1 **Human impacts and aridity differentially alter soil N availability in drylands worldwide**

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99 **Abstract**

100 **Aim** Although very likely to co-occur in the future, it is largely unknown how simultaneous

101 increases in aridity and anthropogenic disturbances will influence the N cycle in dryland soils, the
102 largest terrestrial biome on the planet. Climate and human impacts are changing the inputs to, and
103 losses from, the nitrogen in terrestrial ecosystems. However, our knowledge of how the interaction
104 between these drivers will affect the concentration of available N for plants and microorganisms as
105 well as the dominance of N forms is still scarce and no study has yet explored these interactive
106 effects on the N cycle at global scale.

107 **Location** 224 dryland sites from all continents except Antarctica widely differing in their
108 environmental conditions (from arid to dry-subhumid sites) and human influence (based on distance
109 to towns and roads and population size).

110 **Methods** Using a standardized field survey, we measured the plant cover, aridity, human impacts
111 (i.e., proxies of land uses and air pollution), key biophysical variables (i.e., pH, texture and plant
112 cover) as well as six N cycle important variables: total N, organic and inorganic N and N
113 mineralization rates. We use structural equation modeling to assess the direct and indirect effects of
114 aridity and human impacts together with key biophysical variables on the N cycle.

115 **Results** Human impacts increased the concentration of total N, while aridity decreased it. The
116 effects of aridity and human impacts on the N cycle were spatially disconnected, which may favor
117 N scarcity in the most arid areas and promote N accumulation in the least arid areas. Both
118 increasing aridity and human impacts will enhance the dominance of inorganic N forms.

119 **Main Conclusions** Our findings provide evidence that human impacts will promote the
120 accumulation of N in dryland soils worldwide, while the opposite effect is observed from increasing
121 aridity. Interestingly, we found that these two global change drivers are spatially disconnected in
122 drylands, favoring N losses in the most arid, and accumulation in the least arid ecosystems. Our
123 analyses suggest that both increasing aridity and human impacts will enhance the relative
124 dominance of inorganic N in drylands soils which may negatively impact key ecosystem functions
125 and services at the global scale.

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127 **Keywords:** Aridity, Human impacts, Global change, N cycle, Mineralization,
128 Depolymerization.

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134 **Introduction**

Human activities such as grazing, fertilization, intensive agriculture and fossil fuel combustion are changing the inputs to, and losses from, the nitrogen (N) cycle in terrestrial ecosystems globally (Vitousek *et al.*, 1997; Cui *et al.*, 2013). Anthropogenic N inputs have already doubled the total amount of N fixed naturally by terrestrial and aquatic ecosystems. Current annual rates of both organic and inorganic N deposition are about 124 Tg N per year (Gruber & Galloway, 2008; Schlesinger, 2009; Cornell, 2011). Human pressure on the N cycle is expected to increase during this century because of the predicted increases in global population by 36% over the next 40 years (Charles *et al.*, 2008) and the intensification of land use required to support their demand for food (OECD-FAO 2011), which is estimated to increase by 70-100% by 2050 (World Bank, 2008). For example, human impact such as N deposition derived from fossil fuel combustion and fertilizer production is increasing the availability of N (particularly in inorganic forms) in terrestrial ecosystems (Cui *et al.*, 2013; Gruber & Galloway, 2008; Schlesinger, 2009).

Paralleling the increase of N inputs derived from human activities is an increase in aridity, predicted to increase the total area of drylands (arid, semi-arid and dry-subhumid ecosystems) globally by 10% by the end of this century (Feng & Fu, 2013). Increasing aridity has been predicted to reduce soil N availability in drylands globally and to reduce the pools of organic N in these ecosystems (Schlesinger *et al.*, 1990; Delgado-Baquerizo *et al.*, 2013). These changes are predicted to exacerbate processes leading to land degradation and desertification in drylands, which are estimated to affect more than 250 million people, mostly living in developing countries (Reynolds *et al.*, 2007).

