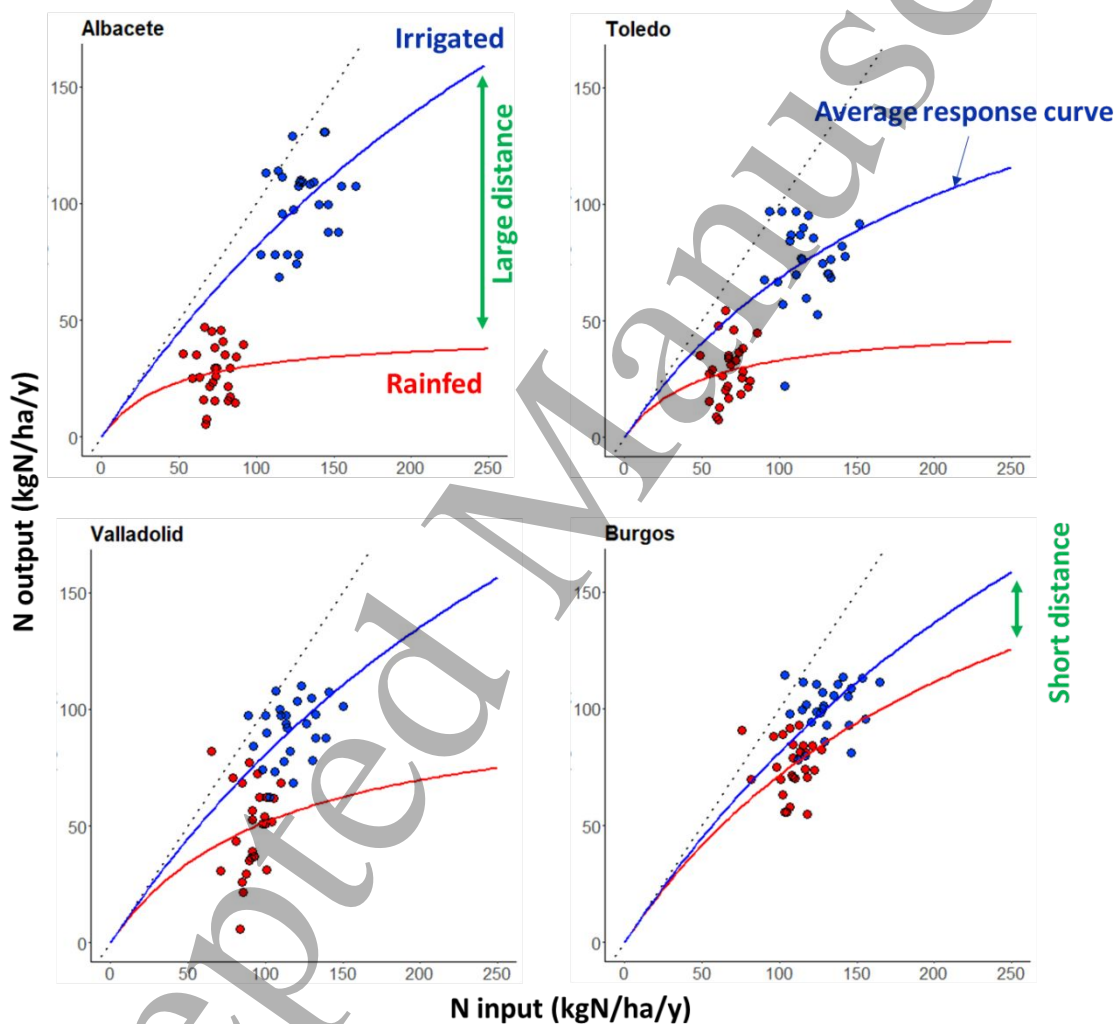
336 Mediterranean climate but different levels of annual precipitation (province range 341-869
 337 mm/yr), temperature (province annual mean temperature range 11-18 °C) and latitude (province
 338 range 36.5-42.8° latitude N). Average yield per province ranged from 0.9-2.5 t/ha/yr for rainfed
 339 systems and 2.6-5.5 t/ha/yr for irrigated systems.

340 Yield instability was significantly lower in irrigated than in rainfed systems in all of the provinces
 341 (Fig. 5a). Thus, irrigation consistently homogenized yields across Mediterranean regions. We
 342 found a strong relationship between mean total water inputs (precipitation and irrigation) and
 343 mean yields in rainfed and irrigated systems ($R^2 = 0.79$), explaining the strong effect of water on
 344 yields (Fig. 5b). In response to the higher yields expected in irrigated systems, N fertilization rates
 345 are significantly higher (Fig. 5c). Water–N interactions in Mediterranean rainfed systems were
 346 revealed through the robust relationship ($R^2 = 0.46$) between precipitation and Y_{\max} in rainfed
 347 systems (Fig. 5d), new water inputs through precipitation increase Y_{\max} and therefore the capacity
 348 of the crop to respond to new N additions. In irrigated systems the relationship is lost; there is no
 349 water limitation and other factors affect crop response to N additions.



350
 351 **Figure 5** Yield instability of wheat in rainfed and irrigated systems for the 40 Spanish provinces under
 352 Mediterranean conditions (1990-2015) (a); relationship between mean water input (precipitation +
 353 irrigation) and mean yield for each of the 40 provinces for rainfed and irrigated systems (b); mean N fertilizer
 354 input (1990-2015) of the 40 provinces for rainfed and irrigated systems (c); and relationship between total
 355 water input and Y_{\max} (the parameter of the hyperbolic response curve that relates yield and N inputs
 356 representing maximum yield at saturating N rates), $R^2 = 0.46$ for rainfed systems, and no significant (n.s.)
 357 relationship for irrigated crops (d).

