Pantanal Basin river muds from source to sink: compositional changes in a tropical back-bulge depozone

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- 28 Short title: Modern fluvial muds in the Pantanal Basin
- 2930 Abstract

31 The Pantanal Basin is a low-gradient back-bulge analog for distal depozones associated with retroarc foreland basin systems in the geological record. Extensive lowland 32 33 environments including fluvial megafans, floodplains, wetlands, and lakes make up the Pantanal Basin today, with detrital sediment sources located along basin-margin plateaus 34 and remnants of ancient orogenic belts. Here, we examine the chemical composition and 35 mineralogy of modern fine-fraction fluvial sediments using X-ray methods to assess the 36 37 influence of chemical weathering on sediment composition in this tropical basin. The abundance of clay minerals follows the rank order pattern of kaolinite > vermiculite > illite 38 39 > smectite. Kaolinite is more abundant in river muds from the north-central than the 40 southern Pantanal, suggesting strong extant chemical weathering plus the potential for clays inherited from siliciclastic parent lithologies that formed under Mesozoic greenhouse 41 42 conditions. Illite occurs in sediments draining the North Paraguay Belt and limited parts of the South Paraguay Belt, and it reflects the influence of mechanical weathering of the 43 metamorphic facies. In the southeastern Pantanal, vermiculite is a dominant constituent 44 of the Miranda River watershed, which drains dacitic parent rocks and rhodic ferralsols. 45 46 The geochemistry of the sediments reveals the interplay of guartz addition and clays inherited from the parent rocks. The most guartzose sediments are encountered at the 47

48 confluence of the Paraguay River and the Taquari River megafan, where the cumulative

49 effect of the 2 – 3-month flood pulse maximizes chemical weathering. Clay plus silt in

50 back-bulge basins are controlled by climate > soils > parent rocks.

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52 Lay summary

53 The compositional controls on clays and silts in tropical rivers of the Pantanal Basin 54 distant from the Andes remain unclear. We collected 74 modern riverbank samples and 55 used X-ray techniques to determine clay mineralogy and chemical elemental composition. 56 The most common clay minerals in rank order of abundance were kaolinite > vermiculite 57 > illite > smectite. Kaolinite was dominant in the north-central Pantanal, whereas vermiculite was dominant in the southeastern Pantanal. The most guartzose clays and 58 59 silts were found in the middle Paraguay River Pantanal clays are controlled first by climate, and secondarily by soils. 60

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62 **Resumo**

63 A Bacia do Pantanal é um sistema sedimentar de baixo relevo na região de back-bulge análoga no registro geológico a outros ambientes deposicionais associados com 64 sistemas de retroarco. A hidrografia da Bacia do Pantanal inclui mega legues fluviais, 65 planície de inundação, áreas alagadiças e lagos, com fontes de detritos provenientes das 66 margens do planalto e resquícios de cinturões orogênicos pré-Cambrianos. Neste 67 estudo, analisou-se a composição química e a mineralogia da fração fina de sedimentos 68 69 fluviais modernos com métodos de raios-X para avaliar a influência do intemperismo 70 químico na composição dos sedimentos nesta bacia tropical. A abundância de minerais de argila segue o padrão de ordem de caulinita > vermiculita > ilita > esmectita. A caulinita 71 72 é mais abundante nos sedimentos fluviais do centro-norte do que no sul do Pantanal, o que sugere forte intemperismo químico recente além do potencial de argilas herdadas 73 de fontes sedimentares siliclásticas formadas em condições de "greenhouse" durante a 74 Era Mesozoica. A ilita ocorre em sedimentos que drenam a Faixa Norte do Paraguai e 75 76 partes limitadas na Faixa Sul do Paraguai, refletindo a influência do intemperismo mecânico das fácies metassedimentares. No sudeste do Pantanal, a vermiculita é um 77 78 constituinte dominante da bacia do Rio Miranda, que drena rochas-fonte dacíticas e 79 latossolos vermelhos. A geoquímica dos sedimentos revela a interação entre a adiação de quartzo e as argilas herdadas das rochas-fonte. Os sedimentos guartzosos são mais 80 81 frequentes na confluência do Rio Paraguai com o mega legue do Taguari onde o efeito 82 cumulativo do pulso de inundação (2 a 3 meses) maximiza o intemperismo químico. Este 83 estudo revela que a argila e o silte que preenchem bacias de back-bulge são controlados 84 pelo clima > solos > rochas-fonte. 85 86 Keywords: Clay mineralogy, Geochemistry, Chemical weathering, Tropical wetlands

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89 **1.** Introduction

90 Modern sands, silts and clays in large watersheds have been studied to reveal the 91 interactions among parent lithology, climate, and tectonics that influence sediment 92 composition (Johnsson, 1993; Jonell et al., 2017; He et al., 2020; Garzanti et al., 2021). 93 For instance, guartz enrichment and kaolinite abundance in lowland settings or intense 94 mechanical weathering on hillslopes are often linked to hot, humid conditions 95 characteristic of the tropics (Oliva et al., 1999; Viers et al., 2000; Aristizábal et al., 2005; 96 Garzanti et al., 2019). Tardy et al. (1973) show that montmorillinite can also be 97 concentrated in the lowlands downstream of granites in humid tropical environments. 98 These diverse floodplain clay mineral compositions suggest that considerable variability 99 surrounds clay mineral development in tropical floodplains. Although studies on clay 100 minerals have been conducted worldwide (e.g., Chamley, 1989), the relationships among 101 fluvial transport, climate, tectonics, and chemical weathering on clay mineralogy are site-102 specific and require localized sediment sampling. Modern fluvial sediment compositions 103 have not been systematically assessed in many South America rivers, with limited 104 research focused on the composition of the suspended sediment load in a few major 105 rivers (Potter, 1994; Guyot et al., 2007; McGlue et al., 2016; Repasch et al., 2020).

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107 Climate gradients are essential controls on the weathering of clay minerals and impact 108 regional vegetation and agricultural production. When hydrolysis is inefficient or incomplete, feldspar minerals persist along with Ca²⁺ and Na⁺, as was observed along 109 110 the Zambezi River system (Garzanti et al., 2022). No clay minerals may be observed in exceptionally arid conditions (e.g., Warr et al., 2022). Where the climate is warm and 111 humid, the 1:1 type clays are predominant with greater kaolinite compared to illite/mica in 112 113 the clay (<2 µm) fraction (Ito & Wagai, 2017). These large-scale trends have been documented and used for understanding the chemical weathering processes globally. 114 115 Changes to the clay mineral assemblage and fine fraction geochemistry along the Pearl 116 River and the Red River were dominantly controlled by climate gradients (He et al., 2020, 117 2022). However, none of these have examined modern fluvial clays within a primarily 118 floodplain or wetland environment. Garzanti et al. (2011) provided the most 119 comprehensive study of floodplain clays on the Indian sub-continent, within the Ganges-120 Brahmaputra foreland basin system.

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122 Parent lithology is a secondary determinant of modern clay minerals. Guyot et al. (2007) examined clay minerals across the Amazon Basin and found that the provenance of the 123 124 areas (shield, Andean cordillera, Piedmont basins) determined the clay mineral constituents. For example, the illite and chlorite reflected erosion of the metapelites and 125 126 metabasites exposed in the Red River basin (He et al., 2022). Kaolinite can originate from 127 virtually any parent rock, given warm and humid environments typical of the tropics (Dill, 128 2016). Tropical and sub-tropical climates commonly result in an under-representation of 129 mafic lithologies relative to their areal extent in modern fluvial sediments (Garzanti et al., 130 2014; Hatzenbühler et al., 2022).

