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

Manuel Delgado-Baquerizo[1,2], Fernando T. Maestre[1], Antonio Gallardo[3], Matthew A. Bowker[4], Santiago Soliveres[1,5], David J. Eldridge[6], Ana Prado-Comesaña[3], Juan Gaitán[7], José L. Quero[8], Victoria Ochoa[1], Beatriz Gozalo[1], Miguel García-Gómez[1], Pablo García-Palacios[1,9], Miguel Berdugo[1], Enrique Valencia-Gómez[1], Cristina Escolar[1], Tulio Arredondo[10], Claudia Barraza-Zepeda[11], Bertrand Boeken[12], Donaldo Bran[7], Omar Cabrera[13], José A. Carreira[14], Mohamed Chaieb[15], Abel A. Conceição[16], Mchich Derak[17], Ricardo Ernst[18], Carlos I. Espinosa[13], Adriana Florentino[19], Gabriel Gatica[20], Wahida Ghiloufi[15], Susana Gómez-González[21], Julio R. Gutiérrez[11,22,23], Rosa M. Hernández[24], Elisabeth Huber-Sannwald[10], Mohammad Jankju[25], Rebecca L. Mau[26], Maria Miriti[27], Jorge Monerris[28], Ernesto Morici[18], Muchai Muchane[29], Kamal Naseri[25], Eduardo Pucheta[20], Elizabeth Ramírez[24], David A. Ramírez-Collantes[30], Roberto L. Romão[16], Matthew Tighe[31], Duilio Torres[32], Cristian Torres-Díaz[21], James Val[33], José P. Veiga[34], Deli Wang[35], Xia Yuan[35], Eli Zaady[36].

1. Área de Biodiversidad y Conservación, Departamento de Biología y Geología, Escuela Superior de Ciencias Experimentales y Tecnología, Universidad Rey Juan Carlos, Calle Tulipán Sin Número, 28933 Móstoles, Spain.

2. Hawkesbury Institute for the Environment, University of Western Sydney, Penrith, 2751, New South Wales, Australia.

3. Departamento de Sistemas Físicos, Químicos y Naturales, Universidad Pablo de Olavide, Carretera de Utrera kilómetro 1, 41013 Sevilla, Spain.

4. School of Forestry, Northern Arizona University, Flagstaff, AZ 86011, USA.

5. Institute of Plant Sciences, University of Bern, Alterbengrain 21, 3013 Bern, Switzerland.

6. School of Biological, Earth and Environmental Sciences, University of New South Wales, Sydney, New South Wales 2052, Australia.

7. Instituto Nacional de Tecnología Agropecuaria, Estación Experimental San Carlos de Bariloche, Casilla de Correo 277 (8400), Bariloche, Río Negro, Argentina.

8. Departamento de Ingeniería Forestal, Campus de Rabanales Universidad de Córdoba, Carretera N- km. 396, 14071 Córdoba, Spain.

9. Centre d'Ecologie Fonctionnelle & Evolutive, CEFE-CNRS, 1919 route de Mende, Montpellier 34293, France.

10. División de Ciencias Ambientales, Instituto Potosino de Investigación Científica y Tecnológica, Código Postal 78210 San Luis Potosí, San Luis Potosí Mexico.

11. Departamento de Biología, Universidad de La Serena, Casilla 599, La Serena, Chile.

12. The Wyler Department of Dryland Agriculture. University of the Negev, Sede Boqer Campus, Midreshet Ben-Gurion, 84990, Israel.

13. Departamento de Ciencias Naturales. Universidad Técnica Particular de Loja, San Cayetano Alto, Marcelino Champagnat, Loja, Ecuador.

14. Universidad de Jaen, Departamento de Biología Animal, Biología Vegetal y Ecología, 23071 Jaen, Spain.

15. University of Sfax, Faculty of sciences, UR Vegetal Diversity and Arid Land Ecosystems, Route de Sokra, kilomètre 3.5, Boîte Postale 802, 3018, Sfax, Tunisia.

16. Departamento de Ciências Biológicas, Universidade Estadual de Feira de Santana, 44036-900, Feira de Santana, Bahia, Brasil.

17. Direction Régionale des Eaux et Forêts et de la Lutte Contre la Désertification du Rif, Avenue Mohamed 5, Boîte Postale 722, 93000 Tétouan, Morocco.

18. Facultad de Agronomía, Universidad Nacional de La Pampa, Casilla de Correo 300, 6300 Santa Rosa, La Pampa, Argentina.

19. Instituto de Edafología, Facultad de Agronomía, Universidad Central de Venezuela, Ciudad Universitaria, Caracas, Venezuela.