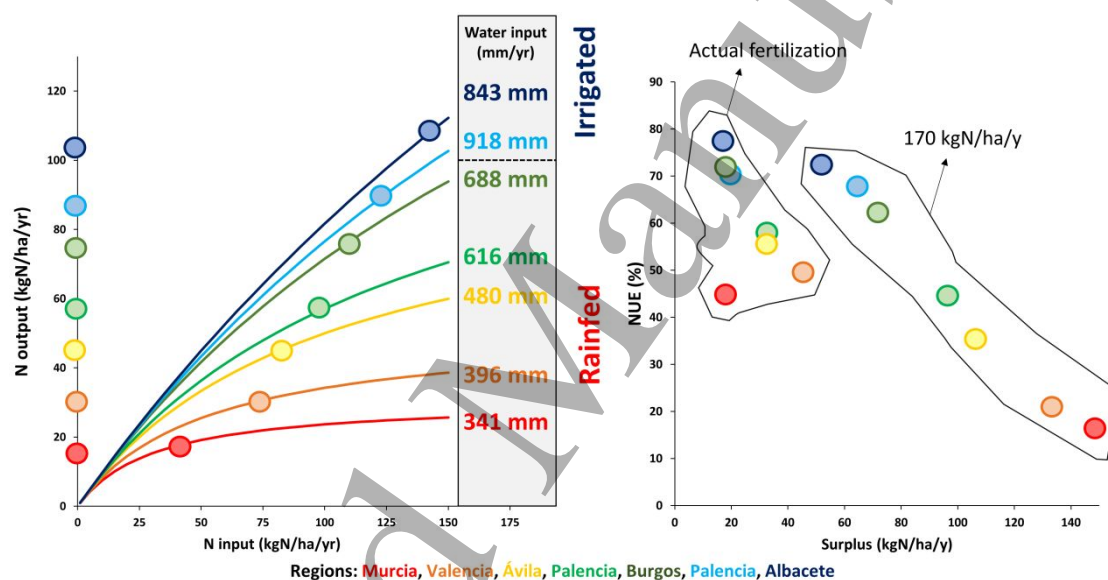
358 This relationship has important implications for agro-environmental management of fertilization
 359 in rainfed systems. In regions with water scarcity, crop N uptake is rapidly saturated and N
 360 surpluses accompanied by pollution (e.g., leaching during rainfall events) could occur at low
 361 fertilization rates (e.g., Albacete province, Fig. 6). However, in areas where water availability is
 362 higher, together with other drivers such as less heat that is common in more northern latitudes,
 363 rainfed systems had higher Y_{max} values that were closer to those from irrigated systems. Here, the
 364 same fertilization rates may lead to higher yield responses (e.g., Burgos province, Fig. 6). Even in
 365 these wetter regions, a dry year can significantly reduce Y_{max} , increasing N surplus and therefore
 366 the risk for N leaching.



367
 368 **Figure 6** Yield vs. total N inputs (1990–2015) in four contrasting Spanish provinces. The curve is the mean
 369 yield fertilization response curve estimated by using the average input and yield during the study period
 370 (1990–2015) for rainfed (red) and irrigated (blue) systems. Annual mean precipitation: Albacete (388
 371 mm/yr), Toledo (426 mm/yr), Valladolid (465 mm/yr), Burgos (688 mm/yr).

372 In summary, for rainfed systems, farmers have the difficult task of adjusting fertilization in order
 373 to maximize yields while reducing N surpluses in a highly variable environment. Mean fertilization
 374 rates per province ranged from 42 to 150 kg N/ha/yr in rainfed systems and from 77 to 186

375 kg N/ha/yr in irrigated systems. In general, actual fertilization rates (estimated as the base for the
 376 national inventories of greenhouse gas emissions) are lower in the rainfed systems of those
 377 provinces with lower precipitation and faster N saturation (Fig. 7), and, with some exceptions,
 378 moderate N surplus (usually lower than 50 kg N/ha/yr). Poor planning with high fertilization rates
 379 in a very dry year entails a disproportionately low NUE and significantly high emissions of Nr to
 380 the environment. As described in Section 3, the increasing livestock production is generating a
 381 growing amount of manure that has to be properly managed and recirculated to crops. This
 382 manure application has to consider the distinct response of cropping systems to N applications.
 383 For example, the EU Nitrates Directive limits the application of N in manure to 170 kg N/ha/yr for
 384 areas considered as Nitrate Vulnerable Zones. These rates, albeit legal, would strongly reduce NUE
 385 in the drier regions, severely impairing the environment (Fig. 7).



386
 387 **Figure 7.** Yield vs. fertilization response curves, NUE and surplus under rainfed and irrigated conditions in
 388 several Spanish provinces in a precipitation gradient (period 1990-2015). Water input (mm/yr) is the mean
 389 annual water input including precipitation and irrigation. Averaged fertilization rates and yields (1990-2015
 390 averages) are provided by the Spanish Ministry of Agriculture as well as the result of a hypothetical
 391 fertilization rate of 170 kg N/ha/yr that is the limit of application of N in manure at the Nitrate Vulnerable
 392 Zones.

393

394 5. Interactions of N use with adaptation to climate change

395 As a summary of the climate change conditions in the Mediterranean described in Section 1,
 396 increases in maximum and minimum temperatures and in atmospheric CO₂ concentration are
 397 expected to be associated with decreased and more unpredictable precipitation. These changes
 398 combined will result in stronger fluctuations in water stress for crops (Fig. 4 Suppl.). Besides these
 399 climatic effects, “elevated atmospheric CO₂ concentration” (hereafter eCO₂) also affects plant N

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3 400 dynamics. Therefore, under climate change scenarios with eCO₂, N dynamics with water and
4 401 temperature interactions have to be comprehensively studied. These effects are in general less
5 402 understood; however, in the past few decades, Free Air CO₂ Enrichment (FACE) experiments
6 403 (O'Leary *et al.*, 2015) and other laboratory or modeling studies indicate that CO₂ might enhance
7 404 photosynthesis in C₃ plant species such as wheat, rice, and barley (Ainsworth and Rogers, 2007),
8 405 especially under dry conditions (Bishop *et al.*, 2014), as well as N fixation by legumes (Parvin *et*
9 406 *al.*, 2018). This might increase total N crop requirements to build more biomass (Ainsworth and
10 407 Rogers, 2007). Under elevated CO₂ conditions, transpiration benefits from reduced stomata
11 408 opening to capture the same amount of CO₂. Therefore, low stomatal conductance, more
12 409 conservative water use, and higher transpiration efficiency may jointly increase WUE as well as
13 410 biomass and total water use (Houshmandfar *et al.*, 2018). On the negative side, a lower N content
14 411 in wheat grain has been reported under high CO₂ conditions (Tausz *et al.*, 2017). For legumes,
15 412 however, reduced N content in grain following a heatwave might be restored with eCO₂ (Parvin
16 413 *et al.*, 2020).