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Mud composition can reflect differences between transport-limited erosion and
 weathering-limited erosion depending on the land surface gradient and hydrology
 (Stallard et al., 1991). Sediment erosion can be considered transport-limited, where

135 weathering creates more clays than can be transported, resulting in profoundly weathered 136 soil profiles (Stallard et al., 1991). Plateaus and floodplains are emblematic of transport-137 limited erosional regimes where the sediments and soils remain in place, subjected to 138 prolonged chemical weathering. Weathering-limited erosion occurs when the bedrock is 139 partially weathered before the sediment is removed, concentrating micas and feldspars 140 (Stallard et al., 1991). Cordilleras erode physically through rockfalls and the breakdown 141 of the rock formation, exacerbated by high slopes. Further understanding of these and 142 other source-to-sink processes requires a close examination of weathering intensity, as 143 recorded in clay mineral composition and elemental geochemistry (He et al., 2020; Cruz 144 et al., 2022). 145

146 Back-bulge basins are an ideal locale to examine how changes in environmental 147 conditions affect clay mineral production in tropical riverine settings. Back-bulges are usually low-gradient zones that store sediment and preserve important environmental 148 149 signals over geologic time (Horton & DeCelles, 1997; Assine et al., 2016; Brewer et al., 150 2020; Caracciolo, 2020). Silt plus clay preservation is excellent in low-gradient back-bulge depositional environments (e.g., in floodplains, lakes, and wetlands) (Quartero et al., 151 152 2015; McGlue et al., 2016; Tineo et al., 2022). Therefore, one of the motivations of this 153 research is to examine the processes that control mud mineralogy and chemistry in a 154 modern tropical setting where the environmental gradients (i.e., climate, soils, vegetation, 155 relief) are relatively well-understood in order to provide insights that may improve 156 interpretations of the geological record. We selected the Pantanal Basin as an exceptional locale to examine the primary sedimentary processes. The Pantanal Basin 157 (Brazil/Bolivia/Paraguay) forms the back-bulge depozone of the Cenozoic Andean 158 159 foreland basin (Chase et al., 2009; Cohen et al., 2015; Horton, 2022). The Pantanal is a tropical savanna extending from the Amazon drainage divide to the Brazilian border with 160 Paraguay (Figure 1A) (Beck et al., 2018). Most hinterland (i.e., basin margin) lithologies 161 162 surrounding the Pantanal are siliciclastic sedimentary rocks, with some pre-Cambrian igneous and metamorphic exposures; these lithologies were recently grouped into six 163 164 provenance regions (Figure 1B, C; Lo et al., 2023).

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In this study, modern mud in the rivers of the Pantanal were collected to evaluate their 166 mineralogy and chemical composition. We employed X-ray diffraction (XRD) for semi-167 168 quantitative clay mineralogy and wavelength-dispersive X-ray fluorescence (WD-XRF) to 169 deduce major elemental geochemistry (Moore & Reynolds, 1989). These data were 170 analyzed along with environmental characteristics of the basin's sub-watersheds (e.g., slope, lithology, precipitation, elevation) to elucidate the processes that control clay 171 composition. We tested the hypothesis that differences in mean annual precipitation 172 173 control the clay mineral assemblage in modern fluvial silt plus clay in the Pantanal. This 174 article is a companion study to a petrographic analysis of contemporary river sands in the 175 Pantanal (Lo et al., 2023), with the end goal of identifying major patterns in sediment generation and transport in this basin. Plata River samples were integrated with this study 176 177 to evaluate the influence of Pantanal Basin clay composition on downstream sediments. 178 Ultimately, our objective is to improve interpretations of ancient sedimentary rocks in 179 similar settings through a detailed set of modern observations and a database of 180 mineralogical and chemical measurements.



Figure 1: (A) Upper Paraguay River Basin (white outline) in South America. Red box denotes area shown in panel C. (B) Pantanal Basin provenance regions with major rivers (dark blue lines). Much of the areas covered at the surface by wetlands are designated as lowlands, in contrast with the fringing cratons and the plateau. White circles represent all sampling sites listed in Table S1. (C) GTOPO30 digital elevation model of South America (USGS, 1996), including a topographic cross-section A – A' from Google Earth©. Modified from Lo et al. (2023).

188189 2. Geological setting of the Pantanal Basin

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190 The Cenozoic Pantanal Basin formed from flexure of the crust as the Andes range arose from the subduction of the Pacific plate beneath the South American plate (Horton & 191 DeCelles, 1997; Assine et al., 2016). The Pantanal Basin has accumulated ~500 m of 192 193 sediment, with the depocenter located near the geographic center of the basin in the area 194 of the Taquari River (Ussami et al., 1999). The Pantanal Basin is occupied by large 195 distributary fluvial systems also known as fluvial megafans (Assine, 2005; Zani et al., 2012; Hartley et al., 2013; Weissmann et al., 2015). Unconsolidated sediments fill the 196 197 lowlands, which span ~150,000 km² within the Upper Paraguay River watershed covering 198 ~465,000 km² of Brazil, Bolivia and Paraguay. The Pantanal Basin can be divided into six 199 hinterland provenance regions: lowlands, Amazon craton, Rio Apa craton, plateau, and the South and North Paraguay Belt (Lo et al., 2023) (Figure 1B). Bedrock in the 200 201 northwestern Pantanal consists of Amazon craton, with granites, granodiorites, schists, 202 and dikes of quartz-diorite and quartz-gabbro making up the bedrock (Figure 2A) 203 (Rizzotto & Hartmann, 2012; Horbe et al., 2013; Braga et al., 2019). The Rio Apa craton 204 in the southwestern Pantanal consists of gneisses, granites, granodiorites, amphibolites, 205 schists, and guartzites (RadamBrasil, 1982; Alvarenga et al., 2011). The plateau region 206 hosts Phanerozoic sedimentary rocks derived from the Paraná Basin: the Aquidauana Formation (arenites, diamictites, siltites, shales), Botucatu Formation (aeolian-207 sandstones), Serra Geral Formation (basalts), Caiuá Group (arenites), and Paraná Group 208 209 (shales, siltites, arenites, arkose) (Lacerda Filho et al., 2004; 2006). The South Paraguay Belt hosts phyllites, schists, metarenites, guartzites, and dolomitic and calcitic marble in 210 211 the Serra da Bodoguena. The North Paraguay Belt includes the Província Serrana, with 212 phyllites, schists, limestones, siltites, and arenites (RadamBrasil, 1982). The point of 213 highest elevation is 1260 m above sea level (m.a.s.l.) on the eastern plateau, and the

lowest point is 70 m.a.s.l. at the basin outlet near the confluence of the Apa and Paraguay

215 Rivers (Figure 1C).



Figure 2: (A) Geology of the Pantanal Basin and drainage network, with major faults. The plateau provenance region is dominated by siliciclastic sedimentary rocks, whereas metamorphic rocks are restricted to the cratons and Paraguay Belt. (B) Soil map for the Pantanal Basin (FAO, 1971). The most widespread soil classes are eutric planosol/fluvisol in the lowlands (Benedetti et al., 2011). Geologic information was obtained for Bolivia (Dirección de Ordenamiento Territorial, Gobierno Autónomo Departamental de Santa Cruz), Paraguay (Vice Ministerio de Minas y Energía), and Brazil (Serviço Geológico do Brasil, CPRM). Faults are based on published studies (Rizzotto & Hartmann, 2012; Warren et al., 2015; Faleiros et al., 2016; Barboza et al., 2018; Rivadeneyra-Vera et al., 2019; Cedraz et al., 2020). White circles represent all sampling sites listed in Table S1. Modified from Lo et al. (2023).





