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Inexorable land degradation due to agriculture expansion in South American Pampa

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27 Abstract

28	From 1985 onwards, South America has undergone a major expansion of agriculture at the
29	expense of native vegetation (e.g. native Pampa grassland). As an emblematic crop, the surface
30	area cultivated with soybeans has increased by 1000% between 1990 and 2020 in Uruguay. The
31	environmental consequences of this massive land use conversion on soil degradation remain poorly
32	documented although the agriculture expansion is projected to continue to increase in the coming
33	years in South America. In this study, sediment cores were collected in reservoirs located
34	downstream of two contrasted agricultural catchments draining the Rio Negro River (Uruguay) for
35	reconstructing the sediment dynamics and the sources of erosion associated with this expansion.
36	Results demonstrated the occurrence of two periods of acceleration of sediment delivery since the
37	1980s. The first period of acceleration was recorded in the mid-1990s and was related to
38	afforestation programs. The second and larger acceleration phase was recorded after 2000 during
39	the soybean crop expansion. This period has been marked by a greater supply of sediment from the
40	native grassland source highlighting the impact of agriculture expansion at the expense of native
41	vegetation. Conservation measures should therefore be urgently taken to preserve biodiversity and
42	soil functions in this region.
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52 Introduction

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Agricultural production has significantly increased in countries of the Northern 54 Hemisphere (e.g. those in Europe and North-America) from the 1950s onwards to respond to the 55 global food demand¹. This increase mainly relied on the modernization of agricultural practices 56 57 (e.g. mechanization, fertilizers, genetic modifications of plants) and the implementation of major 58 landscape changes. In other regions of the world like in South-America, a second phase of agricultural intensification occurred after 1985 with the expansion of the cultivated land area and 59 extensive livestock breeding in zones that were previously not cultivated². This modern cultivation 60 mode has been associated with a greater use of chemicals inputs, the sowing of genetically modified 61 62 species (e.g. with genotypes resistant to glyphosate) and changes in agricultural practices with the 63 implementation of monoculture and no-till farming. Since the mid-1980s, surfaces occupied by pasture, cropland and commercial plantations increased by 23, 160, 288% respectively in South-64 America³. Among these widespread land use changes, the surface devoted to soybean cultivation 65 doubled (from 26.4 Mha to 55.1 Mha) between 2000 and 2019 on grassland that had been 66 previously converted from natural vegetation for cattle production ⁴. By 2018, 40% (713Mha) of 67 the South-American land mass was significantly impacted by human activities ³. Uruguay is one of 68 69 the emblematic countries associated with this agricultural expansion at the expense of natural 70 ecosystems. The generalization of soybean cultivation after 2000 was accompanied by a process of 71 "intensification" of land use, with annual double cropping in both winter and summer, a process 72 that is observed nowadays across more than 30% of the country area⁵. Although agricultural growth occurred mainly in the traditional agricultural areas, it has also affected areas without previous 73 74 agricultural activity that used to be covered with natural vegetation. This agricultural expansion is mainly explained by the arrival of new farmers and internal/external investors. 75

76 Land degradation refers to the processes such as soil erosion that drive the deterioration of all terrestrial ecosystems (IPBES glossary). A recent study (2008) demonstrated that 80% of 77 Uruguayan soils ⁶ are affected by soil degradation (, i.e. the diminishing capacity of the soil to 78 provide ecosystem goods and services as desired by its stakeholders, according to the IPBES 79 80 glossary). To combat erosion, a soil conservation policy was implemented from 2014 onwards in Uruguay. It required the presentation of Responsible Soil Use and Management Plans⁷, the 81 82 adoption of no-tillage while leaving the soil covered with vegetation and the implementation of 83 rotations with pastures in order to mitigate water erosion across the country. The adoption of this 84 policy succeeded in reducing erosion per unit area, although gross erosion continued to increase

as a result of agricultural expansion through the replacement of natural grassland with cropland in 85 other regions of Uruguay⁸. To the best of our knowledge, no regional study quantified changes in 86 soil erosion in this part of the world, although a few studies conducted at the scale of small 87 catchments documented the impact of land use changes and the adoption of no-tillage on soil 88 erosion ⁹. In Southern Brazil, in catchments managed with no-till practices, very high sediment 89 fluxes were recorded for the 2011-2015 period (37-259 t km⁻² yr⁻¹), which demonstrates that the 90 sustainability of the soil cultivation system is threatened ¹⁰. Beyond the *on-site* consequences of 91 soil erosion (e.g. loss of fertility and decrease in crop yields), multiple off-site effects threaten water 92 93 quality with the excessive transfer of potentially contaminated sediment to the hydrosystems ¹¹. In Uruguayan (<20 km²) and Brazilian catchments (respectively <20 and 804 km²), cropland was 94 identified to supply the main source of sediment (>70% of the contribution) 12,13 . This excessive 95 96 supply of sediment from upper catchment parts contributes to the degradation of water quality and 97 to the siltation of water bodies. Accordingly, a significant reduction in the storage capacity of reservoirs used to supply drinking and irrigation water, as well as for hydroelectric power 98 production (respectively 63.5% and 60% of Brazilian and Uruguayan energy production) is 99 100 expected. Beyond the degradation of land and water resources, this second phase of agricultural 101 expansion after 1985 induced a massive loss of biodiversity with the destruction of natural habitats (primary forests, natural grasslands, freshwater)¹⁴, and raised several concerns regarding human 102 health ¹⁵ and ecosystem services ¹⁶. 103

