1 Ecological Impacts of Atmospheric Pollution and Interactions with Climate Change

2 in Terrestrial Ecosystems of the Mediterranean Basin: Current Research and Future

3 Directions

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5 Raúl Ochoa-Hueso*[1#], Silvana Munzi*[2], Rocío Alonso[3], María Arróniz-Crespo[4], Anna Avila[5],

6 Victoria Bermejo[3], Roland Bobbink[6], Cristina Branquinho[2], Laura Concostrina-Zubiri[2], Cristina

7 Cruz[2], Ricardo Cruz de Carvalho[2], Alessandra De Marco[7], Teresa Dias[2], David Elustondo[8], Susana

8 Elvira[3], Belén Estébanez[9], Lina Fusaro[10], Giacomo Gerosa[11], Sheila Izquieta-Rojano[8], Mauro Lo

9 Cascio[12], Riccardo Marzuoli[11], Paula Matos[2], Simone Mereu[12], José Merino[13], Lourdes Morillas[12],

10 Alice Nunes[2], Elena Paoletti[14], Luca Paoli[15], Pedro Pinho[2,16], Isabel B. Rogers[17], Arthur Santos[2],

11 Pierre Sicard[18], Carly J. Stevens[17], Mark R. Theobald[3]

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13 *These authors contributed equally to this work.

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21   Capsule: A coordinated monitoring of air pollution and an assessment network of its effects are

22   needed to improve environmental policy and management decisions in the Mediterranean Basin

1. Autonomous University of Madrid, Department of Ecology, 2 Darwin Street, Madrid, 28049, Spain.

2. cE3c Centre for Ecology, Evolution and Environmental Changes, Faculdade de Ciências, Universidade de Lisboa, Campo Grande, C2, Piso 5, 1749-016 Lisbon, Portugal

3. Air Pollution Division, CIEMAT, Avda. Complutense 22 (edif. 70), Madrid 28040, Spain

4. Escuela Técnica Superior de Ingenieros Agrónomos, Universidad Politécnica de Madrid, Ciudad Universitaria, 28040 Madrid, Spain

5. Center for Ecological Research and Forestry Applications (CREAF), Universitat Autònoma de Barcelona, 08193, Bellaterra, Spain

6. B-WARE Research Centre, Radboud University, PO Box 9010, 6525 ED Nijmegen, The Netherlands

7. ENEA, Via Anguillarese 301, 00123 Rome, Italy

8. LICA, Department of Chemistry and Soil Science, University of Navarre, Irunlarrea, 1-31008 Pamplona, Spain

9. Departamento de Biología, Unidad de Botánica, Universidad Autónoma de Madrid, C/ Darwin 2, 28049, Madrid, Spain.

10. Dept. of Environmental Biology, Sapienza University of Rome, Piazzale Aldo Moro 5, Rome, Italy

11. Dept. of Mathematics and Physics, Catholic University of Brescia, Via dei Musei 41, Brescia, Italy

12. Department of Science for Nature and Natural Resources, University of Sassari, Via Enrico De Nicola 1, 07100 Sassari, Italy

13. Departamento de Sistemas Físicos, Químicos y Naturales, Universidad Pablo de Olavide, Ctra.

46   de Utrera km. 1, 41013 Sevilla, Spain

47   14. IPSP-CNR, Via Madonna del Piano 10, 50019, Sesto Fiorentino, Florence, Italy

48   15. Department of Life Sciences, University of Siena, Via Mattioli 4, I-53100 Siena, Italy

49   16. CERENA-IST-UL, Centro de Recursos Naturais e Ambiente, Instituto Superior Técnico,

50   Universidade de Lisboa, Av. Rovisco Pais 1049-001 Lisboa, Portugal

51   17. Lancaster Environment Center, Lancaster University, Lancaster LA1 4YQ, UK

52   18. ACRI-ST, 260 route du Pin Montard, BP 234, 06904 Sophia Antipolis Cedex, France

53

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Abstract

Mediterranean Basin ecosystems, their unique biodiversity, and the key services they provide are currently at risk due to air pollution and climate change, yet only a limited number of isolated and geographically-restricted studies have addressed this topic, often with contrasting results. Particularities of air pollution in this region include high O_3 levels due to high air temperatures and solar radiation, the stability of air masses, and dominance of dry over wet nitrogen deposition. Moreover, the unique abiotic and biotic factors (e.g., climate, vegetation type, relevance of Saharan dust inputs) modulating the response of Mediterranean ecosystems at various spatiotemporal scales make it difficult to understand, and thus predict, the consequences of human activities that cause air pollution in the Mediterranean Basin. Therefore, there is an urgent need to implement coordinated research and experimental platforms along with wider environmental monitoring networks in the region. In particular, a robust deposition monitoring network in conjunction with modelling estimates is crucial, possibly including a set of common biomonitors (ideally cryptogams, an important component of the Mediterranean vegetation), to help refine pollutant deposition maps. Additionally, increased attention must be paid to functional diversity measures in future air pollution and climate change studies to stablish the necessary link between biodiversity and the provision of ecosystem services in Mediterranean ecosystems. Through a coordinated effort, the Mediterranean scientific community can fill the above-mentioned gaps and reach a greater understanding of the mechanisms underlying the combined effects of air pollution and climate change in the Mediterranean Basin.

Introduction

Human activities and natural processes have shaped each other over ca. eight millennia within Mediterranean Basin ecosystems (Blondel, 2006). This coevolution, together with the heterogeneous orography and geology, the large seasonal and inter-annual climatic variability, the refuge effect during the last glaciations, and the crossroad location between European temperate ecosystems and North African and Asian drylands, has resulted in the high diversification of the flora and fauna that we observe today, making Mediterranean ecosystems a hotspot of biodiversity, but also of vulnerability (Schröter *et al.* 2005; Blondel 2006; Phoenix *et al.* 2006). Moreover, the Mediterranean Basin is one of the world's largest biodiversity hotspots and the only one within Europe, otherwise dominated by temperate natural and semi-natural grasslands, temperate deciduous forests and boreal conifer forests (Myers *et al.*, 2000). Species-rich ecosystems exclusive to the Mediterranean Basin include Spanish *matorrales* and *garrigas*, Portuguese *matos*, Italian *macchias*, Greek *phryganas*, and agrosilvopastoral ecosystems of high natural and economic value such as Spanish *dehesas* and Portuguese *montados* (Cowling *et al.*, 1996; Blondel, 2006). However, the biodiversity and other ecosystem services of this region are currently at risk due to human pressures such as climate change, land degradation and air pollution (Schröter *et al.*, 2005; Scarascia-Mugnozza & Matteucci, 2012). Air pollution in the Mediterranean Basin is primarily in the form of particulate matter, nitrogen (N) deposition and tropospheric ozone (O_3) (Paoletti, 2006; Ferretti *et al.*, 2014; García-Gómez *et al.*, 2014). Production of pollutants is mainly associated with industrial activities, construction, vehicle emissions and agricultural practices and, within the European context, is characteristically exacerbated by more frequent droughts and the typical stability of air masses in the region, with important consequences for ecosystem and human health (Millán *et al.*, 2002; Vestreng *et al.*, 2008; Izquieta-Rojano *et al.*, 2016a). This also has important

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100 social consequences for the Mediterranean region, where approximately 480 million people live,

101 and where more frequent droughts, extreme climatic events and wildfires will only reinforce the

102 current migrant and humanitarian crisis (Werz & Hoffman, 2016).