- Figure 3: (A) Mean annual precipitation (mm/y) from the WorldClim database (Fick & Hijmans, 2017). In the Brazilian Pantanal, the precipitation is 970 1850 mm/y. (B) Vegetation ecoregions regions of the Pantanal Basin (Olson et al., 2001). The primary vegetation of the Pantanal consists of flooded savanna and *cerrado* (tropical savanna). White circles represent all sampling sites listed in Table S1. Modified from Lo et al. (Lo et al., 2023).
- 236 The hydrologic configuration of the Pantanal is responsible for the diversity of lowland 237 environments and the contrast between wet-dry seasons (Figure 3A). The Paraguay 238 River flows along the basin's western margin and is the trunk river of the Pantanal. The 239 Paraguay River is joined by the Jauru River west of the Provincia Serrana in the North 240 Paraguay Belt provenance region. The Cuiabá River discharges into the Paraguay River 241 at 17.9°S latitude, followed by the Taquari River's numerous distributary channels discharging just north of 19°S latitude. The Miranda River joins the Paraguay River at 242 243 ~19.4°S latitude. The Paraguay River flows along the Rio Apa craton between 21°S and 22°S latitude before flowing out of the basin. The waters that flow to the trunk river 244 245 annually depend on the migration of the Intertropical Convergence Zone, which concentrates rainfall in the months of December, January, and February and strongly 246 247 influences patterns of flooding and vegetation (lvory et al., 2019). The peak dry season 248 occurs in June, July, and August, but the dry season varies from 1 – 2 months north of 249 the Taquari River to 4 – 5 months south of the Taquari River (IBGE, 2002). This 250 seasonality of rainfall coupled with the minimal gradient of the lowlands results in a flood 251 pulse effect, because rainfall in the plateau and northern Pantanal takes 2 - 3 months to 252 flow to the basin outlet (Junk et al., 2006). When the waters rise with the flood pulse, the 253 suspended clay particles are delivered to the riverbanks and floodplains (e.g., Hamilton, 254 2002). This results in broad areas of inundation in the lowlands that lasts for several 255 months annually. Mean annual rainfall is ~1800 mm in the northern and eastern Pantanal but diminishes to ~1200 mm along the western and southern Pantanal (Figure 3A). The 256 257 average annual temperature is ~25°C basinwide (Fick & Hijmans, 2017).
- Clay minerals can be transformed in contemporary soils depending on climate, slope 258 259 gradients, and vegetation (Hillier, 1995; Velde & Meunier, 2008) (Figures 2B and 3B). The 260 soils of the Pantanal are dominated by eutric planosols and fluvisols in the lowlands, but 261 the plateau provenance region is more variable (Figure 2B). The plateau region contains mostly ferralsols and arenosols in addition to luvisols in the south (Benedetti et al., 2011). 262 Lithosols are present in the Província Serrana region, and the western Pantanal contains 263 264 mollic planosols, rhodic ferralsols, and orthic solonetz. Figure 3B illustrates the general patterns of vegetation, but additional floral diversity relates to the spatial distribution of 265 266 soil types (de Souza et al., 2021). Broadly, the soils are divided into forest formations, 267 arboreal cerrado, herbaceous cerrado, chaco (woody steppic savanna), monodominant formations, and mixed vegetation (dos Santos Vila da Silva et al., 2021). For example, 268 269 semideciduous (capão) and deciduous forests thrive in vertisols, whereas the savanna 270 woodlands (cerradão) grow best in arenosols (de Souza et al., 2021).
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272 **3. Methods**

3.1. Initial design and fieldwork

Paired sand and silt plus clay samples were collected from river margin bars in the Pantanal in 2019 – 2021. Sampling sites were chosen to maximize spatial coverage and lithological diversity and for ease of access. At several sampling sites (n = 22),

277 accompanying hydrologic flow data was recorded at the closest available stream gauge 278 (Table S2). Samples were collected in both the wet and dry seasons (Table S1). Each 279 site was treated as a pour point, which is the endpoint of a streamflow network (Gleyzer 280 et al., 2004). Pour point analysis was completed with QGIS 3.4.6 to define each sample's contributing watershed using Shuttle Radar Topography Mission (SRTM) digital elevation 281 282 models (DEMs) from USGS EarthExplorer (https://earthexplorer.usgs.gov). Pour point 283 analysis for watersheds > 250,000 km² was completed with 3-arc second resolution DEM 284 (Verdin, 2017). The average watershed slope was calculated from these DEMs, whereas 285 elevation and distance from the Paraguay trunk river were extracted from Google© Earth. 286 The precipitation and temperature for each sampling site were measured from WorldClim 287 (Fick & Hijmans, 2017) (Table S1). Soils were identified in each watershed from global 288 FAO (1971) data and converted to the United States Department of Agriculture 289 classification for the purposes of literature review (Deckers et al., 2003; Souza et al., 290 2018), and vegetation ecoregions were also identified from global data (Olson et al., 291 2001). We did not collect local soil profiles and document local vegetation at each 292 sampling site, because we considered each sample to be the product of cumulative 293 upstream processes rather than localized processes and features. Geologic data were 294 extracted from national geologic maps (Lacerda Filho et al., 2004, 2006; SERGEOMIN, 2005; Spinzi & Ramírez, 2014) (Table S3). Seventy-four (74) distinct silt plus clay 295 296 sampling sites were studied, and of these, 71 samples have mineralogy data, 66 have 297 geochemical data, and 63 have both mineralogy and geochemistry data.

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299 **3.2.** Pretreatment and XRD analyses

The silt plus clay fraction was separated by wet sieving using a 53 µm sieve. We treated 300 301 each sample with 1N sodium acetate (NaOAc) with pH 5 adjusted using glacial acetic acid (HOAc) to dissolve carbonates and replace the exchange sites for Ca and Mg with 302 303 Na. We used 30% hydrogen peroxide (H_2O_2) to dissolve organic matter, followed by 304 washing once with 200 mL NaOAc and 200 mL with 1M sodium chloride (NaCI) (Jackson, 305 1969). To obtain the <2 µm fraction, we centrifuged the samples first at 750 rpm for 3 306 minutes and decanted the supernatant (liquid >2.5 cm from bottom of a 250 mL bottle) 307 containing the <2 µm clays into a separate container for settling. The bottle was refilled 308 with sodium carbonate (Na₂CO₃) and the process repeated until a relatively clear 309 supernatant was achieved. The remaining material was separated as the silt fraction (2 -310 54 µm). Several days to weeks was allowed for the clays to settle, and 50 mL of the clays were transferred to a centrifuge tube for freeze drying (Figure 4). 311

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- Figure 4: Flowchart created with BioRender.com that summarizes all pretreatment steps, modified from Jackson (1969). The process begins with a wet sample, dissolution of organic matter and carbonate, separation of <2 μm clays from silt (2 53 μm), and freeze drying of the <2 μm clays.
- 318 Oriented slides of clay fractions were prepared using the filter peel method (Drever, 1973), with diagnostic treatments of magnesium (Mg), Mg-glycerol, and a potassium (K)-319 320 saturated slide. The use of Mq-saturation and subsequent ethylene glycol is chiefly to 321 identify smectite (Aparicio et al., 2010). The application of K-saturation is to identify 322 vermiculite, and further heating to 550°C confirms the presence of kaolinite. Briefly, we measured 200 mg of freeze-dried clay (<2 µm) for each slide and transferred the sample 323 324 to 50 mL centrifuge tubes. We added 25 mL 0.5 M magnesium chloride (MgCl₂), mixed 325 well, and sonicated. In a separate centrifuge tube, we added 25 mL 0.5 M potassium

chloride (KCl) to 200 mg of freeze-dried clay, mixed well, and sonicated. The tubes were centrifuged at 2000 rpm for 5 minutes, the supernatant was discarded, and the process was repeated twice more. We added deionized water, mixed, sonicated the sample, and poured the mixture onto a Millipore 0.45 µm membrane filter mounted to a vacuum flask. With the clay still moist, we removed the filter and placed the filter clay side down on a glass slide. We lightly rolled a 20 mL glass vial across the back of the filter as we peeled away the filter, leaving behind a uniformly thick oriented clay mount.

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334 The mineralogy of the oriented clays was determined using XRD. A PANalytical X'Pert 335 diffractometer with a Cu tube at 45 kV and 40 mA from 2° - 40° with a step size of 0.03° 336 2-theta (20) step size and scan step time of 10 seconds was employed for the analysis. 337 Total scan time was ~3.5 hours for each treatment: Mg-saturated, K-saturated, Mg-glycol 338 solvated, and K-saturated and heated (550°C). When the first two treatments were scanned, we solvated the Mg-saturated slide with glycol to identify if smectite was 339 340 present, and the K-saturated slide was heated to 550°C for one hour to collapse the 341 kaolinite structure. The major constituent clays identified in X-ray diffractograms used established 20 peak positions (e.g., Moore & Reynolds, 1989). All data were analyzed using X'Pert HighScore software. Semi-quantitative calculation of clay mineral 342 343 344 compositions was accomplished by multiplying the height (counts) by the full width at half 345 maximum (FWHM, ° 20) in the Mg-saturated plot divided by the sum of the calculated 346 areas for the predicted clay minerals and multiplied by 100 (Biscaye, 1965; Moore & 347 Reynolds, 1989) (Table S4). These clay abundances were cross checked in NEWMOD II, in order to confirm the reliability of this semi-quantitative method (Yuan & Bish, 2010). 348 Spatial interpolation of the three primary clay minerals (kaolinite, illite, vermiculite) was 349 performed using the "Spline with barriers" tool in ArcGIS Pro 3.1.2. Smectite, goethite, 350 and gibbsite were reported based on presence or absence at each sampling station. The 351 352 iron content in illite was calculated using the intensity of the illite (001) and (002) peaks: I 353 (001)/I (002) (Brown & Brindley, 1980; Deconinck et al., 1988; Furquim et al., 2010; 354 Nascimento et al., 2015).