104 Although the environmental consequences of the "agricultural revolution" that took place 105 after World War II in Europe and North-America including land degradation were extensively investigated based on modeling, monitoring and paleoenvironmental reconstructions, much less 106 attention has been devoted to the recent and massive expansion of agriculture and its intensification 107 108 across South-America in general, and in the Campos ecosystem in particular (southern Brazil, 109 northeastern Argentina and Uruguay). To the best of our knowledge, the impact of land cultivation on hydro-sedimentary transfers was investigated in this Campos ecosystem through the use of river 110 gauging stations, plots survey, sediment source fingerprinting and modeling ^{17 18} and modeling. 111 These approaches allow to quantify at a high spatial resolution and over relatively short -time scale 112 113 (<5 years) the impacts of modern farming practices on water and sediment exports. However, they do not allow to monitor or reconstruct the inertia, magnitude and resilience of continental 114 115 ecosystems that occurred over longer time scales (>10 years), i.e. including the conversion of 116 Campos into cultivated land and commercial plantations.

117 Collecting and analyzing sediment accumulated in reservoirs provide a powerful tool to 118 achieve these reconstructions ¹⁹. Paleoenvironmental reconstructions were successfully applied at 119 different timescales (e.g. Holocene ²⁰) and in various agricultural contexts for reconstructing the 120 adverse consequences of agriculture on sediment delivery ²¹, transfer of contaminants ²² or 121 eutrophication ²³.

To the best of our knowledge, few paleoenvironmental studies focused on the processes that occurred during the last decades in South-America in general, and in the Campos ecosystems area in particular. One of the reasons for the lack of paleoenvironmental studies in this region highly impacted by anthropogenic pressures is the small number of lakes and reservoirs draining these agricultural areas.

127 In the current research, we have retrospectively investigated the consequences of land use conversion on land degradation by analysing sediment cores (PA-02 and RDB-01 cores) collected 128 in two large reservoirs (the Palmar (PA) and Rincón del Bonete (RDB reservoirs) draining the Rio 129 Negro river in Uruguay (Fig. 1, Fig.2). A limnogeological approach based on dating (using 130 artificial and natural radionuclides including caesium-137 (137 Cs) and excess of lead-210 (210 Pb_{ex})) 131 and characterizing the sedimentary sequences with multiple analyses (e.g. organic matter properties 132 (Total Organic Carbon (TOC), Total Nitrogen (TN) and stable isotope measurements (δ^{13} C and 133 δ^{15} N), X-ray fluorescence core scanner (XRF), tomography scanner (CT-Scan)) was conducted in 134 135 order to (1) reconstruct the changes in sediment delivery based on the mass accumulation rates 136 (MAR) and terrigenous XRF proxy (Fe and Ti) associated with the last and massive expansion of 137 agriculture in Uruguay (1982-2019), (2) identify the main sources of land degradation accumulated 138 in the reservoirs and quantify the respective contributions of cropland (CR) and natural grassland (NG) sources to sediment and, finally, (3) outline the main drivers of land and water degradation. 139 Accordingly, the general goal of this study is to better understand the history and drivers of the 140 141 sediment cascade in this region in order to reduce the deleterious consequences of modern agriculture at a time when this intensification is projected to continue in the coming decades. 142

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- 145 Results
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- 147 Core description and chronology
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The RDB-01 and PA-02 sequences were composed of homogenous fine brown-coloured sediment. No specific facies or layers were visually observed in these cores. CT-Scan images allowed to detect some coarser/denser particle levels at the bottom of the PA-02 core only, whereas such levels were not observed on the RDB-01 core.