103   Environmental pollution interacts synergistically with climate change (Alonso *et al.*, 2001,

104 2014; Bytnerowicz *et al.*, 2007; Sardans & Peñuelas, 2013). This is particularly true for seasonally

105 dry regions like the Mediterranean Basin (Baron *et al.*, 2014), but the effects of this interaction on

106 the structure and function of Mediterranean ecosystems are not adequately quantified and,

107 therefore, the consequences are poorly understood (Bobbink *et al.*, 2010; Ochoa-Hueso *et al.*,

108 2011). Projections for 2100 suggest that mean air temperatures in the Mediterranean Basin region

109 will rise from 2.2°C to 5.1°C above 1990 levels and that precipitation will decrease between –4

110 and –27% (Christensen *et al.*, 2007 and Figure 1). The sea level is also projected to rise, and a

111 greater frequency and intensity of extreme weather events (e.g., drought, heat waves and floods)

112 are expected (EEA, 2005). These changes will exacerbate the already acute water shortage problem

113 in the region, particularly in drylands (Terray & Boé, 2013; Sicard & Dalstein-Richier, 2015),

114 impairing their functionality and ability to deliver the ecosystem services on which society and

115 economy depend (Bakkenes *et al.*, 2002; Lloret *et al.*, 2004). Functions that will be synergistically

116 impaired by air pollution and climate change include reductions in crop yield and carbon

117 sequestration (Maracchi *et al.*, 2005; Mills & Harmens, 2011; Shindell *et al.*, 2012; Ferretti *et al.*,

118 2014). In addition, a higher fire risk is attributed to higher temperatures and more frequent droughts

119 coupled with an N-driven increase of grass-derived highly-flammable fine fuel (Pausas &

120 Fernández-Muñoz 2012).

121   In the last decades, atmospheric concentrations of major anthropogenic air pollutants such

122 as particulate matter and sulphur dioxide (SO_2) have decreased in Southern Europe due to emission

control policies and greener technologies (Querol *et al.*, 2014; Barros *et al.*, 2015; Aguillaume *et al.*, 2016; Àvila & Aguillaume, 2017). However, mitigation strategies have not been equally effective with other compounds such as reactive N and tropospheric O_3 (Figure. 2; Paoletti, 2006; García-Gómez *et al.*, 2014; Sicard *et al.*, 2016). For example, recent increases in N deposition, particularly dry deposition of NO_3, have been detected in North-eastern Spain, where N deposition is estimated in the range of 15-30 kg N ha^{-1} yr^{-1} (Avila & Rodà, 2012; Camarero & Catalan, 2012; Aguillaume *et al.*, 2016). This has been attributed to increased nitrogen oxide (NO_x) and ammonia (NH_3) emissions and changes in precipitation patterns (Aguillaume *et al.*, 2016). Background O_3 pollution is typically high in Mediterranean climates due to the meteorological conditions of the area (Paoletti, 2006) and recent reviews have demonstrated that while O_3 in cities has generally increased, no clear trend, or only a slight decrease, has been detected in rural areas (Sicard *et al.*, 2013; Querol *et al.*, 2014); the annual average at rural western Mediterranean sites over the period 2000-2010 was 33 ppb, with a modest trend of -0.22% $year^{-1}$ (Sicard *et al.*, 2013). The Mediterranean Basin is also exposed to frequent African dust intrusions, which can naturally increase the level of suspended particulate matter and nutrient deposition, changing the chemical composition of the atmosphere (Escudero *et al.*, 2005; Marticorena & Formenti, 2013; Àvila & Aguillaume, 2017). This has profound impacts on the biogeochemical cycles of both aquatic and terrestrial ecosystems (Mona *et al.*, 2006), further exacerbating the negative consequences of air pollution and climate change on ecosystem and human health.

In this review, originated as a result of the 1st CAPER*med* (Committee on Air Pollution Effects Research on Mediterranean Ecosystems; http://capermed.weebly.com/) Conference in Lisbon, Portugal, we (i) summarize the current knowledge about atmospheric pollution trends and effects, and their interactions with climate change, in terrestrial ecosystems of the Mediterranean

146 Basin, (ii) identify research gaps that need to be urgently filled, and (iii) recommend future steps.

147 Due to lack of information for other regions within the Mediterranean Basin, we mainly focused

148 our review on studies carried out in south-western European countries (France, Italy, Portugal and

149 Spain). In contrast, we discuss information generated through a variety of experimental approaches

150 (field manipulation experiments, greenhouse studies, open top chambers [OTCs], observational

151 studies, modelling, etc.) from studies carried out in a wide range of representative natural (e.g.,

152 shrublands, grasslands, woodlands and forests) and semi-natural (e.g., *montados* or *dehesas*)

153 ecosystems.

154

Measurement and modelling of atmospheric pollution and deposition

156 Estimating pollutant deposition loadings, particularly dry deposition, still presents important

157 uncertainties and challenges, both in terms of modelling and measurements (Simpson *et al.*, 2014).

158 This is particularly true in studies at small regional scales and in regions with complex topography

159 or under the influence of local emission sources (García-Gómez *et al.*, 2014), which is very often

160 the case in the Mediterranean Basin. Dry deposition in Mediterranean ecosystems can represent

161 the main input of atmospheric N, contributing up to 65-95% of the total deposition (Figure 2b;

162 Sanz *et al.*, 2002; Avila & Rodà, 2012). For example, wet N deposition at the Levantine border of

163 the Iberian Peninsula can be considered low to moderate (2 - 7.7 kg N ha^{-1} yr^{-1}), but total N

164 deposition loads are comparable to more polluted areas in central and northern Europe (10 - 24 kg

165 N ha^{-1} yr^{-1}) when dry deposition is included (Avila & Rodà, 2012). Given that dry deposition is

166 important in the Mediterranean Basin but is also difficult to measure, we should ideally combine

167 modelled dry deposition with wet deposition measures from representative monitoring stations. A

168 recent modelling analysis has also highlighted that mountain ecosystems in Spain, where

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169   monitoring stations are even scarcer, are frequently exposed to exceedances of empirical critical N

170   loads (García-Gómez *et al.*, 2014, 2017). Moreover, mountain areas of the Mediterranean Basin

171   also frequently register very high O_3 concentrations that are not recorded in air quality monitoring

172   networks (Díaz-de-Quijano *et al.*, 2009; Cristofanelli *et al.*, 2015; Elvira *et al.*, under review). This

173   observation should encourage the inclusion of monitoring stations in mountain areas in air quality

174   networks in the Mediterranean Basin to protect these highly valuable and vulnerable ecosystems

175   (García-Gómez *et al.*, 2017). Another important aspect to be considered in both deposition

176   monitoring networks and model-based estimates is the quantification and characterization of

177   ammonium (NH_4^+) and the organic N fraction (Jickells *et al.*, 2013; Fowler *et al.*, 2015). Dissolved

178   organic N (DON) can represent a significant component of wet and dry deposition fluxes but it is

179   often overlooked and not routinely assessed (Mace, 2003; Violaki *et al.*, 2010; Im *et al.*, 2013;

180   Izquieta-Riojano & Elustondo, 2017). However, DON fluxes may have significant implications in

181   terms of critical loads, reaching up to 34-56% of the total N deposition (12 kg DON ha^{-1} yr^{-1}) in

182   Mediterranean agricultural areas (Izquieta-Rojano *et al.*, 2016a). The quantification of temporal

183   trends in air pollution is equally important for evaluating the impact of changing precursor

184   emissions and informing local and regional air quality strategies.