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356 3.3. WD-XRF geochemistry

357 WD-XRF measurements completed with a Bruker AXS Inc. S4 Pioneer device were used to determine chemical elemental abundances for select bulk sediment samples (Table 358 359 S5) and for the <53 μ m fraction of samples with sufficient material (Table S6, n = 66). 360 Eight duplicate samples were measured to assess the repeatability of the analysis. Following the loss-on-ignition protocol, each sample was heated to 550°C for four hours 361 to remove organic matter and 950°C for two hours to remove carbonate (Heiri et al., 362 363 2001). Samples were disaggregated and homogenized in a mortar and pestle, mixed with borate flux GF-9010 (90% lithium tetraborate and 10% lithium fluoride) in an 8:1 ratio and 364 365 two drops of lithium tetraborate (Li₂B₄O₇), and melted into glass discs using a Katanax X-300 or K1 automatic fusion fluxer machine. The samples were calibrated with a set of 366 eight certified reference samples using linear regression. Molar proportions were utilized 367 to calculate the chemical weathering indices, including the chemical index of alteration 368 369 (CIA; Equation 1) and the Weathering Index of Parker (WIP; Equation 2) (Parker, 1970; 370 Nesbitt & Young, 1982).

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$$CIA = \frac{100 * Al_2 O_3}{Al_2 O_3 + CaO + Na_2 O + K_2 O}$$
.....Equation 1 (Nesbitt & Young, 1982)

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374 $WIP = 100 * \left(\frac{2Na_2O}{0.35} + \frac{MgO}{0.9} + \frac{2K_2O}{0.25} + \frac{CaO}{0.7}\right)$Equation 2 (Parker, 1970)

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376 Both indices are used to determine the extent of weathering (Table S6). Equation 1 (CIA) 377 indicates the extent of feldspar-to-clay conversion, whereas Equation 2 (WIP) measures 378 proportions of alkali and alkaline earth metals, which is suitable for weathering of 379 heterogeneous metasedimentary lithologies (Price & Velbel, 2003). The WIP acts as an 380 index of guartz recycling, whereas the CIA is unaffected by guartz dilution. Calculating 381 the ratio of CIA to WIP allows us to differentiate between weathering and quartz recycling (Garzanti et al., 2019). The weathering indices were spatially interpolated using the 382 383 "Spline with barriers" tool and classified using geometric intervals in ArcGIS Pro 3.1.2. 384 We evaluated the influence of source rock composition on fine-fraction sediment chemistry using ACN, ACNK, ACNKFM plots and molar proportions of the major elements 385 (Nesbitt & Young, 1984; Nesbitt & Wilson, 1992; Fedo et al., 1995). 386

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We examined environmental controls on clay mineralogy and chemistry using canonical correspondence analysis (CCA). The key advantage for using CCA was to distinguish how the environmental variables and major elements affected both the clay abundance and the sampling sites. We also explored other ordination analyses to examine their effectiveness in explaining the distribution of clay minerals in the Pantanal Basin. Finally, we measured pH for a representative set of sediment samples from each region, treating each sample as a 1:2 soil/0.01 M CaCl₂.

396 **4**. **Results**

397 4.1. Basin-wide clay mineralogy

398 At basin-scale, the rank order of clay mineral abundance is kaolinite > vermiculite > illite 399 > smectite (Table S4). Kaolinite was determined to be present if the 7 Å diagnostic (001) 400 peak collapsed when the K-saturated slide was heated to 550°C for one hour. Illite was 401 identified at 10 Å for the (001) peak and 5 Å for the (002) peak. Smectite was identified 402 on the basis of the 14 Å diagnostic peak shifting to 18 Å following Mg-glycerol (Figure 5). 403 Vermiculite was distinguishable from smectite where the 14 Å peak did not shift to 17 – 18 Å following Mg-glycerol treatment. We identified goethite at the (110) peak at 4.18 Å 404 and gibbsite at the (002) peak at 4.85 Å (Moore & Reynolds, 1989). The Cuiabá, Taquari, 405 406 and Paraguay River muds are particularly enriched in kaolinite, representing >50% of the 407 clay mineral composition (Figure 6). Substantial in-channel compositional variability was observed in the Taguari River, which is fed by two large tributary rivers carrying 71% 408 409 kaolinite and 43% kaolinite in the plateau.



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°20 °2A Figure 5: Representative x-ray diffractograms from oriented clay mounts using diagnostic treatments of 413 Mg-, Mg-glycerol, K-25°C, and K-550°C for the six provenance regions detailed in Figure 1B. 414 Diagnostic peaks are labeled for quartz (Q), kaolinite (K), illite (I), smectite (S), vermiculite (V), 415 goethite (G), and gibbsite (Gi). The x-axis showing ° 20 is identical across all six panels of the 416 figure. 417

Spatial interpolation enabled additional basin-wide observations. The highest kaolinite 418 419 percentages (>70%) were found in the medial Pantanal Basin, at the confluence of the 420 Paraguay River with the distal distributary channels of the Taguari River (Figure 6). The 421 medial Pantanal region is known to be regularly inundated with flood waters for some of 422 the longest periods of the year (lvory et al., 2019). The northernmost plateau sampling stations also produced similarly high kaolinite abundances. The northeastern plateau 423 sampling sites were generally enriched in kaolinite downstream of orthic ferralsols and 424 425 ferralic arenosols, whereas the southeastern plateau sampling sites in the Miranda River watershed with extensive ferric luvisols were depleted of kaolinite (Figures 2B and 6). In 426 contrast, the Miranda River watershed has the highest concentrations of vermiculite in 427 428 the Pantanal. As the Paraguay River flows to the basin outlet in the south, the proportion of kaolinite in the river muds decreases noticeably. There, vermiculite was more common 429

430 (>50%) in the clay assemblages, particularly at sampling sites fed by rivers draining the 431 Rio Apa craton (Figure 6) and the South Paraguay Belt. Both the Miranda and Apa rivers 432 drain the Serra Geral Formation dacite, which produce rhodic ferralsols (Figure 2A) 433 (Lacerda Filho et al., 2006). Illite was 40 - 60% in the <2 µm fraction in the Paraguay Belt 434 region where phyllite and amphibolite schist parent lithologies dominate, producing 435 lithosols and orthic acrisols. The Paraguay Belt contained the highest contributions of illite 436 to the Pantanal Basin, followed by the São Lourenço River draining surfaces covered by

- 437 lithosols, acrisols, and ferralsols (Figures 3A and 6).
- 438



- 439
4400%82%0%64%0%93%441
441Figure 6: Spatial interpolation maps of the major clay mineral constituents at 71 sampling stations in the
Pantanal. For each sampling station, the composition was normalized to 100 based on the kaolinite,
illite, and vermiculite percent estimates. Interpolation was not extended to hashed areas.
- 443

The full width at half maximum (FWHM) of the diagnostic peaks shows how crystallinity 444 445 changes along the length of the Taguari River (Aparicio et al., 2006). The kaolinite (001) FWHM remained constant along the length of the megafan, indicating no changes to the 446 447 crystallinity. The gibbsite diagnostic peak at 4.85 Å in the Taguari River sampling station disappears from the proximal to the distal megafan regions. Chlorite was not found, and 448 449 smectite was identified only at limited sites such as sample A26 in the medial Taguari River megafan where the vermiculite and smectite peaks could be clearly disentangled. 450 The average iron content in illite was 2.19 (dimensionless, computed intensity 451 452 (001)/intensity (002)), with much of the highest iron content located in tributaries of the 453 Jauru, Paraguay, and Cuiabá Rivers in the northern Pantanal.

