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1 **Inexorable land degradation due to agriculture expansion in South**
2 **American Pampa**

3

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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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54 Agricultural production has significantly increased in countries of the Northern
55 Hemisphere (e.g. those in Europe and North-America) from the 1950s onwards to respond to the
56 global food demand ¹. This increase mainly relied on the modernization of agricultural practices
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
59 agricultural intensification occurred after 1985 with the expansion of the cultivated land area and
60 extensive livestock breeding in zones that were previously not cultivated ². This modern cultivation
61 mode has been associated with a greater use of chemicals inputs, the sowing of genetically modified
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
64 pasture, cropland and commercial plantations increased by 23, 160, 288% respectively in South-
65 America ³. Among these widespread land use changes, the surface devoted to soybean cultivation
66 doubled (from 26.4 Mha to 55.1 Mha) between 2000 and 2019 on grassland that had been
67 previously converted from natural vegetation for cattle production ⁴. By 2018, 40% (713Mha) of
68 the South-American land mass was significantly impacted by human activities ³. Uruguay is one of
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 area ⁵. Although agricultural growth
73 occurred mainly in the traditional agricultural areas, it has also affected areas without previous
74 agricultural activity that used to be covered with natural vegetation. This agricultural expansion is
75 mainly explained by the arrival of new farmers and internal/external investors.

76 Land degradation refers to the processes such as soil erosion that drive the deterioration of
77 all terrestrial ecosystems (IPBES glossary). A recent study (2008) demonstrated that 80% of
78 Uruguayan soils ⁶ are affected by soil degradation (, i.e. the diminishing capacity of the soil to
79 provide ecosystem goods and services as desired by its stakeholders, according to the IPBES
80 glossary). To combat erosion, a soil conservation policy was implemented from 2014 onwards in
81 Uruguay. It required the presentation of Responsible Soil Use and Management Plans ⁷, the
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

85 as a result of agricultural expansion through the replacement of natural grassland with cropland in
86 other regions of Uruguay⁸. To the best of our knowledge, no regional study quantified changes in
87 soil erosion in this part of the world, although a few studies conducted at the scale of small
88 catchments documented the impact of land use changes and the adoption of no-tillage on soil
89 erosion⁹. In Southern Brazil, in catchments managed with no-till practices, very high sediment
90 fluxes were recorded for the 2011-2015 period (37–259 t km⁻² yr⁻¹), which demonstrates that the
91 sustainability of the soil cultivation system is threatened¹⁰. Beyond the *on-site* consequences of
92 soil erosion (e.g. loss of fertility and decrease in crop yields), multiple *off-site* effects threaten water
93 quality with the excessive transfer of potentially contaminated sediment to the hydrosystems¹¹. In
94 Uruguayan (<20 km²) and Brazilian catchments (respectively <20 and 804 km²), cropland was
95 identified to supply the main source of sediment (>70% of the contribution)^{12,13}. This excessive
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
98 reservoirs used to supply drinking and irrigation water, as well as for hydroelectric power
99 production (respectively 63.5% and 60% of Brazilian and Uruguayan energy production) is
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
102 (primary forests, natural grasslands, freshwater)¹⁴, and raised several concerns regarding human
103 health¹⁵ and ecosystem services¹⁶.

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
106 investigated based on modeling, monitoring and paleoenvironmental reconstructions, much less
107 attention has been devoted to the recent and massive expansion of agriculture and its intensification
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
110 on hydro-sedimentary transfers was investigated in this Campos ecosystem through the use of river
111 gauging stations, plots survey, sediment source fingerprinting and modeling^{17 18} and modeling.
112 These approaches allow to quantify at a high spatial resolution and over relatively short –time scale
113 (<5 years) the impacts of modern farming practices on water and sediment exports. However, they
114 do not allow to monitor or reconstruct the inertia, magnitude and resilience of continental
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.

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

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

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144

145 **Results**

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147 Core description and chronology

148

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

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

162 The $^{210}\text{Pb}_{\text{ex}}$ activity from the PA-02 core decreased according to a linear trend as well ($r^2=0.94$).
163 The age-depth model based on the CF:CS model allowed to date the base of the sequence to 1978
164 (± 4 years) with an average SAR of 5.5 mm yr^{-1} (Fig. 3b). In the PA-02 core, ^{137}Cs was detected
165 along the entire sedimentary sequence with a maximal value recorded between 9 and 12 cm depth
166 (5.1 Bq kg^{-1} , SD 0.8 Bq kg^{-1} , corresponding to the period between 1997 and 2002 according to the
167 $^{210}\text{Pb}_{\text{ex}}$ dating). As for the RDB-01 core, no peak that may have been attributed to the thermonuclear
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
171 were in agreement with the CF:CS model outputs dating the base of the core to 1978 (± 4 years),
172 corresponding approximatively to the age of the dam commissioning (1982).