185

186   **Impacts of atmospheric pollution and climate change on natural and semi-natural terrestrial**

187   **ecosystems**

188   The ecological impacts of air pollution (particularly for N deposition and O_3) on natural and semi-

189   natural ecosystems have been primarily studied in the temperate and boreal regions of Europe and

190   North America and, more recently, in steppe and subtropical areas of China (Paoletti, 2006; Xia &

191   Wan, 2008; Bobbink *et al.*, 2010; Ochoa-Hueso, 2017). In contrast, much less is known for

192   Mediterranean Basin ecosystems, which differ from these better-studied ecosystems in critical

193   aspects that justify their separate consideration, such as their much-higher levels of biodiversity

194   (particularly for plants) and their higher-than-average levels of biologically-relevant spatial and

195   temporal environmental heterogeneity, including the characteristic summer drought period

196   (Cowling *et al.*, 1996; Myers *et al.*, 2000). Most studies on the impacts of atmospheric pollution in

197   terrestrial ecosystems from the Mediterranean Basin have been carried out in just a small part of

198   the geographic area (i.e. certain localities in Italy, Portugal and Spain) and have used different

199   experimental design and methodologies (Fig. 1 and Supplementary Table 1). Similarly, instead of

200   taking advantage of the development of statistical methods to integrate responses at the ecosystem

201   level (e.g., structural equation modelling; Eisenhauer *et al.*, 2015), studies have typically focused

202   solely and independently on plants (community or, more frequently, individual species), lichens

203   (community or, again more frequently, individual species) and soil properties (soil

204   biogeochemistry, structure and functioning; Supplementary Table 1). One notable exception to this

205   is NitroMed, a unique network of three comparable N addition experimental sites (Capo Caccia [0

206   and 30 kg N ha^{-1} yr^{-1}], Alambre [0, 40 and 80 kg N ha^{-1} yr^{-1}], and El Regajal [0, 10, 20 and 50 kg

207   N ha^{-1} yr^{-1}]; see Figure 3b, f and h) that is currently using common experimental methodology and

208   structural equation modelling to understand the cause-effect mechanisms that determine changes

209   in gas (CO_2) exchange and litter decomposition and stabilization rates in response to N deposition

210   in semiarid Mediterranean ecosystems (see Ochoa-Hueso and Manrique 2011 and Dias et al. 2014

211   for further details on experimental methodologies). Preliminary results suggest that N deposition

212   increases soil N availability and reduces soil pH which, in turn, has an effect on microbial

213   community structure (lower fungi to bacteria ratio) and overall enzymatic activity, direct

214   responsible for reduced litter decomposition and higher stabilization rates (Lo Cascio *et al.*, 2016).

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215   Similarly, a new coordinated project is looking at the effects of N addition at realistic doses (20

216   and 50 kg N ha-1 yr-1), in conjunction with P, on alpine ecosystems from five National Parks in

217   Spain. Moreover, most of these studies addressed the impact of one global change driver alone

218   (often increased N availability, mostly the N load, or O_3) and so comprehensive studies on the

219   interaction between global change drivers (e.g., air pollution and climate change) are few. However,

220   recent studies have described a heterogeneous response of annual pasture species to O_3 and N

221   enrichment, with legumes being highly sensitive to ozone but not N, while grasses and herbs were

222   more tolerant to O_3 and more responsive to N (Calvete-Sogo *et al.*, 2016). Thus the interactive

223   effects of O_3 and N can alter the structure and species composition of Mediterranean annual

224   pastures via changes in the competitive relationships among species (González-Fernández *et al.*,

225   2013 and references therein; Calvete-Sogo *et al.*, 2014, 2016). Similarly, only a few studies have

226   addressed the impacts on edaphic fauna and above- and below-ground biotic interactions such as

227   mycorrhiza, biological N fixation, herbivory or pollination in ecosystems from the Mediterranean

228   Basin (Supplementary Table 1 and references therein), despite the relevance of ecological

229   interactions to healthy, functional ecosystems (Tylianakis *et al.*, 2008). For example, Ochoa-Hueso

230   *et al.* (2014a) found that edaphic faunal abundance, particularly collembolans, increased in

231   response to up to 20 kg N ha^{-1} yr^{-1} and then decreased with 50 kg N ha^{-1} yr^{-1}, whereas 10 kg N ha^{-}

232   ^{1} yr^{-1} were enough to completely supress soil microbial N fixation (Ochoa-Hueso *et al.*, 2013a).

233   Another notable exception is Ochoa-Hueso (2016), who showed how even low-N addition levels

234   (10 kg N ha^{-1} yr^{-1}) can completely disrupt the tight coupling of the network of ecological

235   interactions in a semiarid ecosystem from central Spain, despite the lack of evident response of

236   most of the individual abiotic and biotic ecosystem constituents evaluated (i.e., soils, microbes,

237   plants and edaphic fauna). Ozone and N soil availability can also alter volatile organic compound

238 (VOC) emissions, and thus biosphere-atmosphere interactions, of some Mediterranean tree and

239 annual pasture species. The consequences of these interactions need to be further studied (Peñuelas

240 *et al.*, 1999; Llusià *et al.*, 2002; Llusia *et al.*, 2014). Therefore, a more comprehensive and

241 integrative experimental approach is urgently needed to fully capture the real consequences of air

242 pollution in the Mediterranean region.

243

244 *Sensitivity of Mediterranean forests to air pollution and climate change*

245 Mediterranean forest ecosystems have naturally evolved cross-tolerance to deal with harsh

246 environmental conditions (Paoletti, 2006; Matesanz & Valladares, 2014). However, climate

247 change, N deposition and O_3 are currently threatening Mediterranean forests in unprecedented and

248 complex manners, with consistent stoichiometric responses to increased N deposition (higher leaf

249 N:P ratios; Sardans *et al.* 2016), but with physiological and growth-related consequences forecasted

250 to vary among the three main tree functional types (i.e., conifers, evergreen broadleaf trees, and

251 deciduous broadleaf trees). As deposition increases, photosynthesis, water use efficiency, and thus

252 growth, often increase in conifers (Leonardi *et al.*, 2012), although under chronic N deposition,

253 other nutrients such as P can become more limiting, counteracting the initial benefits of more N

254 availability (Blanes *et al.*, 2013). Nitrogen deposition could also increase pine mortality rates in

255 response to drought due to a decline of ectomycorrhizal colonization rates, a phenomenon of

256 widespread occurrence in US dryland woodlands (Allen *et al.*, 2010). On the other hand, their low

257 stomatal conductance and their high stomatal sensitivity to vapour pressure deficit and water

258 availability might limit the diffusion of O_3 to the mesophyll (Flexas *et al.*, 2014). Similarly,

259 conservative strategies of water and nutrient-use may also play a key role in allowing conifers to

260 keep a positive balance between assimilation and respiration in response to climate change (Way

13

261 & Oren, 2010). However, O$_3$ exposure might be impairing their ability to withstand other

262 environmental stresses such as those triggered by drought, high temperature and solar radiation

263 (Barnes *et al.*, 2000; Alonso *et al.*, 2001).

264   In contrast, evergreen broadleaf species inhabiting resource-poor ecosystems might be

265 jeopardized by N deposition by shifting biomass partitioning (Cambui *et al.*, 2011) and altering

266 allometric ratios (e.g., leaf area/sap wood or root/leaf biomass), which may have consequences for

267 their ability to deal with water stress, particularly in the context of the characteristic summer

268 drought period and climate change (Martinez-Vilalta *et al.*, 2003; Mereu *et al.*, 2009).