Human (i.e., air pollution and changes in land use) and climate change impacts are key drivers of ongoing global environmental change (Gruber & Galloway, 2008; Schlesinger, 2009; Canfield *et al.*, 2010; Liu *et al.*, 2010; Bai *et al.*, 2013), and are interrelated in complex ways. These global change drivers may act in opposition, or interact to accelerate their effects on natural communities. The combined impacts derived from human activities and climate change may create a more arid environment that is also characterized by reduced biological control of the N cycle (as explained in Schlesinger *et al.*, 1990). For instance, direct anthropogenic-driven disturbances (e.g. overgrazing) and increases in aridity may have negative impacts on plant growth in drylands (Gruber & Galloway, 2008; Delgado-Baquerizo *et al.*, 2013), thereby reducing inputs of organic N in these ecosystems. The human impacts of N cycle have been largely studied at local scale. For example, Baker *et al.*, (2001) concluded that in Phoenix, the urban and agricultural components of the ecosystem were an order of magnitude higher than inputs to the desert, increasing the amount of N in soil and groundwater pools and promoting losses to rivers. Similarly, nutrient enrichment derived from human activities has been also observed to locally enhance N mineralization in the

169 Sonora desert (Hall *et al.*, 2011). However, little is known on how the interaction between
170 increasing aridity and human impacts will affect the concentration of available N for plants and
171 microorganisms as well as the dominance of N forms and no study has yet explored these
172 interactive effects on the N cycle in global drylands.

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174 Drylands form the largest terrestrial biome on Earth and support over 38% of its population
175 (Reynolds *et al.*, 2007; Schimel, 2010). Nitrogen is, after water, the most important factor limiting
176 net primary production and organic matter decomposition in these areas (Robertson & Groffman,
177 2007; Schlesinger & Bernhardt, 2013). The N cycle is therefore crucial for ecosystem functioning
178 and the provision of ecosystem services in these areas (Robertson & Groffman, 2007; Schlesinger
179 & Bernhardt, 2013; Compton *et al.*, 2011). Knowing how direct and indirect effects from climatic
180 (i.e., aridity), biophysical (i.e., soil texture, pH and plant cover) and anthropogenic (i.e., human-
181 induced climate change, air pollution and land use changes) drivers jointly impact the N cycle is
182 crucial if we are to improve our ability to predict the ecological consequences of climate change for
183 terrestrial ecosystems (Schlesinger *et al.*, 1990, Gruber & Galloway, 2008; Chen *et al.*, 2013).

184 We conducted a global mensurative study of 224 field sites from all continents except Antarctica to
185 evaluate how aridity and human impacts, together with biotic (plant cover) and abiotic (soil texture
186 and pH) factors, will affect total N, dissolved organic N, ammonium and nitrate concentrations,
187 dissolved organic-to-inorganic N (DON:DIN) ratio and the potential net mineralization rate of
188 dryland soils. These variables were selected because they are good proxies of N availability and
189 dominance of N forms within soils (Schimel & Bennett, 2004; Delgado-Baquerizo & Gallardo,
190 2011). We hypothesized that: i) soil total N concentration would be enhanced by human impacts
191 (estimated indirectly using proxies) and decline with aridity (Delgado-Baquerizo *et al.*, 2013); and
192 ii) aridity and human impacts will negatively affect the biological control of the N cycle (e.g.,
193 reducing plant cover), resulting in an increasing dominance of inorganic N forms and processes
194 (i.e., mineralization) in dryland soils (Schlesinger *et al.*, 1990).

196 **Material and Methods**

197 Study area

198 This study was restricted to dryland ecosystems, defined as regions with an aridity index (AI =
199 precipitation/potential evapotranspiration) between 0.05 and 0.65 (UNEP 1992). Original field data
200 were collected at 224 sites located in 16 countries from all continents except Antarctica. The sites
201 surveyed encompass a wide variety of vegetation types typically found in drylands, including
202 grasslands, shrublands, savannas, dry seasonal forests and open woodlands dominated by trees.