454

455 Gibbsite and goethite are not abundant clay minerals in samples from the Pantanal (25%) 456 of samples contained neither of these minerals), but they do constitute important minor 457 components (Figure 7). Sampling sites with both gibbsite and goethite were most 458 frequently present in the north plateau region, where the mean annual precipitation in the 459 basin is the greatest (Figure 3A). Gibbsite was most common in samples along the 460 Taguari River megafan, whereas goethite occurred in samples from the southern plateau and in the medial lowlands (Figure 7A). Samples with neither mineral were common in 461 the Rio Apa craton in southern Pantanal. Smectite was identified in ~30% of the 71 462

463 sampling stations (Figure 7B). The presence of smectite appeared to be distributed 464 across the Pantanal Basin with no specific pattern.

465



Figure 7: (A) Basin-wide map of gibbsite and goethite among 71 stations. Pink (gibbsite + goethite), green (goethite), yellow (gibbsite), and gray (neither). (B) Map of smectite in the Pantanal marked as purple (no smectite) or orange (smectite present). These maps did not account for the intensity or crystallinity of the mineral peaks.

471 472

473 **4.2. Basin-wide geochemistry**

Ternary diagrams showed that most samples exhibited >70% Al₂O₃, and all lowland 474 samples >80% Al₂O₃ (Figure 8A, B). Most samples contained <10% Na₂O (Figure 8B) 475 and 20-30% Fe₂O₃ + MgO (Figure 8C). Geochemically, all samples from watersheds in 476 477 the South Paraguay Belt provenance region and select samples from the Rio Apa craton provenance region were enriched in Ca, Na, and K and low in AI (Figure 8C). Average 478 values for geochemical variables in the six provenance regions further show the greatest 479 480 SiO₂ enrichment in the lowlands provenance region and the least SiO₂ enrichment in the 481 South Paraguay Belt (Table 1). The South Paraguay Belt and plateau provenance regions had average pH 6.6 and 53% kaolinite, whereas the lowland provenance region and the 482 North Paraguay Belt had an average pH of 5.8 and 22% kaolinite (Table 2). 483



Figure 8: Major elemental compositions of fluvial sediments (*n* = 66) plotted as molar proportions on Al₂O₃-CaO-Na₂O (ACN), Al₂O₃-(CaO + Na₂O)-(Fe₂O₃ + MgO) (ACNKFM), and Al₂O₃-(CaO + Na₂O)-K₂O (ACNK). The ACN and ACNK plots form a linear distribution with South Paraguay Belt samples closer to the C- and the CN-pole, respectively. Most mud samples rich in non-mobile Al plot closer to the A-pole. The ACNKFM plot helps distinguish the Mg-rich samples.

492 The geochemical discrimination plots show decreasing Al_2O_3 as SiO_2 increases, 493 consistent with quartz addition relative to the UCC standard (Figure 9) (Taylor & McLennan, 1995). The lowland, Rio Apa craton, and Amazon craton samples followed 494 495 this quartz enrichment trend closely, whereas the plateau and Paraguay Belt samples diverged from this guartz enrichment pattern. Approximately 50% of the samples (Figure 496 9C) contained less SiO₂ than the UCC and were mostly <5 Na₂O+K₂O+MgO+CaO. Most 497 samples followed the guartz addition trend (Figure 9C, D). However, the CIA/WIP plot 498 499 showed mostly a weathering trend concentrated at 70 – 90 CIA and <40 WIP (Figure 9B).





510 511

 Table 1: Chemical elemental abundance summary statistics

Provenance	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	K₂O	Na₂O	CaO	MgO	Kao	Illite	Verm	CIA	WIP
Area	wt%	wt%	wt%	wt%	wt%	wt%	wt%					
A	71.8	15.0	5.6	1.7	0.1	0.4	0.8	56	5.6	35	85.0	18.0
Lowlands	Comments	s: kaolinite	comprised 50) – 60% of t	he Paragua	y River clay	s near th	ne basin	outlet.	Vermicul	ite com	posed
>30% of the lowland clays downstream of the Rio Apa craton.												
В	64.1	18.4	7.4	2.1	0.4	1.1	0.9	51.3	16	32.6	78.7	27.2
Amazon			>									
craton	Comments	s: vermiculi	te varied 15 -	– 59%, and	illite ranged	l 7 – 36%.						
С	64.8	18.2	5.7	3.3	0.8	1.5	0.8	31.4	18.9	38.8	71.3	41.2
Rio Apa	Comments	s: smectite	was commo	on where v	volcanic roc	ks formed	16% of	the wa	tershed	d. Kaolini	ite was	most
craton	common i	n the samp	e where folia	ted metam	orphic rocks	were the s	single lar	gest co	nstituen	t litholog	у.	
D	66.6	13.5	9.5	1.6	0.1	1.0	0.9	41.5	12.1	46.7	79.4	19.2
Plateau	Comments	s: Clay asse	mblages in t	he Miranda	River water	sheds were	almost o	dominar	ntly vern	niculite.	Two san	npling
	stations m	ay drain the	same silicic	lastic litholo	ogies but pro	duce vastly	differen	t propor	tions of	kaolinite	, for exa	mple.
E	59.3	12.5	5.5	1.7	0.1	18.7	1.3	18.5	30.3	51.2	34.6	66.4
South	Comments	s: kaolinite	reached the	lowest prop	ortions of a	ny area in t	he Panta	anal. Bio	ochemic	al litholo	gies we	re the
Paraguay	main pare	ent rock of	the watersh	eds, but cl	ay assembl	ages were	more c	ommon	ly contr	olled by	the ad	acent
Belt	metamorp	hic lithologi	es.									
F	68.1	15.3	7.2	2.5	0.1	0.3	1.1	37.3	42.6	20.2	81.4	25.8
North												
Paraguay												
Belt	Comments	s: many sin	ples contain	ed >50% ill	lite, which ex	ceeds all c	ther reg	ions in t	he Pan	tanal.		
Victo Brovenence regione are lowlands (A) Amazon araten (B) Die Ana araten (C) plateou (D) South							Couth					

512 Note. Provenance regions are lowlands (A), Amazon craton (B), Rio Apa craton (C), plateau (D), South 513 Paraguay Belt (E), and North Paraguay Belt (F). Average smectite values were not determined due to 514 occurrence in few sampling sites. Abbreviations are Kao = kaolinite and Verm = vermiculite.

516 Table 2: pH values for selected samples

Sample	рН	%Kaolinite	Kaolinite FWHM		
A2	5.34	57	0.3542		
A3	5.40	51	0.4723		
F2	5.86	34	0.4723		
F1	5.88	73	0.3542		
C1	5.94	39	0.3542		
C8	5.97	39	0.3542		
A27	5.98	52	0.3542		
B1	5.99	77	0.3542		
C2	6.33	5	0.3542		
D1	6.37	25	0.3542		
B2	6.58	56	0.3542		
E1	6.62	17	0.4723		
E3	6.82	15	0.2952		
D3	6.85	12	0.3542		

517 Note. Samples are ordered by increasing pH representing the six provenance regions. lowlands (A),

518 Amazon craton (B), Rio Apa craton (C), plateau (D), South Paraguay Belt (E), and North Paraguay Belt (F). The sample with the largest watershed in each provenance region was chosen to obtain the net cumulative

519

520 pH value.

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522 Weathering indices and statistical analysis 4.3.

