- 1 Title page
- 2 Back to the future? Conservative grassland management can
- 3 preserve soil health in the changing landscapes of Uruguay
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- 13 **Abstract.** The 'soils of the anthropocene' are predominately agricultural. To understand them,
- 14 we analysed agri- and silvicultural intensification of Uruguayan grasslands in a country wide
- 15 survey on fertility proxies, pH and trace metals in topsoils originating from different land uses.
- 16 We observed a loss of nutrients, trace metals and organic matter from grassland, crops and
- 17 timber plantations. As an example, the cation exchange capacity was 160 percent higher in
- 18 native forests compared to grasslands and lowest in timber plantations, reaching only half of
- 19 the CEC in grasslands acidification of topsoils continues as three fourth of all samples are
- 20 'extremely acidic' and 'very strongly acidic' and lowest in timber plantations. Topsoils of
- 21 riverine forests accumulate more trace metals compared to the other uses. We assume an
- 22 accumulation in the topsoils of giverine forests, where high levels of nutrients, trace metals and

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organic carbon are found. The translocation of nutrients and organic matter across the landscape to the erosion base depends on local land use trajectories. Increasing soil acidification is driving a positive feedback loop, and land use intensification is leading to degradation of local black soils within a few decades. Our data raises questions about the resilience and carrying capacity of Uruguayan soils with regard to currently implemented highly productive management forms, including the use of timber plantation for carbon sequestration, and supports more conservative forms of extensive management on the grassland

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## 1. Introduction

biome\_

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- 40 Human activities alter the bio- and pedosphere, leaving a footprint of such a magnitude that it
- 41 can be verified stratigraphically (Waters et al., 2016). This unprecedented transformational
- 42 force is intimately related to the expansion of societies and its productive frontiers, causing a
- 43 loss of biodiversity, habitat and soil degradation and, consequently, to ecosystem modification
- 44 (Foley et al., 2005, Borrelli et al., 2017). In this context, soil sciences have transitioned from
- 45 studies on natural soil formation to the science of 'anthropedogenesis' (Richter, 2020),
- 46 focussing on the 'soils of the anthropocene' that are predominately agricultural (51 Mio. km²)
- 47 or urban (1.5 Mio km<sup>2</sup>; FAO, 2019).
- 48 The temperate grasslands of South America have historically been characterised by rolling
- 49 plains and low hills that have been extensively exploited for cattle production and its
- 50 derivatives since the arrival of European colonization. The Río de la Plata grasslands are one
- of our planet's four major black soil regions (Durán et al., 2011; Liu et al., 2012), and some of
- 52 the most fertile soils in the world. Playing an important role in the global food production, these
- 53 are characterized by a thick, humus and base cation rich and a high cation exchange capacity

- 56 throughout their profile. Maintaining their properties are therefore crucial to developing
- 57 sustainable and productive agriculture (Durán et al., 2011; Liu et al., 2012).
- 58 Today, the 'Uruguayan savanna' is part of the top-three most critically endangered biomes
- 59 (Veldman et al., 2015). In recent decades, however, the grassland has decreased due to the
- 60 expansion of cash crops and *Eucalyptus* plantations, both of which are promoted by national
- 61 legislation including land grabbing and trans-nationalization (Piñeiro, 2012). This land use
- 62 intensification, with its increased input of energy, nutrients and pesticides, leads to an overall
- 63 loss of soil fertility and increasing toxicity related to acidification, salinization and
- 64 contaminants (Liu et al., 2012; Borrelli et al., 2017). Ecological, economic and cultural
- 65 functions of soils are severely degraded, and the degradation of black soils in South America
- 66 is of particular concern because they have only been heavily exploited for a comparatively
- short period of time (Durán et al., 2011).

- 68 Since the first decades of the twentieth-century, compared to other disciplines, soil sciences
- 69 have received an extraordinary amount of attention in Uruguayan academia, governance, the
- 70 productive sector, and also in the general public, resulting in a national soil inventory program
- 71 in 1965. The subsequent classification of Uruguayan soils and their productivity (CONEAT
- 72 Index) remains an important source for today's land taxation and for management plans by the
- 73 legal conservation regulations, and provides a detailed classification that takes into account soil
- 74 type, texture, natural vegetation, altitude and geology (Lanfranco and Sapriza, 2011).
- 75 As soil degradation is extremely relevant for countries like Uruguay, which are
- 76 socioeconomically dependent on their soils (Zubriggen et al. 2020), it is a topic of discussion
- 77 for local farmers, academia, and the public. An actualization of the state of the art of soils and
- for focus farmers, academia, and the public. The actualization of the state of the art of sons and

related processes is needed (García-Préchac et al. 2004; De Faccio et al. 2021), particularly as

- 79 there has been little study of the impacts of the Uruguayan grassland intensification on soils
- 80 properties (Beretta-Blanco et al., 2019). At the same time, while a new paradigm for grassland
- 81 intensification with a wide set of means including fertilization has been proposed to increase

82 economic and environmental sustainability (Jaurena et al., 2021), it is urgent to get more 83 insights into the dynamics of nutrient in soils of Uruguay and their availability for crops 84 (Beretta-Blanco et al., 2019). 85 Soil classifications are mainly based on subsoils. However, we focus on topsoil as the most 86 relevant and very responsive interface for ecological processes and farmer's management, 87 since understanding the state of the art of topsoils and its processes is crucial for developing 88 recommendations for sustainable land management practices. Due to the diversity of 89 perspectives on soil quality and health and related ecosystem services, operational procedures 90 for evaluation of soil functioning are still lacking (Bonfante et al. 2021). We contribute to a 91 better understanding of globally occurring degradation processes in the field of tension between 92 desired soil productivity, yield limits, especially in erosion sensitive soils, and necessary soil 93 conservation. 94 We therefore explored soil parameters describing current chemical conditions of topsoils that hat gelöscht: actual 95 are parts of different soil groups and orders, and Uruguayan soil categories. Specifically, in 96 order to explore the gains and losses of macro and micro-nutrients and soil organic carbon hat gelöscht: matter across 97 across landscapes and to determine the impact of land use change on acidification and trace 98 metal mobility and related trade-offs with soil degradation and conservation. In detail we 99 address the following question: i) how do fertility proxies such as soil organic carbon and hat gelöscht: Our objective were, we analysed i) analyse hat gelöscht: the variation of 100 content of nutrients, acidification (pH) and trace metals accumulation in topsoils vary across hat gelöscht: organic carbon 101 different land uses? Thus, we expand the knowledge across land uses from more natural to hat gelöscht: or hat gelöscht: 102 strongly modified uses and discuss the results in light of different degradation processes such hat qelöscht: ii) establish the levels of acidification (pH), and iii) 103 as erosion, depletion of nutrients or carbon, acidification and accumulation of pollutants and in analyse the trace metals accumulation. hat gelöscht: W

hat gelöscht: the knowledge and expand the local soil- about the impacts of land use change across Uruguayan topsoils hat gelöscht: from timber plantations, grasslands and native forests using the same methodology as for crops. In detail we answer

the following questions:...

the light on current debates on intensification,

#### 2. Material and Methods

### 2.1 Study area and design

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122 Uruguay covers about 176,000 km<sup>2</sup>, and has a population of 3.5 million, mainly in urban areas. 123 The Western half of Uruguay is dominated by Mollisols, developed on a wide range of sediments from the Devonian to the Cenozoic era (partially associated with Vertisols), while 124 125 the Eastern plains and wetlands have, in addition to Mollisols, other soil types on the slopes, 126 rocks and on flood plans (i.e. Alfisols, Ultisols, Entisols and Inceptisols; Durán et al. 2011). 127 During a country-wide survey from December 2015 to March 2016, we collected 280 topsoil samples of 0-10 cm depth from 101 plots (50x50m in grassland and crops; 100x100m in native 128 129 forests and timber plantations) distributed at 28 monitoring sites throughout Uruguay, South 130 America (Fig. 1a-b), using a stratified random design. In the first step, we randomly selected 131 monitoring sites across the country. In the second step, we contacted landowners to explore 132 their willingness to establish a long-term monitoring site. If the owner agreed, plot selection 133 was stratified by different rural land use types: grassland, timber plantations of *Pinus* and 134 Eucalyptus species, native forest, and crops. Native forests cover mainly riverine and park 135 forests. The later are a savanna like transition zones between riverine forests and the open 136 grasslands. We subdivided grassland plots according to the intensity of use: (i) undisturbed grassland (without grazing), (ii) partially grazed grasslands (with sporadic grazing and low 137 138 animal charge), and (iii) highly grazed grassland (with high animal charge). Land use change 139 from 1986 to 2017 follows basically three different trajectories: i) the expansion of timber 140 plantations over grassland leading to a disaggregation of grassland by timber plantations; ii) 141 cropland expansion where crop cover maintains the open landscape character of former 142 grasslands, grassland conservation where large and regularly interconnected riverine forests in 143 a landscape dominated by grasslands (Ramírez and Säumel 2021) and grassland intensification