20. Departamento de Biología, Facultad de Ciencias Exactas, Físicas y Naturales, Universidad Nacional de San Juan, J5402DCS Rivadavia, San Juan, Argentina.

21. Laboratorio de Genómica y Biodiversidad, Departamento de Ciencias Básicas, Universidad del Bío-Bío, Casilla 447, Chillán, Chile.

22. Instituto de Ecologia y Biodiversida, Casilla 653, Santiago, Chile.

23. Centro de Estudios Avanzados en Zonas Aridas, La Serena, Chile.

24. Laboratorio de Biogeoquímica, Centro de Agroecología Tropical, Universidad Experimental Simón Rodríguez, Apdo 47925, Caracas, Venezuela.

25. Department of Range and Watershed Management, Faculty of Natural Resources and Environment, Ferdowsi University of Mashhad, Azadi Square, Mashhad, 91775–1363, Iran.

26. Center for Ecosystem Science and Society, Northern Arizona University, Flagstaff, AZ 86011, USA.

27. Department of Evolution, Ecology and Organismal Biology, Ohio State University, 318 West 12th Avenue, Columbus, OH 43210, USA.

28. Université du Québec à Montréal Pavillon des sciences biologiques Département des sciences biologiques 141 Président-Kennedy Montréal (Québec) H2X 3Y5 Canada.

29. Zoology Department, National Museums of Kenya, 78420-00500 Ngara Road, Nairobi, Kenya.

30. Production Systems and the Environment Sub-Program. International Potato Center. Apartado

1558, Lima 12, Peru.

31. Department of Agronomy and Soil Science, School of Environmental and Rural Science, University of New England, Armidale, New South Wales 2351, Australia.

32. Departamento de Química y Suelos, Decanato de Agronomía, Universidad Centroccidental "Lisandro Alvarado". Barquisimeto, 3001, Venezuela.

33. Office of Environment and Heritage, Post Office Box 363, Buronga, New South Wales 2739, Australia.

34. Departamento de Ecología Evolutiva, Museo Nacional de Ciencias Naturales, CSIC, José Gutiérrez Abascal, 2, 28006 Madrid, Spain.

35. Institute of Grassland Science, Northeast Normal University, Key Laboratory of Vegetation Ecology, Ministry of Education, Changchun, Jilin 130024, China.

36. Department of Natural Resources, Agriculture Research Organization, Ministry of Agriculture, Gilat Research Center, Mobile Post Negev 85280, Israel.

***Author for correspondence:** Manuel Delgado-Baquerizo. Hawkesbury Institute for the Environment, University of Western Sydney, Penrith, 2751, New South Wales, Australia. E-mail: M.DelgadoBaquerizo@uws.edu.au

Abstract

Aim Although very likely to co-occur in the future, it is largely unknown how simultaneous increases in aridity and anthropogenic disturbances will influence the N cycle in dryland soils, the largest terrestrial biome on the planet. Climate and human impacts are changing the inputs to, and losses from, the nitrogen in terrestrial ecosystems. However, our knowledge of how the interaction between these drivers will affect the concentration of available N for plants and microorganisms as well as the dominance of N forms is still scarce and no study has yet explored these interactive effects on the N cycle at global scale.

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

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

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

Main Conclusions Our findings provide evidence that human impacts will promote the accumulation of N in dryland soils worldwide, while the opposite effect is observed from increasing aridity. Interestingly, we 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 suggest that both increasing aridity and human impacts will enhance the relative dominance of inorganic N in drylands soils which may negatively impact key ecosystem functions and services at the global scale.

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

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 studies 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

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

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

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

Material and Methods

Study area

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

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

Climatic, abiotic, plant and nitrogen variables measured

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

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

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

Assessing human impacts

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

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

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.

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

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

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

Results

Sand content, pH and total plant cover in our study ranged from 5.36 to 97.94%, 4.13 to 9.21 and 2.83 to 82.88% respectively (Table S2). Similarly, for the studied N variables, total N ranged from 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} soil, DON from 1.24 to 43.31mg N kg^{-1} soil and potential mineralization rate from -2.13 to 5.01 mg N kg^{-1} soil day^{-1} (Table S2).

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

Increases in both aridity and human impacts were associated to decreases in the DON:DIN ratio (Figs. 1b, 2b), and increases on potential net mineralization rates (Figs. 1c, 2c). Our different surrogates of anthropogenic disturbances (HC1 and HC2) rendered different and opposite relationships with DON and soil nitrate, although both were associated to increasing ammonium concentrations (Fig. 3e). HC1 showed a positive relationship with the concentrations of DON and soil nitrate whereas HC2 was negatively associated with those N variables.