The ²¹⁰Pb_{ex} activity along the RDB-01 core decreased according to a linear trend ($r^2=0.89$) (Fig. 153 3a). The application of the CF:CS (Constant Flux: Constant Sedimentation) model allowed to date 154 155 the base of this 31 cm long sequence to $1992 (\pm 3 \text{ years})$ with an average sedimentation rate (SAR) of 11 mm yr⁻¹. The first appearance of ¹³⁷Cs was detected between 27 and 31 cm depth in the RDB-156 01 core (3 Bq kg⁻¹, SD 0.4 Bq kg⁻¹), and a very low ¹³⁷Cs peak was observed between 12 and 15 157 cm depth (4.2 Bq kg⁻¹, SD 0.7 Bq kg⁻¹). The characteristic peak typically associated with the 158 maximum ¹³⁷Cs bomb fallout (1964-1965 in South-America ²⁴) was logically not observed in this 159 160 recent sediment sequence (Fig. 3a) in agreement with previous plutonium isotope measurements performed on this core, which demonstrated that sediment deposited well after this period ²⁵. 161

The ²¹⁰Pb_{ex} activity from the PA-02 core decreased according to a linear trend as well ($r^2 = 0.94$. 162 The age-depth model based on the CF:CS model allowed to date the base of the sequence to 1978 163 (\pm 4 years) with an average SAR of 5.5 mm yr⁻¹ (Fig. 3b). In the PA-02 core, ¹³⁷Cs was detected 164 along the entire sedimentary sequence with a maximal value recorded between 9 and 12 cm depth 165 (5.1 Bq kg⁻¹, SD 0.8 Bq kg⁻¹, corresponding to the period between 1997 and 2002 according to the 166 ²¹⁰Pb_{ex} dating). As for the RDB-01 core, no peak that may have been attributed to the thermonuclear 167 168 bomb testing was observed in this post-1965 sequence. The PA-02 age-model was validated by 169 both field and CT-scan observations. During sampling, a paleo-soil was observed at the base of the 170 core and the occurrence of denser particles was detected with CT-Scan images. These observations were in agreement with the CF:CS model outputs dating the base of the core to 1978 (\pm 4 years), 171 corresponding approximatively to the age of the dam commissioning (1982). 172

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174 Organic matter properties

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In both reservoirs, the organic fraction represented only a low proportion of the total material inputs (including both autochthonous and allochthonous inputs). In the RDB-01 sequence, the average TOC content reached $3.0 \pm 0.2\%$ of the dry sediment with a maximal value recorded in 2001 (3.7%) (Fig. S1). In the PA-02 sediment core, the average TOC content amounted to 3.5 $\pm 0.2\%$ with minimal values recorded between 2000-2011 and a maximal value in 1985 (respectively, 3.2 and 3.75%) (Fig. S2). 182 The RDB sequence demonstrated the occurrence of two periods of changes in organic matter 183 sources between 1992 and 2019 (Fig. S1). These periods were associated with simultaneous changes in TN, C:N ratio and δ^{13} C. They took place between 1996-2002 and between 2013-2019. 184 During these periods, TN increased by an average value of 0.31% ($\pm 0.03\%$), whereas C:N ratio and 185 δ^{13} C decreased (respective average values of 10 ±0.7% and -20.1 ±0.55‰). Nitrogen isotope 186 187 variations highlighted the occurrence of distinct periods before and after 2001. Before this period, δ^{15} N amounted on average to 6.2 ±0.7 ‰), whereas after this period it reached around 8 ±0.5‰ 188 189 (Fig. S1).

Both TN and δ^{13} C proxies showed significant changes along the PA-02 sequences (Fig. S2). TN increased in 1984, in 1996 (0.29% rise for both change periods) and even more significantly between 2015 and 2019 (0.31% rise). The δ^{13} C showed an increase during the 1993-2000 period and in 2019. The largest change was observed in 2012 (from -20.1‰ in 2009 to -21.4‰ in 2019). In contrast, no sudden change nor peak in C:N ratio was observed along the sequence. Nevertheless, a constant decrease of the C:N ratio was detected from the base (13.1) to the top (10.7) of the sequence (Fig. S2).

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198 Terrigenous proxies and MAR evolution

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200 Both proxies of the terrigenous fraction (Ti and Fe) were highly correlated in both the RDB-201 01 and PA-02 cores (with respective r^2 values of 0.96 and 0.87). A significant positive trend of terrigenous inputs was recorded in these sequences (MK-test *p*-value >0.01). In the RDB core, three 202 periods of dominant terrigenous delivery were recorded between 1992 and 1998, between 2004 and 203 2008 and between 2012-2019 (Fig. S1). In contrast, the 1998-2004 period recorded a decrease of 204 205 this terrigenous contribution. In the PA-02 sequence, similar periods of increase were observed between 1992 and 1996, between 2004 and 2011 as well as between 2013 and 2019 (Fig. S2), 206 207 whereas no period of decrease was observed in the PA-02 core.