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hat verschoben (Einfügung) [1]

| 144 | changing from natural grassland to so called 'improved' or artificial grasslands (Modernel et                           |   |
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| 145 | al. 2016; Jaurena et al. 2021). Fertilization and application of other agrochemicals is standard                        |   |
| 146 | procedure in timber plantations, artificial grasslands and industrial crops. We sampled top soil                        | hat gelöscht: ¶   |
| 147 | three times at each land use at the edges of the plot, and stored samples below 7°C until                               |   |
| 148 | <u>laboratory</u> processing.   | hat gelöscht: lab   |
| 149 | 2.2 Analysis of Soil Samples  |   |
| 150 | For gravimetric determination of soil moisture, the topsoils samples were dried at 105 °C until                         | hat gelöscht: humidity  |
| 151 | constant weight. Next, lumps in the samples were broken down and the remaining plant                                    | hat gelöscht: content   |
| 152 | material was removed before sieving (2 mm) and ground.  |   |
| 153 | We analysed 280 samples regarding macronutrients, pH and trace metals and 80 samples for                                |   |
| 154 | soluble cations and micronutrients. Among fertility-related variables, we measured the total                            |   |
| 155 | amount of the macronutrients phosphorus (P), organic carbon (SOC) and nitrogen (N), so                                  |   |
| 156 | obtaining the C/N ratio. To determine total carbon and nitrogen, the samples were sieved again                          |   |
| 157 | $(0.5\ \mathrm{mm})$ and analyzed using a LECO TruSpec CN (USA) at a combustion temperature of $950$                    |   |
| 158 | °C with ultra-pure oxygen. In addition, the presence of <u>SOC</u> was tested for by adding                             | hat gelöscht: inorganic carbon  |
| 159 | concentrated hydrochloric acid, the presence of carbonates was assessed to obtain the amount                            |   |
| 160 | of <u>SOC</u> ,   | hat gelöscht: organic carbon  |
| 161 | We determined concentrations of the soluble cations calcium (Ca), magnesium (Mg),                                       | hat gelöscht: (Corg)  hat gelöscht: and soil organic matter (SOM) as Corg x 2 (Chenu et |
| 162 | potassium (K), sodium (Na), and the micronutrients copper (Cu), zinc (Zn), manganese (Mn)                               | al. 2015)   |
| 163 | and iron (Fe) by atomic spectroscopy (Unicam AAS Solaar 969, Thermo Electron Corporation,                               |   |
| 164 | USA), extracting with ammonium acetate (1 mol l <sup>-1</sup> , pH 7), and with DTPA-CaCl <sub>2</sub> -TEA at (pH      |   |
| 165 | 7.3). We calculated the cation-exchange capacity (CEC). Acidity was measured by adding                                  |   |
| 166 | calcium chloride (0.01 $\underline{\mathbf{M}}$ ) to the samples in a 2.5:1 proportion, and after shaking and two hours | hat gelöscht: mol   |
| 167 | rest, read with a pH meter (HI2550 meter, Hanna Instruments, USA)). For these variables, each                           |   |

178 tenth sample was duplicated. We categorized acidity using the USDA Natural Resources 179 Conservation Service classification (Kellogg, 1993). 180 To determine the concentrations of arsenic (As), cadmium (Cd), chromium (Cr) and lead (Pb), 181 samples were further sieved as before (0.5 mm), weighed out into a digesting container, and 182 extracted with a mixture of nitric acid (70%) and hydrochloric acid (30%) in a 1:3 proportion. 183 The digestion took place in a Titan MPS (Perkin Elmer, USA) microwave at a programme 184 suitable for this digestion (Method 3051A). During the proceedings, only deionized water Type 185 I ASTM1193 (EC max 0.06 to 0.1 μS/cm) was used. Reagents were used to eliminate traces of 186 other materials and to avoid contamination of the samples. The trace metals were determined 187 by inductively coupled plasma optical emission spectroscopy using the Optima 8000 (Perkin 188 Elmer) with metal-associated wavelengths of 193.696 nm for As, 228.802 nm for Cd, 267.716 189 nm for Cr and 220.353 nm for Pb. 190 Total P concentration of phosphorus was determined calorimetrically after microwave-assisted 191 digestion with Unicam spectrometer at a wavelength of 660 nm. For all these variables, a 192 repetition for each round of the microwave digestion was made.

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2.3 Soil classification

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We intersected the coordinates of the centre of the plots with maps containing geospatial

information on the classification of the Uruguayan soils using ArcGIS 10.3 (ESRI, 2018). For

Soil Groups classification, we used the of the World Reference Base for Soil Resources (WRB;

IUSS Working Group, 2015); for Soil Orders, the USDA soil taxonomy (Soil Survey Staff,

1999); and for the local Uruguayan classification, Soil CONEAT (Comisión Nacional de

Estudio Agronómico de la Tierra) Groups categories, which include productive capacity of

cattle and sheep (MGAP, 2020). The CONEAT groups are defined by their productive capacity

in terms of beef, sheep and wool expressed by an index relative to the average productive

205 capacity of the country, to which the index 100 corresponds. The classification is based on 206 photo-interpretation at a scale of 1:40,000, field verifications and physico-chemical analysis of 207 the soils. The productivity indices correspond to soil groups. The CONEAT groups have been 208 defined by the dominant and associated soils according to the Soil Classification of Uruguay. 209 The groups are related to the units of the Soil Reconnaissance Chart of Uruguay at a scale of 210 1:1,000,000. For each group, some important soil properties and associated landscape 211 characteristics are indicated. The nomenclature of the CONEAT groups correlates with the Soil 212 Use and Management Zones of Uruguay. The Soil Groups are superimposed on the rural parcel 213 and are represented in the CONEAT cartography at a scale of 1:20,000 (for more details see 214 MGAP, 2020).

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## 2.4 Data Analysis

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216 In a first step, we explored and prepared our database for further analysis. Exploring the 217 distribution of the soil parameters in R (R Core Team 2021), ruled out the normality of the data 218 using the Shapiro-Wilk Test and the homogeneity of variances with the Fligner-Killeen Test. 219 We tested for outliers using the 1.5-3 IOR threshold and the function *outlierTest* from the R package car (Fox and Weisberg 2019), reviewing the flagged observations case by case in the 220 221 experimental context. The variables on soils characteristics showed generally positive skewed 222 distributions, in some cases multimodal, and tests showed evidence contrary to assumptions of 223 spatial autocorrelation, homoscedasticity and normality in most cases (Supplementary 224 Material: Table S1-S2; Fig. S1-S2). 225 Spearman's rank correlations ( $\rho$ ) were calculated to explore linear associations between soil 226 parameters across all single samples and within different land uses. We used the adonis 227 function of the R package vegan v2.5-7 (Oksanen et al., 2020) with a Euclidean dissimilarity 228 matrix from our normalized soil data to perform PERMANOVA tests with 9999 permutations

- 229 to analyse the multivariate homogeneity of group dispersions based on differences on soil
- parameters between land uses.
- 231 To compare the general effects of land use type on topsoil parameters, non-parametric Kruskal-
- Wallis tests were carried out in R. When significant ( $p \le 0.05$ ), we used Pairwise Wilcoxon
- 233 Rank Sum tests with Benjamini & Hochberg correction to evaluate pairwise differences among
- 234 land uses.
- 235 We used non-metric multidimensional scaling (NMDS) as a robust unconstrained ordination
- 236 method to visualize patterns of top soil characteristics among all samples and within
- 237 subsamples (intersected with different soil Orders) across different land uses using the Bray-
- 238 Curtis dissimilarity matrix. The matrix was constructed with the vegan package to depict
- 239 patterns of all soil parameters in two dimensions (Fig. 3a, c-f) and for the dataset without soluble
- 240 cations and micronutrients variables comparing subcategories within single land use types (i.e.
- 241 within grasslands in 'undisturbed', 'partially grazed' and 'highly grazed' plots; timber
- plantations in 'Eucalyptus' or 'Pinus' plots; Fig. 3b).

## 3. Results

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- Nearly 75% of the sampled 101 plots intersected with the soil Group category 'Luvic Phaeozem'
- 245 (WRB; IUSS Working Group, 2015). A quarter of our plots were distributed among four other
- 246 Groups (i.e. Haplic Luvisols, Eutric Vertisols, Planosols, Lithic Leptisols; Fig. 1c). Half of our
- 247 plots intersected with the Orders 'Argiudolls' or 'Argiudolls & Hapluderts' (Soil Survey Staff,
- 248 1999), and about a quarter with 'Hapludalfs & Hapludults' or 'Argiudolls, Hapludolls &
- 249 Hapludalfs' (Fig. 1d). Our plots intersected with 32 different CONEAT categories (MGAP,
- 250 2021). The most frequent categories were '10.2', '9.3' and '2.12' (25% of our plots) and another
- 251 quarter with '9.1', '8.8', '9.6' and 'G03.11' (Fig. 1e).

#### 3.1 General characteristics of Uruguayan topsoils

hat nach oben verschoben [1]: We subdivided grassland plots according to the intensity of use: (i) undisturbed grassland (without grazing), (ii) partially grazed grasslands (with sporadic grazing and low animal charge), and (iii) highly grazed grassland (with high animal charge).

