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1 **Unraveling late Quaternary atmospheric circulation in the Southern Hemisphere**
2 **through the provenance of Pampean loess**

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25
26 **Abstract**

27 The Pampean loess is the most extensive continental paleo-record of aeolian material in the
28 Southern Hemisphere, recording the deposition of dust transported by two major zonal wind
29 systems: the southern westerly winds and the subtropical jets. In order to increase the

30 understanding on paleo-atmospheric circulation over the Southern Hemisphere, we evaluate dust
31 provenance through REE, Nd, Sr and Pb isotopes in three sections deposited during late
32 Pleistocene-early Holocene across 700 km in the loess belt of the Pampean region in central
33 Argentina. The isotopic comparison of loess from the three sections with southern South
34 American (SSA) potential dust sources show that (1) sources from the southern Altiplano to
35 latitudes of northern Patagonia supplied dust to the Pampas, (2) the slight Sr-Nd isotopic
36 difference between fine and coarse loess may be attributable to grain size effects rather than to
37 differences in provenance, and (3) higher mass accumulation rates in the Pampas are associated
38 with an increased presence of dust originated in the southern Altiplano and southern Puna during
39 the spans of 43-41 ka BP, 20-18 ka BP, 14.6-12.6 ka BP and 11.4-8.9 ka BP. We associate these
40 rises in continental dust fluxes with climatic transitions from wetter to drier periods in the Puna-
41 Altiplano Plateau related to synchronous climatic shifts to humid conditions at the Pampean Plain,
42 probably triggered by El Niño-like conditions. The isotopic comparison with modern SSA dust
43 indicates similar provenance compared to paleo-dust records, suggesting almost constant dust
44 sources from MIS 3 to modern times and/or modest changes in the geochemical signature under
45 the activation/deactivation of the different dust sources. Moreover, contrasting the isotopic
46 signature of the loess sections with more distal palaeoarchives (i.e., South Atlantic Ocean marine
47 sediment cores and Antarctic ice cores), the new data suggest that contrary to previous ideas, the
48 Pampean Loess was not an important source of dust to these regions. Also, a common dust
49 provenance during cold periods (e.g., Last Glacial Maximum and Antarctic Cold Reversal)
50 supports the idea that changes in atmospheric transport efficiency can better explain dust flux
51 variations observed over glacial/interglacial periods in distant palaeoarchives than changes in
52 provenance.

53

54 **Keywords**

55 Radiogenic isotopes, unmixing model, South America, Pampean loess, provenance, dust,
56 westerly, East Antarctica, Southern Ocean

57

58 **1. Introduction**

59

60 Atmospheric circulation is the major mechanism of heat transport on the Earth's surface, and
61 tracing windblown dust from sources to depositional locations is the best means available for
62 characterizing past wind circulation. Moreover, geological records of mineral dust enable the
63 reconstruction of past dust fluxes and are therefore key to assessing its impact on the Earth system.
64 The role of dust in forcing climate remains poorly understood and represents one of the largest
65 uncertainties in climate model simulations (Heavens et al., 2012; Lee et al., 2016; Adebisi and
66 Kok, 2020). For the modeling and observational communities, paleo-dust records represent a
67 large archive of information on the magnitude and spatial variability of dust deposition for the
68 pre-observational era, as well as a way to validate dust models under different climate scenarios
69 (e.g., Mahowald et al. 2006). Furthermore, clastic sediments have enormous potential to unravel
70 past geological processes. Dust deposits provide valuable clues on paleoenvironmental settings
71 such as continental aridity, glacial conditions, and dominant wind directions (e.g., An et al., 2001;
72 Soreghan et al., 2002; 2008; 2014) and may provide analogues for desertification due to the
73 ongoing global warming (e.g., D'Odorico et al., 2013). The provenance of wind-blown dust and
74 its characteristics are key issues to understand climate, atmospheric and environmental changes
75 (Lanci et al., 2020). Although sedimentary processes like weathering, erosion, transport,
76 sedimentation and diagenesis are likely to modify to some extent the composition of sediments,
77 it has been generally accepted that the chemical and isotopic compositions of sediments are still
78 dominantly influenced by their source rocks (e.g., Ahmad et al., 2016). Accordingly, the chemical
79 and isotopic compositions of paleo-dust records have been extensively used to decipher
80 provenance (e.g., Bhatia, 1985; McLennan et al., 1993; Biscaye et al., 1997; Grousset et al., 1988,
81 1992, 1998; Grousset and Biscaye, 2005; Sun et al., 2005, 2010).