203 Mean annual precipitation and temperature of the study sites ranged from 66 to 1219 mm and from
204 -1.8 to 27.8°C, respectively. See Maestre *et al.*, (2012) for additional details on the study sites.

205 Climatic, abiotic, plant and nitrogen variables measured

206 Data collection was carried out between February 2006 and December 2010 according to a
207 standardized sampling protocol. The cover of vascular plants at each site was measured using four
208 30-m transects and the line-intercept method, as described in Maestre *et al.*, (2012). The coordinates
209 of each plot were recorded *in situ* with a portable Global Positioning System, and were standardized
210 to the WGS84 ellipsoid for visualization and analyses. Aridity (1-aridity index) was estimated using
211 data from the Worldclim global database (Hijmans *et al.*, 2005). Soils (0-7.5 cm depth) were
212 sampled during the dry season under the canopy of the dominant perennial plants, and in open
213 plant-free areas (10-15 samples were sampled per site, over 2600 samples in total). After field
214 collection, the soil samples were taken to the laboratory, where they were sieved (2 mm mesh), air-
215 dried for one month and stored in this condition until laboratory analyses. All the soil analyses in
216 this study were carried out with air-dry samples for logistical reasons. Previous studies have shown
217 that in drylands such as those we studied, air drying and further storage of soils does not
218 appreciably alter the functions of interest in this study (Zornoza *et al.*, 2006, 2009). It is also
219 important to note that our sampled soils were collected when the soil was in this dry state. Thus, the
220 potential bias induced by our drying treatment is expected to be minimal.

221 Soil texture was measured in two to three composite samples per site, as preliminary analysis
222 revealed that within-site variability was very low. One composite sample each per microsite (open
223 areas or soil under the canopy of the dominant perennial plants) and site were analyzed for sand,
224 clay and silt content according to Kettler *et al.*, (2001). Soil pH was measured in all the soil samples
225 with a pH metre, in a 1: 2.5 mass: volume soil and water suspension. We also measured multiple
226 variables from the nitrogen (N) cycle (total N, mineralization rate, dissolved inorganic N [DIN; sum
227 of NH_4^+ and NO_3^-] and DON) as described by Maestre *et al.*, (2012). In brief, soil samples (2.5 gr of
228 soil) were extracted with K_2SO_4 0.5 M in a ratio 1:5. Soil extracts were shaken in an orbital shaker
229 at 200 rpm for 1 h at 20°C and filtered to pass a 0.45- μm Millipore filter (Jones & Willett, 2006).
230 The filtered extract was kept at 4°C until colorimetric analyses. Using the indophenol blue method
231 (Sims *et al.*, 1995), we estimated concentrations of ammonium and nitrate (colorimetrically) and
232 available N (after potassium persulphate digestion in an autoclave at 121°C over 55 minutes; Sollins
233 *et al.*, 1999). DON was determined as the difference between available N and inorganic N (sum of
234 ammonium and nitrate). The ratio DON:DIN was determined from these data. Regarding potential
235 mineralization rate, air-dried soil samples were re-wetted to reach 80% of their water holding
236 capacity and incubated in the laboratory for 14 days at 30° C (Allen *et al.*, 1986). The potential net

237 N mineralization rate was estimated as the difference between initial and final inorganic N by
238 following Delgado-Baquerizo & Gallardo (2011). Total N was obtained using a CN analyzer (Leco
239 CHN628 Series, LECO Corporation, St Joseph, MI, USA). The N variables used here were selected
240 because they are good proxies of N availability and dominance of N forms within soils (Schimel &
241 Bennett 2004; Delgado-Baquerizo & Gallardo, 2011). All of these variables were then averaged to
242 obtain site-level estimates by using the mean values observed in bare ground and vegetated areas,
243 weighted by their respective cover at each site.