Like the terrigenous Ti and Fe proxies, the MAR calculated for both reservoirs showed a significant positive trend (MK-test *p*-value >0.01) in sediment delivery (Fig. 4). In the RDB sediment core, three main periods of greater accumulation of sediment were observed, in 1992-1998, 2004-2008 and 2012-2019 (with corresponding average MAR values of 0.16, 0.17 and 0.18 g cm⁻² year⁻¹) as well as one period of decrease between 1999 and 2002 (with a minimum in 2001, and a MAR of 0.11 g cm⁻² year⁻¹) (Fig.4). In the Palmar reservoir, three main periods of high mass accumulation of sediment were also recorded between 1991 and 1995, 2006 and 2011 and between 215 2013 and 2019 (with average MAR values for these periods of 0.15, 0.16 and 0.18 g cm⁻² year⁻¹).

A major period of decrease of sediment inputs was also observed in 2002 (with a MAR of 0.14 g

217 $cm^{-2} year^{-1}$ (Fig. 4).

- 218
- 219 Sediment sources
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221 Among the potential organic matter properties used for tracing the sources of sediment, TN 222 allowed to reclassify 76% of the source samples whether they originated from natural grassland or cropland sources. The sediment fingerprinting results indicate that in both reservoirs, the dominant 223 224 sources of sediment were associated with the cropland sources (in average $75.0 \pm 5.0\%$ and 74.5225 $\pm 1.5\%$) for the RDB-01 and PA-02 sequences) (Fig. 4). In the PA sequences, the lowest 226 contribution of natural grassland source was recorded around 1993 (22%) while the highest was found around 1984 and 1997 (28%). No significant trend of change in source contributions was 227 observed in this reservoir. In the RDB-01 core, the lowest contribution of natural grassland was 228 229 observed in 2012 (18%) whereas the maximal contribution of this source was detected in 2018 230 (39%). In addition, four peaks of the natural grassland source contribution were detected in 1994, 2001, 2013, 2015 (supplying respectively 29, 33, 31 and 32% of sediment). The natural grassland 231 232 contribution has increased on average by 58% between 2002 and 2018 (Fig. 4) in the RDB 233 sequence. Nevertheless, this trend was found not to be significant (MK-test *p*-value <0.01).

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236 Discussion

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During its recent history (<70 years), the Palmar catchment was impacted as the southern and 238 239 western parts of Uruguay by a first expansion phase of agriculture during the 1950s, whereas both Palmar and Rincón del Bonete catchments experienced another period of intensification and 240 expansion of agriculture after the 1980s²⁶. In response to this land conversion, the reservoirs 241 recorded a sediment delivery increase of 33 and 7% on average since 1982 and 1992 and until 2019 242 243 in the PA-02 and the RDB-01 cores (Fig. 4). This period was associated with a dominant contribution of the cropland source (>70%) as previously reported in other studies conducted in 244 Uruguay and Southern Brazil^{12,13} and characterised by a strong correlation between the sediment 245 246 dynamics (MAR) and the cultivated surface area available from the FAO database (e.g. r^2 of 0.84 247 for the 1982-2019 period in the PA reservoir).