259 and classification to different soil orders (Table 1, Table A1-A4). The soil organic carbon in 260 topsoils ranged between 0.8 to 16 percent, and was highest in native forests and lowest in timber 261 plantations. The mean of the C/N ratio was about 13, and lowest in crop soils. Phosphorous 262 ranged between 43 to 1009 mg kg<sup>-1</sup>. We also observed a high variability for the micro- and 263 macronutrients and trace metals. The average cation-exchange capacity (CEC) was 12.41 cmol 264 kg<sup>-1</sup>, highest in topsoils of native forest, followed by crops and grasslands and timber 265 plantations (Table 1, Table A1-A2). 266 For the whole data set, high correlation was found between P with SOC and Zn ( $\rho$ =0.82 and 267 0.76, respectively), and between Mg with Ca and Na ( $\rho$ =0.82 and 0.76, respectively; Fig. 2). 268 Similar results were observed within particular land uses, although in native forests, a negative 269 moderate correlation between Ca and Fe was observed (Fig. S3). There was high correlation between pH and Ca ( $\rho$ =0.89; Fig. 2). In native forests, we also found similar correlation with 270 271 other soluble cations like Mg and K (Fig. S3). In park forests topsoils, there was a negative correlation between pH and As and Pb ( $\rho$ =-0.81 and  $\rho$ =-0.84, respectively). In highly grazed 272 273 pastures and crops pH was highly correlated with Cr, and in crops also with As ( $\rho$ =0.93; Fig. 274 S3). Among trace metals, Spearman correlation was moderate between As with Cr and Pb and 275 high between Cr and Pb (Fig. 2). These correlation trends increased in grasslands, timber 276 plantations and crops. We also found a high correlation between cadmium and Cr in crops 277  $(\rho=0.81)$ . Phosphorus was highly correlated with Cr and As in pine plantations, while in 278 soybean crops the correlation was between phosphorus with Cr and Pb (Fig. S3). 279 3.2 Topsoil characteristics clustered by land use

The measured topsoil parameters vary widely across Uruguay, between the different land uses

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We found differences in multivariate distribution of samples according to the different land

uses (i.e., grassland, timber plantations or native forests; Fig. 3a-f; Table S3-S8). All pairwise

comparison of land uses showed significant differences with all variables (p=0.0001; Fig. 3a;

287 cations and micronutrients variables, only finding significant differences between Eucalyptus 288 and *Pinus* stands (p=0.0001; Fig. 3b; Table S4) but not among different grassland subtypes. 289 We also found differences analysing subsamples of different soil order classification to the 290 different land uses (Fig. 3c-f; Table S5-S8). The samples intersected with 'Argiudolls' included 291 all plots on crops, and we found significant differences in all pairwise comparisons of land uses 292 except between grasslands and timber plantation (p=0.0004; Fig. 3c; Table S5). We further 293 found differences between timber plantations and native forests at soils of the 'Argiudolls & 294 Hapluderts' Orders (p=0.0009; Fig. 3d; Table S6) and between timber plantations and 295 grasslands or native forests in soils of the 'Argiudolls, Hapudolls & Hapludalf' Orders 296 (p=0.0284; Fig. 3e; Table S7). Results for samples in 'Hapludalfs & Hapludults' soils were 297 similar to those obtained at country scale (p=0.0001; Fig. 3f; Table S8). 298

## 3.3 Differences in fertility proxies

forests (Fig. 5a-o; Table A3-A4).

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299 We found significantly higher fertility proxies for SOC, P, Ca, Mg and Zn in topsoils from 300 native forests compared to grasslands and timber plantations (Fig. 4b-e, h). Phosphorous was 301 significantly higher in topsoils of grassland compared to those of timber plantations (Fig. 4c). 302 Potassium was significantly higher in topsoils of native forests compared to timber plantations 303 (Fig. 4f). At subcategories level, we found significantly higher amounts of K in partially used 304 grasslands in comparison to samples from highly grazed pastures, and higher SOC (p=0.002), 305 P (p=0.059), Na (p=0.043), K (p=0.012) and Zn (p=0.048) in Eucalyptus compared to Pinus 306 plantations (Fig. S4). Among native forests, samples from riverine forests contain more Mg 307 (p=0.023) and Na (p=0.023) in comparison to park forests. 308 Considering all samples within the Order 'Argiudolls', C/N ratio in agricultural topsoils are 309 lower compared to all other land uses. Phosphorus was higher in topsoils of native forests and 310 crops compared to grasslands and timber plantations. Soil organic carbon was highest in native hat gelöscht: values for the hat gelöscht: SOM

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### 3.4 Soil Acidification

- 319 Topsoil samples showed a markedly acid profile (median pH=4.66), with nearly 75% classified
- 320 as 'extremely acidic' and 'very strongly acidic' (Fig. 6a). We found significant differences across
- land uses, with less acidic native forests, and lower pH in timber plantations (Fig. 6b).
- 322 Comparing between land uses, we found more samples with neutral acidity in grasslands and
- 323 more with higher acidity in timber plantations (Fig. 6b-c). In addition, we found lower pH in
- samples from *Pinus* compared to *Eucalyptus* stands (p=0.018). Results of analysis inside soil
- 325 Orders showed similar variations observed at country scale, with timber plantations being more
- acid and native forest closer to neutral pH (Fig. 6d-f).

#### 327 3.5 Trace metal accumulation across land uses

- 328 For As, Cd, Cr and Pb, we found significantly higher concentrations in topsoils originating
- 329 from native forests compared to the grassland and timber plantation samples (Fig. 4l-o and 5l-
- 330 o; Table A1-A4). At the same time, samples from Eucalyptus plantations had higher
- 331 concentrations of both Cr (p<0.005) and Pb (p<0.005) than *Pinus* topsoils, while the same was
- observed for Cr (p<0.001) and Pb (p<0.05) in riverine forests compared to park forests (Fig.
- 333 S5).

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## 4. Discussion

- 335 The vicious circle between the wish to stop soil degradation and concurrent increases in land
- 336 productivity to satisfy the increasing demand for food, fibres and energy has not been broken
- 337 since green revolution. Socio-economic and conventional management practices that drive soil
- 338 degradation have generated several traps, such as the 'inputs trap' where a reduced yield per
- 339 area is followed by higher fertilizer application or the 'credit or poverty trap' where economic
- pressure forces the farmer to intensification (Gomiero, 2016). Caught in this loop, the soils of

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the temperate grasslands of Uruguay have suffered strong degradation from erosion, acidification, contamination, salinification and compaction (Zurbriggen et al., 2020). This is clearly reflected in the results of our topsoil survey, which also adds interesting insights from timber plantations, grasslands and native forests to an existing database consisting mainly of crops and pastures samples from 2002-2014, which demonstrated the loss of organic matter by 25% and an increasing loss of nutrients (Beretta-Blanco et al. 2019). We contribute deeper insights on fertility, acidification and trace metals accumulation in topsoils from a wide range of different land uses, which is, to our knowledge, unique for the region since the CONEAT classification (CONEAT Index, 1976).

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## 4.1 Translocation of elements in topsoils within landscape

356 Our data demonstrate the high amounts of organic carbon, nutrients and trace metals in topsoil 357 samples from riverine forests, suggesting transport of soil particles from the surrounding land 358 uses (e.g., grasslands, crop or timber plantations) to the borders of rivers, streams and creeks. 359 Therefore, we assume that organic carbon, nutrients and trace metals are displaced within the 360 landscape and accumulate in the floodplains. Regional soil erosion models estimate the loss of 2-5 tons ha-1 year-1 for a third of the country depending on precipitation, topography, soil 361 362 erodibility and land management (Carrasco-Letelier and Beretta-Blanco 2017). One possible 363 direct impact is the increasing eutrophication reported for larger local rivers, although the 364 models used by these authors did not link Chlorophyll-a concentrations with agricultural land 365 use (Beretta-Blanco and Carrasco-Letelier 2021 and replies). However, other vertical processes 366 and differences across land uses can be hidden deeper in the soil profile, and have not been 367 analysed in this study.

Organic carbon content and the exchangeable cations are strongly reduced in topsoils of grasslands, timber plantations and crops compared to native forests (Fig. 4b, d-h and 5b, d-h).

Of all the fertility proxies assessed, phosphorus in topsoil was most significantly affected by

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378 different land uses, being highest in native forests (Fig. 4c and 5c). The cation exchange 379 capacity (CEC) was highest in native forests (more than 160 percent of the CEC in grasslands) 380 and lowest in timber plantations, reaching only half of the CEC in grasslands (Table A1-A2). 381 Lower average nutrient concentrations and corresponding CEC reported for two timber 382 plantations in the East and one in the Norwest of Uruguay (Hernández et al., 2009 and 2016; 383 Céspedes-Payret et al., 2012) may result from a combined effect of topsoil degradation in 384 timber plantations due to management practices and to soil texture (Sandoval-Lopez et al., 385 2020). The trees' uptake and the general export of nutrients from fast growing timber 386 plantations through harvesting is higher than the natural input into those systems (Merino et 387 al., 2005). This effect is particularly relevant because timber plantations in Uruguay are on 388 'forestry priority soils', which are generally soils with low fertility, superficial to moderate 389 depth and good drainage (OAS, 1994), so afforestation might reduce even more their soil 390 fertility. 391 Comparing neighbouring sites of grassland and afforested grasslands, our data set shows no 392 significant depletion of nutrients by timber plantations (p=0.208) but a slightly higher average 393 of CEC at grasslands (Table A1). In general, plants are expected to compensate the extraction 394 of cations from upper soil by the 'uplift of nutrient' from deeper horizons (Jobbagy and Jackson, 395 2004). This has been questioned for *Eucalyptus* plantations on sandy soils, where, for example, 396 an increase of potassium in the topsoil was not observed compared to the neighbouring 397 grassland (Céspedes-Payret et al., 2012). Phosphorous and cations decline with sand content, 398 so physicochemical factors such as the percentage of sand and organic matter, influence soil 399 fertility (Sandoval-Lopez et al., 2020). A study of Eucalyptus plantations in South-East Brazil 400 did not show a significant depletion of nutrients and carbon; in contrast, carbon, potassium and 401 calcium increased in the topsoil after twelve years over one or two harvest and fertilization 402 cycles (McMahon et al., 2019). To sum up, the patterns observed in the various sites in our 403 survey and the other studies indicate very complex interactions of numerous factors. Removal