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27 414 Adaptation to climate change in Mediterranean cropping systems needs to consider these
28 415 interactions and how they are modified by heat and water stress, as both of these adverse
29 416 conditions for crop growth are projected to occur more frequently in the future (Trnka *et al.*,
30 417 2014). Briefly, positive (crop growth stimulation and water saving) and negative (grain N dilution)
31 418 eCO₂ effects alike may be diminished under stressful conditions (Ainsworth *et al.*, 2008; van der
32 419 Kooi *et al.*, 2016). Also, stresses limit the crop response to N, as some part of the fertilizer cannot
33 420 be used while crop growth is water- or temperature-limited (Savin *et al.*, 2019). This implies low
34 421 NUE and high N surplus and greenhouse gas emissions (Quemada and Gabriel, 2016) (Section 4).
35 422 Under these conditions, adaptation of annual crops in Mediterranean environments usually
36 423 implies using varieties with high transpiration efficiency traits and early sowing dates (Porter *et*
37 424 *al.*, 2014). The aim of such strategies is to increase water accumulation over the crop cycle and to
38 425 restore the duration of the vegetation period shortened by warming, which accelerates crop
39 426 development. The target then is to identify the optimum combination of crop phenology and
40 427 sowing date, selecting the earliest possible sowing date ensuring that: (1) there is enough soil
41 428 water for successful germination, emergence, and canopy establishment, (2) the crop will flower
42 429 after the last spring frosts and before late spring–early summer heatwaves, and (3) most of grain
43 430 filling happens before the dry summer period (Kirkegaard and Hunt, 2010; Christy *et al.*, 2018;
44 431 Ruiz-Ramos *et al.*, 2018). These are already the goals of current strategies recommended in
45 432 Mediterranean systems, together with management for conserving soil moisture and reducing
46 433 soil and canopy temperature. For climate change adaptation, the main novelty is that the suitable
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3 434 sowing window could become narrower in the future, as may the range of suitable varieties in
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5 435 terms of phenology, potential yield, and resistance to stresses. Therefore, in these systems the
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7 436 fulfilment of mitigation targets to reduce Nr emissions within the limits of effective adaptation
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9 437 will require the combination of new or existing tailored varieties, optimized management for soil
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11 438 and water conservation, and improved efficiencies, supported by knowledge and technological
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13 439 solutions (Springmann *et al.*, 2018).

14 440 Farm-level management options such as agroforestry are currently studied in detail for their
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16 441 potential to improve the sustainability of Mediterranean farming system, especially under climate
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18 442 change (Arenas-Corraliza *et al.*, 2016; Mosquera-Losada *et al.*, 2018). Among others, they offer
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20 443 access to deeper soil water tables (Cardinael *et al.*, 2015) and they reduce evaporation due to the
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22 444 wind-break effect of trees (Campi *et al.*, 2012), thereby enhancing groundwater recharge (Kay *et*
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24 445 *al.*, 2019), which are important issues regarding the climate projections for the Mediterranean.
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26 446 While it has been shown that agroforestry can increase soil fertility and nutrient availability in
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28 447 Europe (mostly Mediterranean cases) and North Africa (Torralba *et al.*, 2016; Zayani *et al.*, 2020),
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30 448 and that tree–legume interactions maintain a sufficient level of N fixation under the shade of trees
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32 449 in a Mediterranean agroforestry system (Querné *et al.*, 2017), the implications of agroforestry for
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34 450 nutrient acquisition under water and heat stress are still poorly understood (Guillot *et al.*, 2019).
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36 451 As stated by (Isaac and Borden, 2019), extensive research is needed on how agroforestry practices
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38 452 stabilize key nutrient acquisition patterns in the face of environmental change.

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40 453 Conservation agriculture (CA) – with no (or reduced) tillage, permanent organic soil cover, and
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42 454 crop diversification – is another innovative agricultural system that has been suggested for climate
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44 455 mitigation and adaptation in the Mediterranean due to its soil and water conservation capability
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46 456 (Kassam *et al.*, 2012). The physical, chemical, and biological properties of soil are improved,
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48 457 resulting in higher yields for dry conditions due to improved soil moisture availability (Mrabet,
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50 458 2002). Although authors logically hypothesize on improved nutrient availability, studies indicate
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52 459 that extractable N is not significantly greater under CA (López-Garrido *et al.*, 2011).
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54 460 Measurements at all soil depths are needed to better understand the effect of CA on nutrient
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56 461 distribution and its potential contribution to sustainable Mediterranean agriculture (Mrabet *et*
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58 462 *al.*, 2012). Moreover, in an Italian dry environment, the limited amount of wheat crop residues
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60 463 during the transition phase to CA required high N fertilization to maintain yields (Galieni *et al.*,
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465 2016).

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468 6. Emission of Nr compounds: drivers and adapted solutions

469 6.1 Ammonia

470 Similar to temperate agroecosystems, the main pathways of Nr loss from fertilized crops under
471 Mediterranean conditions are NH_3 volatilization to the atmosphere and NO_3^- leaching to
472 hydrosystems (Jarvis, 2011; Quemada *et al.*, 2013; Sanz-Cobena *et al.*, 2014). Ammonia (NH_3)
473 volatilization in cropping systems mainly occurs following NH_3 production through mineralization
474 or application of NH_4^+ -containing fertilizers, urea- or manure- and other organic-based N
475 fertilizers. The process of NH_3 volatilization is physicochemical (Walker *et al.*, 2013).
476 Meteorological conditions (air temperature, wind speed near the soil surface, atmospheric
477 stability, and rainfall), soil properties (e.g., texture, cation exchange capacity, and pH), and N
478 management options are the main drivers involved (Bouwman *et al.*, 1997). Most of these factors
479 are vastly determined by Mediterranean unique pedoclimatic conditions and management
480 practices, the latter being shaped by the dominant types of crop and livestock systems present in
481 these areas (Sanz-Cobena, 2010).