244 Assessing human impacts

245 Quantitative estimates of the magnitude of human impacts in natural ecosystems at global scales are
246 difficult to obtain due to the lack of available data and the wide range of processes affected by
247 human activities (e.g., N deposition, grazing, soil erosion), their different spatial scales, and the
248 interactions among them (Beelen *et al.*, 2013). We therefore estimated such impacts indirectly by
249 measuring four variables at each study site: average proximity (in km) to the nearest northern,
250 southern, eastern and western paved roads from each plot, average proximity (in km) to the four
251 nearest towns/cities from each plot, average population of the four nearest towns/cities to each plot
252 in the last census available (number of people; Table S1), and population density of the province or
253 region of each plot in the most recent available census (number of people·km⁻²; Table S1). Due to
254 the large distances between some of our study sites and the nearest towns/cities, we considered the
255 four closest cities to our plots, as an average value of the local human impact. Distances to nearest
256 roads, urban centres and human population are classic proxies of human perturbation on ecosystem
257 health and services (Schlesinger & Harley, 1992; Gill *et al.*, 1996; Drechsel *et al.*, 2001; Liu *et al.*,
258 2010; Beelen *et al.*, 2013). We assumed that the size of the negative effects of humans on the N
259 cycle, such N deposition and/or soil erosion, would be directly related to the distance of each site to
260 the nearest city/town and paved road, or in densely populated areas (Drechsel *et al.*, 2001; Gadsdon
261 & Power, 2009; Gilbert *et al.*, 2009; Liu *et al.*, 2010; Beelen *et al.*, 2013). Similarly, soil N
262 depletion derived from land use changes have been observed to be linked to increasing local human
263 population size (Drechsel *et al.*, 2001; Canfield *et al.*, 2010).

264 As the four surrogates of human impacts considered were highly correlated, we conducted a
265 principal component analysis (PCA) to reduce them to independent components. Before conducting
266 the PCA, all the human impact proxies were log-transformed to normalize them. We retained the
267 two first components from the PCA for further analyses. These had an eigenvalue higher than 1, and
268 together explained 80.5% of the variance in the PCA. The first component of the PCA (HC1) was
269 highly related to the average distance to the four nearest towns/cities from each plot (Pearson's $r =$
270 0.96), average distance to the nearest northern, southern, eastern and western paved roads from each

plot (Pearson's $r = 0.76$) and population density of the province of each plot in the most recent available census (Pearson's $r = 0.71$). The HC1 was positively related to other indexes of human influence (Fig. S1a) and footprint (Fig. S1b). In addition, our HC1 was positively related to estimates of inorganic N deposition (Fig. S2a), and fertilizer application (Fig. S2b), and to the amount of N in livestock manure production (Fig. S2c). Similarly, our HC1 was positively related to the percentage land areas used as cropland (Fig. S3a) and to estimates of soil degradation (Fig. S4a). The second component of the PCA (HC2) was highly related to the average population size of the four nearest towns/cities during the most recent census (Pearson's $r = 0.90$). This component was positively related to the previous human influence and footprint indexes (Fig. S1b). In addition, our HC2 was positively related to estimates of N in manure production (Fig. S2c), soil degradation (Fig. S4a) and infiltration of water, determined at our study sites (Fig. S4b). We acknowledge that variables such as fire frequency (Durán *et al.*, 2009), N deposition (Ochoa-Hueso *et al.*, 2011) and/or grazing intensity (Qiu *et al.*, 2013) at each study site would have provided better estimates of human impacts on the N cycle. However, these data were not available for most countries, as the available historical archives do not have the resolution required to obtain such data at the spatial scale of the sampled plots. Geographic distances were obtained with Google Earth® (www.google.com/earth/index.html), while population data were gathered from official statistics of each country (see Table S1).