Table 1 provides summary statistics across all six provenance regions. The CIA and WIP 523 values showed that samples from the lowlands provenance region were the most 524 weathered, especially near the Taguari megafan where high values (83 - 94) of the CIA 525 were recorded. Weathering intensities measured by WIP were more variable in the 526 lowlands (10 – 36) (Figure 10). The Itiquira and Piquiri Rivers draining the plateau 527 highlands immediately north of the Taquari River produced silt plus clay minerals that 528 were consistently highly weathered as indicated by the CIA and WIP. The weathering 529 indices reflected the clay mineral proportions deduced by XRD; the areas with the 530 greatest kaolinite proportions coincided with the highest CIA values and the lowest WIP 531 values for the Piquiri, Cuiabá, São Lourenço, and the Paraguay River at the Taquari 532 megafan (Figure 10). In contrast, the lowest CIA values and highest WIP values were 533 recorded in the Rio Apa and South Paraguay Belt regions (Figure 10). 534

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Figure 10: Spatial interpolation maps of chemical weathering indices based on the molar proportions of the major elemental data from 66 sampling stations.

539 Fe₂O₃ and metamorphic parent lithologies loaded positively on axis 1 of the canonical 540 correspondence analysis (Figure 11). The SiO₂, K₂O, and average watershed slope loaded negatively on axis 1. The average watershed slope, MgO, and Fe₂O₃ loaded 541 542 positively whereas watershed area, sedimentary parent rocks, and SiO₂ loaded negatively on axis 2. All lowland samples plotted in negative axis 2 space, whereas most 543 of the Paraguay Belt samples plotted in positive axis 2 space. Two large clusters of data 544 545 points are distinguishable. The first cluster is oriented diagonally along a continuum 546 formed by the SiO₂ and Fe₂O₃ rays with n = 44 sampling sites. The samples with more SiO₂ are also closer to the abundance of kaolinite, and samples with more Fe₂O₃ are 547 548 associated with vermiculite. The quadrant with vermiculite is characterized by significantly more MgO and slightly more Al_2O_3 , as suggested by the length of the shorter ray for Al_2O_3 . 549 The second cluster of data is oriented perpendicular to the first cluster, composed mostly 550 551 of Paraguay Belt samples and an association with illite, K₂O, average watershed slope, 552 and elevation of the sampling stations.



enriched in K₂O, whereas the sampling stations in the Miranda River basin in the plateau region

are enriched in Fe_2O_2 in the first axis. In the second axis, most of the Rio Apa craton and Paraguay

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5. Discussion

561 **5.1.** Insights from clay mineralogy

Belt sampling stations show enriched MgO and CaO.

562 5.1.1. Climate control

563 The Pantanal Basin is warm and seasonally wet with open *cerrado* savanna vegetation 564 in the hinterland areas (Cole, 1960), where a pronounced hydroclimate gradient in rainfall 565 and seasonality controls modern clay distribution. The Taquari River forms a weathering 566 hinge between the increased weathering intensity to its north and reduced weathering 567 intensity to its south. The greater rainfall and shorter dry season north of the Taquari River 568 results in high kaolinite production as bedrock and soils are leached (Goldich, 1938; 569 Depetris & Griffin, 1968; Singer, 1980; Garzanti et al., 2014; Guinoiseau et al., 2021). 570 Most neoformed kaolinite in soils is subsequently transported downstream towards the 571 Paraguay River in the suspended sediment fraction (e.g., Depetris & Probst, 1998). The 572 fluvial sediment samples in the medial Pantanal reflect the cumulative climate-driven 573 weathering north of the Taquari River hinge. On the basis of greater kaolinite abundance 574 in the northern Pantanal, our data broadly support the hypothesis that mean annual 575 precipitation controls clay mineralogy.

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577 As the clay minerals are carried as suspended loads, their composition is subsequently 578 transformed (Setti et al., 2014). Detrital clays such as vermiculite and illite can be 579 compared with transformed clays such as kaolinite and smectite as a measure of 580 chemical weathering and mechanical erosion (Shover, 1963; Vanderaveroet et al., 2000; 581 Setti et al., 2014). The Jauru and Paraguay River clays west of the Província Serrana are 582 mostly kaolinite, plus illite from the North Paraguay Belt. These clays likely originated from the Amazon craton and the Paraguay Belt lithologies, in addition to the siliciclastic plateau 583 584 at the northernmost end of the basin. The illite (> 60%) in the uppermost Cuiabá River 585 and along small watersheds of the North Paraguay Belt points to rapid mechanical 586 weathering (e.g., Selvaraj & Chen, 2006). Mechanical weathering breaks down outcrops of the muscovite-bearing Cuiabá Group rocks (Alvarenga et al., 2011; Vasconcelos et al., 587 588 2015). This interpretation is consistent with the lithosols, which are thin and poorly 589 developed (Camargo & Bennema, 1966). Following the confluence with the Cuiabá River, 590 the Paraguay River carries more kaolinite, similar to levels recorded in the tributaries of 591 the Cuiabá River.

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Lower kaolinite proportions in samples from sites south of the Taquari River are interpreted to be linked to reduced rainfall (~1200 mm/y) and increased length of the dry season (4 – 5 months). In contrast, areas north of the Taquari River are characterized by ~1800 mm/y and 1 – 2 month-long dry season. This pattern of reduced weathering intensity that produces more detrital clays (e.g., illite and vermiculite) relative to transformed clays (e.g., kaolinite and smectite) supports our hypothesis.

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600 The 2:1 type clays were primarily vermiculite, commonly a byproduct of incomplete 601 weathering of biotite (Cleaves et al., 1970; Ojanuga, 1973; Johnsson & Meade, 1990). 602 The Paraguay River clays downstream of the confluence with the Taguari and Miranda 603 Rivers begin to incorporate substantial vermiculite. The South Paraguay Belt region and 604 the plateau provenance region south of the Taguari River weathering hinge contain ferric 605 luvisols overlying carbonate and foliated metamorphic rocks, a unique combination of 606 geological factors in the Pantanal Basin. As the Paraguay River flows along the Rio Apa 607 craton towards the basin outlet at the confluence with the Apa River, the clay composition 608 is modified by the illite and vermiculite chemically and mechanically eroded from the 609 craton. The intensity of weathering inferred from the relative proportions of 50% kaolinite 610 and 50% illite + vermiculite would indicate incomplete weathering closer to the outlet 611 (samples A2 and A3; Table S1) than in the medial Pantanal Basin (samples A6 – A9; 612 Table S1). The modification of the Paraguay River suspended clays attests to non-linear 613 compositional changes downstream.

615 **5.1.2**. Soil control

The composition of extant soils in the provenance regions is interpreted to be an important secondary control on modern clay mineralogy and chemistry. Although we did not examine the mineralogy of soil profiles adjacent to each sample, we can infer soil properties and clays based on the soil classification map (Figure 2B). Using the soil classification map is sufficient for this study, because each sample is an integrated result of the cumulative processes in the entire upstream watershed.

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623 The extensive availability of kaolinite in the dominant soils helps to explain higher 624 proportions of kaolinite in modern fluvial samples north of the Taquari weathering hinge. 625 Soils in the northern Pantanal Basin were described as acrisols, arenosols, and ferralsols. 626 The acrisols and ferralsols are known to have high amounts of kaolinite and gibbsite clays 627 in the topsoil and subsoil, whereas the arenosols contain kaolinite and illite primarily in 628 the subsoil (Ito & Wagai, 2017). The abundant kaolinite usually occurs in lateritic soils (e.g., Truckenbrodt et al., 1991), which may appear as ferralsols or ferralic arenosols in 629 630 the Pantanal (Figure 2B) (Righi & Meunier, 1995; Mathian et al., 2020). Some of the kaolinite formed from laterite is instead replaced by hematite (Ambrosi et al., 1986), as 631 632 observed by the iron-rich concretions (~1 cm in diameter) on the armored, wind-deflated surface of the Taquari River's lateritic soils (Figure 12). Although not shown on the map, 633 634 gleysols and plinthosols also contribute to the high occurrence of kaolinite in the northern 635 Pantanal soils (Coringa et al., 2012).