482 Atmospheric conditions greatly influence NH_3 volatilization rates (Cape *et al.*, 2009). Typical stable
483 atmospheric conditions with calm periods of wind (commonly associated with high pressures)
484 under Mediterranean conditions may lead to low volatilization rates following fertilization, mainly
485 in winter and during the night (Theobald *et al.*, 2015). Lower emission factors (EF: the percentage
486 of applied N that is lost as NH_3) are expected for winter crops as compared with irrigated summer
487 crops, even if similar agricultural practices are used. The rainfall regime is also critical for the
488 enhancement (or mitigation) of NH_3 losses. Sanz-Cobena *et al.* (2011) showed that more than 7-
489 14 mm water application immediately after fertilizer application is an effective strategy for
490 abating NH_3 volatilization due to enhanced solubilization and incorporation of the fertilizer into
491 the upper soil. By contrast, under low rainfall (i.e., 3 mm) conditions, NH_3 volatilization increases
492 because high concentrations of soluble N forms might be kept at the soil surface and hydrolysis
493 rates triggered following urea application (Sanz-Cobena *et al.*, 2011). Based on this, applying
494 water immediately after surface fertilization would be an efficient NH_3 abatement strategy in
495 irrigated Mediterranean agroecosystems. In rainfed crops, as the Mediterranean climate is
496 characterized by a limited number of rainfall events, the possibility of NH_3 mitigation due to
497 rainfall requires a good weather forecast.

498 Regarding fertilization practices, in Mediterranean cropping systems, even under conditions of
499 high atmospheric stability, the risk of NH_3 volatilization is greater if urea or liquid manure (i.e.,
500 slurries) is surface-applied (Marchetti *et al.*, 2004; Sanz *et al.*, 2010; Abalos *et al.*, 2012; Yagüe and

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3 501 Bosch-Serra, 2013; Bosch-Serra *et al.*, 2014; Recio *et al.*, 2018; Sanz-Cobena *et al.*, 2019). The use
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5 502 of urea as a synthetic N fertilizer has increased in the past decades in Mediterranean cropping
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7 503 systems, as in the rest of the world (IFASTAT, 2020). However, the large NH₃ losses associated
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9 504 with its application, triggered by the hydrolysis-mediated increase in soil pH, necessitate the
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11 505 implementation of specific NH₃ abatement for this fertilizer (Bittman *et al.*, 2014). The most
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13 506 common abatement practices are the incorporation of urea immediately following application,
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15 507 mechanically or by means of water (Sanz-Cobena *et al.*, 2011; Rochette *et al.*, 2013), or the use of
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17 508 urease inhibitors (Sanz-Cobena *et al.*, 2014; Recio *et al.*, 2020). These synthesized compounds
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19 509 reduce the rate of urea hydrolysis thus allowing for its incorporation into the soil (Sanz-Cobena *et*
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21 510 *al.*, 2008; Turner *et al.*, 2010; Sanz-Cobena *et al.*, 2017; Fu *et al.*, 2020). Pollution swapping (such
22
23 511 as N leaching) might also occur if these strategies are improperly managed (Sanz-Cobena *et al.*,
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25 512 2017). On the contrary, due to the predominant nitrifying conditions in rainfed systems under
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27 513 Mediterranean conditions, the decreased NH₄⁺ pool induced by urease inhibitors can have a co-
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29 514 benefit due to mitigated N₂O emissions from this microbiological process (Sanz-Cobena *et al.*,
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31 515 2016). The incorporation of urea by water in irrigated systems is facilitated (Sanz-Cobena *et al.*,
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33 516 2011), thus avoiding excessive soil compaction as well as greenhouse gas emissions from
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35 517 machinery. Increasing generation of animal manures in certain Mediterranean areas (e.g. Italy
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37 518 and Spain) has triggered ammonia emissions due to surface application of slurries. Although less
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39 519 effective than injection (Sanz-Cobena *et al.*, 2019), the possibility of adapting existing on-farm
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41 520 equipment to mix the slurry with the soil makes this option technically suitable and cost-effective
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43 521 (Bittman *et al.*, 2014).

44 522 6.2 Nitrous oxide

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46 523 Studies on N₂O emissions in Mediterranean cropping systems are underrepresented compared to
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48 524 other temperate areas. (Aguilera *et al.*, 2013b), and studies deciphering N₂O formation pathways
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50 525 are conspicuously low. The few published studies to date point at nitrification and nitrifier-
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52 526 denitrification as particularly relevant mechanisms in Mediterranean soils (Zhu *et al.*, 2013). The
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54 527 prevalence of distinct formation pathways in Mediterranean soils, driven by the specific climatic
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56 528 conditions and agronomic practices, will ultimately determine the best N₂O mitigation strategies.
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58 529 There is ample evidence that N₂O emission factors (EF: the percentage of applied N that is lost as
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60 530 N₂O) in Mediterranean rainfed cropping systems (EF: 0.27% according to Cayuela *et al.*, 2017) are
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532 531 significantly lower than in other temperate areas (0.9% according to Stehfest and Bouwman
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534 532 2006). However, as irrigation is a common practice in Mediterranean cropping systems in
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536 533 summer, it changes the moisture pattern and substantially increases the total amount of water in

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3 534 soil. As a consequence, EFs in irrigated crops (0.63%) are significantly higher than in rainfed crops
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5 535 (0.27%) (Aguilera *et al.*, 2013b; Cayuela *et al.*, 2017).

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7 536 In both rainfed and irrigated systems, the adjustment of N application to crop needs is one of the
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9 537 best recommended strategies to mitigate N₂O emission (Guardia *et al.*, 2018). Additionally, the
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11 538 use of nitrification inhibitors with both synthetic and organic N fertilizers (Sanz-Cobena *et al.*,
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13 539 2012; Abalos *et al.*, 2014; Gilsanz *et al.*, 2016; Guardia *et al.*, 2017b) is also an effective mitigation
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15 540 strategy.

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17 541 The type of irrigation directly determines N₂O emissions (Cayuela *et al.*, 2017). Localized irrigation
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19 542 systems such as drip irrigation significantly abate N₂O losses due to a highly localized moist area.
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21 543 On the contrary, sprinkler-irrigated fertilized systems are an important source of N₂O emissions
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23 544 due to the increased soil moisture in large surface areas (Guardia *et al.*, 2017b). Nevertheless,
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25 545 recent studies show how optimized sprinkler irrigation systems can considerably reduce N₂O
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27 546 emissions in intensive maize crops (Franco-Luesma *et al.*, 2019; Franco-Luesma *et al.*, 2020).