289 Statistical analyses

We used structural equation modeling (SEM) to determine the relative importance of human impacts (HC1 and HC2), aridity, pH, sand content, plant cover and the spatial influence (distance from equator and longitude) on the different N variables evaluated. We first established an *a priori* model (Fig. S5), based on the known effects and relationships among the drivers of the N cycle (Supplementary Methods S1). Total N, concentrations of ammonium, nitrate and DON, DON:DIN ratios, and pH were log-transformed to improve linearity in the relationships between the variables in our SEM models. Similarly, plant total cover and sand content were square root transformed. We found that all N metrics, sand content and HC1 showed unimodal relationships with aridity. To introduce these second-order polynomial relationships into our SEM model, we calculated the square of aridity and introduced it into our model using a composite variable (Fig. S5). Similarly, the human impact and spatial influence metrics were also included as composite variables. The use of composite variables does not alter the underlying SEM model, but collapses the effects of multiple conceptually-related variables into a single composite effect, aiding interpretation of model results (Grace, 2006). We also examined the distributions of all of our endogenous variables (those with arrows pointing to them within the *a priori* model structure), and tested their normality.

305 Because some of the variables introduced were not normally distributed, the probability that a path
306 coefficient differs from zero was tested using bootstrap tests (Schermerle-Engel *et al.*, 2003). Our
307 *a priori* model structure satisfactorily fitted to our data, as suggested by non-significant χ^2 values (χ^2
308 = 4.740; $P = 0.315$; $d.o.f = 4$ in all cases), non-parametric Bootstrap $P = 0.302$ and by values of
309 RMSEA = 0.029 with a $P = 0.569$.

310 To aid final interpretation in light of this ability of SEM, we calculated the standardized total
311 effects (direct plus indirect effects from the structural equation model) of human impacts (HC1 and
312 HC2), aridity, pH, sand content, plant cover and spatial influence (longitude and distance from
313 equator) on the selected N metrics (Grace, 2006). The net influence that one variable had upon
314 another was calculated by summing all direct and indirect pathways between two variables. All the
315 SEM analyses were conducted using the software AMOS 20 (IBM SPSS Inc, Chicago, IL, USA).

316 Finally, we explored the relationship between the different N variables and human impacts
317 (HC1 and HC2) within each of the studied dryland ecosystems: arid, semiarid and dry-subhumid.
318 By doing this, we wanted to check what dryland ecosystems suffer the highest impact on N cycle
319 derived from human activities. Because our data were not normal, we determined our cross-validate
320 R^2 (CV R^2 ; percent of squared error explained by the model compared to the null model) and P -
321 values using the A3 package from R (Fortmann-Roe *et al.* 2013).

322

323 Results

324 Sand content, pH and total plant cover in our study ranged from 5.36 to 97.94%, 4.13 to 9.21 and
325 2.83 to 82.88% respectively (Table S2). Similarly, for the studied N variables, total N ranged from
326 0.01 to 0.45%, ammonium from 0.82 to 55.86 mg N kg⁻¹ soil, nitrate from 0.00 to 92.07 mg N kg⁻¹
327 soil, DON from 1.24 to 43.31 mg N kg⁻¹ soil and potential mineralization rate from -2.13 to 5.01 mg
328 N kg⁻¹ soil day⁻¹ (Table S2).

329 Aridity was directly and negatively related to soil total N whereas human impacts (HC1 and
330 HC2) were directly positively related to the latter (Fig. 1a). Interestingly, HC1 was negatively
331 related to aridity (Fig 1; Fig. 2), however, aridity and HC2 were unrelated (Fig. 2). Aridity and
332 human impacts, together with sand content, were the most important factors controlling soil total N
333 as shown by the size of their total effects (Fig. 3a). Moreover, the total (direct plus indirect) effect
334 of distance to towns and roads (HC1) and population size (HC2) showed opposite effects on soil
335 total N (Fig. 3a). In absolute terms, however, the impact of HC1 was higher than that of HC2,
336 resulting in a net total positive effect of human impacts on this variable (Fig. 3a).