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Soils in the southern Pantanal Basin are distinguished by the extensive development of 637 luvisols that contain greater proportions of illite in both the topsoil and subsoil (Ito & 638 Wagai, 2017; Warr, 2022). The South Paraguay Belt soils are dominantly mollisols, 639 containing primarily vermiculite, followed by illite (Warr, 2022). Mollisols are interpreted 640 641 as key contributors to the higher proportions of vermiculite in the southern Pantanal. Soil 642 clay mineralogy and the processes may be altered due to human-induced land use changes (e.g., Céspedes-Payret et al., 2012; Fink et al., 2014; Austin et al., 2018). 643 644 However, the relationship between land use and fluvial clay minerals remains unclear, so 645 disentangling the anthropogenic land use effect on clays from each sample is not feasible. 646 The presence of dibbsite is an indicator of intense weathering and desilication (Certini et 647 al., 2006; Reatto et al., 2008). Gibbsite is an aluminum hydroxide associated with strong 648 hydrolysis and bauxitization processes (Chamley, 1989; Velde & Meunier, 2008). 649 Bauxitization, or the formation of aluminum ore, occurs when extensive hydrolysis leads to gibbsite authigenesis. We infer that the gibbsite was eroded primarily from the 650 surrounding soil cover (phaeozems, luvisols, and ferralsols). The northeastern Pantanal 651 652 including the São Lourenço and Cuiaba Rivers are sources of gibbsite. Gibbsite peaks were also identified in the Amazon craton rivers and the South Paraguay Belt samples. 653 654 The gibbsite identified in the medial Paraguay River, upstream of the confluence with the 655 Taguari River, were likely derived from erosion of lateritic soils in the uppermost regions of the plateau provenance region. Iron-bearing minerals, particularly hematite and 656 goethite, are common in lateritic soils (Madeira et al., 1997). However, the occurrence of 657 658 gibbsite and goethite was not consistently related to higher or lower kaolinite proportions. 659 This observation suggests that the highest proportions of kaolinite are independent of 660 goethite and gibbsite occurrence.



Figure 12: Uppermost hinterland of the Taquari River watershed consists of (A) deeply incised gullies facilitating sediment export to the lowlands. The surfaces are commonly characterized by friable iron-rich concretions, known as ferricretes (B, C, D). Photo credit: E. Lo.

666 5.1.3. Geological and slope control

When illite is generated from metamorphic rocks, rapid removal of material is often 667 668 implicated, which is expected in a tropical environment with heavy seasonal rainfall like 669 the Pantanal (Selvaraj & Chen, 2006; Velde & Meunier, 2008; Wang et al., 2011). The close spatial relationship between illite abundances and the North and South Paraguay 670 671 Belt provenance regions suggests a direct contribution from the muscovite-rich greenschist facies (Almeida et al., 1976). The broad spatial occurrence of illite in the North 672 673 Paraguay Belt region is evidence of mechanical bedrock erosion observed in regions of 674 high precipitation (e.g., Liu et al., 2012). Calculation of iron content in the mica structure 675 yielded a relatively high dimensionless average value of 2.19 (dimensionless, computed intensity (001)/intensity (002)) (Brown & Brindley, 1980; Deconinck et al., 1988). High iron 676 677 availability is a prerequisite for authigenesis of ferric illite (Furguim et al., 2010), consistent with tropical environments that generate extensive iron oxides (Liptzin & Silver, 2009). 678 679 The largest values for Fe content in the illite were mostly concentrated north of the Taquari 680 weathering hinge. The increased distribution of illite in the North Paraguay Belt is consistent with present-day weathering conditions. 681

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The weathering of phyllite and amphibolite schist outcrops along the Salobra River, the Miranda River, and the uppermost Apa River are the best candidates for vermiculite generation. Select sampling stations in the Miranda River contain as much as 90% vermiculite, which we attribute to the erosion of adjacent Cuiabá Group phyllites (Lacerda Filho et al., 2006). Dacites such as the Serra Geral Formation in the study area have generated vermiculite clays in other regions (Harvey & Beck, 1962). Illite may also be altered to vermiculite as K is released in the soils, creating an interlayered illite-vermiculite mineral similar to Amazon Basin soils and verified with NEWMOD II fitting (Han et al.,
 2014; Delarmelinda et al., 2017). Sample E1 in the South Paraguay Belt region contained
 an intermediate peak at 11.9 Å suggesting the presence of hydroxy-interlayered
 vermiculite (HIV), implicating a mixed layer illite-HIV.

694

695 The average watershed slope regulates fluvial incision and channel behavior. Steeper 696 slopes favor higher mass wasting rates, incised river channels, and minimal pedogenic 697 development. In contrast, the low-slope floodplains act as temporary sinks for 698 unconsolidated, highly weathered fine sediment subject to fluvial channel migration. The 699 Taquari River at the distal Zé da Costa avulsion (sample A25; Table S1) contained greater 700 amounts of vermiculite than at the medial Caronal avulsion (samples A26-A27; Table S1), 701 suggesting that reworked floodplain sediments may be an important contribution of vermiculite in the distal Taquari River. The exhumation of floodplain deposits can 702 703 remobilize clays that were deposited during drier Holocene climatic conditions where 704 transformation of clays was less efficient (McGlue et al., 2015, 2017; Novello et al., 2017). 705 Vermiculite is diluted by dominantly kaolinitic tributary inputs when the Taguari River 706 discharges into the Paraguay River.

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708 Clay minerals may represent inherited weathering phases from recycling of more ancient 709 sedimentary rocks that are exhumed to the surface environment (Eberl et al., 1997; 710 Wilson, 1999; Bhattacharyya et al., 2000). For example, inherited clays may come from 711 clay coats that formed prior to lithification of aeolian sands into arenites (Wilson, 1992). The Mesozoic Botucatu Formation in the plateau provenance region is an aeolian 712 sandstone with amorphous silica, pore-filling, and kaolinite and smectite grain coatings 713 714 (França et al., 2003; Hirata et al., 2011; Bertolini et al., 2020; 2021). The Mesozoic Era 715 was characterized by hothouse conditions (Holz, 2015) followed by diagenetic processes 716 that contributed to the generation of kaolinite in the Botucatu Formation (Corrêa et al., 717 2021). This formation may have contributed an unknown amount of inherited kaolinite to 718 the silt plus clay fraction recovered in modern river samples (Balan et al., 2007). Inherited 719 kaolinite is commonly more ordered than neoformed kaolinite (Balan et al., 2007; Bauluz 720 et al., 2008), such that higher crystallinity with lower FWHM can indicate inheritance. Samples with the most disordered (neoformed) kaolinite (FWHM 0.45 ° 20) were located 721 at the confluence of the Taguari and Paraguay Rivers, where elevations are very low and 722 723 the annual floodwater inundation period is high (lvory et al., 2019). Furian et al. (2002) 724 likewise encountered kaolinite in poorly drained areas of the Pantanal. The high levels of 725 kaolinite at the confluence of the distal Taquari River and the Paraguay River further attest to kaolinite authigenesis associated with strong hydrolysis (Chamley, 1989). Estimating 726 727 inherited versus neoformed kaolinite remains challenging, because studies such as Balan 728 et al. (2007) examined these processes primarily in soil profiles, not in modern fluvial 729 samples.

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731 **5.2.** Insights from geochemistry

Clay minerals generated today across the Pantanal Basin are controlled primarily by
 climate-induced chemical weathering and secondarily by soil and parent lithology. Nearly
 all Rio Apa craton samples were relatively enriched in Na, K, and Al compared to samples
 from rivers draining the other provenance regions. Similarly, the Amazon craton samples

736 had high relative Na, K, and Al, but less than that of the Rio Apa craton samples. South 737 Paraguay Belt samples were all enriched in Fe and Ca. Most of the lowland samples were 738 high in Si, reflecting the guartzose nature of muds where repeated cycles of flooding and channel avulsions enhance sediment reworking (e.g., Louzada et al., 2021). The rivers 739 740 draining provenance regions with the lowest CIA values were the Rio Apa craton and the 741 South Paraguay Belt, suggesting that the weathering effect for these metamorphic and 742 carbonate rocks was low, most likely due to the reduced mean annual rainfall (~1200 743 mm/y) (Fick & Hijmans, 2017). These two regions also had WIP >40, indicating reduced guartz recycling relative to the other four provenance regions. Most samples from the 744 745 Pantanal had CIA 75 – 95 and had WIP <20, attesting to both high guartz recycling and 746 extensive weathering effects (Figure 9B). Our spatial distribution maps show that this 747 effect was most concentrated in the medial Pantanal Basin, supplied mainly by the 748 Cuiabá, São Lourenço, and Piguiri Rivers (Figure 10). Maximum guartz recycling and 749 weathering effects were consistent with the highest quartz compositions observed in 750 Paraguay River fine fraction samples (n = 7) near the confluence of the Paraguay River 751 with the Cuiabá and Taguari Rivers. The lowest WIP values in the basin are likely linked 752 to the Cretaceous Botucatu Formation and the Cretaceous Bauru Formation guartz 753 arenites of the plateau provenance region (Fernandes & Magalhães Ribeiro, 2015; Bertolini et al., 2021). Because quartz arenite weathering contributes little to the clay 754 755 fraction in extant river muds, we interpret that most of the depletion of mobile ions 756 occurred through kaolinite authigenesis by transformation. This view is consistent with 757 the presence of goethite and gibbsite in unconsolidated sediments of the Botucatu 758 Formation (Fagundes & Zuguette, 2011).