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

Discussion

Global change impacts on soil total N

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

Inorganic N accumulation derived from global change

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

Shifts in the different N forms derived from human impacts

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

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 N_2O.

Acknowledgments

This research is supported by the European Research Council (ERC) under the European Community's Seventh Framework Programme (FP7/2007-2013)/ERC Grant agreement n° 242658 (BIOCOM), and by the Ministry of Science and Innovation of the Spanish Government, Grant nº CGL2010-21381. CYTED funded networking activities (EPES, Acción 407AC0323). S.G. was funded by CONICYT/FONDAP/15110009.

References

Allen SE, Grimshaw HM, Rowland AP (1986) *Chemical analysis. Methods in plant ecology*. Blackwell Scientific, Oxford, UK.

Anderson N, Strader R, Davidson C (2003) Airborne reduced nitrogen ammonia emissions from agriculture and other sources. *Environment International*, **29**, 277-289.

Aznar-Sánchez JA, Galdeano-Gómez E (2011) Territory, Cluster and Competitiveness of the Intensive Horticulture in Almería, Spain. *Open Geography Journal*, **4**, 103-114.

Bai E *et al.* (2013) A meta-analysis of experimental warming effects on terrestrial nitrogen pools and dynamics. *New Phytologist*, **199**, 441-451.

Beelen R *et al.* (2013) Development of NO_2 and NO_x land use regression models for estimating air pollution exposure in 36 study areas in Europe - The ESCAPE project. *Atmospheric Environment*, **72**, 10-23.

Bouwman L *et al.* (2011) Exploring global changes in nitrogen and phosphorus cycles in agriculture induced by livestock production over the 1900–2050 period. *Proceedings of the National Academy of Sciences USA* 10.1073/pnas., 1012878108.

Canfield DE, Glazer AN, Falkowski PG (2010) The evolution and future of Earth’s nitrogen cycle. *Science,* **330**, 192–196.

Charles H *et al.* (2010) Food Security, The Challenge of Feeding 9 Billion People. *Science*, **327**, 812-818.

Chen H *et al.* (2013) The impacts of climate change and human activities on biogeochemical cycles on the Qinghai-Tibetan Plateau. *Global Change Biology*, **19**, 2940–2955.

Clarisse L, Clerbaux C, Dentener F, Hurtmans D, Coheur, P-F (2009) Global ammonia distribution derived from infrared satellite observations. *Nature Geoscience*, **2**, 479-483.

Compton JE *et al.* (2011) Ecosystem services altered by human changes in the nitrogen cycle, a new perspective for US decision making. *Ecology Letters*, **14**, 804-15.

Cornell SE (2011) Atmospheric nitrogen deposition, Revisiting the question of the importance of the organic component. *Environmental Pollution,* **159**, 2214-2222.

Cui S *et al.* (2013) Centennial-scale analysis of the creation and fate of reactive nitrogen in China 1910-2010. *Proceedings of the National Academy of Sciences USA* doi, 10.1073/pnas.1221638110.

Delgado-Baquerizo M, Gallardo A (2011) Depolymerization and mineralization rates at 12 Mediterranean sites with varying soil N availability A test for the Schimel Bennett model. *Soil Biology and Biochemistry,* **43**, 693–696.

Delgado-Baquerizo M *et al.* (2013a) Decoupling of nutrient cycles as a function of aridity in global dryland soils. *Nature,* **502**, 672–676.

Delgado-Baquerizo M *et al.* (2013b) Biocrusts control the nitrogen dynamics and microbial functional diversity of semi-arid soils in response to nutrient additions. *Plant Soil,* **372**, 643-654.

Delgado-Baquerizo M *et al.* (2013c) Biological soil crusts promote N accumulation in response to dew events in dryland soils. *Soil Biology and Biochemistry,* **62**, 22-27.

Dentener F *et al.* (2006) Nitrogen and sulfur deposition on regional and global scales, A multimodel evaluation. *Global Biogeochemical Cycles,* **20**, doi,10.1029/2005GB002672.

Drechsel P *et al.* (2001) Population density, soil nutrient depletion, and economic growth in sub-Saharan Africa. *Ecological Economics,* **38**, 251–258.

Durán JA *et al.* (2009) Changes in net N mineralization rates and soil N and P pools in a pine forest wildfire chronosequence. *Biology and Fertility Soils,* **45**, 781-788.

Gadsdon SR, Power SA. 2009) Quantifying local traffic contributions to NO_2 and NH_3 concentrations in natural habitats. *Environmental Pollution,* **157**, 2845–2852.

Galloway JN *et al.* (2008) Trends, Questions, and Potential Solutions Transformation. *Science,* **320**, 889-892.