337 Increases in both aridity and human impacts were associated to decreases in the DON:DIN
338 ratio (Figs. 1b, 2b), and increases on potential net mineralization rates (Figs. 1c, 2c). Our different

339 surrogates of anthropogenic disturbances (HC1 and HC2) rendered different and opposite
340 relationships with DON and soil nitrate, although both were associated to increasing ammonium
341 concentrations (Fig. 3e). HC1 showed a positive relationship with the concentrations of DON and
342 soil nitrate whereas HC2 was negatively associated with those N variables.

343 Dry-submid were the dryland ecosystem with the highest positive and negative relationship
344 between HC1 and total N and HC1 and DON:DIN ratio, respectively (Fig. 4). However, the
345 opposite effect was observed from HC1 on total N in dry-subhumid ecosystems (Fig. S6). In
346 addition, the dry-submid ecosystems showed the highest positive relationship between HC1 and
347 potential mineralization and nitrate concentration (Fig. 4). Again, the opposite effect was observed
348 from HC2 on nitrate and mineralization for dry-subhumid ecosystems (Fig. S6).

349

350 **Discussion**

351 *Global change impacts on soil total N*

352 Although human activity should increase the N budget worldwide (Galloway *et al.*, 2008), our
353 results suggest that the increases in aridity forecasted for large areas of the planet will counteract
354 such increment in total N. Of particular interest was the observed negative relationship between
355 aridity and human impacts in our models. This is likely derived from the constraints that aridity, and
356 hence shortage in water availability, generally impose on human activities and urban development
357 (Whitford, 2002; Schwinning & Sala, 2004). In particular, we found a quadratic negative
358 relationship between aridity and HC1. This result suggests that there is a current spatial disconnect
359 between the impacts of aridity, which may favour N losses, and those of human activities, which
360 may favor N accumulation, in different dryland regions (Liu *et al.*, 2012). Thus, at the global scale,
361 the driest regions will tend to become more N limited, but N enhancement due to human activities
362 in the least arid drylands may counteract any trend towards greater N limitation. In addition, aridity
363 and HC2 were unrelated, suggesting that increasing aridity is related to more scattered urban areas
364 (HC1), but do not population density in general (HC2; Mainguet, 1999). We stress that the spatial
365 distribution of our plots did not cover areas where this pattern may not hold, such as large, rapidly -
366 growing desert urban areas (e.g. Phoenix or Las Vegas in USA; Kane 2014) or semi-arid areas with
367 intensive agricultural activities (e.g. Almería in SE Spain; Aznar-Sánchez & Galdeano-Gómez,
368 2011). We also would like to acknowledge the limitations of the observational approach followed,
369 however we believe that our study provide a good snapshot of the status of N cycle at a global scale,
370 and show from an integrative point of view how interactive effects derived from aridity and human
371 impacts can globally affect N concentrations and dominance of relative N forms.

372

373 *Inorganic N accumulation derived from global change*

374 Increasing human impacts and aridity resulted in direct and total negative impacts on the DON:DIN
375 ratio, and a positive direct effect on potential net mineralization rates. Thus, any increase in human
376 impacts and aridity derived from global change will lead to a greater dominance of inorganic N
377 forms. This scenario is compatible with both the observed loss of biological control on N cycle
378 derived from climate change suggested by Schlesinger *et al.*, (1990) and Delgado-Baquerizo *et al.*,
379 (2013), and the trend to an inorganic N saturation stage predicted by models in terrestrial
380 ecosystems as a consequence of anthropogenic N deposition (Fig. S2a; Gruber & Galloway 2008;
381 Schlesinger, 2009; Chen *et al.*, 2013). An increase in aridity has been suggested to result in a world
382 with a lower net depolymerization rate (DON production) in the most arid areas, likely linked to the
383 low precipitation and plant cover of these environments (Schlesinger *et al.*, 1990), which would
384 increase the dominance of inorganic N forms. This was supported by the direct negative relationship
385 between aridity and DON:DIN found. However, this direct negative effect was counteracted by the
386 indirect positive effects mediated through sand content and pH, both increasing the ratio DON:DIN
387 (Fig. 1b). As a consequence of the interplay between direct negative and indirect positive effects,
388 the total effect of aridity on the dominance of dissolved organic versus inorganic N forms was
389 negligible (Fig. 2b). Conversely, proximity to human populations (HC1) was the most important
390 factor controlling the DON:DIN ratio as shown by its total effect size, which was greater than for
391 any other factors evaluated (Fig. 2b). This decrease in the DON:DIN ratio with increasing human
392 impact may be driven by the increase of inorganic N inputs linked to human activities such as
393 fertilizer production, accumulation of livestock wastes and fossil fuel combustion in the vicinity of
394 our sites (Dentener *et al.*, 2006; Cornell, 2011). An increase in inorganic N in soils may have a
395 negative impact on the functioning and services provided by drylands worldwide. For example,
396 Delgado-Baquerizo *et al.*, (2013b) found that inorganic N inputs were negatively linked to
397 microbial functional diversity and N depolymerization (production of DON), and may also reduce
398 the organic N uptake by plants and microorganisms in these ecosystems (Warren, 2009).