173

174 Organic matter properties

175

176 In both reservoirs, the organic fraction represented only a low proportion of the total material
177 inputs (including both autochthonous and allochthonous inputs). In the RDB-01 sequence, the
178 average TOC content reached $3.0 \pm 0.2\%$ of the dry sediment with a maximal value recorded in
179 2001 (3.7%) (Fig. S1). In the PA-02 sediment core, the average TOC content amounted to 3.5
180 $\pm 0.2\%$ with minimal values recorded between 2000-2011 and a maximal value in 1985
181 (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
184 changes in TN, C:N ratio and $\delta^{13}\text{C}$. They took place between 1996-2002 and between 2013-2019.
185 During these periods, TN increased by an average value of 0.31% ($\pm 0.03\%$), whereas C:N ratio and
186 $\delta^{13}\text{C}$ decreased (respective average values of $10 \pm 0.7\%$ and $-20.1 \pm 0.55\%$). Nitrogen isotope
187 variations highlighted the occurrence of distinct periods before and after 2001. Before this period,
188 $\delta^{15}\text{N}$ amounted on average to $6.2 \pm 0.7\%$, whereas after this period it reached around $8 \pm 0.5\%$
189 (Fig. S1).

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

197

198 Terrigenous proxies and MAR evolution

199

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
202 terrigenous inputs was recorded in these sequences (MK-test p -value > 0.01). In the RDB core, three
203 periods of dominant terrigenous delivery were recorded between 1992 and 1998, between 2004 and
204 2008 and between 2012-2019 (Fig. S1). In contrast, the 1998-2004 period recorded a decrease of
205 this terrigenous contribution. In the PA-02 sequence, similar periods of increase were observed
206 between 1992 and 1996, between 2004 and 2011 as well as between 2013 and 2019 (Fig. S2),
207 whereas no period of decrease was observed in the PA-02 core.

208 Like the terrigenous Ti and Fe proxies, the MAR calculated for both reservoirs showed a
209 significant positive trend (MK-test p -value > 0.01) in sediment delivery (Fig. 4). In the RDB
210 sediment core, three main periods of greater accumulation of sediment were observed, in 1992-
211 1998, 2004-2008 and 2012-2019 (with corresponding average MAR values of 0.16, 0.17 and 0.18
212 $\text{g cm}^{-2} \text{ year}^{-1}$) as well as one period of decrease between 1999 and 2002 (with a minimum in 2001,
213 and a MAR of $0.11 \text{ g cm}^{-2} \text{ year}^{-1}$) (Fig.4). In the Palmar reservoir, three main periods of high mass
214 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).
216 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

220

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
223 cropland sources. The sediment fingerprinting results indicate that in both reservoirs, the dominant
224 sources of sediment were associated with the cropland sources (in average 75.0 ±5.0% and 74.5
225 ±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
227 found around 1984 and 1997 (28%). No significant trend of change in source contributions was
228 observed in this reservoir. In the RDB-01 core, the lowest contribution of natural grassland was
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,
231 2001, 2013, 2015 (supplying respectively 29, 33, 31 and 32% of sediment). The natural grassland
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).