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406 of nutrients by high-yield timber plantations may exceed the capacity of nutrient exchange or 407 turnover of forest litter and wastes. Both stand management and environmental conditions (e.g., 408 precipitation) influence nutrient and carbon stocks (McMahon et al., 2019; Sandoval-Lopez et 409 al., 2020). 410 The C/N ratio in topsoils ranged within values reported for grassland and timber plantations of 411 the region (see Berthrong et al., 2012). In contrast to that study, however, we observed no 412 differences between grassland and timber plantations (Fig. 4a), but between topsoils of timber 413 plantations, native forests and crops (Fig. 5a). Organic matter and nitrogen decrease in topsoils 414 after grassland afforestation (Berthrong et al., 2009) and C/N ratio increases with plantation 415 age and decreases with precipitation (Berthrong et al., 2012). The topsoils of timber plantations 416 have, on average, lower N concentrations compared to other land uses (Table A2) and data in 417 literature (Jobbagy and Jackson 2003). 418 Although in cropland, nutrients are regularly compensated for by increased application of 419 fertilizers (Beretta-Blanco et al., 2019), timber plantations are usually fertilized only in the first year of planting (e.g., Binkley et al., 2017; Sandoval-Lopez et al., 2020). The CEC and the 420 average ratio between K<sup>+</sup>(Ca<sup>2+</sup> +Mg<sup>2+</sup>)<sup>-1/2</sup> in crops and grasslands are in the ranges reported by 421 422 Beretta-Blanco et al. (2019) with lower availability of potassium for crops. A shortage of 423 potassium and calcium is also most likely for future timber plantations after harvest, especially 424 if the logging process do not include bark stripping on site (Hernández et al., 2009) and due to 425 short time spans between harvest and new planting in the same area (Sandoval-Lopez et al., 426 2020), but see contrasting results of McMahon et al. (2019). The extraction of nutrients and 427 biomass due to grazing (Fernandez et al., 2017), timber (Hernández et al., 2009 and 2016) and crop production (Beretta-Blanco et al., 2019) lowers organic matter content and exchangeable 428

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cations, affects the physical, chemical and biological soil properties, and drives degradation.

4.2 Acidification in Uruguayan topsoils across land uses

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433 A further dimension of the soil degradation directly linked to the cation extraction is the 434 acidification of soils. This has been demonstrated for crops and pastures (Beretta-Blanco et al., 435 2019), and a study site with Eucalyptus (Céspedes-Payret et al., 2012) and is now broadly 436 supported by our topsoil samples originating from a wide range of different land uses across 437 hat gelöscht: values Uruguay. The pH of our topsoil samples are mainly in the category of very strongly to 438 extremely acidic and lowest in timber plantations (Fig. 6), which are below the means reported hat gelöscht: with hat gelöscht: values for 439 so far (Jobbagy and Jackson, 2003; Céspedes-Payret et al., 2012). Moreover, as our data on 440 topsoil pH fall short compared to the values estimated by the Food and Agriculture 441 Organization of the United Nations and the Intergovernmental Technical Panel on Soils (Caon 442 and Vargas, 2017), acidification and the deterioration of topsoil quality continues. 443 Acidification results from intensified land uses with nitrogen fertilization, with biological N 444 fixation by the legumes both used in the improved pastures (Modernel et al., 2016), or with hat gelöscht: so-called " hat gelöscht: ' 445 cation extraction by crop or timber harvest (Jobbagy and Jackson, 2003). So far, although forest 446 soils tend to be more acidic than agricultural soils due to acid-neutralizing treatments in the 447 latter (Baize and van Oort, 2014), we found no differences between topsoils of crops and native 448 forests (Fig. 6d) as the high organic carbon content in native forests buffers the process to a hat gelöscht: matter 449 certain extent. 450 4.3 Riverine Forest soils as sink for trace metals 451 In general, the average concentration of Zn, Cu, Cd, Cr, As and Pb in our topsoils were within 452 the expected ranges of samples from crops, pastures and grassland in the region (Lavado et al., 453 1998, 2004; Roca, 2015). Some samples, especially from orchards and crops, exceeded the 454 background values of copper, cadmium and arsenic (Table A2; Roca, 2015). Little is known 455 about the pedo-geochemical background of the Pampean soils (Roca, 2015): for example, high 456 hat gelöscht: content arsenic concentration in Uruguayan ground waters has been hypothesized to be due to 457 quaternary ash deposits (Wu et al., 2021). In addition, the lateral heterogeneity of Pampean

soils over short distances makes separating geochemical and anthropic signatures difficult

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466 (Roca, 2015). However, that the main risk of soil contamination in the region is from the 467 application of fertilizers and agrochemicals is uncontested (Kaushal et al., 2018). 468 To our knowledge, there has been no regional study of trace metals in the native riverine forests 469 or timber plantations. Our work thus expands the evidence base for these land uses.. The 470 topsoils of riverine forests accumulate more trace metals compared to those of timber 471 plantations and crops (Fig. 4l-o). The higher amount of soil organic matter in riverine forests 472 favours the retention of cations, including trace metals. Although the origin of potentially 473 harmful elements in forest soils have been primarily attributed to atmospheric deposition 474 (Baize and van Oort, 2014), atmospheric deposition only plays a major role in vicinity of urban 475 or industrial development, and our data from rural sites suggests a different entry path from the 476 surrounding land uses to the riverine forests. High acidification and low amounts of organic 477 matter reduce the retention of trace metals in the soil of timber plantations, and elements leach 478 out of the soil towards the water table (Baize and van Oort, 2014). The acidification strongly 479 contributes to the overall mobility of base cations into the 'chemical cocktail of the 480 Anthropocene' (Kaushal et al., 2018), including trace metals. We thus observe positive 481 feedback in already impoverished soils with high acidity favouring cations solubility, in 482 addition to the uptake by trees intensifies this effect. Timber plantations extract trace metals 483 from soil and also accumulate it in the bark or leaves, so they have been used for 484 phytoremediation (Li et al., 2020). This may explain the higher concentration of cadmium in 485 grassland compared to timber plantations (Fig. 4m). Differences between Eucalyptus and Pinus 486 stands may be related to different age classes, as the later may have extracted more lead and 487 chromium from the soil due to their older stand age with rotation periods of about 20 years (Li

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Our study provides evidence that the loss of soil organic carbon limits not only the productivity

4.4 Carbon storage in topsoils of *Eucalyptus* plantations?

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et al., 2020).

| 496 | Grassland conversion to cropland decreases soil organic carbon, storage compared to adjacent                | hat gelöscht: SOC    |
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| 497 | native grasslands (Hernandez-Ramirez et al., 2021).   |                      |
| 498 | Afforestation of croplands has been also discussed as a carbon sequestration measure to                     |                      |
| 499 | proactively address and effectively mitigate ongoing climate change within a person's lifetime              |                      |
| 500 | (Hernandez-Ramirez et al., 2021; Mayer et al. 2020). Although carbon stocks of four                         |                      |
| 501 | Eucalyptus stands in the Brazilian Cerrados increased but did not change in four Eucalyptus                 |                      |
| 502 | stands within the Atlantic Forest (McMahon et al., 2019), this may not be the case for short                |                      |
| 503 | rotation of Eucalyptus plantation on Campos grasslands. In our topsoil samples originating                  |                      |
| 504 | from 28 different stands across Uruguay, organic carbon is lowest in topsoils of timber                     | hat gelöscht: matter |
| 505 | plantations (Fig. 4b). Similar amounts have been reported for a timber plantation in the East of            |                      |
| 506 | Uruguay (Céspedes-Payret et al., 2012), and in North-Eastern Argentine (Sandoval-Lopez et                   |                      |
| 507 | al., 2020). Our data therefore provide clear evidence that rather than contributing to carbon               |                      |
| 508 | sequestration in the topsoil, the carbon release from the transformation of native grasslands to            |                      |
| 509 | plantation with these fast-growing species has several adverse effects depending on                         |                      |
| 510 | precipitation and soil type (reviewed by Mayer et al., 2020).   |                      |
| 511 | Several trade-offs between carbon sequestration through afforestation and local water yield and             |                      |
| 512 | soil fertility have been demonstrated, including nutrient and soil organic matter depletion,                |                      |
| 513 | acidification, and biodiversity loss and corresponding challenges for landscape conservation                |                      |
| 514 | (e.g., Jackson et al., 2005; Veldman et al., 2015; Friggens et al., 2020). Soil carbon changes              |                      |
| 515 | dynamically during the first decade of afforestation. Remaining grassland carbon declines,                  |                      |
| 516 | while tree carbon gain starts (e.g. Paul et al., 2002). Although a net gain of carbon is expected           |                      |
| 517 | when the new forests approached equilibrium after decades (Hernández-Ramirez et al., 2021),                 |                      |
| 518 | in contrast to long-lasting forests, <i>Eucalyptus</i> harvest in Uruguay takes place after less than a     |                      |
| 519 | decade (appr. 7-10 years). Soil organic <u>carbon</u> does not differ between <i>Eucalyptus</i> plantations | hat gelöscht: matter |
| 520 | and neighbouring grasslands (Appendix A; Table A1 and A2). Another study near our sites 12                  |                      |
| 521 | and 13 also did not find a significant difference between the soil organic carbon of the upper              | hat gelöscht: SOC    |