Gilbert NL *et al.* (2007) The influence of highway traffic on ambient nitrogen dioxide concentrations beyond the immediate vicinity of highways. *Atmospheric Environment,* **41**, 2670–2673.

Gill JA, Sutherland WJ, Watkinson AR (1996) A method to quantify the effects of human disturbance on animal populations. *Journal of Applied Ecology*, **33**, 786-792.

Gruber N, Galloway JN. 2008) An Earth-system perspective of the global nitrogen cycle. *Nature,* **451**, 293–296.

Feng S, Fu Q (2013) Expansion of global drylands under a warming climate. *Atmospheric Chemistry and Physics,* **13**, 10081-10094.

Finzi AC *et al.* (2011) Coupled biochemical cycles, Responses and feedbacks of coupled biogeochemical cycles to climate change, examples from terrestrial ecosystems. *Frontiers in Ecology and Environment,* **9**, 61–67.

Grace JB (2006) *Structural Equation Modeling*. Natural Systems Cambridge University Press, New York, USA.

Hijmans RJ *et al.* (2005) Very high resolution interpolated climate surfaces for global land areas. *International Journal of Climatology,* **25**, 1965-1978.

Jones DL, Willett VB (2006) Experimental evaluation of methods to quantify dissolved organic nitrogen DON and dissolved organic carbon DOC in soil. *Soil Biology and Biochemistry,* 38, 991-999.

Kane K *et al.* 2014) A spatio-temporal view of historical growth in downtown Phoenix, Arizona. *Landscape and Urban Planning,* **121**, 70-80.

Kettler TA, Doran JW, Gilbert TL (2001) Simplified method for soil particle-size determination to accompany soil-quality analyses. Soil Science Society of America Journal, **65**, 849

Liu J *et al.* (2010) A high-resolution assessment on global nitrogen flows in cropland. *Proceedings of the National Academy of Sciences USA,* **107**, 8035–8040.

Maestre FT *et al.* 2012) Plant species richness ecosystem multifunctionality in global drylands. *Science,* **335**, 214-218.

Mainguet M (1999) Aridity, Droughts and Human Development Springer, NY, USA.

OECD/FAO 2011. OECD-FAO Agricultural Outlook 2011-2020, OECD Publishing and FAO. http,//dx.doi.org/10.1787/agr_outlook-2011-en.

Peñuelas J *et al.* (2012) The human-induced imbalance between C, N and P in Earth’s life system. *Global Change Biology,* **18**, 3–6.

Qiu L *et al.* (2013) Ecosystem Carbon and Nitrogen Accumulation after Grazing Exclusion in Semiarid Grassland. *PLoS ONE*, **8**, e55433. doi,10.1371/journal.pone.0055433.

Ravishankara AR, Daniel JS, Portmann RW (2009) Nitrous oxide N_2O, The dominant ozone-depleting substance emitted in the 21st century. *Science,* 326,123–125.

Reynolds JF. *et al.* (2007) Global desertification, Building a science for dryland development. *Science,* **316**, 847–851.

Robertson, G.P., Groffman, P (2007) Soil Microbiology, *Biochemistry, and Ecology.* Springer, New York, New York, USA.

Schermelleh-Engel K, Moosbrugger KH, Müller H (2003) Evaluating the fit of structural equation models, tests of significance descriptive goodness-of-fit measures. *Methods of Psychological Research Online*, **8**, 23-74.

Schimel JP, Bennett J (2004) Nitrogen mineralization, challenges of a changing paradigm. *Ecology*, **85**, 591-602.

Schimel DS. (2010) Drylands in the earth system. *Science*, **327**, 418-419.

Schlesinger W.H *et al.* (1990) Biological Feedbacks in Global Desertification. *Science*, 247, 1043-1048.

Schlesinger WH, Harley PC (1992) A global budget for atmospheric NH_3. *Biogeochemistry*, **15**, 191-211.

Schlesinger WH (2009) On the fate of anthropogenic nitrogen. *Proc. Natl. Acad. Sci. USA*, **106**, 203–208.

Schlesinger WH, Bernhardt ES (2013) *Biogeochemistry, an analysis of global change*. Academic Press, CA, USA.

Schwinning S, Sala OE (2004) Hierarchy of responses to resource pulses in arid and semi-arid ecosystems. *Oecologia*, **141**, 211–20.

Sims GK, Ellsworth TR, Mulvaney RL (1995) Microscale determination of inorganic nitrogen in water and soil extracts. *Communications in Soil Science and Plant Analysis*, 26, 303–316.

Sollins P, Glassman C, Paul EA, Swantston C, Lajtha K, Heil JW, Ellikott ET (1999) S*oil carbon and nitrogen: Pools and fraction. Standard Soil Methods for Long-Term Ecological Research*. Oxford University Press, Oxford.