234

235

236 Discussion

237

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

248 In details, a first acceleration was identified in the PA reservoir between 1990 and 1995 (peaking
249 in 1993) and was characterized by an increase of the MAR, the Fe proxy and by a decrease in $\delta^{13}\text{C}$
250 values. In the RDB sequence, the 1990s displayed the same trend (increase of the MAR and of the
251 Fe proxy) with an increase of sediment accumulation during the 1992-1998 period. In Uruguay,
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
255 law encouraging investors to convert land into profitable forests, while major constraints were
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
258 expansion of afforestation zones in “better quality” soil areas²⁸. In the RDB catchment,
259 approximately 6,000 km² of non-indigenous trees were planted between 1990 and 2001 with an
260 increase of the planted area from 500 km² in 1988 to 6,610 km² in 2001²⁹. A similar situation was
261 observed in the PA basin, although to a lesser extent. As a response to the eucalyptus and pine
262 planting, the mass accumulation rates of sediment increased by 27% and 21% in the RDB and PA
263 reservoirs. To the best of our knowledge, only few records of erosion rates are available at the plot
264 or at the catchment scale in areas affected by eucalyptus afforestation in Uruguay⁹. Nevertheless,
265 in the neighboring State of Rio Grande do Sul in southern Brazil, soil erosion rates were measured
266 in such plots and values ranging between 3 and 6379 t km⁻² year⁻¹ were found depending on the soil
267 properties and the number of years since the plantation³⁰. As reported by the authors, after the
268 plantation, erosion rates decreased rapidly as the canopy covered the soil surface by reducing the
269 soil exposure to raindrops and runoff. Erosion was measured to decrease from 620 to 110 t km⁻²
270 year⁻¹ from 2 to 7 years after the plantation³⁰ under the effect of the tree growth and the progressive
271 cover of the soils with vegetation. The sediment delivery trajectories recorded for the RDB and PA
272 cores are in agreement with these findings, which may explain the decrease of sediment inputs into
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
276 increase of cropland source was observed in the PA sequence (contribution of 78% in 1993). These
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,

281 in the RDB catchment, the natural grassland (e.g. used for livestock farming) was converted for
282 afforestation purposes. Despite some studies suggest a decrease of soil erosion after the conversion
283 of degraded grassland into artificial forests ³¹, the current research recorded a significant
284 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
289 a sharp drop of the accumulation rates in response to the reduction of the cultivated areas (Fig. 4)
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
293 cultivation and afforestation of exotic trees ³⁴. In the RDB catchment, this crop was implemented
294 at the expense of native vegetation and it occupied more than 700 km² in 2019 ²⁹. This expansion
295 had led to a 58% increase of the natural grassland source contribution to the Bonete sediment
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
299 despite their cultivation during the first years that followed this land use conversion ³⁶. The greater
300 contribution of the NG and the acceleration of MAR was associated with major changes in organic
301 matter properties in the RDB sequence, which is demonstrated by low values of C:N, $\delta^{13}\text{C}$ and a
302 strong increase of TN (Fig. S1). These changes in organic matter properties support the sediment
303 tracing results showing the impact of the extension of the soybean cultivation on the sediment
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
306 – unfortunately, no data is available at the catchment scale) (FAO Database) supporting the finding
307 that the soybean expansion was the main driver of land degradation. Other modes of cultivation
308 can also explain this acceleration of sediment inputs such as the increase in the artificial forest
309 surfaces. A more detailed sediment fingerprinting approach that may rely on novel environmental
310 DNA metabarcoding ³⁶ and the analysis of historical data on land use changes may contribute to
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
315 vegetation. Consequently, this likely explains why no significant change in the natural grassland
316 source contribution to sediment was observed in the PA sequence (Fig. 4), contrary to the results
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
320 of a decrease of the C:N ratio, low values of $\delta^{13}\text{C}$ and high values of TN corresponding to a greater
321 contribution of areas cultivated with soybean to sediment (Fig. S2). This expansion of soybean in
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 (r^2
324 of 0.91 between the soybean surfaces at the scale of Uruguay and the MAR of the PA core) (FAO
325 database).

326 As the soybean production is projected to further increase by 50% by 2050 in South-America
327 ³⁷, the results obtained in the current study lead us to anticipate a further acceleration of land and
328 water degradation in this part of the world. This question is crucial for a country like Uruguay,
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
333 compensate the decrease of soil fertility. The import of fertilizers has already tripled between 2000
334 and 2015 ³⁸. Some of these nutrients can locally increase the soil acidity in association with a strong
335 carbon print ³⁹ and may reach the hydrographic system where they may further increase
336 eutrophication problems and the frequency and intensity of cyanobacterial blooms. This type of
337 phenomenon is already observed in Uruguay and its occurrence is expected to increase in the
338 coming years, thereby threatening the sustainability and the quality of water resources ^{40,41}. In
339 addition, a particular attention should be focused on the fate of pesticides massively applied in this
340 area ⁴² and more specifically on the interaction between the greater use of herbicides (import of
341 herbicides increased by 30% for the 2000-2015 period ³⁸) and soil erosion, although this
342 relationship needs to be further investigated ²¹. The environmental impacts of the massively applied
343 *Mirex* insecticides in eucalyptus plantations should also be looked into. Understanding the adverse
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

347 characterise the persistence and the potential remobilization of pesticides which could lead to a
348 further degradation of soil and water quality.