526 soil (0-30cm) compared to afforested versus native grasslands (Hernández et al., 2016). A study 527 near our sites 7, 8, 10 reported higher top soil carbon of grassland compared with afforested 528 sites in their vicinity only if sand content is lower than sixty percent (Sandoval-Lopez et al., 529 2020). McMahon et al. (2019) identified a greater carbon gain under Eucalyptus stands 530 compared to (potential) carbon losses in neighbouring degraded Cerrado grasslands. There is 531 evidence that the retention of residues after harvest increases the carbon stock in soil (Mayer 532 et al., 2020). Long term monitoring of carbon stocks is needed to verify the influences of 533 management and environmental changes. A regional study on Eucalyptus plantations across 534 different biomes in Brazil shows both decreases and no changes depending on precipitation in 535 the dry season, clay content and on the initial stock of carbon in the soil (Cook et al., 2016) 536 The simplistic solution of huge tree plantations to compensate anthropogenic CO<sub>2</sub> emissions 537 has been challenged in the last decade, and some crucial lessons learnt have been summarized 538 (Di Sacco et al., 2020). Since grasslands are more sustainable carbon sinks compared to 539 climate-vulnerable monocultures such as timber plantations (Dass et al. 2018), avoiding 540 afforestation of previously non-forested lands is important. This is the case for Eucalyptus 541 afforestation in the originally forestless grasslands of Uruguay: models on carbon sequestration 542 and dynamics in Mollisols and Oxisols under South American grasslands estimated a higher 543 carbon retention efficiency under grasslands compared to afforested sites, suggesting that 544 silvopastoral systems are a potential solution for soil carbon sequestration in tropical soils 545 (Berhongaray and Alvarez, 2019). 546 Connecting or expanding existing forest and using native species for plantings (Di Sacco et al., 547 2020) is also recommended (see also Alvaro et al. 2020, Ortiz et al. 2020). Our topsoil data 548 indicates that carbon sequestration occurs mainly in the topsoils of native riverine forests that 549 cover less than five percent of the Uruguayan territory. Consequently, the expansion of native 550 forests and the use of native species in forestry project for long term establishment can reduce adverse effects of timber plantations. While there is preliminary evidence that N-fixing tree 551

552 species can help increase local C stocks of afforestation, because of their potential for invasion, 553 exotic N-fixers should be avoided (Mayer et al. 2020). 554 Recent studies indicated that novel techniques such as perennial grain cropping can help to turn 555 around cropland degradation into beneficial cropland aggradation by using the advantage of 556 perennial vegetation of conserving and even enhancing short term and long-term soil carbon 557 storage and other ecosystems services (Kim et al. 2022). hat gelöscht: 558 4.5. Back to more conservative grassland management? 559 Our soil survey data shows strong soil degradation of Uruguayan black soils from erosion, 560 acidification and contamination, and suggests a translocation of nutrients and organic carbon hat gelöscht: indicates hat gelöscht: matter across the landscape from grassland, timber and crop plantations to the riverine forests. The 561 562 potential of grasslands as cropland reserve have been largely overestimated (Lambin et al., 563 2013), and they have already degraded during the last decades by inappropriate land management techniques (Jaurena et al., 2021, De Faccio et al., 2021), lack of mainstreaming 564 soil conservative techniques (García-Préchac, 2004; Fernandez et al., 2017), decoupling of crop 565 566 and livestock (De Faccio et al, 2021) plus climate change impacts with storm water events and hat gelöscht: f 567 drought, all of which trigger soil erosion (Wingeyer et al. 2015). From the although very limited point of view on topsoils, the concept of conserving 'old growth grasslands' with extensive use 568 569 (Veldman et al. 2015) appears a more promising strategy to put the 'grasslands at the core' in 570 the Campos region than the use intensification strategies envisioned by Jaurena et al. (2021). 5. Conclusions 571 572 The land use intensification in Uruguay associated with increasing inputs of energy, nutrients and pesticides leads to an overall loss of soil fertility and increasing toxicity related to 573 574 acidification, salinization and trace metal contaminants. Our data demonstrate the high amounts of organic carbon, nutrients and trace metals in topsoil samples from riverine forests, 575

suggesting transport of soil particles from the surrounding grasslands, crop or timber plantations to the borders of rivers, streams and creeks. Of all the fertility proxies assessed, phosphorus in topsoil was most significantly affected by different land uses, being highest in native forests. Cation exchange capacity was also highest in native forests and lowest in timber plantations, where only half that of grasslands was measured. Our study highlights that soil acidification is ongoing and probably also mobilizing trace metals and their accumulation in riverine forest topsoils.

hat gelöscht: Our study allowed us to corroborate soil acidification processes as a result of soil change. Furthermore, based on the probable accumulation of heavy metals in riverine forests, we infer that management and conservation in the vicinity of productive land uses should be a priority.

## Data availability

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- 588 All data generated or analysed in this study are included in this article (and its supplementary
- 589 materials). Further data are available from the corresponding authors upon reasonable request.

### 590 Author contributions

- 591 IS, LRR, ST and MB conceptualized the study, performed the soil sampling and data analysis.
- 592 EZ advised the laboratory analyses. IS wrote the initial draft. LRR, ST, MB and EZ contributed
- 593 to generating and reviewing the subsequent versions of the manuscript. IS received the funding
- 594 for the study.

595

## **Competing interests**

596 The authors declare that they have no conflict of interest

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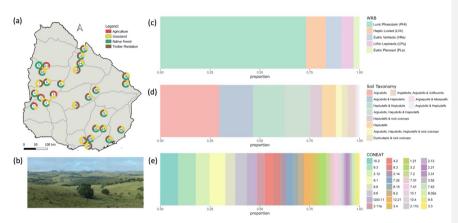
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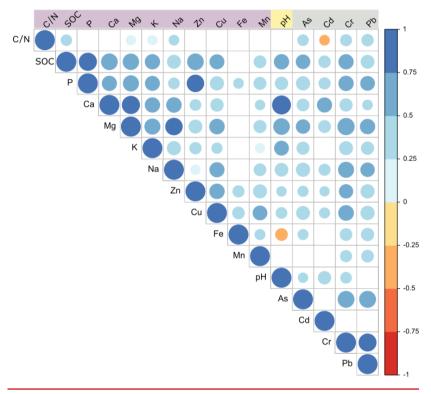
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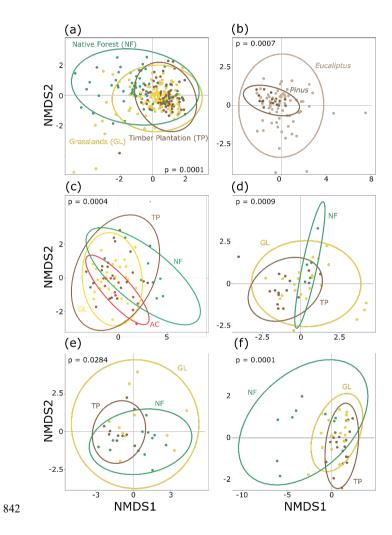
**Figure 1:** Study design and sampling. (a) Location of the 28 monitoring sites across Uruguay including land use types sampled (grassland (b); timber plantations, native forest and agricultural land. Proportion of plots with particular category of soil classification according to the World Reference Base (WRB, c), Soil Taxonomy (d) and CONEAT (Comisión Nacional de Estudio Agronómico de la Tierra, e). Photo: RuralFutures.

C/N SOM P Ca Mg + W Na Zn

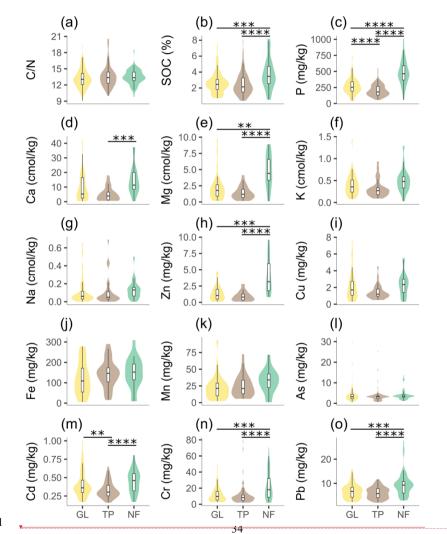
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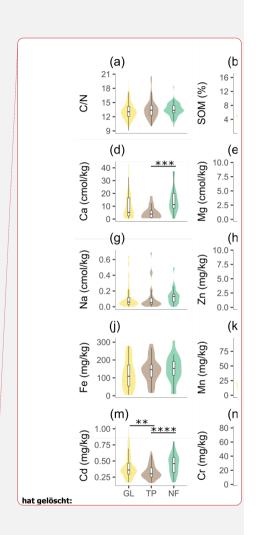


**Figure 2.** Spearman's rank correlations for parameters of topsoils (n=80) regarding (a) fertility proxies, (b) acidity and (c) trace metals. Colour intensity and the size of the circle are proportional to the correlation coefficients ( $\rho$ ). Empty slots show correlations with p>0.05.

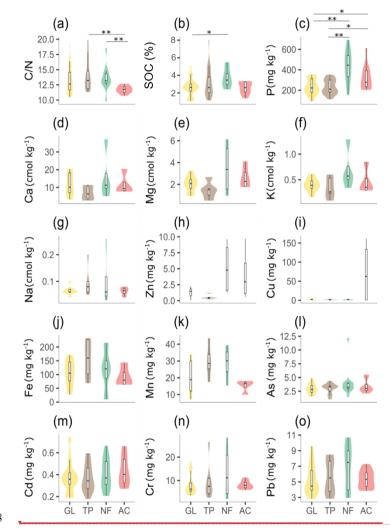


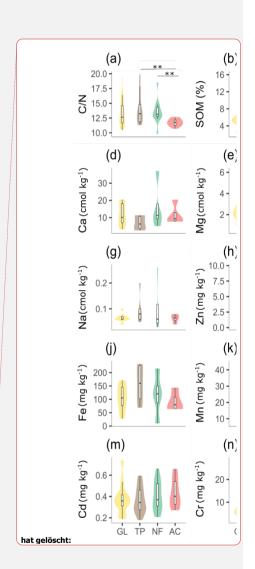
**Figure 3.** Non-metric multidimensional scaling showing significant clustering differences among samples from (a) grassland (GL), timber plantations (TP) and native forests (NF); (b) among samples from *Pinus* and *Eucalyptus* plantations; (c) among land uses (including agriculture AC) in Argiudolls; (d) among land uses in Argiudolls & Hapluderts; (e) among land uses in Argiudolls, Hapudolls & Hapludalfs & Hapludults.