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29 547 Precipitation following long drought periods promotes high N₂O emissions. This is very common
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31 548 in Mediterranean soils in summer or following the first rainfall in autumn (Cárdenas *et al.*, 1993).
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33 549 A recent meta-analysis demonstrates that flooding the soil after extended periods of drought
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35 550 produces a larger pulse in N₂O emissions, compared with soils that were already moderately wet
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37 551 before the flood event (Barrat *et al.*, 2020). These pulses are likely more common under
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39 552 Mediterranean conditions than in temperate agroecosystems where pulses are usually due to
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41 553 freeze–thaw events (Sanchez-Martin *et al.*, 2010).

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43 554 Another important factor modulating N₂O emissions in Mediterranean cropping systems is soil
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45 555 pH. Mediterranean soils are predominantly neutral or alkaline (Slessarev *et al.*, 2016) and this has
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47 556 important implications for the final ratio of denitrification products. (Cayuela *et al.*, 2013) found
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49 557 an average N₂O/(N₂+ N₂O) ratio of 0.4 in five Mediterranean high-pH soils, whereas the average
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51 558 value for other temperate soils was 0.8. This implies that the last step of denitrification (the
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53 559 transformation of N₂O to N₂) was enhanced in Mediterranean soils. A recent global meta-analysis
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55 560 of 1104 field measurements found soil pH to be the most relevant parameter defining regional
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57 561 N₂O emissions, with significantly lower EFs for soils with increasing pH (Wang *et al.*, 2018).

562 6.3 Nitric oxide

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57 563 Together with N₂O, nitric oxide (NO) is a by-product of nitrification (Medinets *et al.*, 2015), thus
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59 564 particularly common in Mediterranean rainfed systems (Vallejo *et al.*, 2005; Meijide *et al.*, 2007;
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3 565 Meijide *et al.*, 2009; Sanchez-Martín *et al.*, 2010; Sanz-Cobena *et al.*, 2012; Guardia *et al.*, 2017a;
4 566 Guardia *et al.*, 2017b; Recio *et al.*, 2019). Significant emissions of NO, a precursor of tropospheric
5 567 ozone (O₃), have been reported after N fertilization with either organic or synthetic N fertilizers
6 568 and in both rainfed and irrigated systems, particularly if drip-irrigation is performed (Guardia *et al.*
7 569 *et al.*, 2017a). These authors associated increased NO fluxes with enhanced nitrification in the
8 570 proximity of the wet front that surrounds the irrigation bulb. There is evidence that NO_x (both NO
9 571 and NO₂) emission exponentially increases with temperature and that, after soil rewetting, strong
10 572 pulses of NO_x emission are produced in high-temperature regions (Garrido *et al.*, 2002). In the
11 573 Mediterranean region, as mentioned earlier, the pulses that occur after rewetting result in high
12 574 contributions to the total NO budget. Analyzing the different agroecosystems, irrigated crops are
13 575 also important contributors of NO, because the importance of nitrification and the temporal
14 576 pattern of soil moisture occurred when the soil temperature is high, with frequent irrigation–
15 577 drying cycles. Abating soil NO_x emissions should be considered in future efforts to improve
16 578 regional air quality in the surrounding of irrigated crops, because these emissions contribute to
17 579 the high concentration of O₃ found in many rural areas in the Mediterranean region (Calvete-Sogo
18 580 *et al.*, 2014).

31 581 *6.4 Nitrate and losses to the hydrosphere*

32 582 In Mediterranean irrigated agriculture, nitrate leaching is strongly dependent on water
33 583 application. Intensive irrigation together with over-fertilization is the main cause of nitrate
34 584 leaching (Isidoro *et al.*, 2006; Caverro *et al.*, 2012; Laspidou and Samantzi, 2014; Lasagna and De
35 585 Luca, 2019). Excessive water is often applied either to avoid salt accumulation in soil or to
36 586 compensate for the heterogeneity of the water-delivery system, enhancing nitrate leaching
37 587 during crop growth (Díez *et al.*, 2000). Low crop N availability is then compensated by increasing
38 588 fertilizer rates leading to a vicious circle characterized by low WUE and NUE. Therefore, the two
39 589 main drivers of nitrate leaching are water percolation and soil inorganic N accumulation, and
40 590 strategies to mitigate losses are oriented toward keeping these processes under sustainable
41 591 thresholds.

42 592 Strategies to improve water management rely on adapting water application to crop
43 593 requirements and are mainly based on improving irrigation schedules or technologies. Irrigation
44 594 should be scheduled to avoid over-irrigation and limit water percolation (Zinkernagel *et al.*, 2020).
45 595 Additionally, the water delivery technology may have a major impact on the main N processes
46 596 governing N losses. Drip and sprinkle irrigation have the potential to localize water in the root
47 597 zone and limit percolation and nitrate leaching (Causape *et al.*, 2006; Vazquez *et al.*, 2006; Caverro

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3 598 *et al.*, 2012; Garcia-Garizabal *et al.*, 2014; Malik *et al.*, 2019), whereas with flood irrigation it is
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5 599 difficult to achieve a high water efficiency and control leaching losses (Garcia-Garizabal *et al.*,
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7 600 2012).