82 The southern portion of South America (SSA) features a number of environmental
83 characteristics that allow the efficient production of fine particles: high relief, (semi)arid
84 conditions with intermittent fluvial activity (Prospero et al., 2002; Ginoux et al., 2012; Gaiero,
85 2007; Cosentino et al., 2020; Lamy et al., 2019), and increased glacier activity during glacial

86 intervals (Ammann et al., 2001; Zech et al., 2017). The rain shadow effects caused by the last
87 Andean uplift (i.e., since 26–28 Ma) created the ‘South American arid diagonal’ with average
88 annual precipitation of roughly 250 mm (Blisniuk et al., 2005). This is a long and narrow region
89 that extends from $\sim 2^\circ$ S in the Gulf of Guayaquil to $\sim 53^\circ$ S on the northern Tierra del Fuego
90 island, following the coast in Ecuador to northern Chile, crossing into Argentina north and east
91 of Santiago, and continuing southward through Patagonia. The major present-day dust source
92 areas in SSA are located in a continuous N–S band of arid and semi-arid terrains coinciding with
93 the arid diagonal (Prospero et al., 2002). Furthermore, SSA intersects two major zonal wind
94 systems of the Southern Hemisphere: the Southern Westerly Winds (SWW) and the SubTropical
95 Jet (STJ) (Fig. 1a). These expressions of the tropospheric circulation in the Southern Hemisphere
96 are a key component of the global climate system, affecting precipitation, the Antarctic
97 Circumpolar Current, and the global carbon cycle (e.g., Rojas, 2013; Toggweiler et al., 2006;
98 Lamy et al., 2001; Menviel et al., 2018). In particular, the SWW play an important role in mid-
99 and high-latitude atmosphere-ocean dynamics (Yuan et al., 2018), together with Southern Ocean
100 buoyancy (Watson et al., 2015). These atmosphere-ocean processes control the upwelling of
101 carbon-rich deep-water masses in the Southern Ocean, thereby affecting atmospheric CO_2
102 changes over glacial cycles (Anderson et al., 2009; Menviel et al., 2018) and the Holocene (Lamy
103 et al. 2010; Saunders et al. 2018).

104 Based exclusively on the Antarctic paleo-dust archive it is only possible to evaluate the
105 spatial and temporal variability of mass deposition rates far from sources (e.g., Lambert et al.,
106 2008). The attribution of this long-range paleo-dust to specific sources requires the chemical
107 and/or physical characterization of potential source areas (PSAs) of dust during the past (e.g.,
108 Gaiero, 2007; Gili et al., 2017; Scheuven et al., 2013; Chen et al., 2007; Aleinikoff et al., 2008).
109 Reconstructions of dust provenance in ice cores from the Southern Hemisphere benefit from the
110 geochemical differences between individual PSAs, revealing that SSA sources dominated the dust
111 supply to the East Antarctic Plateau (Gaiero, 2007; Sudgen et al., 2009; Gili et al., 2016, 2017,
112 2022 under review; Koffmann et al., 2021; DeDeckker et al., 2021) and the South Atlantic Ocean
113 (e.g., Noble et al., 2012; Anderson et al 2009; Peleari et al., 2019) during the last several glacial-

114 interglacial cycles. However, dust mass deposition rate variability in distal environments does not
115 depend only on changes in dust provenance, but also on changes in the intensity of emissions
116 from PSAs (Lambert et al., 2008; Delmonte et al., 2017) and in the intensity of the hydrologic
117 cycle along atmospheric dust transport pathways (Markle et al., 2018). To disentangle the
118 contributions of changes in dust provenance, dust emission intensity, and the hydrologic cycle
119 intensity, it is necessary to complement the characterization of distal paleo-dust with that of close-
120 to-source records.

121 In this contribution, our main objective is to constrain Late Quaternary SSA dust provenance
122 to improve the interpretation of coeval dust records located in middle to distal environments, thus
123 increasing knowledge on past atmospheric circulation over the Southern Hemisphere. To achieve
124 this, we present for the first time a systematic analysis of rare earth elements (REE), strontium
125 (Sr), neodymium (Nd) and lead (Pb) isotopes of Marine Isotope Stage 3 (MIS 3) to early Holocene
126 loess samples from three sites that together describe proximal, intermediate and distal Pampean
127 loess environments with respect to their PSAs. Furthermore, we revised the whole geochemical
128 data set representing dust PSAs associated with the main winds affecting atmospheric circulation
129 of SSA (i.e., SWW and STJ). We also considered other suggested sources (e.g., Zárate et al.,
130 2003) associated with more restricted atmospheric circulation, as for example, the low-surface N-
131 S circulation potentially deflating sediments from the Paraná River system, the Pampean Ranges,
132 or the shores of Mar Chiquita Lake. Our new geochemical data and unmixing model reveal
133 changes in the temporal and spatial distribution of fine dust deposited between MIS 3 and the
134 early Holocene, refining the existing knowledge of dust sources and wind circulation changes in
135 SSA.

136

137 **2. Background: The origin of Pampean loess**

138

139 Modern dust data single out the Pampean plains of SSA as a powerful dust sink (Cosentino
140 et al., 2020). This has also been the case during the Quaternary, as evidenced by the extensive
141 Pampean loess deposits across SSA (Zárate, 2003). Since the pioneering work of Teruggi (1957),

142 many researchers have been interested in understanding the Pampean loess origin (e.g., González-
143 Bonorino 1966; Iriondo, 1997; Krohling et al., 1999; Sayago et al., 2001; Zárata and Blasi, 1991,
144 1993; Zárata, 2003 and references therein). Based on mineralogical and/or stratigraphic features,
145 Patagonia, the Andean piedmont, the Pampean Ranges, Mar Chiquita Lake, and the Paraná River
146 Basin have been suggested as possible wind-borne sediment suppliers to the Pampean loess (e.g.,
147 González Bonorino 1965; Cantú and Degiovanni, 1984; Iriondo 1990; Cantú, 1992; Morrás 1997;
148 Krohling, 1999; Kirschbaum et al., 2000; Torre et al., 2020a). Apart from the source areas
149 previously mentioned, dust from the Altiplano region was also suggested to be entrained by slope
150 winds and carried aloft and eastward by the STJ, eventually being deposited across the eastern
151 plains (Bloom, 1990).