269 Ecophysiological responses to O$_3$ vary from down-regulation of photosystems (Mereu *et al.*, 2009)

270 to reduced stomatal aperture and increased stomatal density (Fusaro *et al.*, 2016) and sluggishness

271 (Paoletti & Grulke, 2005, 2010). However, Mediterranean vegetation usually has efficient

272 antioxidant defences (Nali *et al.*, 2004), which are key factors in O$_3$ tolerance (Calatayud *et al.*,

273 2011; Mereu *et al.*, 2011), and is usually known to be more O$_3$-tolerant than mesophilic broadleaf

274 trees (Paoletti, 2006). Nevertheless, biomass losses and allocation shifts cannot be excluded,

275 especially as a consequence of synergistic effects of N deposition and drought, although local

276 differentiation may result in significant intraspecific tolerance differences (Alonso *et al.*, 2014;

277 Gerosa *et al.*, 2015).

278   Responses of deciduous broadleaf species to N deposition may be modulated by water and

279 background nutrient availability (mainly P) but, in general terms, growth is favoured over storage

280 (Ferretti *et al.*, 2014). In contrast, broadleaf tree species are highly sensitive to climate change,

281 particularly to the combination of drought and increased temperature (Lopez-Iglesias *et al.*, 2014),

282 which also suggests relevant interactions between air pollution and climate change. In this

283 direction, De Marco *et al.* (2014) predicted that crown defoliation will increase in Mediterranean

284   environments due to drought events and higher temperatures by 2030, a phenomenon that could be

285   exacerbated by excessive N. Deciduous broadleaf species also have lower capacity to tolerate

286   oxidative stress than evergreen broadleaf species due to traits such as thinner leaves and higher

287   stomatal conductance (Calatayud *et al.*, 2010). Gas exchange and antioxidant capacity in deciduous

288   broadleaves are, therefore, generally more affected by high O_3 concentrations than in evergreen

289   broadleaves (Bussotti *et al.*, 2014). Based on their levels of visible foliar injury and expert

290   judgement, deciduous broadleaf species range from highly to moderately sensitive species such as

291   *Fagus sylvatica* and *Fraxinus excelsior*, respectively (Baumgarten *et al.*, 2000; Tegischer *et al.*,

292   2002; Gerosa *et al.*, 2003; Deckmyn *et al.*, 2007; Paoletti *et al.*, 2007; Sicard *et al.*, 2016), to O_3-

293   tolerant species like some *Quercus* species (*Q. cerris*, *Q. ilex* and *Q. petraea*; Gerosa *et al.* 2009;

294   Calatayud *et al.* 2011; Sicard *et al.* 2016).

295   Relatively little is known about the effects of O_3 on annual, perennial and woody understory

296   vegetation of Mediterranean forest ecosystems. Under experimental conditions, some species

297   characteristic of the annual grasslands associated with *Q. ilex dehesas* have high O_3 sensitivity.

298   Interestingly, N fixing legumes, of higher nutritional value, are more O_3 sensitive than grasses

299   (Bermejo *et al.*, 2004; Gimeno *et al.*, 2004), particularly in terms of flower and seed production

300   (Sanz *et al.*, 2007), which could affect their competitive fitness and, ultimately, reduce the

301   economic value of the pasture. Nitrogen availability can partially counterbalance O_3 effects on

302   aboveground biomass when the levels of O_3 are moderate, but O_3 exposure reduces the fertilization

303   effect of higher N availability (Calvete-Sogo *et al.*, 2014). Anyhow, given that O_3 levels are higher

304   in summer, when herbaceous species are dormant, Mediterranean species that are summer-active

305   such as pines and oaks are more likely to be directly affected by O_3 than forbs and grasses. This

306   suggests that the seasonality of O_3 concentrations as well as plant phenology and functional type

307   must be considered if we are to fully understand the consequences of air pollution on the highly

308   diverse Mediterranean plant communities. A unique ozone FACE (free air controlled experiment)

309   is now available in the Mediterranean Basin (Figure 3) to help fill this gap (Paoletti et *al.*, in

310   preparation).

311

312   *Role of environmental context in the response of biodiversity and C sequestration*

313   The local abiotic (e.g., climate, soil properties) and biotic (e.g., vegetation type, community

314   attributes, etc.) contexts are known to modulate ecosystem responses to environmental drivers at

315   different temporal and spatial scales (Bardgett *et al.*, 2013). Given that plant biodiversity at the

316   regional ($10\text{-}10^6$ km^2) and local (< 0.1 ha) scales in Mediterranean ecosystems ranks among the

317   highest in the world (Cowling *et al.*, 1996), this is a particularly relevant aspect for the region.

318   Various studies in Mediterranean ecosystems have shown that increased N availability may have a

319   positive (Pinho *et al.*, 2012; Dias *et al.*, 2014), negative (Bonanomi *et al.*, 2006; Bobbink *et al.*,

320   2010) or even no effect (Dias *et al.*, 2014) on plant species richness, which is probably due to

321   cumulative effects and modulating factors such as the ecosystem type, the initial N status of the

322   system, the dominant form of mineral N in the soil (NH_4^+, NO_3^-), and/or the N form added. Positive

323   effects on species richness, however, have only been observed in areas characterized by strong

324   environmental stress and low nutrient availability (e.g., open arid and semiarid Mediterranean

325   ecosystems) and are often associated with an increase in nitrophytic and weedy species (Bobbink

326   *et al.*, 2010; Pinho *et al.*, 2011; Dias *et al.*, 2014). The presence and density of shrubs, as well as

327   the availability of inorganic phosphorus (P) and other macro and micronutrients, can also modulate

328   the response of the herbaceous vegetation to N addition and plant invasion in semiarid

329   Mediterranean areas (Ochoa-Hueso *et al.*, 2013b; Ochoa-Hueso & Stevens, 2015). For example,

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330 Ochoa-Hueso & Manrique (2014) found that N addition increased the nitrophytic element,

331 particularly native crucifers, only when these species were present in the seed bank in relevant

332 densities and there was sufficient P, whereas a closed scrub vegetation is known to be less

333 susceptible to invasion by N-loving species than open shrublands, woodlands and grasslands (Dias

334 *et al.*, 2014). The role of soil nutrient availability, typically lower than in other Mediterranean-type

335 ecosystems such as those from Chile (Cowling *et al.*, 1996), in the ecosystem response to extra N

336 can also be linked to induced nutrient imbalances, particularly N in relation to P, and therefore to

337 an alteration of ecosystem stoichiometry (Ochoa-Hueso *et al.*, 2014b; Sardans *et al.*, 2016).