349

350

351

352 **Materials and Methods**

353

354 **Sampling**

355

356 Sediment cores were collected in the Rincon del Bonete (RDB-01, 31 cm long, IGSN number:
357 IEFOU0007) and Palmar (PA-02, 22.5 cm long, IGSN number: IEFOU0006) reservoirs using a 60
358 mm Uwitec gravity corer equipped with a 0.6 meter PVC liner tube available at the Universidad de
359 la República (Montevideo, Uruguay). Coring sites were selected in the vicinity of the dams in order
360 to avoid sampling of coarser deposits classically found in deltaic areas associated with coarse-
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
363 good representativeness of all the river inputs transported from the drainage areas into the reservoirs
364 (Fig. 1).

365

366 **Sediment core dating**

367

368 Core chronology was established based on the measurements of short-lived fallout
369 radionuclides (caesium-137 (^{137}Cs) and excess of lead-210 ($^{210}\text{Pb}_{\text{ex}}$)) 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 ($\approx 10\text{g}$) collected along the sedimentary sequences.

372 Gamma spectrometry measurements were obtained using coaxial N- and P- type HPGe
373 detectors (Canberra/Ortec) available at the Laboratoire des Sciences du Climat et de
374 l'Environnement (Gif-sur-Yvette, France). Radionuclide data was decay-corrected to June 2020.
375 Sediment ages were determined using the CF:CS model (Constant Flux: Constant Sedimentation),
376 which assumes a constant rate of $^{210}\text{Pb}_{\text{ex}}$ from atmospheric fallout with a constant rate of
377 sedimentation⁴³. These models were automatically computed using the R package SERAC⁴⁴. Age
378 model validation was conducted through the identification of the ^{137}Cs time marker originating

379 from the thermonuclear weapon testing (maximal emissions in 1962) assumed to peak in 1964-65
380 in sediment archives of South America ²⁴ and through the identification of the underlying paleosoil
381 in the PA sediment core attributed to correspond to the dam commissioning in 1982 for PA.
382 Radionuclide measurements used for the establishment of the core chronology are provided in an
383 open access Zenodo dataset (<https://zenodo.org/record/6630369#.YqiGkOzP2Uk>).

384

385

386 Non-destructive laboratory analyses

387

388 The Avaatech X-ray fluorescence core scanner (XRF) available at the Laboratoire des
389 Sciences du Climat et de l'Environnement was used to obtain high resolution (0.5 cm) and semi-
390 quantitative (cps) values of geochemical element contents along the core. This data was used for
391 characterizing potential changes in sediment sources throughout time. Titanium (Ti) and iron (Fe)
392 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
395 Discovery CT750 HD) available at the DOSEO platform (French Atomic Energy and Alternative
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})
399 ³) in 20 samples collected along the cores (obtaining a r^2 of 0.8 and 0.85 between DBD and CT-
400 number for PA-02 and RDB-01 cores, respectively).

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

407

408 Destructive laboratory analyses

409

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

412 – TOC, Total Nitrogen –TN, both expressed in %) and stable isotope measurements ($\delta^{13}\text{C}$ and $\delta^{15}\text{N}$,
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

421

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
424 (TOC, TN, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$) measured in the sediment cores with those analyzed in potential soil
425 sources (soil samples from cropland and natural grassland, under the form of composite samples -
426 10 subsamples collected in a radius of 100 meters – from the upper 0-2 cm surface soil layer) from
427 previous studies conducted in the Campos biome in Southernmost Brazil ⁴⁶. Soil composite samples
428 were taken in representative areas of each land use, i.e. cropland including rice, soybean and mixed
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
434 accumulated sediment originating from other sources, to prevent the collection of transient
435 material. The source sampling sites were selected in order to cover all the soil types and the
436 variability in slope positions of a 5,943-km² basin. More details and a map showing the soil
437 sampling distribution can be found in Ramon (2021) ⁴⁶.