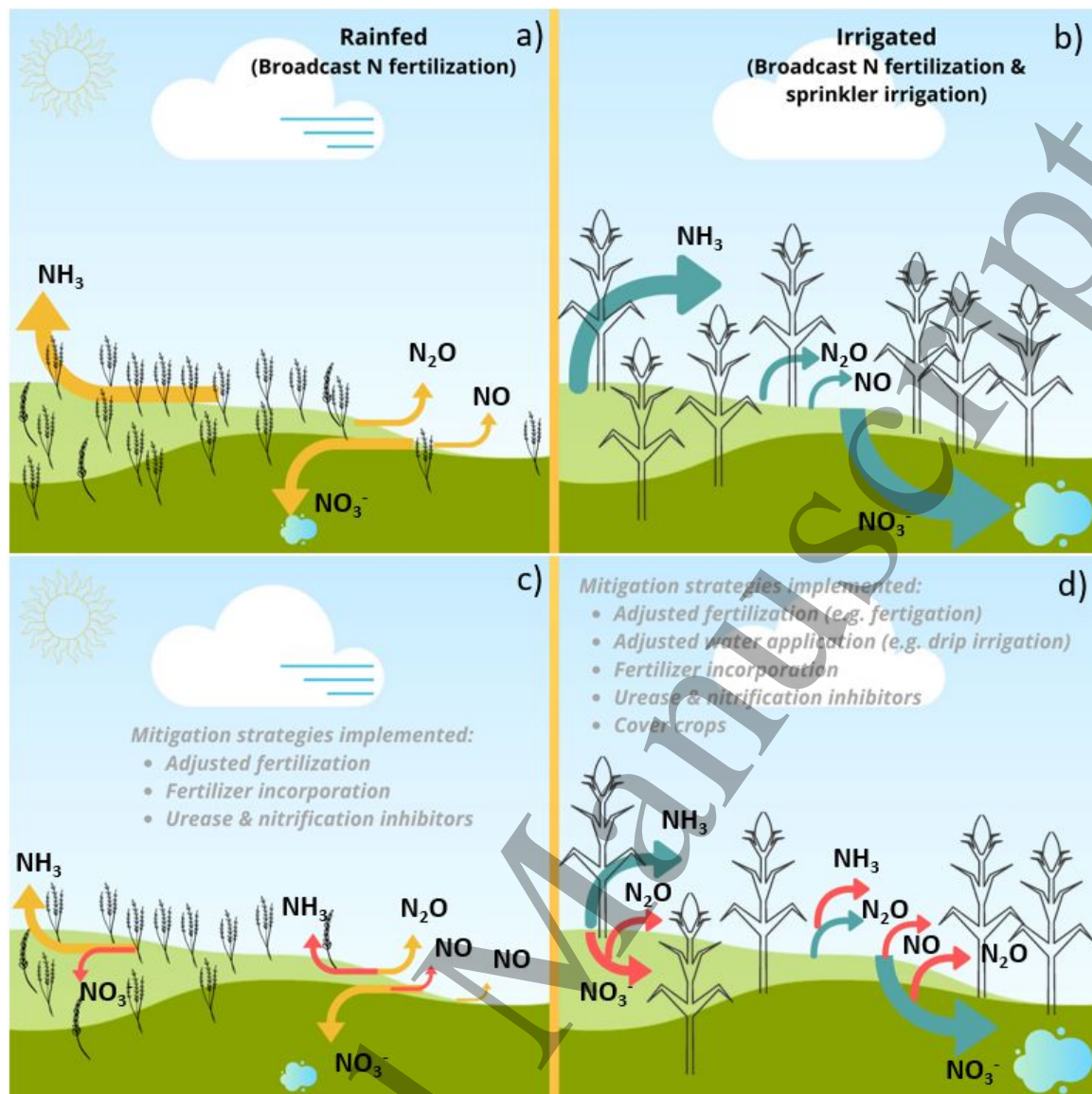
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9 601 Adjusting N fertilization, both synthetic and organic, to crop requirements remains crucial in order
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11 602 to avoid soil N accumulation and mitigate nitrate leaching (Quemada *et al.*, 2013). Intensive
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13 603 livestock associated with irrigated land is common in Mediterranean areas and avoiding excessive
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15 604 application of manure to cropland is highly recommended (Mantovi *et al.*, 2006; Nikolaidis *et al.*,
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17 605 2009; Yague and Quilez, 2010). In the Po valley, establishing a N balance to minimize N surplus
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19 606 was identified as the first step toward mitigating the effect of high livestock concentration that
20
21 607 leads to poor water quality (Perego *et al.*, 2012).

22 608 Replacing the traditional fall–winter fallow by cover crops in irrigated areas reduced nitrate
23
24 609 leaching by 35% on average in a meta-analysis of irrigated land (Quemada *et al.*, 2013). Compare
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26 610 to fallow soils, cover crops minimize the risk of N leaching by reducing water percolation due to
27
28 611 an increase in evaporation, assimilating the soil residual N after harvesting the summer crop thus
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30 612 reducing nitrate concentration in the leachate, and enhancing microbial N immobilization
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32 613 associated to C inputs from cover crops roots (Gabriel *et al.*, 2012; Benoit *et al.*, 2016; Thapa *et al.*,
33
34 614 2018). Replacing the fallow with a non-legume is more efficient than with a legume in reducing
35
36 615 leaching, and mixes of grass/legume greatly reduce leaching with respect to the fallow (Tosti *et al.*,
37
38 616 2014). Additionally, the combination of N recycling and the boost of soil N mineralization in
39
40 617 the short and long term after cover crops may allow to reduce fertilization rates without affecting
41
42 618 yields. Indeed, the increase in N₂O emissions caused by legume cover crops could be offset by the
43
44 619 N fertilizer savings and its associated indirect CO₂ emissions (Guardia *et al.*, 2019; Quemada *et al.*,
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46 620 2020b). In general, Mediterranean irrigated soils are prone to high mineralization rates, and
47
48 621 strategies that enhance soil stable organic compounds, allow for the recovery of soil physical
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50 622 properties and fertility in the long term and contribute toward mitigating nitrate leaching losses
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52 623 (Diacono and Montemurro, 2010; García-González *et al.*, 2018).