152 The first attempts to determine the provenance of the Pampean loess (~30-36° S, Fig. 1)
153 using geochemical data (e.g., REE and Sr-Nd isotopes) were made by Gallet et al. (1998) and
154 Smith et al. (2003). These works, however, lacked chronological control and hence, did not allow
155 for time-resolved provenance constraints. Also lacking a chronological framework, other studies
156 reported Sr-Nd isotopes (Delmonte et al., 2004) and Pb isotopic ratios (Gili et al., 2016) for
157 surface loess samples. Carter-Stiglitz et al. (2006) determined the first optically stimulated
158 luminescence (OSL) ages and Nd isotopes in loess-paleosols sequences located at lower latitudes
159 at the core of the Pampean loess (i.e., ~26°S) (Fig. 1b), where the main atmospheric circulation
160 depends on the STJ (Smith et al., 2003). The first age model of Pampean loess was that of Torre
161 et al. (2019), which allowed time-resolved provenance determinations of the western Pampean
162 loess through REE (Torre et al., 2020a), and of an E-W Pampean loess transect based on grain
163 size characteristics (Torre et al., 2020b).

164 A primary conclusion from previous studies on the Pampean loess origin is that efforts
165 should be focused on the mineralogical/chemical/isotopic characterization of PSAs of SSA in
166 order to improve loess provenance studies. In this sense, Gili et al. (2016, 2017) have
167 characterized more than 50 samples from PSAs of SSA through Sr, Nd, Pb isotopes and REE with
168 the aim of constraining the provenance of paleo-dust records in the Southern Hemisphere. They
169 collected topsoil samples from desert terrains along a 4000-km-long latitudinal transect, which is

170 perpendicular to the main zonal atmospheric circulation (e.g., SWW and STJ) and includes the
171 main sources of dust (i.e., Patagonia, the Andean piedmont in central-western Argentina and the
172 Puna-Altiplano Plateau) to downwind settings such as the Pampas, the South Atlantic Ocean, the
173 Southern Ocean and the East Antarctic Plateau. Moreover, geochemical data from other PSAs
174 mentioned in the literature (e.g., Pampean Range rocks, Paraná suspended sediments, Mar
175 Chiquita Lake sediments) are now included in the discussion about the origin of Pampean loess.

176

177 **3. Sample locations and analytical procedures**

178

179 *3.1 Regional setting and loess sampling*

180

181 The Pampean plains are located to the east of the Andes and are bounded to the west by the
182 foothills of the Pampean Ranges and to the east by the South Atlantic Ocean (Fig. 1b). We based
183 our sampling approach on previous age models (Kemp et al., 2004; 2006, Zárate et al., 2009)
184 published for three loess sections across the Pampean loess belt, which we used to perform a 1-
185 m vertical sampling centered at a depth corresponding to the previously reported Last Glacial
186 Maximum (LGM, ~19-26 ka BP) for each site. Unfortunately, new ages have been determined
187 for the new samples with significant differences compared to the previously published ones, and
188 we now know that cover together a time span from MIS 3 to the early Holocene, with only one
189 site capturing the LGM (see the discussion in Torre et al., 2019).

190 Loess sampling for isotopes and REE was performed using thin-walled plastic cylindrical
191 corers driven into the face of the sections at an average resolution of ~2.3 cm which is controlled
192 by the external diameter of the plastic cylinders (see Supplementary Material, S1). The age of
193 each sampled level for isotopes and REE determination was assigned through linear interpolation
194 considering the mass accumulation rates for each dated interval (Torre et al., 2019). Numerical
195 ages obtained by OSL methods are reported in Torre et al. (2019) (Supplementary Material, S2).
196 The westernmost studied profile is Lozada (Lz, ~31°39'S; 64°08'W) (Fig. 1). This site is the
197 closest to SSA's main dust sources to the west and is located in the foothills of the Pampean

198 Ranges, occupying a gentle slope (<3–5%) dissected by rivers and streams that drain the
199 mountains. From the total of 9.3-m of vertical outcrop, we sampled the section between 2.8 to 1.8
200 m below the surface. This loess section was deposited during the transition from the last
201 deglaciation to the early Holocene, between ~16.3 ka and 8.9 ka BP (Torre et al., 2019). The
202 Tortugas site is located 250 km SE of Lozada and represents the type section for the loess of the
203 central Pampas (Tr, ~32°43'S; 61°48'W) (Fig. 1). Here we sampled a 1-meter section between
204 2.2 and 1.2 m depth below the surface, deposited during the last deglaciation, between 19.5 ka
205 and 12.6 ka BP (Torre et al., 2019). The easternmost profile is exposed at the Gorina quarry (Gn,
206 ~34°54'S; 58°01'W) (Fig. 1), which has a more complex regional stratigraphic sequence (Tonni
207 et al., 1999) alternating between discrete loess and paleosol units (Zárate et al., 2002). The 1-
208 meter sampled section (e.g., between 1.6 and 0.6 m below the surface) was deposited during mid-
209 MIS 3 and the late LGM, between 47 ka and 22.1 ka BP (Torre et al., 2019).