In details, a first acceleration was identified in the PA reservoir between 1990 and 1995 (peaking 248 in 1993) and was characterized by an increase of the MAR, the Fe proxy and by a decrease in $\delta^{13}C$ 249 values. In the RDB sequence, the 1990s displayed the same trend (increase of the MAR and of the 250 Fe proxy) with an increase of sediment accumulation during the 1992-1998 period. In Uruguay, 251 252 planting of non-indigenous trees and installation of farms for agroforestry has been common for a 253 long time (e.g. for providing windbreaks and shelters for livestock, poles, firewood production)²⁷. 254 However, the 1990s paved the way to industrial planting with the entry into force of the forestry law encouraging investors to convert land into profitable forests, while major constraints were 255 256 introduced for farming (cultivation) and livestock grazing areas, which impacted in turn the 257 productive capacity. The higher internal and external demands in wood products also facilitated the expansion of afforestation zones in "better quality" soil areas ²⁸. In the RDB catchment, 258 approximately 6,000 km² of non-indigenous trees were planted between 1990 and 2001 with an 259 increase of the planted area from 500 km² in 1988 to 6,610 km² in 2001 ²⁹. A similar situation was 260 observed in the PA basin, although to a lesser extent. As a response to the eucalyptus and pine 261 planting, the mass accumulation rates of sediment increased by 27% and 21% in the RDB and PA 262 reservoirs. To the best of our knowledge, only few records of erosion rates are available at the plot 263 or at the catchment scale in areas affected by eucalyptus afforestation in Uruguay ⁹. Nevertheless, 264 265 in the neighboring State of Rio Grande do Sul in southern Brazil, soil erosion rates were measured in such plots and values ranging between 3 and 6379 t km⁻² year⁻¹ were found depending on the soil 266 properties and the number of years since the plantation ³⁰. As reported by the authors, after the 267 plantation, erosion rates decreased rapidly as the canopy covered the soil surface by reducing the 268 soil exposure to raindrops and runoff. Erosion was measured to decrease from 620 to 110 t km⁻² 269 year⁻¹ from 2 to 7 years after the plantation ³⁰ under the effect of the tree growth and the progressive 270 271 cover of the soils with vegetation. The sediment delivery trajectories recorded for the RDB and PA cores are in agreement with these findings, which may explain the decrease of sediment inputs into 272 273 the reservoirs several years after the implementation of afforestation programs, after an initial phase 274 of increase (Fig. 4). This afforestation period was associated with a greater contribution of the 275 natural grassland source to the RDB core (contribution of 29% in 1994) whereas only a slight increase of cropland source was observed in the PA sequence (contribution of 78% in 1993). These 276 277 different sediment source signatures suggest the occurrence of two strategies of land conversion in 278 relation with the catchment history. In the PA catchment, the historically cultivated land (e.g. 279 mostly mixed-farming with the alternation of crops and livestock) was converted into artificial 280 forests (e.g. mainly on the soils with the largest constraints to produce annual crops). In contrast, in the RDB catchment, the natural grassland (e.g. used for livestock farming) was converted for
afforestation purposes. Despite some studies suggest a decrease of soil erosion after the conversion
of degraded grassland into artificial forests ³¹, the current research recorded a significant
acceleration of erosion in both historically and recently farmed catchments.

285 The highest acceleration was observed between 2002 and 2019 with an increase of sediment 286 delivery by 20 and 67% and that of terrigenous inputs by 19 and 77%, respectively, for the PA and 287 RDB reservoirs. This acceleration occurred during the last phase of agricultural expansion observed 288 just after the economic crisis in Uruguay (2000-2002), which is reflected in the sediment cores by a sharp drop of the accumulation rates in response to the reduction of the cultivated areas (Fig. 4) 289 290 After 2002, land conversion in Uruguay was strongly impacted by internal (favorable financial 291 climate, decrease of exportation taxes in Uruguay) and external policies (2002 financial collapse 292 and increase of taxes in Argentina), which have resulted in a sudden expansion of soybean cultivation and afforestation of exotic trees ³⁴. In the RDB catchment, this crop was implemented 293 at the expense of native vegetation and it occupied more than 700 km² in 2019²⁹. This expansion 294 had led to a 58% increase of the natural grassland source contribution to the Bonete sediment 295 296 sequence. Although grassland is less sensitive to erosion than cultivated soils, the increase of NG 297 contribution to sediment may be explained by the mobilization of particles from ploughed grassland 298 that had been recently converted into cropland and that maintained temporarily their NG signature despite their cultivation during the first years that followed this land use conversion ³⁶. The greater 299 contribution of the NG and the acceleration of MAR was associated with major changes in organic 300 matter properties in the RDB sequence, which is demonstrated by low values of C:N, δ^{13} C and a 301 strong increase of TN (Fig. S1). These changes in organic matter properties support the sediment 302 tracing results showing the impact of the extension of the soybean cultivation on the sediment 303 304 supply observed in the RDB sequence. During the 2002-2019 period, the acceleration of sediment 305 dynamics was strongly correlated to the soybean surfaces cultivated at the country scale (r^2 of 0.67 - unfortunately, no data is available at the catchment scale) (FAO Database) supporting the finding 306 307 that the soybean expansion was the main driver of land degradation. Other modes of cultivation can also explain this acceleration of sediment inputs such as the increase in the artificial forest 308 309 surfaces. A more detailed sediment fingerprinting approach that may rely on novel environmental DNA metabarcoding ³⁶ and the analysis of historical data on land use changes may contribute to 310 311 further improve the unambiguous identification of the main drivers of erosion in this catchment. 312 The PA catchment is located in one of the most productive soybean areas in Uruguay (concentrating 313 more than 50% of the national soybean production). The soybean expansion mainly occurred in 314 former cultivated plots without a massive expansion of agriculture at the expense of native vegetation. Consequently, this likely explains why no significant change in the natural grassland 315 source contribution to sediment was observed in the PA sequence (Fig. 4), contrary to the results 316 317 found in the RDB sequence, with the increase of sediment supply from NG material recently 318 converted into cropland. Nevertheless, sediment accumulated in the PA reservoir demonstrated as 319 in the RDB core a major increase of sediment accumulation rates associated with the combination of a decrease of the C:N ratio, low values of δ^{13} C and high values of TN corresponding to a greater 320 contribution of areas cultivated with soybean to sediment (Fig. S2). This expansion of soybean in 321 322 former cultivated plots induce a less important increase of sediment dynamics in PA sequence that 323 one observed in the RDB core associated to the grassland conversion between 2002 and 2019 (r² of 0.91 between the soybean surfaces at the scale of Uruguay and the MAR of the PA core) (FAO 324 325 database).