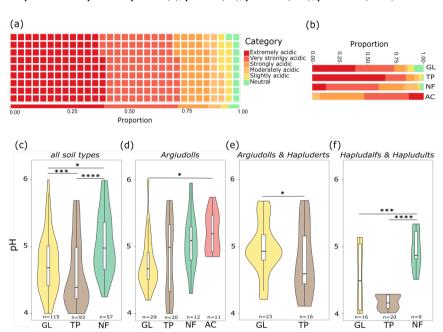


**Figure 4.** Violin box plots for significant Kruskal-Wallis Tests across evaluated land uses (i.e. grassland (GL), timber plantation (TP) and native forest (NF)) for each variable. Significance in posterior Wilcoxon pairwise comparisons is depicted as p<0.05 (\*), p<0.01 (\*\*\*), p<0.001 (\*\*\*\*).





**Figure 5**. Violin box plots for significant Kruskal-Wallis Tests in 'Argiudolls' Soil Taxonomy category for fertility variables across available land uses (GL: Grassland, TP: Timber plantation, NF: Native forest, AC: Agriculture). Significance in posterior Wilcoxon pairwise comparisons is depicted as p<0.05 (\*), p<0.01 (\*\*), p<0.001 (\*\*\*), p<0.0001 (\*\*\*\*).



**Figure 6.** Acidity of all topsoils samples (a) and (b) according to different land use (GL: Grassland, TP: Timber plantation, NF: Native forest, AC: Agriculture). Proportion of samples with a given acidic category (Kellogg, 1993). pH of topsoil given as violin box plots for

significant Kruskal-Wallis Tests across different land uses independent from soil Order (c) and separated per soil Orders (d-f). Sample number (n) is given. Significance in posterior Wilcoxon pairwise comparisons is depicted as p<0.05 (\*), p<0.01 (\*\*\*), p<0.001 (\*\*\*\*), p<0.0001 (\*\*\*\*).

Table 1. General characteristics of Uruguayan topsoils: descriptive statistics for the parameters
 of all single samples (n) across different land uses and classification to different soil types (for
 details on different land uses i.e. grassland, timber plantations, native forests and crops see

Appendix A1-A2 and on different soil types see Appendix A3-4). SD = standard derivation.

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| Variable                         | n   | Mean | SD   | Minimum | Maximum |
|----------------------------------|-----|------|------|---------|---------|
| Soil moisture (%)                | 280 | 18.9 | 10.8 | 1.34    | 51.5    |
| N_total (%)                      | 280 | 0.2  | 0.14 | 0.04    | 1.2     |
| C_total (%)                      | 280 | 3.0  | 2.3  | 0.4     | 25.7    |
| C/N ratio                        | 279 | 13.3 | 1.8  | 9.1     | 20.6    |
| <u>SOC (%)</u>                   | 267 | 5.6  | 3.0  | 0.8     | 16.4    |
| P (mg kg <sup>-1</sup> )         | 278 | 295  | 166  | 43      | 1008    |
| Ca (cmol kg <sup>-1</sup> )      | 82  | 9.4  | 8.9  | 0.3     | 42.7    |
| Mg (cmol kg <sup>-1</sup> )      | 82  | 2.5  | 2.2  | 0.1     | 9.7     |
| K (cmol kg <sup>-1</sup> )       | 82  | 0.4  | 0.25 | 0.03    | 1.44    |
| Na (cmol kg-1)                   | 82  | 0.1  | 0.14 | 0       | 0.69    |
| Zn (mg kg <sup>-1</sup> )        | 82  | 1.9  | 2.1  | 0.1     | 9.7     |
| Cu (mg kg <sup>-1</sup> )        | 82  | 5.4  | 22.1 | 0.3     | 161.2   |
| Fe (mg kg <sup>-1</sup> )        | 82  | 134  | 75   | 5       | 309     |
| Mn (mg kg <sup>-1</sup> )        | 81  | 25   | 17   | 0.7     | 93      |
| pH                               | 279 | 4.8  | 0.8  | 3.6     | 7.34    |
| As (mg kg <sup>-1</sup> )        | 279 | 3.9  | 3.6  | 0.6     | 30.7    |
| Cd (mg kg <sup>-1</sup> )        | 274 | 0.4  | 0.2  | 0.2     | 1       |
| Cr (mg kg <sup>-1</sup> )        | 280 | 13.6 | 12.6 | 2       | 78.9    |
| Pb (mg kg <sup>-1</sup> )        | 280 | 7    | 3.4  | 2       | 27.6    |
| CEC (cmol kg-1)                  | 82  | 12.4 | 10.7 | 0.5     | 50.1    |
| $K^+/(Ca^{2+}+Mg^{2+})$          | 82  | 0.07 | 0.08 | 0.01    | 0.43    |
| $K^{+}/(Ca^{2+}+Mg^{2+}+Na^{+})$ | 82  | 0.07 | 0.07 | 0.01    | 0.42    |

| hat gelöscht: is | s  |
|------------------|--|
| hat gelöscht: a  | and 'humidity' stands for the gravimetric moisture |
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# **Appendices**

# Appendix A

**Table A1.** Descriptive statistics of topsoil variables for all single soil types at grassland and native forests. Number of samples (n), mean, standard deviation (SD), minimum (Min) and maximum (Max) of each variable are given<sub>e</sub>

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|                           |     |      | Grasslar    | nd   |      | Native forests |      |      |      |      |  |  |
|---------------------------|-----|------|-------------|------|------|----------------|------|------|------|------|--|--|
| Variable (unit)           | n   | Mean | Mean SD Min |      |      | n              | Mean | SD   | Min  | Max  |  |  |
| Soil moisture (%)         | 115 | 20.4 | 11.5        | 1.5  | 51.5 | 57             | 25.8 | 9.3  | 6.3  | 50.3 |  |  |
| N_total (%)               | 115 | 0.22 | 0.13        | 0.06 | 0.80 | 57             | 0.30 | 0.19 | 0.04 | 1.20 |  |  |
| C_total (%)               | 115 | 2.9  | 1.9         | 0.7  | 10.8 | 57             | 4.2  | 3.6  | 0.5  | 25.7 |  |  |
| C/N ratio                 | 115 | 13.2 | 1.7         | 9.1  | 19.4 | 56             | 13.5 | 1.4  | 9.8  | 18.5 |  |  |
| <u>SOC (%)</u>            | 109 | 5.3  | 2.6         | 1.4  | 15.2 | 53             | 7.5  | 3.8  | 1.1  | 16.4 |  |  |
| P (mg/kg)                 | 114 | 272  | 124         | 43   | 749  | 57             | 484  | 194  | 56   | 1009 |  |  |
| Ca (cmol kg-1)            | 32  | 9.5  | 10.0        | 0.3  | 42.7 | 18             | 15.1 | 10.0 | 0.3  | 37.3 |  |  |
| Mg (cmol kg-1)            | 32  | 2.2  | 2.1         | 0.18 | 9.7  | 18             | 4.8  | 2.6  | 0.12 | 8.9  |  |  |
| K (cmol kg-1)             | 32  | 0.40 | 0.26        | 0.08 | 1.44 | 18             | 0.50 | 0.28 | 0.03 | 1.29 |  |  |
| Na (cmol kg-1)            | 32  | 0.12 | 0.15        | 0.00 | 0.65 | 18             | 0.14 | 0.12 | 0.00 | 0.50 |  |  |
| Zn (mg kg-1)              | 32  | 1.4  | 1.2         | 0.10 | 4.7  | 18             | 4.0  | 2.7  | 0.80 | 9.60 |  |  |
| Cu (mg kg-1)              | 32  | 2.2  | 1.8         | 0.30 | 7.7  | 18             | 2.4  | 1.5  | 0.30 | 5.50 |  |  |
| Fe (mg kg <sup>-1</sup> ) | 32  | 121  | 81          | 5    | 279  | 18             | 149  | 76   | 10   | 309  |  |  |
| Mn (mg kg <sup>-1</sup> ) | 32  | 22   | 18          | 2    | 93   | 17             | 33   | 18   | 1    | 72   |  |  |
| pН                        | 114 | 4.9  | 0.9         | 3.6  | 7.3  | 57             | 5.1  | 0.7  | 3.6  | 7.2  |  |  |
| As (mg kg <sup>-1</sup> ) | 115 | 4.3  | 4.5         | 0.6  | 30.7 | 56             | 4.1  | 2.5  | 1.2  | 13.6 |  |  |
| Cd (mg kg-1)              | 111 | 0.41 | 0.17        | 0.17 | 1.00 | 57             | 0.45 | 0.15 | 0.17 | 0.81 |  |  |
| Cr (mg kg <sup>-1</sup> ) | 115 | 12.3 | 9.2         | 2.6  | 49.5 | 57             | 22.8 | 17.3 | 2.0  | 70.7 |  |  |
| Pb (mg kg <sup>-1</sup> ) | 115 | 6.8  | 3.0         | 2.3  | 15.9 | 57             | 9.5  | 4.8  | 2.9  | 27.7 |  |  |
| CEC (cmol/ kg-1)          | 32  | 12.2 | 11.5        | 0.8  | 50.1 | 18             | 20.4 | 11.4 | 0.5  | 44.6 |  |  |
| $K^+/(Ca^{2+}+Mg^{2+})$   | 32  | 0.08 | 0.10        | 0.01 | 0.43 | 18             | 0.03 | 0.02 | 0.01 | 0.07 |  |  |
| K+/(Ca2++Mg2++Na+)        | 32  | 0.08 | 0.10        | 0.01 | 0.42 | 18             | 0.03 | 0.02 | 0.01 | 0.07 |  |  |

hat gelöscht: . 'Humidity' stands for the gravimetric moisture content

hat gelöscht: Humidity

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**Table A2.** Descriptive statistics for of topsoil variables for all single soil types at timber plantations and crops. Number of samples (n), mean, standard deviation (SD), minimum (Min) and maximum (Max) of each variable are given.