438 TOC and TN in Brazilian grassland ⁴⁶ varied between $46.2 \pm 16.1 \text{ g kg}^{-1}$ and $4.5 \pm 1.5 \text{ g kg}^{-1}$,
439 respectively. In cropland, TOC and TN concentrations amounted to $32.7 \pm 9.1 \text{ g kg}^{-1}$ 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 $29 \pm 19 \text{ g kg}^{-1}$ (n = 115), while TN amounted to $2.2 \pm 1.3 \text{ g kg}^{-1}$ (n = 115). Under cropland, TOC
443 reached $26 \pm 7 \text{ g kg}^{-1}$ (n = 15), and TN amounted to $2.2 \pm 0.6 \text{ g kg}^{-1}$ (n = 15). These concentrations
444 were slightly lower than those obtained by Ramon (2021) ⁴⁶, which is very likely explained by the

445 fact that different soil depths were targeted by both studies (i.e. 0–10 cm layer for S amuel 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 $42 \pm 36 \text{ g kg}^{-1}$ ($n = 15$), and TN amounted to 3.0 ± 1.9
448 g kg^{-1} ($n = 15$). Finally, in forest plantations, TOC was $25 \pm 15 \text{ g kg}^{-1}$ ($n = 93$), 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;
451 ii) a Mann Whitney *U* test (MW *U* test); and iii) a linear discriminant function analysis (LDA).
452 For passing the range test, mean parameters values for sediment must fall within the range between
453 the maximum and minimum values observed in the sources. The MW *U* test was then performed
454 to test the null hypothesis ($p < 0.05$) that the sources belong to the same population. A forward
455 stepwise LDA ($p < 0.1$) was applied to the variables that provided significant discrimination
456 between the sources to reduce the number of variables to a minimum that maximizes source
457 discrimination ⁴⁸. The statistical analyses were performed with R software ⁴⁹.

458 A mass balance un-mixing model was applied to calculate the source contributions to individual
459 sediment layers by minimizing the sum of squared residuals (SSR).

460

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

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 .

465 To ensure that the contributions did not result in negative values and that the sum of the
466 contributions from all sources was equal to 1, restrictions were applied in the optimization process.
467 To evaluate the uncertainty, the un-mixing model was solved by a Monte Carlo simulation with
468 2500 iterations, following the methods described in Batista et al. ⁵⁰.

469

470 **Data availability Statement**

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

473

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475

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480

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)

493

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

498

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

503

504 **Fig 4.** Evolution of the Mass Accumulation Rate (MAR), the terrigenous fraction proxy (Fe) and
505 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 agriculture in RDB catchment vs. the expansion of agriculture alone in PA catchment.

511

512

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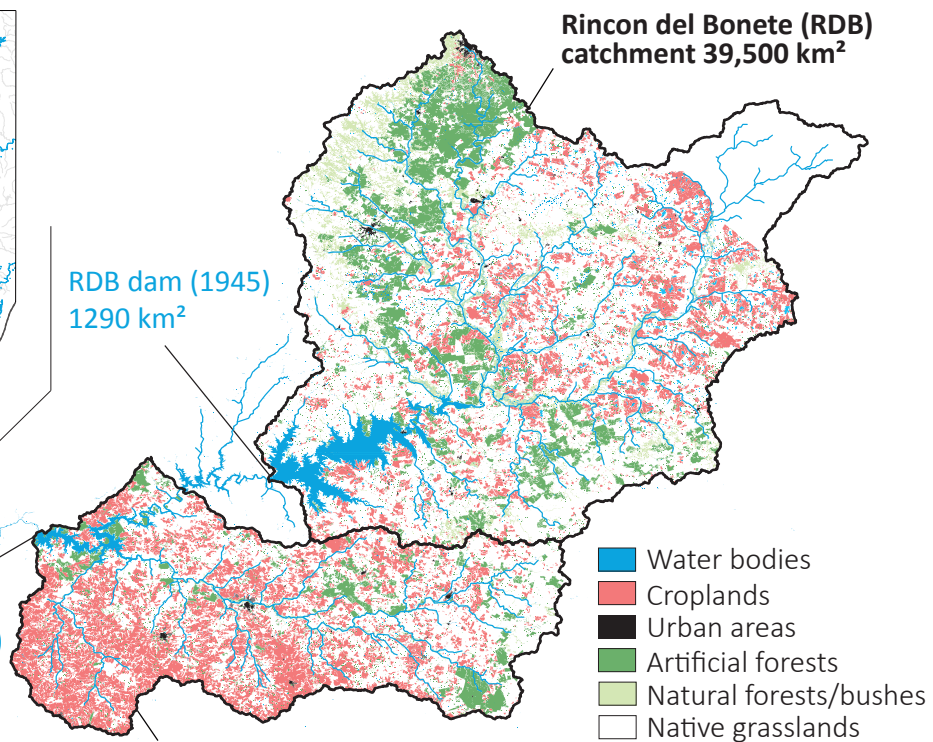
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0 120 240 Km



0 25 50 100 Kilometers