UNEP 2012) *United Nations Environment Programme World Atlas of Desertification*. Edward Arnold, London, UK.

Vitousek PM *et al.* (1997) Human alteration of the global nitrogen cycle, Sources and consequences. *Ecological Applications*, **7**, 737–750.

Warren CR (2009) Does nitrogen concentration affect relative uptake rates of nitrate, ammonium, and glycine?. *Journal of Plant Nutrition and Soil Science*, **172**, 224-229.

Whitford WG (2002) *Ecology of Desert Systems*. Academic Press, San Diego, CA.

World Bank (2008) *World Development Report, Agriculture for Development*. World Bank, Washington, DC.

Zornoza R. *et al.* (2006) Assessing air-drying and rewetting pre-treatment effect on some soil enzyme activities under Mediterranean conditions. *Soil Biology and Biochemistry*, **38,** 2125

Zornoza R. *et al.* (2009) Storage effects on biochemical properties of air-dried soil samples from southeastern Spain. *Arid Land Restauration and Management*, **23,** 213

Supporting Information legends

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

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

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

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

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

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

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

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

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

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

Figure legends

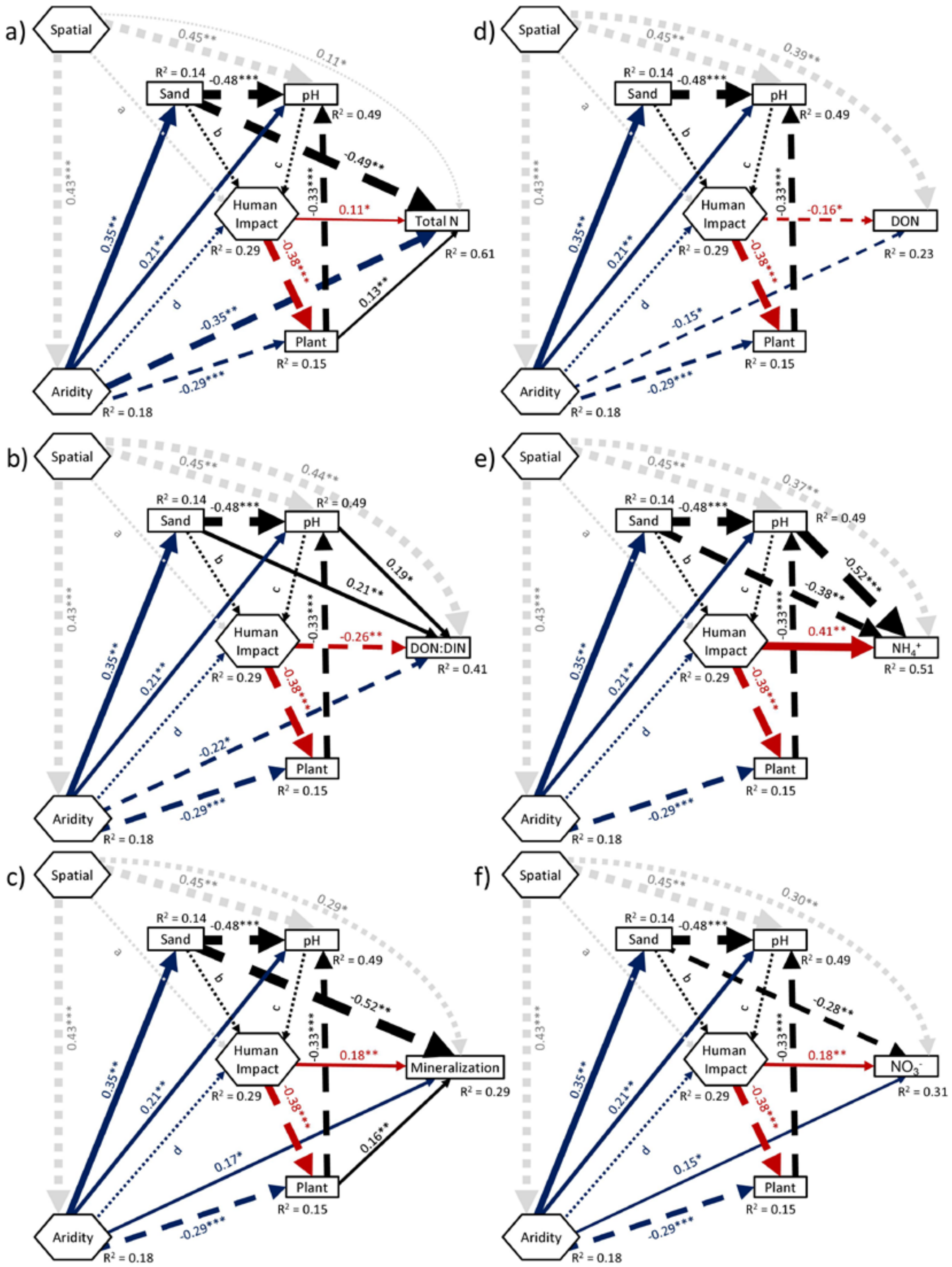
Figure 1. Effects of aridity (blue arrows), human impacts (red arrows), pH, sand content, plant cover and spatial influence (grey arrows) on: total N (a), DON:DIN ratio (b), mineralization rate (c), DON (d), NH_4^+ (e) and NO_3^- (f). Numbers adjacent to arrows indicative of the effect size of the relationship.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