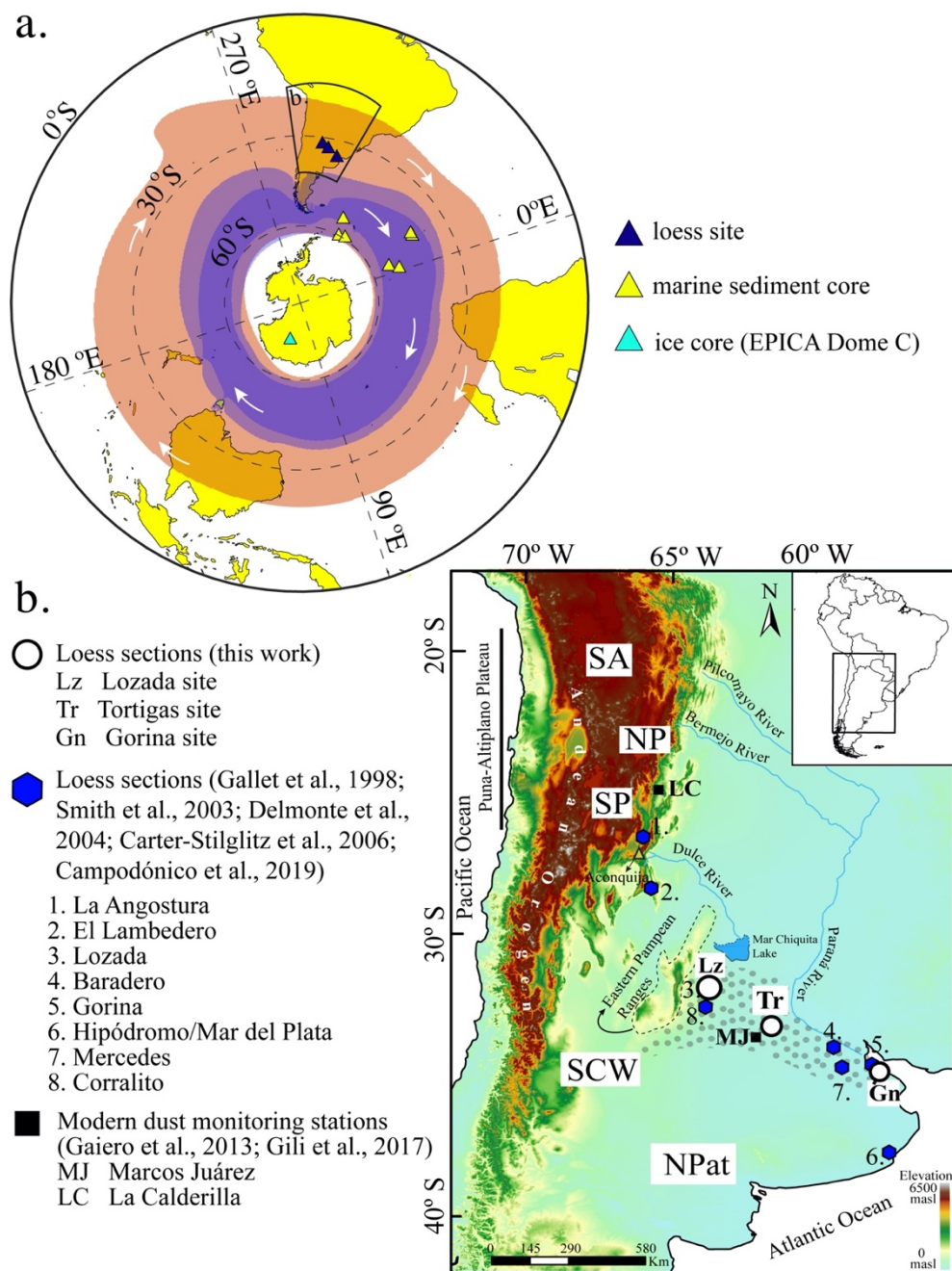


Figure 1. (a) Map showing the location of dust records and main wind belts from the Southern Hemisphere discussed in this paper. The lighter and darker blue-purple areas are delimited by the 7 m s^{-1} and 10 m s^{-1} contours of the E-W wind component at 850 hPa, respectively, and represent the South Westerly Winds. In turn, the reddish area is delimited by the 18 m s^{-1} contours of the E-W wind component at 200 hPa, and represents the SubTropical Jet. In all cases, contours represent the 1979-2020 climatological means calculated from monthly ERA5 reanalysis data. White arrows indicate general wind direction. (b) Map showing the location of the three loess sections studied in this work (white dots): Lz, Lozada; Tr, Tortugas and Gn, Gorina. The dotted area indicates the limits of the “loessic/loessoid” or “loess belt” sector of the Pampean region. The black squares are the present-day dust monitoring stations. Blue symbols indicate previously studied loess sites with available geochemical data.