| 1 | hat gelöscht: . | 'Humidity' | stands | for the | gravimetric | moisture |
|---|-----------------|------------|--------|---------|-------------|----------|
| l | content         |            |        |         |             |          |

|                                  |    | Ti   | mber plan | tations |      | Crops |      |      |      |       |  |  |
|----------------------------------|----|------|-----------|---------|------|-------|------|------|------|-------|--|--|
| Variable (unit)                  | n  | Mean | SD        | Min     | Max  | n     | Mean | SD   | Min  | Max   |  |  |
| Soil moisture (%)                | 93 | 13.2 | 7.0       | 1.7     | 38.0 | 15    | 18.1 | 12.0 | 1.3  | 50.5  |  |  |
| N_total (%)                      | 93 | 0.18 | 0.10      | 0.04    | 0.61 | 15    | 0.22 | 0.06 | 0.13 | 0.29  |  |  |
| C_total (%)                      | 93 | 2.5  | 1.5       | 0.4     | 8.4  | 15    | 2.6  | 0.7  | 1.4  | 3.3   |  |  |
| C/N ratio                        | 93 | 13.6 | 2.1       | 10.0    | 20.6 | 15    | 11.7 | 0.7  | 10.5 | 12.8  |  |  |
| <u>SOC (</u> %)                  | 90 | 4.9  | 2.8       | 0.8     | 16.1 | 15    | 5.2  | 1.3  | 2.8  | 6.7   |  |  |
| P (mg kg <sup>-1</sup> )         | 93 | 206  | 91        | 56      | 551  | 14    | 310  | 120  | 142  | 613   |  |  |
| Ca (cmol kg <sup>-1</sup> )      | 27 | 5.2  | 4.8       | 0.6     | 18   | 5     | 10.8 | 5.5  | 7.2  | 20.4  |  |  |
| Mg (cmol kg <sup>-1</sup> )      | 27 | 1.4  | 1.0       | 0.16    | 4.1  | 5     | 2.5  | 1.0  | 1.76 | 4.1   |  |  |
| K (cmol kg <sup>-1</sup> )       | 27 | 0.32 | 0.21      | 0.09    | 0.93 | 5     | 0.46 | 0.23 | 0.29 | 0.85  |  |  |
| Na (cmol kg <sup>-1</sup> )      | 27 | 0.11 | 0.16      | 0.00    | 0.69 | 5     | 0.06 | 0.02 | 0.03 | 0.08  |  |  |
| Zn (mg kg <sup>-1</sup> )        | 27 | 0.94 | 0.74      | 0.10    | 2.8  | 5     | 3.7  | 3.7  | 0.60 | 9.7   |  |  |
| Cu (mg kg <sup>-1</sup> )        | 27 | 1.5  | 0.9       | 0.5     | 4.5  | 5     | 57.7 | 78.8 | 0.8  | 161.0 |  |  |
| Fe (mg kg <sup>-1</sup> )        | 27 | 143  | 69        | 16      | 290  | 5     | 115  | 62   | 63   | 210   |  |  |
| Mn (mg kg <sup>-1</sup> )        | 27 | 25   | 16        | 6       | 72   | 5     | 17   | 5    | 10   | 23    |  |  |
| pН                               | 93 | 4.5  | 0.7       | 3.6     | 6.8  | 15    | 5.1  | 0.4  | 4.2  | 5.7   |  |  |
| As (mg kg <sup>-1</sup> )        | 93 | 3.4  | 3.0       | 0.7     | 25.5 | 15    | 3.1  | 1.2  | 1.1  | 5.5   |  |  |
| Cd (mg kg <sup>-1</sup> )        | 91 | 0.33 | 0.11      | 0.16    | 0.65 | 15    | 0.41 | 0.14 | 0.21 | 0.66  |  |  |
| Cr (mg kg <sup>-1</sup> )        | 93 | 10.6 | 11.3      | 2.0     | 78.9 | 15    | 7.8  | 1.8  | 5.0  | 11.3  |  |  |
| Pb (mg kg <sup>-1</sup> )        | 93 | 6.1  | 2.3       | 2.0     | 11.7 | 15    | 5.1  | 1.2  | 3.4  | 7.3   |  |  |
| CEC (cmol kg <sup>-1</sup> )     | 27 | 7.0  | 5.6       | 1.0     | 20.6 | 5     | 13.8 | 6.5  | 9.5  | 25.0  |  |  |
| $K^{+}/(Ca^{2+}+Mg^{2+})$        | 27 | 0.08 | 0.07      | 0.02    | 0.25 | 5     | 0.04 | 0.03 | 0.02 | 0.09  |  |  |
| $K^{+}/(Ca^{2+}+Mg^{2+}+Na^{+})$ | 27 | 0.08 | 0.07      | 0.02    | 0.25 | 5     | 0.04 | 0.03 | 0.02 | 0.08  |  |  |

hat gelöscht: SOM

hat gelöscht: Humidity

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**Table A3.** Descriptive statistics of topsoil variables for Argiudolls and for grassland and native forests within of Argiudolls. Number of single samples (n), mean, standard deviation (SD), minimum (Min) and maximum (Max) of each variable are given.

| hat | gelöscht: | Humidity  |  |  |
|-----|-----------|-----------|--|--|
|     |           |           |  |  |
|     |           |           |  |  |
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|     |           |           |  |  |
| _   |           |           |  |  |
| hat | gelöscht: | SOM       |  |  |
| hat | gelöscht: | Organic C |  |  |
| _   | -         |           |  |  |