338   The behaviour of terrestrial ecosystems as a global C sink or source under increased N

339 deposition or O_3 pollution scenarios is currently a research hot-topic and is of paramount

340 importance for the mitigation of climate change (Felzer *et al.*, 2004; Reich *et al.*, 2006; Pereira *et

341 al.*, 2007). Recent studies have suggested that seasonally water-limited ecosystems, such as those

342 typically found in the Mediterranean Basin, may have a disproportionately big role in the inter-

343 annual C sink-source dynamics at the global scale due to higher C turnover rates (Poulter *et al.*,

344 2014); this is attributed to their large inter-annual climatic variability, with unusually wet years

345 contributing to strengthen the terrestrial C sink but where multiple processes like fire or rapid

346 decomposition could result in a rapid loss of most of the accumulated C. These aspects are,

347 however, still poorly understood in Mediterranean ecosystems, where different studies have

348 reported contrasting results (Ochoa-Hueso *et al.*, 2013a, 2013c; Ferretti *et al.*, 2014). In

349 Mediterranean ecosystems, ecosystem C storage should, therefore, be evaluated in terms of altered

350 abundance and patterns of rainfall (both within and between years) (Pereira *et al.*, 2007), in relation

351 to the levels of N saturation (NO_3^-) and toxicity (NH_4^+) in soil (Dias *et al.*, 2014), as well as other

352 site-dependent characteristics such as dominant vegetation, soil type (texture and pH), and stand

353 history and age (Ferretti *et al.*, 2014). Experimental and observational field studies suggest that, at

354 least in the short-term, seasonal and inter-annual dynamics may override any potential effect of

355 atmospheric N pollution, despite potential cumulative negative impacts in the long-term due to an

356 overall decline in ecosystem health (Ochoa-Hueso *et al.*, 2013c; Ferretti *et al.*, 2014).

357 Although within the Mediterranean Basin there is still a large gap in the knowledge of the

358 impacts of atmospheric pollution and climate change on natural and semi-natural ecosystems, taken

359 together, all the scattered information available suggests the particularly key role of spatial and

360 temporal environmental heterogeneity, biotic interactions, and ecosystem stoichiometry in

361 mediating the ecosystem response to air pollution.

362

363 *Critical loads and levels*

364 The concepts of critical loads and critical levels were developed within the United Nation

365 Economic Commission for Europe (UNECE) Convention on Long-Range Transboundary Air

366 Pollution (CLRTAP) for assessing the risk of air pollution impacts to ecosystems and defining

367 emission reductions. This tool is commonly used to anticipate negative effects of air pollution and,

368 therefore, to protect ecosystems before the changes become irreversible. The derivation of

369 empirical critical loads for nutrient N is based on experimental activities performed on different

370 vegetation types and they are assigned to habitat classes, while the derivation of NH_3 and NO_x

371 critical levels is based on the responses of broad vegetation types such as higher plants or lichens

372 and bryophytes. The pan-European critical level for atmospheric NH_3 is currently set at an annual

373 mean of 1 $\mu g\ m^{-3}$ for lichens and bryophytes and 3 $\mu g\ m^{-3}$ for higher plants, while the NO_x critical

374 level for all vegetation types is an annual mean of 30 $\mu g\ m^{-3}$ (CLRTAP, 2011). Although some

modelling approaches exist to define N critical loads, the identification of empirical critical loads
is recommended for Mediterranean ecosystems due to its particularities such as co-occurrence with
other pressures and high seasonality (de Vries *et al.*, 2007; Fenn *et al.*, 2011). Empirical critical
loads of N for European-Mediterranean habitats have only been proposed for four ecosystems: (1)
Mediterranean xeric grasslands (EUNIS [European Nature Information System] E 1.3), 15-25 kg
N ha^{-1} yr^{-1}; (2) Mediterranean maquis (F5), 20-30 kg N ha^{-1} yr^{-1}; (3) Mediterranean evergreen
(*Quercus*) woodlands (G 2.1), 10-20 kg N ha^{-1} yr^{-1}, and (4) Mediterranean *Pinus* woodlands (G
3.7), 3-15 kg N ha^{-1} yr^{-1} (Bobbink & Hettelingh, 2011). However, these critical loads are based on
very little information and are thus classified as expert judgement. Similarly, NH_3 critical levels
have only been set for Mediterranean evergreen woodlands and dense holm oak forests. Critical
levels of atmospheric NH_3 of < 1.9 and 2.6 µg m^{-3} have been estimated for evergreen woodlands
surrounded by intensive agricultural landscapes (Pinho *et al.*, 2012; Aguillaume, 2015), while for
evergreen woodlands under little agricultural influence but strong oceanic influence, the critical
level was estimated to be 0.69 µg m^{-3} (Pinho *et al.*, 2014). Nevertheless, the N critical loads and
NH_3 critical levels for many European-Mediterranean ecosystems remain unstudied, despite their
relevance for protecting relatively undisturbed and oligotrophic ecosystems. Therefore, long-term
manipulation experiments across a range of typical Mediterranean terrestrial ecosystems are
desperately needed to obtain a more complete set of reliable empirical critical N loads and levels
for the Mediterranean Basin (Bobbink *et al.*, 2010; Bobbink & Hettelingh, 2011). Ozone critical
levels have also been proposed for the protection of natural vegetation at European level for two
vegetation types, forests and semi-natural vegetation (CLRTAP, 2011). The new flux-based O_3
critical levels allow species-specific physiological conditions and O_3 uptake mechanisms to be

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397   included considering the particularities of Mediterranean species. Interestingly, multiple studies

398   performed with Mediterranean tree species recommend higher O_3 critical levels for the protection

399   of Mediterranean forests than the values currently accepted (Calatayud et al., 2011; Alonso et al.,

400   2014; Gerosa et al., 2015). The possible definition of different O_3 critical levels for different

401   biogeographical regions or vegetation types is currently under analysis within the Convention

402   (CLRTAP, 2011).

403

404   *Cryptogams as indicators of the impact of air pollution and climate change*

405   Lichens and bryophytes (i.e., cryptogams), very often used in the definition of critical loads and

406   levels, are important components of the vegetation in Mediterranean ecosystems. These organisms

407   are key drivers of ecosystem properties (soil aggregation and stability) and processes (C and N

408   fixation and nutrient cycling), particularly in the case of biological soil crusts (hereafter biocrusts),

409   a functionally-integrated association of cyanobacteria, protists, fungi, mosses and lichens

410   inhabiting the first millimetres of soil (Cornelissen *et al.*, 2007; Maestre *et al.*, 2011). Cryptogams

411   are usually extremely sensitive to environmental changes and so they often provide early-warning

412   indicators of impacts before any other constituent of the ecosystem, particularly in the case of N

413   (Pardo *et al.*, 2011; Munzi *et al.*, 2012). For example, mosses have been used in N deposition

414   surveys under the ICP-Vegetation framework (Harmens *et al.*, 2014). The results showed that N

415   concentration in mosses can potentially be used as an indicator of total atmospheric N deposition.

416   Similarly, Root *et al.* (2013) showed that lichens can be a suitable tool for estimating throughfall

417   N deposition in forests. However, the relationship between N deposition and tissue N concentration

418   can also be affected by environmental factors such as local climate and the form of N deposition.

419   Mosses and lichens have been instrumental to the evaluation of the impacts of global change

420   drivers on temperate and boreal ecosystems (e.g., Arróniz-Crespo *et al.* 2008), though the number

421   of studies carried out in Mediterranean ecosystems is very limited. Recent studies have, however,

422   reported significant impacts of increased N deposition on Mediterranean biocrust and epiphytic

423   communities. For example, two studies carried out in the Iberian peninsula found higher tissue N

424   content and a shift from N to P limitation in the terricolous moss *Tortella squarrosa* (=*Pleurochaete*

425   *squarrosa*; Ochoa-Hueso & Manrique 2013; Ochoa-Hueso *et al.* 2014a). Similarly, an alteration of

426   physiological and chemical responses in lichen transplants (Branquinho *et al.*, 2010; Paoli *et al.*,

427   2010, 2015) and a shift in epiphytic lichen communities from oligotrophic-dominated to

428   nitrophytic-dominated species have also been reported in Portugal (Pinho *et al.*, 2008, 2009) and

429   Spain (Aguillaume, 2016). Recent studies have also observed a change in the isotopic N

430   composition of mosses due to the impact of N from fuel combustion sources (shift to more positive