211 3.2 Grain size separation

212

213 Highly time-resolved grain size analysis of loess samples reveals that loess deposited during
214 the late Pleistocene-early Holocene has multimodal distributions consistent with the existence of
215 multiple source areas and/or transport pathways (Torre et al., 2020b). When isotopic analyses are
216 made on bulk aeolian sediments (finer than 75 μm), the isotopic signature could be the result of
217 a mixture of sources with different grain sizes. To overcome this limitation, 10 loess samples
218 from Lozada and 10 from Tortugas were separated by a 44- μm sieve. The sieve diameter was
219 selected such that the fine and coarse grain size subpopulations were fully separated. Loess
220 samples from Gorina section were not split as they consist mainly of fine sediments (Torre et al.,
221 2020b).

222

223 3.3 Isotopic analysis

224

225 Sediments deposited in different environments are composed of mixtures of materials
226 (subpopulations) from different sources and/or transported by different mechanisms (Weltje,
227 1997 and references therein). In a previous work (Torre et al., 2020b), it was concluded that loess
228 samples from the Pampean region have multimodal grain size distributions consistent with the
229 existence of multiple source areas and/or atmospheric transport pathways. Following this idea, in
230 the present work we analyzed the isotopic composition of loess samples in two main grain size
231 subpopulations, in order to investigate possible isotopic changes according to changes in the
232 source areas and/or in the regional atmospheric circulation. For this purpose, fifty samples from
233 three loess records were analyzed for Nd and Sr isotopes and 18 for Pb isotopes. For Lozada and
234 Tortugas, isotopic analyses were performed for 10 fine (<44 μm) and 10 coarse (>44 μm) samples,
235 and 10 bulk samples were analyzed for Gorina. In order to better characterize the dust available
236 for long-range transport, Pb isotope ratios were analyzed on six and seven fine (<44 μm) samples
237 from Lozada and Tortugas, respectively, and five bulk samples from Gorina.

238 Prior to samples digestion, an aqueous solution with ~3% H₂O₂ was used to eliminate
239 organics, followed by 1 M HCl leach (overnight) to remove carbonates, then samples were
240 washed with Milli-Q water and dried. After this procedure, an aliquot of 100 mg of each sample
241 was dissolved by tri-acid attack (HF-HNO₃-HCl) in Teflon Savillex[®] containers. Once in solution,
242 Pb, Sr, and Nd were separated from the isotope aliquot using a sequence of ion chromatographic
243 columns. We used Eichrom AG1-X8[®] 100-200 mesh anion exchange resin (for Pb), Eichrom
244 TRU[®] resin (to separate REEs from major cations), Eichrom Sr[®] resin, and Eichrom LN[®] resin
245 (for Nd).

246 Neodymium and Sr isotopic composition was determined by Thermal Ionization Mass
247 Spectrometry (TRITON[™] Plus) at the *Observatoire Midi-Pyrénées* (Toulouse, France). Isotope
248 ratios were measured in dynamic mode, corrected for instrumental mass bias using $^{88}\text{Sr}/^{86}\text{Sr} =$
249 0.1194 and $^{146}\text{Nd}/^{144}\text{Nd} = 0.7219$. Chemical blanks for Sr and Nd isotopic measurements were
250 below the detection limit. Pb isotope ratios were measured on a ThermoScientific NeptunePlus
251 Inductively Coupled Plasma-Mass Spectrometer (ICP-MS) at Lamont-Doherty Earth
252 Observatory (New York, USA). We used an Elemental Scientific APEX desolvator for Pb
253 isotopic analyses. Measurements used Faraday static mode with 60 ratios per sample, with ~200
254 ppb Pb in 3% HNO₃ solutions, and ^{208}Pb beam were intensities between ~1.3 to 9×10^{-11} A. Pb
255 samples were spiked with thallium (Tl) to achieve a Pb/Tl ratio of ~5.4. Tl isotopes were used to
256 correct for mass fractionation using the exponential fractionation law and assuming $^{203}\text{Tl}/^{205}\text{Tl} =$
257 0.41844 (Thirlwall 2002). Pb isotopes were measured at concentrations of 200 ng mL^{-1} , in 3%
258 nitric acid solution.