399 *Shifts in the different N forms derived from human impacts*

400 The relatively strong total positive relationship between HC1 and DON concentrations may suggest
401 that atmospheric deposition of organic N, which has rarely been considered a significant source of
402 atmospheric N (Cornell *et al.*, 2011), may be affecting DON concentration in dryland soils. In
403 addition, HC1 was positively related to the concentrations of soil nitrate and ammonium, suggesting
404 the importance of both reduced and oxidized N deposition in global drylands. Because our sites are
405 not located in agricultural areas, the effect of highly populated towns surrounding our plots (HC2)
406 should be related more to the use of these drylands for grazing and wood harvesting than to more

intensive human uses. Overgrazing can lead to losses of soil organic matter and nutrients through the conversion of semiarid grasslands to arid shrublands (Schlesinger *et al.*, 1990). However, HC2 was positively related to N in manure production at the global scale (Fig. S2c). This constitutes one of the most important sources of reduced N to the atmosphere (Bouwman *et al.*, 2011), and may explain why the observed negative effect of HC2 on DON and nitrate by intensive agriculture is not found with ammonium. Intensive land management may result in DON and nitrate leaching into streams and the groundwater, which may pollute them (Gruber & Galloway 2008; Schlesinger 2009; Chen *et al.*, 2013). However, both HC1 and HC2 were positively related to the concentration of ammonium in soil (Fig. 2e). Ammonium is one of the most common N sources associated with human activities, as intensive agriculture and livestock are significant sources (Anderson *et al.*, 2003; Clarisse *et al.*, 2009; Canfield *et al.*, 2010). Increases in the concentration of soil ammonium with increasing human impacts in this study suggest that at least a part of the ammonium present in dryland soils may come from human-derived activities. Overall, this increase in soil ammonium concentrations may increase the potential of N to cross ecosystem boundaries by ammonia volatilization or through ammonium conversion to nitrate followed by leaching from soil, all of which are common phenomena in drylands and may cause eutrophication and reduce water quality (Schlesinger *et al.*, 1990; Schlesinger & Harley, 1992; Robertson & Groffman, 2007; Ravishankara *et al.*, 2009). For example, as processes such as nitrification usually require small amounts of water (Schwinning & Sala 2004; Delgado-Baquerizo *et al.*, 2013c), the accumulation of ammonium in the less arid drylands may quickly promote its conversion to nitrate after even small rainfall events (Schwinning & Sala, 2004). Our study supports this, as we observed an increase in the potential net nitrification rate in our soils with increasing ammonium ($P < 0.001$; Fig. S7). The overall dominance of inorganic forms of N resulting from increasing aridity and human impacts may enhance nitrification and denitrification rates in drylands, (e.g. releasing N_2O ; Schlesinger *et al.*, 2009; Canfield *et al.*, 2010), potentially enhancing the emission of greenhouse gases from these ecosystems.