431    $\delta^{15}N$ signature) and agricultural activities (shift to more negative $\delta^{15}N$ signature; Delgado *et al.*,

432   2013; Varela *et al.*, 2013; Izquieta-Rojano *et al.*, 2016b). Cryptogam traits (e.g., morphology,

433   anatomy, life form) are also strongly connected to water availability. For example, mosses from

434   dry habitats are organized in dense cushions, naturally retaining water by capillarity and

435   dehydrating slowly, whereas mosses from moist habitats have a less dense morphology and require

436   the activation of specific mechanisms to survive during dry periods (Arróniz-Crespo *et al.*, 2011;

437   Cruz de Carvalho *et al.*, 2011, 2012, 2014). Similarly, lichen growth form and photobiont type have

438   been shown to be relevant traits in the response to water availability in Mediterranean areas

439   (Concostrina-Zubiri *et al.*, 2014; Matos *et al.*, 2015). Cryptogam traits related to water availability

440   could, therefore, be equally effective biomarkers to detect climate-induced hydrological changes

441   in Mediterranean ecosystems but the application of biomonitoring techniques using cryptogams in

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442 the Mediterranean region may be complicated by the fact that cryptogam species are

443 simultaneously exposed to both severe water restriction and pollution, and some biomarkers (e.g.,

444 ecophysiological responses) are similarly affected by both stress factors (Pirintsos *et al.*, 2011).

445 Thus, we need to disentangle the multiple environmental drivers (Munzi *et al.*, 2014a), possibly by

446 integrating physiological and ecological data to understand the specific response mechanisms to

447 different ecological parameters and environmental changes (Munzi *et al.*, 2014b).

448

449 *Anticipating global tipping points using ecological indicators*

450 The fact that ecosystem responses to air pollution and climate change are very often non-linear

451 may complicate the use of bioindicators in the Mediterranean Basin. Non-linear dynamics often

452 manifest in the form of tipping points, defined as ecosystem thresholds above which a larger-than-

453 expected change happens, shifting ecosystems from one stable state to another stable state (Scheffer

454 & Carpenter, 2003). Due to its climatic peculiarities, tipping points may be particularly relevant

455 for the Mediterranean Basin. One example is the ability of soils to store extra mineral N. Above a

456 certain N deposition value, N-saturated soils will start leaching N down into the soil profile. This

457 excessive N can also accumulate as inorganic N in seasonally dry soils and be leached by surface

458 flows that, as in the case before, will eventually reach and, therefore, pollute aquifers and

459 watercourses (Fenn *et al.*, 2008). Another relevant example is related to increased fire risk due the

460 accumulation of highly flammable leaf litter, particularly from exotic grasses, as a consequence of

461 N deposition; above a certain N deposition threshold the probability of a fire to occur increases

462 exponentially, priming the ecosystem for a state change (Rao *et al.*, 2010).

463 Despite the potential prevalence of tipping point-like dynamics in Mediterranean

464 ecosystems in response to air pollution and climate change, we are not aware of any vegetation-

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465   based tools available to predict ecosystem thresholds in the Mediterranean Basin context. A notable

466   exception is the work by Berdugo *et al.* (2017), who suggested that changes in the spatial

467   configuration of drylands may be an early-warning indicator of desertification. However, we

468   suggest that if we are to aim for universal indicators of environmental change (i.e., at wide

469   geographical ranges) and to account for the role of the environmental context as a driver (i.e., across

470   ecosystem types), functional trait-based approaches (e.g., functional diversity and community

471   weighted mean trait values [CWM]) should be preferred over other widely used indicators,

472   including species richness (Jovan & McCune, 2005; Valencia *et al.*, 2015). Functional diversity

473   and CWM are independent of species identity and may be functionally linked to the environmental

474   variable of interest (e.g., oligotrophic species, nitrophytic species, or subordinate species

475   responding to eutrophication, species-specific leaf litter traits, etc.). More research is, however,

476   needed to integrate these concepts (ecological indicators, ecological thresholds and functional

477   diversity) in a meaningful way.

478

479   **Linking functional diversity to the provision of ecosystem services**

480   The universal applicability and ecological relevance of the functional trait diversity concept makes

481   it equally valuable to establish possible connections between global environmental change and the

482   loss of ecosystem services. Ecosystem services that may be impaired by air pollution and climate

483   change and that may be particularly associated with changes in functional diversity include C

484   sequestration, soil fertility and nutrient cycling and pollination, among many others. However,

485   research on the link between functional diversity and ecosystem services is lagging behind in the

486   Mediterranean region where only a few controlled experiments exist (Hector *et al.*, 1999; Pérez-

487   Camacho *et al.*, 2012; Tobner *et al.*, 2014; Verheyen *et al.*, 2016), species trait databases are still

488 incomplete (Gachet *et al.*, 2005; Paula *et al.*, 2009), and field surveys along climatic and air

489 pollution gradients are only recently starting to emerge (De Marco *et al.*, 2015; Sicard *et al.*, 2016).

490   The few studies available within the Mediterranean Basin context have shown that N

491 deposition has already induced changes in functional diversity of epiphytic lichens along a NH_3

492 deposition gradient in Mediterranean woodlands, with a drastic increase and decrease of nitrophytic

493 and oligotrophic species, respectively, (Pinho *et al.*, 2011). Similarly, a continuous increase of

494 nitrophytic species (plants, lichens, mosses) has been detected in the Iberian Peninsula for the

495 period 1900-2008 using the Global Biodiversity Information Facility (GBIF) database (Ariño *et*

496 *al.*, 2011). Increased N availability in nutrient-poor ecosystems like Mediterranean maquis can also

497 alter plant functional composition (e.g., higher proportion of short-lived species in relation to

498 summer semi-deciduous and evergreen sclerophylls), leading to changes in litter amount and

499 quality (e.g. higher proportion of evergreen sclerophyll litter from affected shrubs and a general

500 increase in lignin and N content in litter and a decrease in lignin/N ratio) and microbial community

501 (e.g., reduction in biomass and activity), thus affecting nutrient cycling (an ecosystem function)

502 and, therefore, soil fertility (including soil C accumulation, an ecosystem service) (Dias *et al.*, 2010,

503 2013, 2014). In another study, Concostrina-Zubiri *et al.* (2016) showed that livestock grazing

504 greatly affected the abundance and functional composition of moss–lichen biocrusts in a

505 Mediterranean agro-silvo-pastoral system, with direct negative consequences on microclimate

506 regulation and other ecosystem processes (CO_2 fixation, habitat provision and soil protection). This

507 also affected the cork-oak regeneration processes, one of the traditional and most economically

508 valuable services in these systems. Given the negative impacts of air pollution on cryptogamic

509 biocrusts, a similar effect of air pollution on the cork-oak regeneration processes mediated by

510 biocrusts might be expected.