259 All the isotopic data are corrected using the following values for international reference
260 materials: NBS 987 for $^{87}\text{Sr}/^{86}\text{Sr}$; La Jolla for $^{143}\text{Nd}/^{144}\text{Nd}$ and BCR-2 for $^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$,
261 and $^{208}\text{Pb}/^{204}\text{Pb}$ ratios. These same reference materials bracketed each measured sample to assess
262 instrumental drift and fractionation, and to determine data reproducibility. They were measured
263 several times per run, yielding the following 2σ external errors. For $^{87}\text{Sr}/^{86}\text{Sr}$, two runs gave
264 external errors of ± 0.000009 . For $^{143}\text{Nd}/^{144}\text{Nd}$, the reproducibility varied during two measurement
265 intervals with external errors of ± 0.000017 . For $^{206}\text{Pb}/^{204}\text{Pb}$, three runs gave external errors of

266 0.00017, 0.00019 and 0.00013; for $^{207}\text{Pb}/^{204}\text{Pb}$, 0.00017, 0.00020 and 0.00014; for $^{208}\text{Pb}/^{204}\text{Pb}$,
 267 0.00044, 0.00048 and 0.00040; for $^{207}\text{Pb}/^{206}\text{Pb}$, 0.000008, 0.000007; and for $^{208}\text{Pb}/^{207}\text{Pb}$, 0.000003
 268 and 0.000004. Our measured NBS 987 $^{87}\text{Sr}/^{86}\text{Sr}$ is 0.710213 ± 0.000009 ($n=3$, 2σ). We also
 269 measured the La Jolla Nd standard twice, which gave $^{143}\text{Nd}/^{144}\text{Nd} = 0.511869 \pm 0.000017$. These
 270 values are within the 2σ error envelopes of the preferred values for these standards. Our measured
 271 BCR-2 Pb isotope values of $^{206}\text{Pb}/^{204}\text{Pb} = 18.7555 \pm 0.0001$, $^{207}\text{Pb}/^{204}\text{Pb} = 15.6216 \pm 0.0001$, and
 272 $^{208}\text{Pb}/^{204}\text{Pb} = 38.7107 \pm 0.0127$ ($n = 6$, 2σ), are within the 2σ error envelopes of unleached BCR-
 273 2 values: $^{206}\text{Pb}/^{204}\text{Pb} = 18.7529 \pm 0.0195$, $^{207}\text{Pb}/^{204}\text{Pb} = 15.6249 \pm 0.0040$, and $^{208}\text{Pb}/^{204}\text{Pb} =$
 274 38.7237 ± 0.0004 , thus giving us confidence in the accuracy of our measurements.

275 For convenience, Nd isotope data are also reported here in units of $\epsilon\text{Nd} = [(^{143}\text{Nd}/^{144}\text{Nd}_{\text{measured}}$
 276 $/ ^{143}\text{Nd}/^{144}\text{Nd}_{\text{CHUR}}) - 1] * 10^4$, where in this work we use $^{143}\text{Nd}/^{144}\text{Nd}_{\text{CHUR}} = 0.512638$ (Jacobsen
 277 and Wasserburg, 1980), rather than updated values (Bouvier et al., 2008) in order to be consistent
 278 with decades of published Nd isotope data. Also, for the three loess sites, grain size distributions
 279 were obtained for bulk samples (Torre et al., 2020b), which permits the calculation of a bulk
 280 isotopic composition that allows comparison between the isotopic signature of the three loess
 281 sections. For Tortugas and Lozada we averaged the isotope ratios of each grain size fraction for
 282 every analyzed sample, where fine and coarse isotopic ratios were previously multiplied by the
 283 relative abundance of each fraction.

284

285 *3.4 Major and trace elements*

286

287 Bulk loess samples from the Pampean Plain are finer than $75 \mu\text{m}$ (Torre et al., 2020b). A
 288 total of 61 bulk samples from the three studied loess sections were analyzed for REE.
 289 Determinations of REE for 22 samples from the Lozada section are reported in Torre et al.
 290 (2020a), while determinations for the samples of the other two sections (i.e., Tortugas and Gorina)
 291 are first reported in this study. Approximately 100 mg of each sample was introduced in carbon
 292 crucibles and were exposed to sodium peroxide (Na_2O_2) attack for 0.5 h in an oven at 490°C
 293 (Meisel et al., 2002). Once the fusion of the samples was achieved, they were taken into solution

294 by means of the addition of ultrapure HNO₃. The concentrations of REE in were determined by
295 ICP-MS Agilent 7500cx at National University of Córdoba (Córdoba, Argentina). Repeated
296 analyses (n = 7) of the AGV-2 and GSP-2 (USGS) rock standards show significant accuracy of
297 values to be on the order of 3%. The Eu anomaly (Eu/Eu*) was calculated as follows:
298 $Eu_N / (Sm_N \cdot Gd_N)^{0.5}$ (McLennan and Xiao, 1998), where the subindex N represents the element
299 normalized to the North American Shale Composite (NASC) (Wu et al., 2020). Sixteen loess
300 samples were dissolved by the alkaline fusion method (Li₂B₄O₇, 1050 °C, with HNO₃ dissolution)
301 and analyzed for major elements (i.e., P₂O₅ and TiO₂) by inductively coupled plasma with optical
302 emission spectroscopy (ICP-OES) at Lamont-Doherty Earth Observatory (New York, USA).