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

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

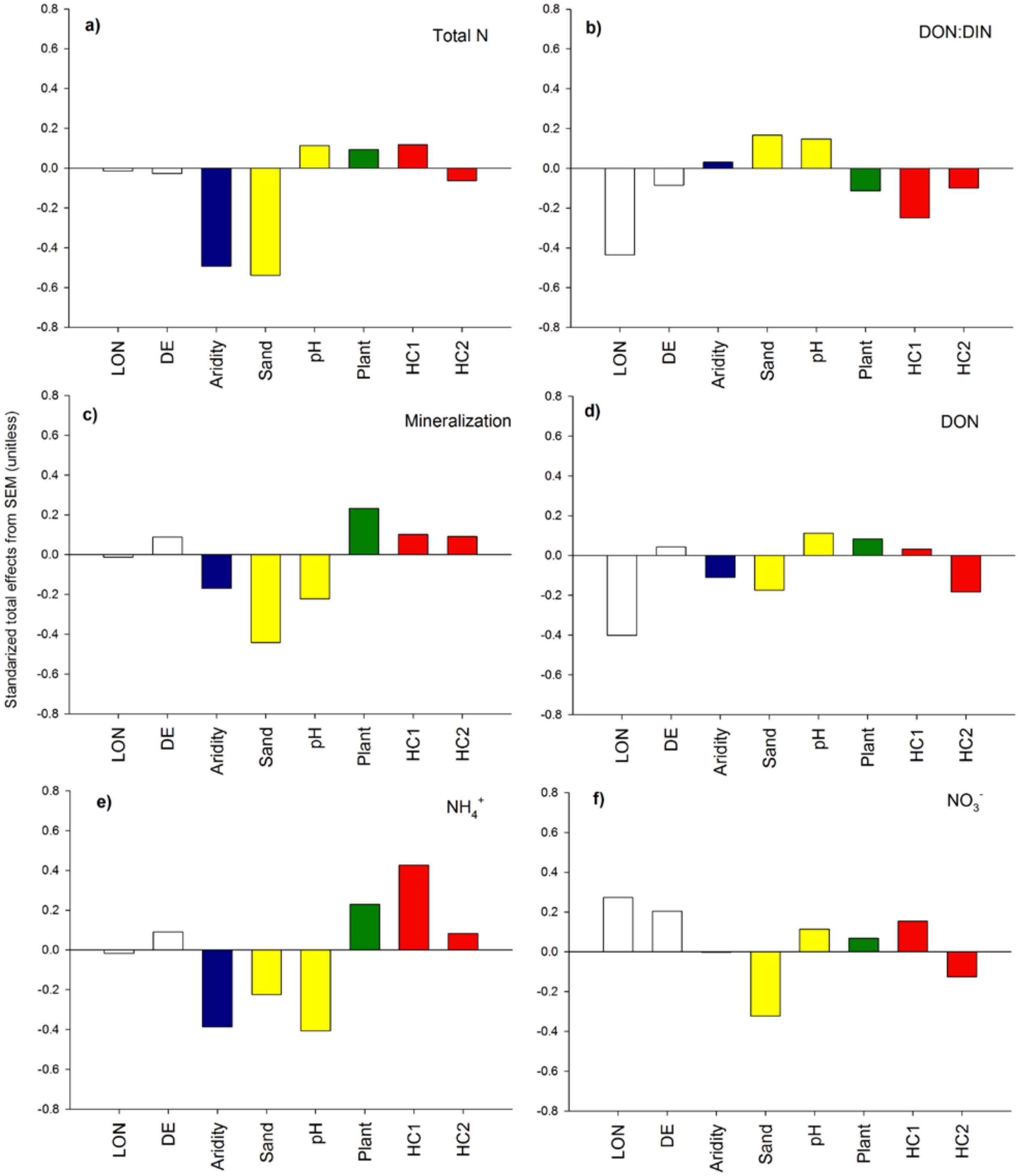
Figure 3. Standardized total effects (direct plus indirect effects) derived from the structural equation modeling, including the effects of aridity (Aridity), percentage of sand (sand), pH, plant cover (Plant), distance from equator (DE) and longitude (LON) and human impact (HC1 and HC2) on the total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