As the soybean production is projected to further increase by 50% by 2050 in South-America 326 ³⁷, the results obtained in the current study lead us to anticipate a further acceleration of land and 327 water degradation in this part of the world. This question is crucial for a country like Uruguay, 328 329 which is socio-economically very much dependent on the quality of soil resources. Beyond the 330 problems directly associated with soil degradation, this agricultural expansion induced a large 331 number of multiple deleterious consequences on terrestrial and aquatic ecosystems, which remain 332 poorly understood. Consumption of NPK fertilizers should be promote in the coming years to compensate the decrease of soil fertility. The import of fertilizers has already tripled between 2000 333 and 2015³⁸. Some of these nutrients can locally increase the soil acidity in association with a strong 334 carbon print 39 and may reach the hydrographic system where they may further increase 335 eutrophication problems and the frequency and intensity of cyanobacterial blooms. This type of 336 phenomenon is already observed in Uruguay and its occurrence is expected to increase in the 337 coming years, thereby threatening the sustainability and the quality of water resources ^{40,41}. In 338 addition, a particular attention should be focused on the fate of pesticides massively applied in this 339 area ⁴² and more specifically on the interaction between the greater use of herbicides (import of 340 herbicides increased by 30% for the 2000-2015 period ³⁸) and soil erosion, although this 341 relationship needs to be further investigated ²¹. The environmental impacts of the massively applied 342 *Mirex* insecticides in eucalyptus plantations should also be looked into. Understanding the adverse 343 344 consequences of the agriculture expansion in South-America is of paramount importance in order 345 to design and implement mitigation strategies to protect water and soil resources. Furthermore, 346 additional research will be needed to identify in more details the sources of sediment and to

characterise the persistence and the potential remobilization of pesticides which could lead to afurther degradation of soil and water quality.

349 350 351 352 **Materials and Methods** 353 354 Sampling 355 Sediment cores were collected in the Rincon del Bonete (RDB-01, 31 cm long, IGSN number: 356 IEFOU0007) and Palmar (PA-02, 22.5 cm long, IGSN number: IEFOU0006) reservoirs using a 60 357 358 mm Uwitec gravity corer equipped with a 0.6 meter PVC liner tube available at the Universidad de la República (Montevideo, Uruguay). Coring sites were selected in the vicinity of the dams in order 359 to avoid sampling of coarser deposits classically found in deltaic areas associated with coarse-360 361 grained material deposited during extreme events. Other goals of this strategy were to ensure the 362 collection of sediment at places where the accumulation of fine material is maximal, and to provide good representativeness of all the river inputs transported from the drainage areas into the reservoirs 363 364 (Fig. 1).

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366 Sediment core dating

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368 Core chronology was established based on the measurements of short-lived fallout 369 radionuclides (caesium-137 (137 Cs) and excess of lead-210 (210 Pb_{ex})) conducted on 17 samples 370 (respectively 10 samples for the RDB-01 core and 7 samples for the PA-02 core) of dry sediment 371 (\approx 10g) collected along the sedimentary sequences.

Gamma spectrometry measurements were obtained using coaxial N- and P- type HPGe detectors (Canberra/Ortec) available at the Laboratoire des Sciences du Climat et de l'Environnement (Gif-sur-Yvette, France). Radionuclide data was decay-corrected to June 2020. Sediment ages were determined using the CF:CS model (Constant Flux: Constant Sedimentation), which assumes a constant rate of ²¹⁰Pb_{ex} from atmospheric fallout with a constant rate of sedimentation ⁴³. These models were automatically computed using the R package SERAC ⁴⁴. Age model validation was conducted through the identification of the ¹³⁷Cs time marker originating from the thermonuclear weapon testing (maximal emissions in 1962) assumed to peak in 1964-65
in sediment archives of South America ²⁴ and through the identification of the underlying paleosoil
in the PA sediment core attributed to correspond to the dam commissioning in 1982 for PA.
Radionuclide measurements used for the establishment of the core chronology are provided in an
open access Zenodo dataset (https://zenodo.org/record/6630369#.YqiGkOzP2Uk).