303

304 *3.5 Source end-member mixing calculation*

305

306 To quantify the contributions of dust from various sources, the isotope mixing model
307 SIMMR was used (Parnell, 2020). This model solves mixing equations for isotope data using a
308 Bayesian framework. Stable isotope mixing models have been widely used in zoology to provide
309 probabilistic estimates of source contributions to dietary mixtures (e.g., Kadye et al., 2020).
310 Recently, Pb isotope tracers gave satisfactory results using a Bayesian mixing model to analyze
311 dust provenance from the Pacific Ocean (Erhardt et al., 2020). This mixing model can be used to
312 determine the isotopic contribution of sources in a sediment mix and determine relative
313 contributions to the sediment provenance. For n isotopes, a unique solution can be calculated for
314 up to n+1 sources. The package SIMMR allows for the possible contribution of additional sources,
315 using an iterative approach to calculate all possible feasible solutions for any given sources and
316 isotopic mixtures, generating a distribution of feasible solutions (Inger et al., 2006). End members
317 were chosen as those regions likely contributing dust to the Pampean loess based on previous
318 isotopic studies (Gili et al., 2017; Gaiero et al., 2013).

319

320 **4. Results**

321

322 The REE concentrations of bulk (i.e., <75 μm) loess samples are listed in Table S1. Figure
323 2a-c shows REE loess composition normalized to the North American Shale Composite (NASC)
324 (Wu et al., 2020). The loess samples from the three sections show similar flat REE NASC-
325 normalized patterns, with positive Eu anomalies and negative Ce anomalies. The spider diagrams
326 indicate that aeolian sediments from Tortugas (Fig. 2b) have homogeneous temporal REE
327 compositions. On the other hand, loess samples from Lozada (Fig. 2a) and Gorina (Fig. 2c) show
328 greater variability. The three sites show similar mean values for La/Yb: 0.83 for Lozada, 0.88 for
329 Tortugas and 0.77 for Gorina, indicating slightly enrichments of heavy REEs compared to NASC.
330 The UCC-normalized europium anomaly (Eu/Eu^*) averages 1.13 ± 0.06 , 1.20 ± 0.05 and $1.21 \pm$
331 0.13 for the <75 μm (bulk) fraction of Lozada, Tortugas and Gorina samples, respectively.

332 The radiogenic isotope data is shown in Table S2. The data indicates that Tortugas' $^{87}\text{Sr}/^{86}\text{Sr}$
333 is the highest in the fine (<44 μm) fraction ranging between 0.7080 and 0.7089, with an average
334 value of ~ 0.7083 (Fig. 2d). For the same grain size fraction, the Lozada samples are slightly less
335 radiogenic, ranging between 0.7078 and 0.7088 and with an average value of ~ 0.7081 (Fig. 2d).
336 It is observed that for increasing loess particle size the $^{87}\text{Sr}/^{86}\text{Sr}$ is lower (Fig. 2e), this being more
337 evident in particular for the Tortugas samples. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios for the Tortugas coarse fraction
338 range between 0.7073 and 0.7076 with an average of ~ 0.7075 , which is slightly lower than data
339 from Lozada, which varies between 0.7077 and 0.7084, with an average ratio of 0.7081. The ϵNd
340 values of the fine-grained Tortugas sediments range between -5.82 and -4.33, with an average of
341 -5.08 ± 0.46 , while the coarse-grained sediments range between -4.95 and -2.92 with an average
342 of -3.99 ± 1.45 (Fig. 2e). Comparatively, ϵNd data for Lozada are slightly lower, showing fine
343 loess ranging between -5.77 and -3.10 (average of -4.65 ± 1.73), while coarse sediments vary
344 between -4.72 and -2.86 (average of -3.87 ± 1.45). For the three sections, the bulk isotopic
345 composition of loess is quite similar, with $^{87}\text{Sr}/^{86}\text{Sr}$ (ϵNd) averages for Lozada, Tortugas and
346 Gorina of 0.7081 (-4.36), 0.7079 (-4.58) and 0.7080 (-4.41), respectively (Fig. 2f).

347 The fine loess fractions from Lozada and Tortugas and bulk loess from Gorina display
348 $^{208}\text{Pb}/^{207}\text{Pb}$ between 2.47-2.49 and $^{206}\text{Pb}/^{204}\text{Pb}$ between 1.195-1.205 (Fig. 2g-h). Unlike Sr and Nd,

349 Pb isotopes show a distinguishable signature among the loess samples from the three sites,
 350 showing a NW-SE trend of higher to lower values from Lozada to Gorina.