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54 624 An additional pathway of N losses that is influenced by practices such as irrigation and cover
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56 625 cropping is leaching of dissolved organic N (DON). Soil water content regulates the ratio of DON
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58 626 and inorganic N forms in Mediterranean soils, being DON dominant under various plant
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60 627 communities during wet periods (Delgado-Baquerizo *et al.*, 2011). In an irrigated maize field of
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62 628 central Chile, DON leaching under a grass cover crop was twice the rate of nitrate leaching, even
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64 629 if the total N losses were reduced compared with the fallow (Salazar *et al.*, 2019). Although the
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66 630 full ecological significance of DON loss remains unclear, it may represent a large part of the total

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3 631 dissolved N, and monitoring DON leaching may contribute toward mitigating diffuse pollution
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5 632 from agricultural systems.
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7 633 Losses to hydrosystems from rainfed agriculture in the Mediterranean basin are mainly associated
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9 634 with dissolved N in runoff or attached to soil-eroded particles (Section 7). In rainfed cropland,
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11 635 nitrate leaching showed to be relevant after intense rainfall events or wet periods that are
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13 636 common in semi-arid (Ruiz-Ramos *et al.*, 2011) or humid (Arregui and Quemada, 2006)
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15 637 Mediterranean areas. High winter nitrate flows from upland cereals are very common when
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17 638 winter fertilization is applied (Lassaletta *et al.*, 2009, 2010). Finally, transformation of rainfed into
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19 639 irrigated fields may greatly enhance nitrate leaching and become a major risk for water pollution
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21 640 if it concerns large areas (Merchan *et al.*, 2015). During the transition period, N mineralization
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23 641 may release a large amount of nitrate until the soil organic matter content reaches a new
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25 642 equilibrium (Quemada and Gabriel, 2016). Promoting temporary soil mining by minimizing N
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27 643 fertilizer application may help mitigate the risk of nitrate pollution (Vazquez *et al.*, 2006). The
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29 644 main pathways of environmental N losses from rainfed and irrigated cropland are summarized in
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31 645 Figure 8.
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647 **Figure 8.** Schematic representation of the main pathways of reactive N losses in Mediterranean
 648 agroecosystems, both rainfed (a, c) and irrigated (b, d) under broadcast N fertilization. The size of the arrow
 649 is proportional to the net amount of N being lost to both the atmosphere and water bodies. Bottom figure
 650 (c, d) shows the impact of the most recognized Nr abatement strategies in the area focusing on N fertilizer
 651 management, irrigation (e.g. drop or well-planned sprinkler irrigation), and cover cropping. Red arrows
 652 indicate the potential pathways of pollution swapping. * Similar to temperate areas, high nitrate leaching
 653 can occur in upland cereal croplands during intensive precipitation in winter. Our own elaboration based
 654 on the scientific literature referred to in the text.

655

656 7. From the croplands to the aquatic continuum

657 7.1 Soil erosion and N flows

658 Mediterranean soils are particularly vulnerable to water erosion, due to frequent intense rainfall
 659 events (Cortesi *et al.*, 2012; Merheb *et al.*, 2016), the typical steep slopes (Tarolli *et al.*, 2012) and
 660 the climatic conditions that make Mediterranean regions prone to wildfire, often associated with

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3 661 soil erosion (Shakesby, 2011). While annual precipitation in many Mediterranean regions is
4 expected to decrease under climate change (Cramer *et al.*, 2018; Brogli *et al.*, 2019), several
5 662 studies highlight an expected increase in extreme precipitation events (Hagos *et al.*, 2016; Polade
6 663 *et al.*, 2017; Trambly and Somot, 2018) which could lead to increased soil erosion (Morán-
7 664 Ordóñez *et al.*, 2020).
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12 666 It has been estimated that globally 23–42 Tg N is moved by soil erosion each year, which is in the
13 667 same order of magnitude as N applied as chemical fertilizer and N removed by harvested crops
14 668 (Quinton *et al.*, 2010). In spite of this, the transport of N has been largely neglected in soil erosion
15 669 studies, which mostly focus on the impact of erosion on the redistribution of soil organic matter
16 670 (SOM) and associated phosphorus (Garnier *et al.*, 2015). N is a structural component of SOM
17 671 (Tipping *et al.*, 2016), hence, the transport of SOM after extreme rainfall events affects soil N
18 672 stocks. The redistribution of SOM is associated with the dynamics of sediment transport and
19 673 deposition (Boix-Fayos *et al.*, 2015). As such, SOM may be deposited in common depositional
20 674 landscape units, including river bars, floodplains, and reservoirs (Fissore *et al.*, 2017) (see also
21 675 Section 7.2). The impact of soil erosion on SOM dynamics is still open to debate. Eroded SOM may
22 676 be subject to mineralization during transport, potentially leading to increased SOM
23 677 decomposition (Lal, 2004), depending on the travel time and distance (Quinton *et al.*, 2010). On
24 678 the contrary, the deep burial of SOM-rich sediments may lead to reduced SOM decomposition
25 679 rates (Berhe *et al.*, 2007). Due to heterogeneous land use configurations, the transport of
26 680 sediment and SOM in Mediterranean catchments is highly complex (Boix-Fayos *et al.*, 2015),
27 681 making it more difficult to assess how SOM, and consequently N, is redistributed in Mediterranean
28 682 environments.
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42 683 *7.2 N retention in Mediterranean river basins*

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45 684 Nitrogen losses from agricultural land reach water bodies by surface runoff and leaching to
46 685 groundwater, as well as by soil erosion and atmospheric deposition. Once in the aquatic system,
47 686 N undergoes different processes of transport, storage, transformation and elimination to gaseous
48 687 forms, and part of it is finally exported to coastal areas.
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53 688 Mediterranean climatic features, including high temperatures and low rainfall, often
54 689 concentrated in a few large precipitation events, favor N retention in the watersheds –i.e.
55 690 considering retention as the sum of storage, transformation and elimination processes–. High
56 691 temperatures are accompanied by high evapotranspiration rates, so runoff mainly occurs after
57 692 intense events. Moreover, the irregular precipitation patterns and the recurrence of summer
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3 693 droughts have driven intensive hydraulic management to secure water availability (Castaldelli *et*
4 694 *al.*, 2013; Soana *et al.*, 2017; Soana *et al.*, 2019; Romero *et al.*, 2021), and most rivers comprise a
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6 695 large number of dams, reservoirs and irrigation channels (Sadaoui *et al.*, 2018). Reservoirs and
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8 696 channels increase land-water contact surface and residence time in the watersheds, and allows
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10 697 for enhanced nutrient processing via denitrification, uptake by riparian vegetation and freshwater
11 698 macrophytes, infiltration and leaching to groundwater (Bartoli *et al.*, 2012; Lassaletta *et al.*, 2012;
12 699 Romero *et al.*, 2016). Besides, reservoirs are built to contend with water scarcity and buffer great
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14 700 episodes of rain and flooding, which means that they also decrease the transport of N to coastal
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16 701 areas during flashy rainfall events.
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19 702 All these waterwork developments contribute to increasing N retention in the river basin with
20 703 potentially higher pressure on inland waters (including groundwater) and less export to coastal
21 704 systems. Higher retention percentages in the Mediterranean with respect to other climatic
22 705 conditions –and especially in arid and semiarid Mediterranean basins– have been reported in
23 706 several recent studies that include different geographical scales (the Iberian Peninsula, European
24 707 Mediterranean basins, and the entire Mediterranean basin) and different modeling approaches
25 708 (Romero *et al.*, 2016; Grizzetti *et al.*, 2021; Romero *et al.*, 2021).
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32 709 Despite high retention and hence high elimination rates, N concentration in water bodies might
33 710 be increased in regions affected by water scarcity due to the low dilution capacity by water flow,
34 711 as has been observed in southern Europe, mainly in Spain, Italy and Greece (Grizzetti *et al.*, 2017).
35 712 In addition, as N retention includes permanent and temporal components, high values of
36 713 retention might hide an accumulation of N in soils and groundwater in the river basin, and a delay
37 714 between sources and impacts in aquatic ecosystems (Grizzetti *et al.*, 2015).
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46 716 **8. Lessons learned and policy dimensions**