hat gelöscht: . 'Humidity' stands for the gravimetric moisture content

|                              | Total Argiudolls |      |      |      |      |    | Grassland |      |      |      |    | Native forests |      |      |      |  |
|------------------------------|------------------|------|------|------|------|----|-----------|------|------|------|----|----------------|------|------|------|--|
| Variable (Unit)              | n                | Mean | SD   | Min  | Max  | n  | Mean      | SD   | Min  | Max  | n  | Mean           | SD   | Min  | Max  |  |
| Soil moisture (%)            | 77               | 18.3 | 10.9 | 1.3  | 50.5 | 31 | 19.0      | 9.2  | 5.0  | 49.0 | 12 | 26.4           | 13.4 | 9.0  | 48.2 |  |
| N total (%)                  | 77               | 0.24 | 0.15 | 0.07 | 1.20 | 31 | 0.21      | 0.11 | 0.07 | 0.66 | 12 | 0.34           | 0.28 | 0.13 | 1.20 |  |
| C_total (%)                  | 77               | 3.3  | 3.0  | 1.1  | 25.7 | 31 | 2.9       | 1.8  | 1.1  | 10.8 | 12 | 5.3            | 6.5  | 1.7  | 25.7 |  |
| C/N ratio                    | 76               | 13.3 | 2.2  | 9.8  | 19.9 | 31 | 13.3      | 2.1  | 10.5 | 19.4 | 11 | 13.6           | 2.1  | 9.8  | 18.5 |  |
| <u>SOC (%)</u>               | 73               | 5.8  | 2.5  | 2.1  | 16.1 | 30 | 5.2       | 1.9  | 2.1  | 10.0 | 9  | 7.4            | 2.1  | 4.8  | 11.0 |  |
| P (mg kg <sup>-1</sup> )     | 76               | 280  | 122  | 93   | 693  | 30 | 234       | 78   | 93   | 353  | 12 | 438            | 155  | 155  | 693  |  |
| Ca (cmol kg <sup>-1</sup> )  | 21               | 11.4 | 8.0  | 2.6  | 37.1 | 8  | 11.5      | 6.7  | 3.2  | 20.5 | 4  | 16.2           | 14.3 | 5.1  | 37.1 |  |
| Mg (cmol kg-1)               | 21               | 2.3  | 1.4  | 0.5  | 6.1  | 8  | 2.0       | 0.8  | 0.9  | 3.2  | 4  | 3.5            | 2.5  | 1.0  | 6.1  |  |
| K (cmol kg-1)                | 21               | 0.45 | 0.26 | 0.1  | 1.3  | 8  | 0.4       | 0.14 | 0.16 | 0.61 | 4  | 0.69           | 0.41 | 0.34 | 1.29 |  |
| Na (cmol kg <sup>-1</sup> )  | 21               | 0.08 | 0.05 | 0.03 | 0.26 | 8  | 0.07      | 0.02 | 0.04 | 0.10 | 4  | 0.10           | 0.11 | 0.03 | 0.26 |  |
| Zn (mg kg <sup>-1</sup> )    | 21               | 2.4  | 3.0  | 0.1  | 10   | 8  | 1.3       | 0.8  | 0.1  | 2.2  | 4  | 5.1            | 4.3  | 1.3  | 10   |  |
| Cu (mg kg <sup>-1</sup> )    | 21               | 15.5 | 42.7 | 0.8  | 161  | 8  | 2.6       | 1.6  | 1.0  | 6.0  | 4  | 1.9            | 1.0  | 0.8  | 3.0  |  |
| Fe (mg kg <sup>-1</sup> )    | 21               | 117  | 62   | 10   | 232  | 8  | 104       | 50   | 28   | 171  | 4  | 117            | 84   | 10   | 216  |  |
| Mn (mg kg <sup>-1</sup> )    | 21               | 23.3 | 10.4 | 7.1  | 43.6 | 8  | 20.5      | 10.4 | 7.1  | 33.8 | 4  | 28.8           | 10.4 | 15.4 | 39.4 |  |
| pH                           | 77               | 5.0  | 0.7  | 3.8  | 6.6  | 31 | 4.9       | 0.7  | 3.9  | 6.6  | 12 | 5.2            | 0.5  | 4.3  | 6.1  |  |
| As (mg kg <sup>-1</sup> )    | 76               | 3.2  | 1.4  | 1.0  | 12.2 | 31 | 2.9       | 0.9  | 1.5  | 4.9  | 11 | 4.0            | 2.8  | 1.5  | 12.2 |  |
| Cd (mg kg <sup>-1</sup> )    | 76               | 0.4  | 0.1  | 0.2  | 0.7  | 30 | 0.4       | 0.1  | 0.2  | 0.7  | 12 | 0.4            | 0.1  | 0.2  | 0.7  |  |
| Cr (mg kg <sup>-1</sup> )    | 77               | 9.3  | 6.2  | 2.2  | 28.3 | 31 | 8.1       | 4.1  | 3.6  | 18.5 | 12 | 13.2           | 9.5  | 2.2  | 28.3 |  |
| Pb (mg kg <sup>-1</sup> )    | 77               | 5.6  | 2.1  | 2.8  | 10.7 | 31 | 5.2       | 1.8  | 2.8  | 9.6  | 12 | 6.8            | 2.9  | 2.9  | 10.7 |  |
| CEC (cmol kg <sup>-1</sup> ) | 21               | 14.2 | 9.3  | 3.4  | 44.6 | 8  | 14.0      | 7.3  | 4.5  | 23.1 | 4  | 20.4           | 16.7 | 6.4  | 44.6 |  |
| $K^+/(Ca^{2+}+Mg^{2+})$      | 21               | 0.04 | 0.02 | 0.02 | 0.09 | 8  | 0.03      | 0.02 | 0.02 | 0.07 | 4  | 0.04           | 0.01 | 0.03 | 0.06 |  |
| K+/(Ca2++Mg2++Na+)           | 21               | 0.04 | 0.02 | 0.02 | 0.08 | 8  | 0.03      | 0.02 | 0.02 | 0.07 | 4  | 0.04           | 0.01 | 0.03 | 0.06 |  |

921 T: 922 cr 923 m

**Table A4.** Descriptive statistics of topsoil variables for Argiudolls and timber plantations and crops within of Argiudolls. Number of single samples (n), mean, standard deviation (SD), minimum (Min) and maximum (Max) of each variable are given.

|   | hat gelöscht: | 'Humidity' | stands for | the | gravimetric moisture |
|---|---------------|------------|------------|-----|----------------------|
| Į | content.      |            |            |     |                      |

hat gelöscht: Humidity

|                             |    | Tot  | al Argiu | idolls |      | Timber plantations |      |      |      |      | Crops |      |      |      |      |  |
|-----------------------------|----|------|----------|--------|------|--------------------|------|------|------|------|-------|------|------|------|------|--|
| Variable (unit)             | n  | Mean | SD       | Min    | Max  | n                  | Mean | SD   | Min  | Max  | n     | Mean | SD   | Min  | Max  |  |
| Soil moisture (%)           | 77 | 18.3 | 10.9     | 1.3    | 50.5 | 22                 | 14.0 | 7.8  | 5.5  | 38.0 | 12    | 16.5 | 13.0 | 1.3  | 50.5 |  |
| N_total (%)                 | 77 | 0.24 | 0.15     | 0.07   | 1.20 | 22                 | 0.22 | 0.10 | 0.10 | 0.45 | 12    | 0.22 | 0.06 | 0.13 | 0.29 |  |
| C_total (%)                 | 77 | 3.3  | 3.0      | 1.1    | 25.7 | 22                 | 3.2  | 1.6  | 1.2  | 8.0  | 12    | 2.6  | 0.7  | 1.4  | 3.3  |  |
| C/N ratio                   | 76 | 13.3 | 2.2      | 9.8    | 19.9 | 22                 | 14.0 | 2.6  | 11.3 | 19.9 | 12    | 11.8 | 0.6  | 10.7 | 12.7 |  |
| <u>SOC (%)</u>              | 73 | 5.8  | 2.5      | 2.1    | 16.1 | 22                 | 6.3  | 3.3  | 2.3  | 16.1 | 12    | 5.2  | 1.4  | 2.8  | 6.7  |  |
| P (mg kg <sup>-1</sup> )    | 76 | 280  | 122      | 93     | 693  | 22                 | 234  | 70   | 111  | 353  | 12    | 321  | 119  | 204  | 613  |  |
| Ca (cmol kg-1)              | 21 | 11.4 | 8.0      | 2.6    | 37.1 | 5                  | 6.9  | 4.1  | 2.6  | 11.4 | 4     | 11.7 | 5.9  | 7.7  | 20.4 |  |
| Mg (cmol kg-1)              | 21 | 2.3  | 1.4      | 0.5    | 6.1  | 5                  | 1.5  | 0.8  | 0.5  | 2.7  | 4     | 2.6  | 1.1  | 1.8  | 4.1  |  |
| K (cmol kg-1)               | 21 | 0.45 | 0.26     | 0.09   | 1.29 | 5                  | 0.34 | 0.21 | 0.09 | 0.60 | 4     | 0.46 | 0.27 | 0.29 | 0.85 |  |
| Na (cmol kg <sup>-1</sup> ) | 21 | 0.08 | 0.05     | 0.0    | 0.3  | 5                  | 0.1  | 0.06 | 0.05 | 0.20 | 4     | 0.06 | 0.02 | 0.04 | 0.08 |  |
| Zn (mg kg <sup>-1</sup> )   | 21 | 2.4  | 3.0      | 0.1    | 9.7  | 5                  | 0.5  | 0.4  | 0.2  | 1.2  | 4     | 4.1  | 4.2  | 0.6  | 9.7  |  |
| Cu (mg kg <sup>-1</sup> )   | 21 | 15.5 | 42.7     | 0.8    | 161  | 5                  | 2.0  | 1.5  | 1.0  | 4.5  | 4     | 71.8 | 83.4 | 0.8  | 161  |  |
| Fe (mg kg <sup>-1</sup> )   | 21 | 117  | 62       | 10     | 232  | 5                  | 158  | 73   | 69   | 232  | 4     | 91   | 38   | 63   | 143  |  |
| Mn (mg kg <sup>-1</sup> )   | 21 | 23.3 | 10.4     | 7.1    | 43.6 | 5                  | 29.8 | 9.8  | 17.5 | 43.6 | 4     | 15.3 | 3.7  | 9.9  | 18.1 |  |
| pH                          | 77 | 5.0  | 0.7      | 3.8    | 6.6  | 22                 | 4.9  | 0.8  | 3.8  | 6.4  | 12    | 5.2  | 0.3  | 4.9  | 5.7  |  |
| As (mg kg <sup>-1</sup> )   | 76 | 3.2  | 1.4      | 1.0    | 12.2 | 22                 | 3.0  | 1.0  | 1.0  | 4.4  | 12    | 3.4  | 1.2  | 2.1  | 5.5  |  |
| Cd (mg kg <sup>-1</sup> )   | 76 | 0.39 | 0.13     | 0.18   | 0.74 | 22                 | 0.37 | 0.12 | 0.18 | 0.60 | 12    | 0.43 | 0.13 | 0.27 | 0.66 |  |
| Cr (mg kg <sup>-1</sup> )   | 77 | 9.3  | 6.2      | 2.2    | 28.3 | 22                 | 9.5  | 7.3  | 2.7  | 26.4 | 12    | 8.2  | 1.7  | 6.0  | 11.3 |  |
| Pb (mg kg <sup>-1</sup> )   | 77 | 5.6  | 2.1      | 2.8    | 10.7 | 22                 | 5.6  | 2.1  | 2.8  | 8.5  | 12    | 5.3  | 1.2  | 3.9  | 7.3  |  |
| CEC (cmol kg-1)             | 21 | 14.2 | 9.3      | 3.4    | 44.6 | 5                  | 8.8  | 4.6  | 3.4  | 13.6 | 4     | 14.8 | 7.0  | 9.9  | 25.0 |  |
| $K^{+}/(Ca^{2+}+Mg^{2+})$   | 21 | 0.04 | 0.02     | 0.02   | 0.1  | 5                  | 0.04 | 0.03 | 0.02 | 0.07 | 4     | 0.04 | 0.03 | 0.02 | 0.09 |  |
| K+/(Ca2++Mg2++Na+)          | 21 | 0.04 | 0.02     | 0.02   | 0.08 | 5                  | 0.04 | 0.02 | 0.02 | 0.07 | 4     | 0.04 | 0.03 | 0.02 | 0.08 |  |

hat gelöscht: SOM
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