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



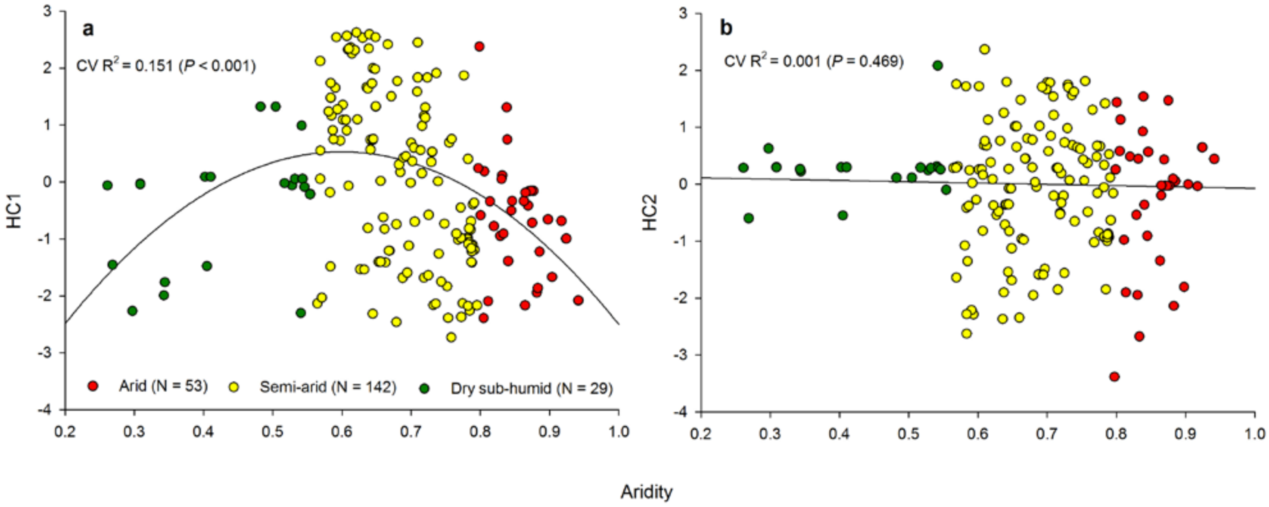
a)
Spatial
0.45**
0.11*
R² = 0.14
-0.48***
Sand
pH
R² = 0.49
-0.49**
a
b
c
0.43***
-0.33***
Human Impact
0.11*
Total N
R² = 0.29
-0.38***
R² = 0.61
0.35**
0.21**
d
-0.35**
0.13**
Plant
R² = 0.15
Aridity
-0.29***
R² = 0.18
b)
0.44**
0.19*
0.21**
-0.26**
DON:DIN
R² = 0.41
-0.22*
c)
0.29*
-0.52**
0.18**
Mineralization
R² = 0.29
0.17*
0.16**
d)
0.39**
-0.16*
DON
R² = 0.23
-0.15*
e)
0.37**
-0.52***
-0.38**
0.41**
NH4+
R² = 0.51
f)
0.30**
-0.28**
NO3-
R² = 0.31
0.15*

Figure 1



a)
Total N
b)
DON:DIN
c)
Mineralization
d)
DON
e)
NH_4^+
f)
NO_3^-
Standarized total effects from SEM (unitless)
0.8
0.6
0.4
0.2
0.0
-0.2
-0.4
-0.6
-0.8
LON
DE
Aridity
Sand
pH
Plant
HC1
HC2