47 717 The climatic and agronomic specificities of the Mediterranean region worldwide demand N
48 718 management techniques specifically suited for this region. Agronomic practices (rainfed vs.
49 719 irrigated crops; annual crops vs. permanent crops) or soil characteristics (acidic vs. calcareous; soil
50 720 carbon content) could limit the beneficial effects of sustainable practices successfully developed
51 721 and implemented in other regions, and even prevent their implementation. In any case, they
52 722 should be adapted considering the climate change context and potential adverse side effects, such
53 723 as pollution swapping. The definition of the most suited management practices at the local level
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3 724 should be defined under a co-creation process considering the end-users' needs and constraints
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5 725 with the support of researchers and the various implicated agents.
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7 726 The systemic approach considering the overall N budget within the agro-food system in the
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9 727 Mediterranean Basin has highlighted those processes demanding urgent action. Anyhow,
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11 728 accounting, surveillance, and control of the different elements of the agro-food system need to
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13 729 be implemented. Although measures taken at the farm level eventually impact the overall system,
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15 730 multilateral, subregional, and national actions are needed to control N surpluses and their
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17 731 environmental impacts. This study has identified important differences in the agro-food system
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19 732 across the Mediterranean Basin, mostly related to climatic factors (coastal vs. in-land areas;
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21 733 subarid vs. arid areas). The study of these systems at the national and province level has identified
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23 734 where governmental actions via incentives or regulations are needed, considering the geopolitical
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25 735 angle in the degree of intensification or import–export of feed and food.

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27 736 In the Mediterranean biome, management of water and N should be jointly appraised developing
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29 737 N fertilization schemes tailored not only to the potential N crop demand but also to water
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31 738 availability (quantity) and quality, the soil chemical and physical characteristics, as well as the
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33 739 fertilizer resources locally available. The case of the agro-food system transformation observed in
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35 740 the countries of the Mediterranean Basin is paradigmatic. The increasing manure production and
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37 741 the associated crop–livestock disconnection are critical in other regions of the world (Garnier *et*
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39 742 *al.*, 2016; Jin *et al.*, 2020; Mueller and Lassaletta, 2020), but they are of particular concern in the
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41 743 Mediterranean Basin. The best use of the available local manure has to consider the much lower
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43 744 response of rainfed systems and the high leaching potential of irrigated systems. Thus, the
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45 745 consideration not only of the two contrasting worlds –namely, rainfed and irrigated systems – but
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47 746 also of the precipitation gradient to rainfed systems is important. Analysis of potential maximum
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49 747 yields and yield vs. fertilization relationships, adapted to local conditions, should be made to set
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51 748 sustainable N application rates.

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53 749 In conclusion, bottom–up and top–down approaches should go hand in hand, combining (1) local
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55 750 assessments of the most suited farming systems that jointly improve N and water use efficiency
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57 751 considering the crop specificities, water availability for irrigation, and the edapho-climatic
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59 752 conditions up-scaled to national and continental level, and (2) a systemic analysis of N inputs into
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753 the systems fueled by food demand. Indicators for monitoring should be derived in order to
754 suggest policies and governance systems tailored to the local specificities. In addition, structural
755 measures promoting the reconnection of crop and livestock farming, including the relocation of
756 feed production, the reduction of food waste, and the consideration of the key effect of human
757 consumption patterns (diet, reconnection of production and consumption in time and space) have

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3 758 to be considered (Billen *et al.*, 2019). This integrated vision is aligned with the spirit of the recent
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5 759 Farm to Fork Strategy (and Biodiversity Strategy) of the European Union that aims to halve N
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7 760 pollution in agricultural systems and also considers recommendations at the consumption level..

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9 761 Markets are not currently considering N-related environmental externalities in the region, which
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11 762 also have socio-economic implications. However, the lack of proper connection of our scientific
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13 763 findings with socio-economic indicators prevents defining relevant guidelines integrating
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15 764 agronomic, environmental and social aspects of the Mediterranean farming systems.
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17 765 Nevertheless, farmers' adoption of locally suited best sustainable practices could very likely result
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19 766 in yield losses compared to "business as usual" practices, demanding the implementation of
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21 767 compensating schemes to account for the environmental services they provide.

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23 768 As a whole, the implementation of these practices also demands modifications in rural planning,
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25 769 considering the local biophysical, climatic, and socioeconomic conditions but also the
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27 770 heterogeneity of different land uses at the landscape and catchment levels. The integration of
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29 771 sustainable practices at the regional scale may help address coexisting environmental challenges
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31 772 such as land degradation, biodiversity loss, food security, climate change mitigation and
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33 773 adaptation, and water scarcity (Ruiz *et al.*, 2020). These features should be taken into account
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35 774 when developing future policies by integrating top-down system analysis with bottom-up
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37 775 initiatives aiming to identify the most urgent problems and feasible solutions, based on the
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39 776 quantification of ecosystem services as well as sustainable economic issues and the well-being of
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41 777 farmers (van Leeuwen *et al.*, 2019).

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