433

434 **Conclusions**

Our findings provide evidence that human impacts promote the accumulation of N in dryland soils worldwide, but that these effects are offset by increases in aridity. We also found that these two global change drivers are spatially disconnected in drylands, favoring N losses in the most arid, and accumulation in the least arid ecosystems. Our analyses indicate that both increasing aridity and human impacts linked to the intensity of anthropogenic disturbance will enhance the inorganic control of the N cycle in drylands soils. This increase in inorganic N dominance in dryland soils

may have negative effects on key ecosystem functions (e.g. microbial functionality) and services (e.g. quality of water and air) at the global scale, and may enhance the emission of important greenhouse gases such as N₂O.

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577 **Supporting Information legends**

578 Supplementary information can be found in the online version of this article

579 **Supplementary Methods S1.** Analyzing our structural equation model: rationale for the variables
580 included.

581 **Figure S1.** Relationships between our human impacts and previous human impact indices.

582 **Figure S2.** Relationships between our human impacts and global inorganic N deposition, N
583 fertilizer application and the N in manure production.

584 **Figure S3.** Relationships between our human impacts and global land area used as cropland and
585 pasture.

586 **Figure S4.** Relationships between our human impacts and the global human-induced soil
587 degradation, and field assessed infiltration and stability.

588 **Figure S5.** A priori generic structural equation model (SEM) used in this study.

589 **Figure S6.** Relationships between aridity and our human impacts in this study.

590 **Figure S7.** Relationship between ammonium concentration and the potential net nitrification rate.

591 **Table S1.** Information about the population data used to estimate human impacts at our study sites.

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603 **Figure legends**

604 **Figure 1.** Effects of aridity (blue arrows), human impacts (red arrows), pH, sand content, plant
605 cover and spatial influence (grey arrows) on: total N (a), DON:DIN ratio (b), mineralization rate (c),
606 DON (d), NH_4^+ (e) and NO_3^- (f). Numbers adjacent to arrows indicative of the effect size of the
607 relationship. Continuous and dashed arrows indicate positive and negative relationships,
608 respectively. R^2 denotes the proportion of variance explained. For graphical simplicity, factors
609 influencing human impacts are: a. Spatial $\rightarrow$ HC1 = 0.13, Spatial $\rightarrow$ HC2 = -0.35***; b. Sand $\rightarrow$

610 HC1 = -0.05, Sand $\rightarrow$ HC2 = -0.16**; c. pH $\rightarrow$ HC1 = 0.34, pH $\rightarrow$ HC2 = -0.37**; d. Composite
611 aridity $\rightarrow$ HC1 = -0.43***, Aridity $\rightarrow$ HC2 = 0.28**. Significance levels are as follows: * $P < 0.05$,
612 ** $P < 0.01$ and *** $P < 0.001$.

613 **Figure 2.** Relationships between aridity (1- aridity index) and the first (a; HC1) and second (b;
614 HC2) components of a principal component analysis from four proxies of human impacts:
615 proximity to urban areas, paved roads, population density and population size. The fitted lines
616 correspond to quadratic (a) and (b) linear models. Because our data were not normal, we determined
617 our cross-validate R2 (CV R2; percent of squared error explained by the model compared to the null
618 model) and P-values using the A3 package from R (Fortmann-Roe et al. 2013).

619 **Figure 3.** Standardized total effects (direct plus indirect effects) derived from the structural equation
620 modeling, including the effects of aridity (Aridity), percentage of sand (sand), pH, plant cover
621 (Plant), distance from equator (DE) and longitude (LON) and human impact (HC1 and HC2) on the
622 total N (a), DON:DIN ratio (b), potential mineralization rate (c), DON (d) NH4+ (e) and NO3- (f).

623 **Figure 4.** Relationships between the HC1 component and the different N variables: total N (a),
624 DON:DIN ratio (b), potential net mineralization (c), DON (d), ammonium (e) and nitrate (f) for
625 each of the studied dryland ecosystems: arid (n = 53), semiarid (n = 142) and dry-subhumid (n =
626 29).

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629 **Figure 1**

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 635 **Figure 4**
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