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760 **5.3.** Clay transformation in the Plata River

The clav composition of the Pantanal back-bulge is distinguishable from the Andean 761 foreland basin clays. The Paraguay, Paraná, and Uruguay Rivers are the primary sources 762 763 of kaolinite to the Plata River estuary (Table S7), which ranges from 50 - 75% in the suspended load (Figure 13) (Depetris & Griffin, 1968; Manassero et al., 2008). Samples 764 765 downstream of the Pantanal outlet were commonly 15 - 20% kaolinite, indicating dilution 766 of the kaolinite by sub-Andes-derived illite. The Bermejo River is an example of 767 concentrated illite supply to the Paraguay River (Bertolino & Depetris, 1992; McGlue et al., 2016; Repasch et al., 2021). Bermejo clays were <5% kaolinite near the thrust front, 768 769 and the kaolinite remained <5% as far as 40 km downstream (Bertolino & Depetris, 1992). 770 Illite comprised ~60% of clay composition throughout the length of the Bermejo River to 771 its confluence with the Paraguay River. Other rivers such as the Pilcomayo and the Salado Rivers that drain the Andean thrust belt were similarly enriched in illite (Bertolino 772 773 & Depetris, 1992; McGlue et al., 2016). We ascribe the dilution of kaolinite in the Paraguay 774 and Paraná Rivers to these illite-rich clay compositions draining the Andean foothills. The 775 back-bulge and interior craton are dominated by kaolinite, thereby creating ancient wetland deposits that are also rich in kaolinite (e.g., Tineo et al., 2022). In contrast, the 776 thrust front sediments are dominated by illite and smectite, with the latter influenced by 777 778 the dry climate of the Chaco Seco (McGlue et al., 2016).



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- Figure 13: Summary of mud transport from the Paraguay River to the confluence with the Pilcomayo, Bermejo, Paraná, and Salado Rivers to the Plata River mouth. Data (n = 84) from past studies 784 (Depetris & Griffin, 1968; Bertolino & Depetris, 1992; Ronco et al., 2001; Manassero et al., 2008; 785 McGlue et al., 2016) (Table S7). Pantanal samples in the hatched area are from this study. The 786 rivers were obtained from the HydroSheds database (Lehner et al., 2008), and the Plata River 787 watershed was downloaded from the Transboundary Freshwater Diplomacy Database, College of 788 Earth, Ocean, and Atmospheric Sciences, Oregon State University. Additional information about the TFDD can be found at: https://transboundarywaters.oregonstate.edu.
- 789 790

791 We find that the dominantly kaolinite clay composition at the Pantanal outlet is controlled by climate > soil > lithology. We interpret that because vermiculite clays were not present 792

793 in downstream clay fractions, this observation suggests that vermiculite might be diluted 794 by illite as it exits the Pantanal Basin. In addition to this dilution effect, we identify three 795 potential factors for the rapid change in clay composition. First, the decreased kaolinite in 796 the Paraguay and the Paraná Rivers roughly coincide with the boundary between tropical 797 savanna climate (Aw) and the humid subtropical (Cfa) zones (Beck et al., 2018). 798 Campodonico et al. (2016) demonstrate that the CIA decreases downstream in the higher 799 latitude and sub-tropical climate regions. Second, the adjacent lithologies may be 800 supplying illite locally to the fluvial clays. Illite was formed from burial diagenesis (Lanson 801 et al., 2002) and locally eroded into the Paraguay River as it flowed past the Rio Apa 802 craton, which diluted the sediment samples. The lower Paraguay River flows adjacent to 803 the Carboniferous Coronel Oviedo Group, consisting of shale, arenite, diamictite, and 804 glacial tills (Orué, 1996). Third, kaolinite-rich clays might be preserved near the wetland 805 but not preserved in much farther downstream sediments. Further systematic 806 investigations of downstream clay compositions and heavy mineral suites to constrain the 807 controls on clay composition in the entire Plata River catchment is warranted. This study 808 of modern fluvial clays is an important contribution to understanding clay distribution in 809 modern river sediments and provides a key source of information to improve the accuracy 810 of global clay distribution models (Ito & Wagai, 2017; Warr, 2022).

812 6. Conclusions

This study of modern fluvial clays plus silt from 74 sampling sites revealed the spatial distribution of clay minerals and major fine-fraction chemical elements across an extant tropical back-bulge basin. Mineralogy and chemical weathering indices (CIA and WIP) showed distinct areas of clay generation among the provenance regions. The controls on fine-fraction mineralogy were systematically assessed, and the implications for the downstream fine sediment in the Plata River were summarized.

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The clay proportions follow the rank order pattern of kaolinite > vermiculite > illite >
smectite, but these clays are not evenly distributed. The Taquari River forms a prominent
E-W trending hinge across the Pantanal Basin, where more intensive leaching and soil
authigenesis produce more kaolinite north of the river. Vermiculite was more common
south of the Taquari River, and illite was most common along the North Paraguay Belt.
Gibbsite and goethite in the clay-sized fraction signaled contribution from heavily
weathered soils such as laterites.

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Major elemental geochemistry of the clay plus silt was used to calculate average CIA =
76.4 and average WIP = 27.6 throughout the Pantanal. The medial Pantanal Basin is
highly weathered at the confluence of the Taquari and Paraguay Rivers, representing the
cumulative weathering effects of the northern Pantanal. The southern Pantanal fine
sediments along the Rio Apa craton and the South Paraguay Belt were poorly weathered,
displaying greater values of CaO, Na₂O, and K₂O consistent with climate and parent
rocks.

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- The main controls on modern fluvial clay plus silt were climate > soil > parent rock. This
 interpretation was supported by the Taquari River weathering hinge, where kaolinite-rich
 clays north of the river were linked to greater precipitation and shorter dry season. This

- same region also contained more kaolinite-rich soils such as acrisols and ferralsols. In
 contrast, mollisols and luvisols coupled with reduced precipitation and longer dry seasons
 in the southern Pantanal allowed for more detrital clays: illite and vermiculite. Illite was
 especially linked to low-grade metamorphic lithologies present only in the Paraguay Belt.
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The Pantanal Basin's clay mineral composition near the basin outlet is primarily kaolinite
and vermiculite, contrasting sharply with detrital back-bulge clays from the sub-tropics
(Bermejo, Pilcomayo, etc.), which are dominated by illite and smectite. The illite
transported from the sub-Andean regions significantly dilutes the proportion of kaolinite
in the Plata River. This composition likely generates distinct mudstones in the
stratigraphy, with implications for interpretations of the rock record.

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- 878 8. Conflict of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

- 881882 9. Data availability statement
- All our relevant datasets are included in supplementary materials.

884 **10.** Authors contribution

Conceptualization: E.L.L., M.M.M., and A.S. Logistics and fieldwork: G.G.R., E.L.L., A.S.,
 S.K., and R.O.L. Lab analyses: E.L.L. and K.C.H., with access and training to the soil
 chemistry lab provided by C.J.M. Writing and figure development: E.L.L., M.M.M., C.J.M.,
 and G.G.R. All authors have read and agreed to the published version of the manuscript.

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