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511

Common experimental design, data sharing and global networks

The understanding of the ecological impacts of pollution and climate change across the Mediterranean region would improve through co-ordinated efforts and networks, which could take several forms. One possible approach is the use of large-scale regional surveys on existing pollution gradients representative of the current range of pollution loads (e.g., from big cities and/or extensive agricultural areas to their periphery). This approach was successfully used to survey 153 acid grasslands in ten countries across the Atlantic biogeographic zone of Europe (significantly less biodiverse than their Mediterranean counterparts) (Stevens *et al.*, 2010), where each partner surveyed sites in their local area according to an agreed protocol. Other networks have been successful using experimental approaches. For example, the Nutrient Network (NutNet) is a global network of over 90 sites following a common experimental protocol for nutrient addition and grazing (Borer *et al.*, 2014). Similarly, the previously presented NitroMed network, originated within the CAPER*med* platform, aims at using the same experimental protocols to integrate results from three comparable experiments in semiarid Mediterranean ecosystems. Other experimental networks have not used common experimental protocols, but through coordinated analyses have added value to individual experiments (Phoenix *et al.*, 2012). Co-ordinated experimental networks (e.g., low-cost N addition experiments) bring many advantages such as the ability to assess the general applicability of results, additional statistical power resulting from well-established and robust statistical methods (e.g., linear mixed effects models, hierarchical Bayesian models, structural equation modelling), and opportunities to explore interactions with other natural and human-caused gradients such as climate, ecosystem and soil type, land use, atmospheric pollution (including O_3 gradients), etc. They can also provide support and collaboration for individual

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534   scientists. An inventory of the existing sites with manipulation experiments in the Mediterranean

535   Basin would provide added value to the individual sites through the implementation of common

536   protocols and experiments.

537   In the Mediterranean region, another path to follow may be to build upon existing research

538   and to participate more in already existing large-scale initiatives, in which the Mediterranean

539   research community is not particularly well-represented. For example, interacting with the

540   International Long Term Ecological Research (ILTER) network or with the International

541   Cooperative Programme (ICP), established under the United Nation Economic Commission for

542   Europe (UNECE) "Convention on Long-Range Transboundary Air Pollution" (CLRTAP) that

543   includes several initiatives such as ICP Forest, ICP-Vegetation, and ICP-IM, would facilitate the

544   collection of large-scale spatial and temporal data series. Cooperation with other more specific

545   networks like NitroMed (N deposition), ICOS (C cycle), and GLORIA (Alpine environments)

546   would also help to establish a wider and more collaborative research community focused on air

547   pollution impacts in Mediterranean terrestrial ecosystems.

548   The need of more coordination and investment to better understand the Mediterranean

549   responses to climate change and air pollution has already been acknowledged by several groups of

550   scientists both at the European (e.g. CAPER*med*) and global scales (e.g. MEDECOS). These

551   groups not only represent suitable arenas to discuss scientific results, but can also provide leading

552   members able to manage the above-mentioned research and networking activities. However, all the

553   above mentioned presented approaches require considerable funding and determined political

554   support to foster the exchange of information and best practices across the entire Mediterranean

555   region and, thus, to promote the development of concrete projects and initiatives. In this context,

556   the European Commission, through funding programs like Horizon 2020, could and should have,

557 in our opinion, a pivotal role in supporting research projects (as it happened with the CIRCE

558 project) and to provide the logistic means for transferring the scientific knowledge to the society.

559 Increasing awareness about the effects of climate change and pollution among stakeholders

560 and society is encouraging the development of several European and Pan-European Programs (e.g.

561 UNECE/ICP, Climate-ADAPT). One important step towards the coordinated action of the

562 Mediterranean-basin countries in relation to Adaptation to climate change was the creation of "The

563 Union for the Mediterranean Climate Change Expert Group" (UfMCCEG), a partnership

564 promoting multilateral cooperation between 43 countries (28 EU Member States and 15

565 Mediterranean countries). These initiatives show that opportunities do exist for countries to make

566 progress. Due to campaigning, and partially because of the considerable losses from extreme

567 weather events in recent years, public awareness in Mediterranean countries about risks associated

568 with climate and air pollution increased. Governments and organisations at the EU level, national

569 and sub-national level, have developed or are in the process of developing adaptation strategies.

570 Therefore, there is an opportunity to make progress by actively engaging actors from all sections

571 of the Mediterranean society.

572 **Conclusions and future directions**

573 The comparatively fewer number of studies on the effects of air pollution and its interactions with

574 climate change on terrestrial ecosystems from the Mediterranean Basin is particularly noteworthy

575 considering the high biodiversity, cultural value, and unique characteristics of this region such as

576 high O_3 levels, dominance of dry deposition over wet deposition, and long dry periods. Therefore,

577 we emphasize the need to urgently implement common and coordinated research and experimental

578 platforms in the Mediterranean region along with wider and more representative environmental

579 monitoring networks. In particular, a robust connection between N deposition monitoring networks

580 and modelling estimates is crucial. Ideally, monitoring and assessment programs should regularly

581 include a set of common biomonitors such as local and/or transplanted cryptogams to identify local

582 pollutant sources and, thus, help refine pollutant deposition maps (physiological indicators) and to

583 provide early warning indication of potential critical thresholds (community shifts). Only by filling

584 these gaps can the scientific community reach a full understanding of the mechanisms underlying

585 the combined effects of air pollution and climate change in the Mediterranean Basin and,

586 consequently, provide the science-based knowledge necessary for the development of sustainable

587 environmental policies and management techniques and the implementation of effective mitigation

588 and adaptation strategies. Finally, CAPERmed, a bottom-up initiative (from the researchers to the

589 institutions), can be the longed-for catalyst that brings the Mediterranean community together and,

590 therefore, represents an excellent opportunity to make all this happen.

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1065 **Figure 1.** Modeled nitrogen deposition for the Mediterranean region based on the European

1066 Monitoring and Evaluation Programme (EMEP) model at 0.1º-0.1º longitude-latitude resolution

1067 (EMEP MSC-W chemical transport model [version rv4.7; www.emep.int]). Modelled N deposition

1068 is based on 2013 emissions data. (a) Total N deposition (oxidized + reduced; dry + wet), (b)

1069 percentage of dry deposition, (c) percentage of wet deposition, (d) percentage of oxidized