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386 Non-destructive laboratory analyses

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The Avaatech X-ray fluorescence core scanner (XRF) available at the Laboratoire des Sciences du Climat et de l'Environnement was used to obtain high resolution (0.5 cm) and semiquantitative (cps) values of geochemical element contents along the core. This data was used for characterizing potential changes in sediment sources throughout time. Titanium (Ti) and iron (Fe) proxies were specifically used for identifying changes in detritical material contributions ⁴⁵.

393 Non-calibrated sediment density was recorded every 0.6 mm along the sediment sequences 394 using Computer Tomography scanner (CT-Scan) images obtained using the equipment (GE Discovery CT750 HD) available at the DOSEO platform (French Atomic Energy and Alternative 395 396 Energy Commission, CEA Paris-Saclay, France). Relative density values (derived from CT-397 number) were extracted from the reconstructed scanner images using the free software ImageJ. The 398 relative values of dry bulk density (DBD) were calibrated by measuring the absolute DBD (g cm⁻ ³) in 20 samples collected along the cores (obtaining a r² of 0.8 and 0.85 between DBD and CT-399 number for PA-02 and RDB-01 cores, respectively). 400

The Mass Accumulation Rate (MAR, expressed in g cm⁻² yr⁻¹) was reconstructed for each individual core by multiplying the high resolution DBD extracted from the calibrated tomography scanner profiles (expressed in g cm⁻³) with the Sediment Accumulation Rate (SAR, cm yr⁻¹) provided by the age model following the procedure described by Foucher et al. ¹⁹.The MAR corresponds to the amount of organic and terrigenous material deposited at each individual coring site and its evolution throughout the time.

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408 Destructive laboratory analyses

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Isotope-Ratio Mass Spectrometry (IRMS) analyses were conducted on dry sediment for
 determining organic matter properties, including elemental concentrations (Total Organic Carbon

412 – TOC, Total Nitrogen –TN, both expressed in %) and stable isotope measurements (δ^{13} C and δ^{15} N, 413 expressed in ‰). These measurements were performed with a continuous flow Elementar® 414 VarioPyro cube analyzer coupled to a Micromass® Isoprime IRMS available at the Alysés platform 415 of the Institut de Recherche pour le Développement (Bondy, France) on a selection of 41 samples 416 (respectively 29 samples for the RDB-01 core and 12 samples for the PA-02). Organic matter 417 properties measured in both cores are provided in an open access Zenodo dataset 418 (https://zenodo.org/record/6630479#.YqiFLuzP2Uk).

419

420 Mixing model

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422 Temporal reconstruction of the sediment source contributions accumulated in the reservoirs and 423 their evolution with time was achieved through the comparison of the organic matter properties (TOC, TN, δ^{13} C and δ^{15} N) measured in the sediment cores with those analyzed in potential soil 424 425 sources (soil samples from cropland and natural grassland, under the form of composite samples -10 subsamples collected in a radius of 100 meters – from the upper 0-2 cm surface soil layer) from 426 previous studies conducted in the Campos biome in Southernmost Brazil⁴⁶. Soil composite samples 427 were taken in representative areas of each land use, i.e. cropland including rice, soybean and mixed 428 429 farming fields (n=36), and native grassland (n=31) which is generally used to extensively feed 430 livestock. Soil from the upper 0-2 cm layer was collected, as this layer is the most likely to be 431 eroded and transported to the river network. For each composite source sample, around 10 sub-432 samples were collected within a radius of approximately 50 meters, mixed in a bucket and 433 approximately 500 grams of material were stored. Care was taken to avoid sites that have accumulated sediment originating from other sources, to prevent the collection of transient 434 material. The source sampling sites were selected in order to cover all the soil types and the 435 variability in slope positions of a 5,943-km² basin. More details and a map showing the soil 436 sampling distribution can be found in Ramon (2021)⁴⁶. 437

438 TOC and TN in Brazilian grassland 46 varied between 46.2 ±16.1 g kg⁻¹ and 4.5 ±1.5 g kg⁻¹,

439 respectively. In cropland, TOC and TN concentrations amounted to 32.7 ± 9.1 g kg⁻¹ and 3.3 ± 0.8

440 g kg⁻¹. These results are consistent with those of Sämuel et al. ⁴⁷ who conducted similar

441 measurements at 28 monitoring sites across Uruguay. They found that in grassland, TOC reached

442 29 \pm 19 g kg⁻¹ (n = 115), while TN amounted to 2.2 \pm 1.3 g kg⁻¹ (n = 115). Under cropland, TOC

- 443 reached 26 ± 7 g kg⁻¹ (n = 15), and TN amounted to 2.2 ± 0.6 g kg⁻¹ (n = 15). These concentrations
- 444 were slightly lower than those obtained by Ramon $(2021)^{46}$, which is very likely explained by the