Figure 2



632

633 **Figure 3**

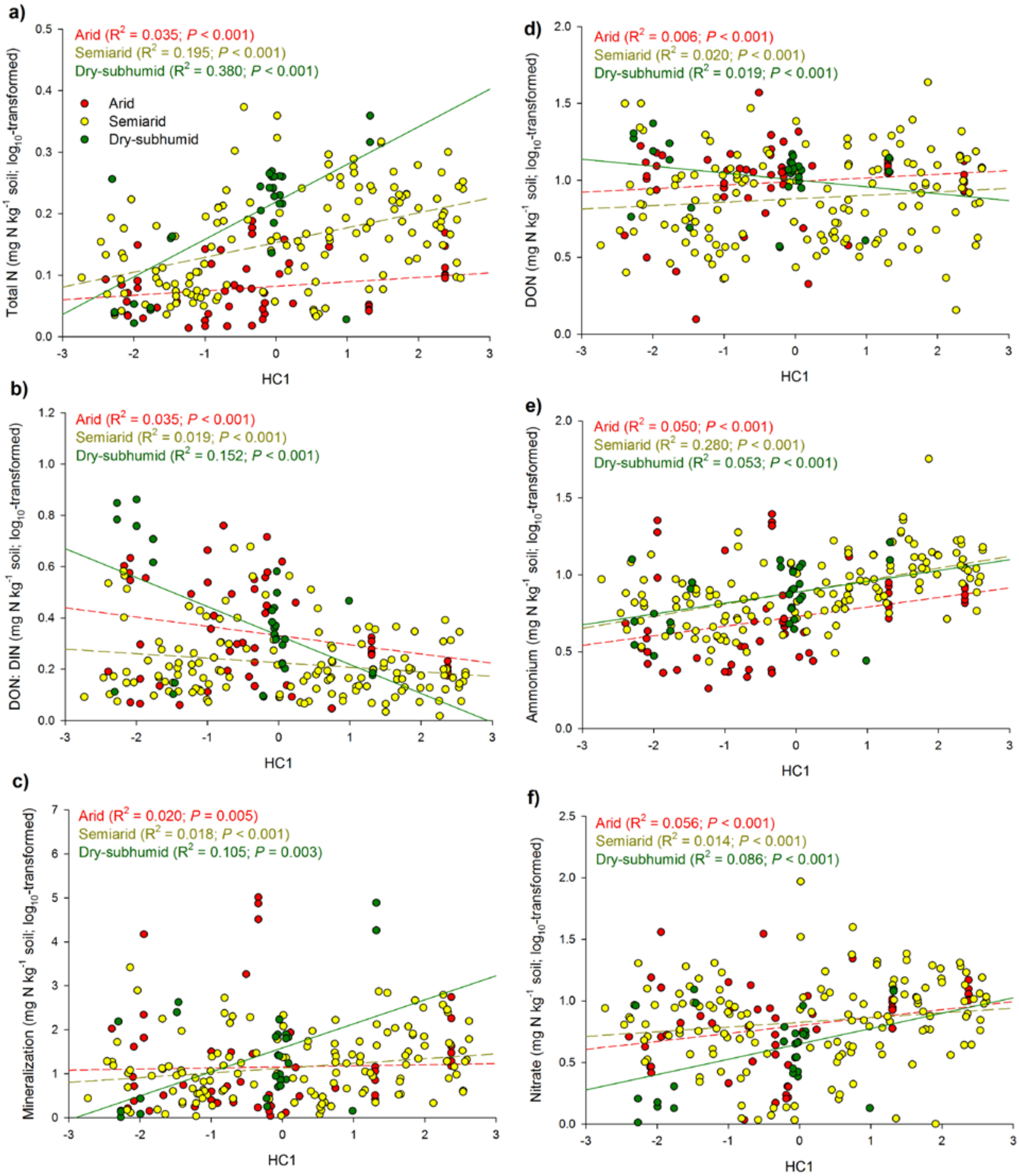

a)
Arid (R² = 0.035; P < 0.001)
Semiarid (R² = 0.195; P < 0.001)
Dry-subhumid (R² = 0.380; P < 0.001)
Arid
Semiarid
Dry-subhumid
Total N (mg N kg⁻¹ soil; log₁₀-transformed)
HC1
b)
Arid (R² = 0.035; P < 0.001)
Semiarid (R² = 0.019; P < 0.001)
Dry-subhumid (R² = 0.152; P < 0.001)
DON: DIN (mg N kg⁻¹ soil; log₁₀-transformed)
HC1
c)
Arid (R² = 0.020; P = 0.005)
Semiarid (R² = 0.018; P < 0.001)
Dry-subhumid (R² = 0.105; P = 0.003)
Mineralization (mg N kg⁻¹ soil; log₁₀-transformed)
HC1
d)
Arid (R² = 0.006; P < 0.001)
Semiarid (R² = 0.020; P < 0.001)
Dry-subhumid (R² = 0.019; P < 0.001)
DON (mg N kg⁻¹ soil; log₁₀-transformed)
HC1
e)
Arid (R² = 0.050; P < 0.001)
Semiarid (R² = 0.280; P < 0.001)
Dry-subhumid (R² = 0.053; P < 0.001)
Ammonium (mg N kg⁻¹ soil; log₁₀-transformed)
HC1
f)
Arid (R² = 0.056; P < 0.001)
Semiarid (R² = 0.014; P < 0.001)
Dry-subhumid (R² = 0.086; P < 0.001)
Nitrate (mg N kg⁻¹ soil; log₁₀-transformed)
HC1

Figure 4