1070 deposition and (e) percentage of reduced deposition.
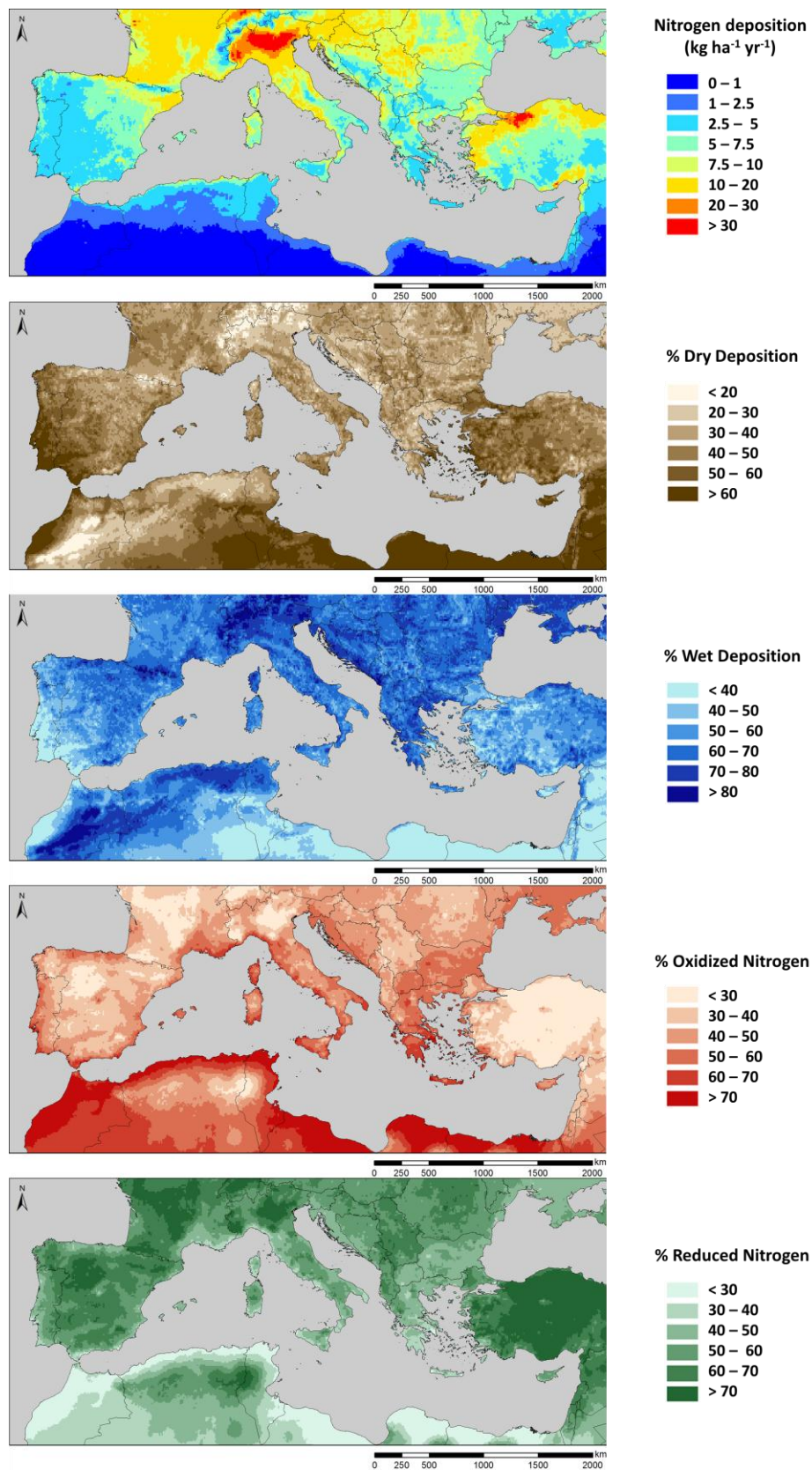
1072 **Figure 2.** (a) Mean annual precipitation (MAP) and (b) temperature (MAT) for the year range between 1960-1990. Projected (c)

1073 MAP and (d) MAT for the year 2070 based on predictions from the CCSM4 model considering the RCP 8.5 (no mitigation of

1074 emissions) IPCC5 scenario. Data obtained from http://www.worldclim.org/version1 (Hijmans *et al.*, 2005).

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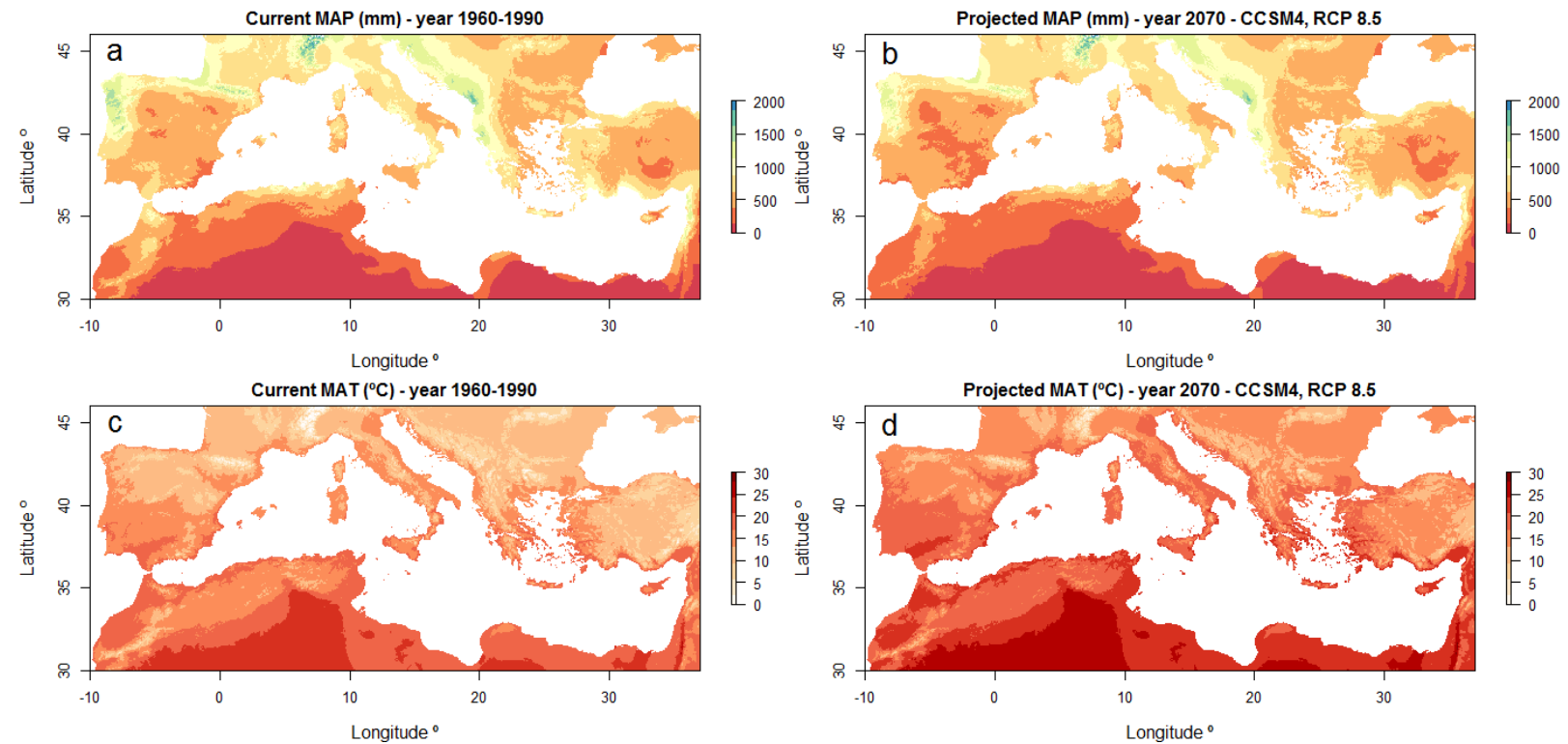
Figure 3. Examples of terrestrial ecosystems and experimental facilities set up to investigate the effects of air pollution and climate change in the Mediterranean Basin (see Supplementary Table 2 for details): a) Companhia das Lezírias, Samora Correia, Portugal; b) Alambre, Serra da Arrábida, Portugal; c) Herdade da Coitadinha, Barrancos, Portugal; d) Alto de Guarramillas, Madrid, Spain; e) La Higueruela, Toledo, Spain; f) El Regajal, Madrid, Spain; g) Tres Cantos, Madrid, Spain; h) Capo Caccia, Sardinia, Italy; i) La Castanya, Spain; j) Ozone FACE (Free-Air Controlled Exposure) facility, Florence, Italy; k) Fontblanche, Provence, France.

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1087 **Figure 4.** The biomonitoring chain: from the source of stress to ecological impacts. Measurements

1088 closer to the source of stress (e.g. bioaccumulation of pollutants) have a stronger link to source

1089 attribution, provide an account of exposure, and can be seen as an early warning system for

1090 potential impacts. On the other hand, biological effects (biomarkers) and species-based

1091 measurements commonly have a close link to impacts on the ecosystem but can have a weaker link

1092 to source attribution. Dark frame indicates those levels and measurements most commonly

1093 considered in biomonitoring studies.

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