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fact that different soil depths were targeted by both studies (i.e. 0–10 cm layer for Sämuel et al. ⁴⁷,

- 446 vs. 0-2cm for Ramon (2021)⁴⁶) and the uppermost layer is known to be the most enriched in
- 447 organic matter. Under native forest, TOC was 42 ± 36 g kg⁻¹ (n = 15), and TN amounted to 3.0 ± 1.9
- 448 g kg⁻¹ (n = 15). Finally, in forest plantations, TOC was 25 ± 15 g kg⁻¹ (n = 93), and TN was 1.8 ± 1.0
- 449 g kg⁻¹ (n = 93). The source database used in this study can be found in Ramon (2021) ⁴⁶.
- 450 Tracer selection and source apportionments were performed following three steps: i) a range test; ii) a Mann Whitney U test (MW U test); and iii) a linear discriminant function analysis (LDA). 451 For passing the range test, mean parameters values for sediment must fall within the range between 452 the maximum and minimum values observed in the sources. The MW U test was then performed 453 454 to test the null hypothesis (p < 0.05) that the sources belong to the same population. A forward stepwise LDA (p < 0.1) was applied to the variables that provided significant discrimination 455 456 between the sources to reduce the number of variables to a minimum that maximizes source discrimination ⁴⁸. The statistical analyses were performed with R software ⁴⁹. 457
- A mass balance un-mixing model was applied to calculate the source contributions to individual
 sediment layers by minimizing the sum of squared residuals (SSR).
- 460

$$SSR = \sum_{i=1}^{n} \left(\left(C_i - \left(\sum_{s=1}^{m} P_s S_{si} \right) \right) / C_i \right)^2$$
(1)

- 461 where *n* is the number of variables/elements used for modelling, C_i is the value of the parameter 462 *i* in the target sediment (i.e. succession of sediment layers in both reservoirs), *m* is the number of 463 sources, P_s is the optimized relative contribution of source *s* obtained by the SSR minimizing 464 function, and S_{si} is the concentration of element *i* in the source *s*.
- To ensure that the contributions did not result in negative values and that the sum of the contributions from all sources was equal to 1, restrictions were applied in the optimization process. To evaluate the uncertainty, the un-mixing model was solved by a Monte Carlo simulation with 2500 iterations, following the methods described in Batista et al. ⁵⁰.
- 469

470 Data availability Statement

471 Dataset are available in ZENODO repository at <u>https://zenodo.org/record/6630369</u> and
472 https://zenodo.org/record/6630479

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474	Acknowledgments
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481 Author Contributions Statement: M.T, G.C, M.C, J.G, P.C have collected the sediment cores. 482 AC. S, M.A have carried out CT-Scan measurements. A.F has performed gamma and organic 483 matter analyses as well as the XRF core scanner measurements. R.R and T.T have developed the 484 sediment tracing approach. A.F, PA. C and O.E drafted the manuscript. All co-authors have 485 participated to the redaction and review.

486 **Competing Interest Statement:** The authors declare no competing interest.

487

488 Figure caption

489

490 Fig 1. Location of the study sites within the Rio Negro Basin, Uruguay – Land use pattern in the
491 Rincón del Bonete and Palmar catchments base on the open access Land Use Atlas of Uruguay
492 (FAO)

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Fig 2. Evolution of the main land uses between 2000 and 2015 for the Palmar (A) and Rincón del
Bonete (B) catchments based on the Land Use Atlas of Uruguay (FAO) (left panel). Evolution of
the soybean and natural grassland surface areas (C) as estimated from the FAO database at the
Uruguayan scale (right panel).

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Fig 3. Age-depth models of the Rincón del Bonete (RDB-01) and Palmar (PA-02) sediment cores
based on the CF:CS model computed from 10 and 7 gamma spectrometry measurements
(respectively A and B). Vertical error bars correspond to the thickness of the analyzed layer. The
horizontal error bars represent the analytical error.

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Fig 4. Evolution of the Mass Accumulation Rate (MAR), the terrigenous fraction proxy (Fe) and
the contribution of the natural grassland source to sediment accumulated in the Rincon del Bonete

506	(RDB) and Palmar (PA) reservoirs. The grey zones show the periods of change in accumulation of		
507	sediment and terrigenous material corresponding to (1) the planting of exotic tree species in RDB		
508	catchment (mostly eucalyptus), (2) the economic crisis in Uruguay (2000-2002) associated with a		
509	drop of the cultivated areas, (3) the planting of a mix of exotic trees associated with the expansion		
510	of agri	culture in RDB catchment vs. the expansion of agriculture alone in PA catchment.	
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