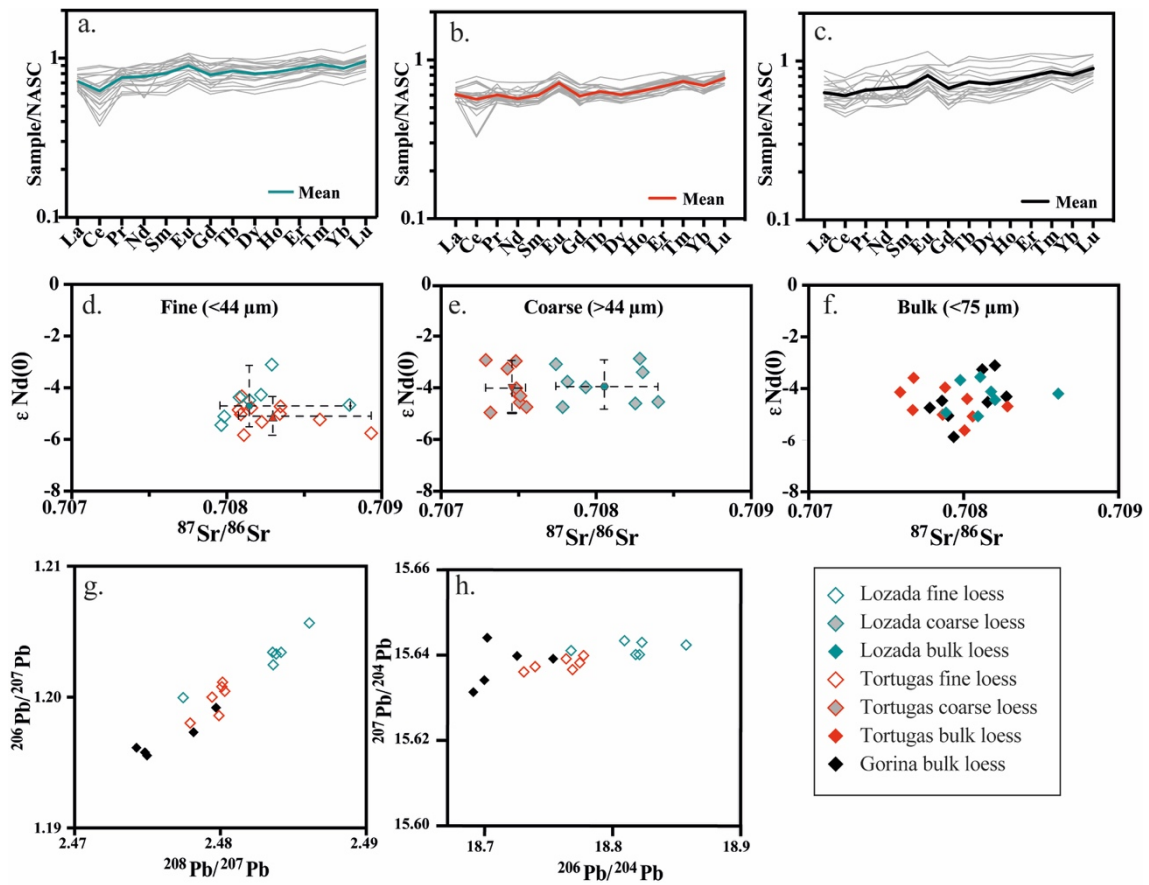


Figure 2. Rare Earth Element (REE) concentrations normalized to North American Shale Composite (NASC) (Wu et al., 2020) (a) Lozada, (b) Tortugas and (c) Gorina loess sections. $^{87}Sr/^{86}Sr$ and ϵ_{Nd} results are shown for (d) fine fractions of Lozada and Tortugas sections, (e) coarse fractions of Lozada and Tortugas and (f) bulk fractions of Lozada, Tortugas and Gorina loess sections. Lead isotope ratios for fine fractions (<44 μm) of Lozada and Tortugas sections, and bulk (<75 μm) sediments from the Gorina profile (g-h).

351

352 5. Discussion

353

354 5.1. Rare earth elements as provenance tools

355

356 Rare earth elements have been widely used in provenance studies of atmospheric dust (e.g.,
 357 Gallet et al., 1996, 1998; Humphries et al., 2020; Jahn et al., 2001; Pease and Tchakeria, 2002;
 358 van der Does et al., 2018; Zdanowicz et al., 2006; Campodonico et al., 2019). The importance of

359 this group of elements for sediment provenance studies lies in the fact that they are relatively
360 immobile during physical and chemical processes, serving as important markers in tracing
361 material sources and for characterizing the geochemical evolution of various systems (Khan et
362 al., 2017; Gwenzi et al., 2018). For distinguishing different aeolian dust sources, REE ratios are
363 more diagnostic than individual concentrations, as the former do not depend on dilution effects
364 of certain minerals (Ferrat et al., 2011).

365 Although the compositional field of most dust PSAs show good differentiation between
366 them, there is considerable overlapping between potential sources in the Paraná River basin,
367 southern central-western Argentina (S-CWA) and the Puna Plateau, thus precluding a distinctive
368 interpretation for the Pampean loess provenances (Fig. 3). The whole REE data set indicate
369 similar geochemical composition for the three loess sections (Fig. 2a-c, Fig. 3), and in agreement
370 with published data (i.e., Gallet et al., 1998; Smith et al., 2003; Campodónico et al., 2019),
371 pointing out that multiple sources could have supplied dust to the Pampean region between 47 ka
372 to 9 ka BP. Contrasting the geochemical composition of Lozada and the nearby Corralito section
373 (Campodónico et al., 2019) with those more local PSAs (e.g., shore sediments from Mar Chiquita
374 Lake, and riverine and lake sediments from the Pampean Ranges), Torre et al. (2020a) and
375 Campodónico et al. (2019) concluded that these sources were not significant dust contributors in
376 the past, pointing out that the Pampean loess of these localities derives mainly from Andean
377 sources. In the next section we further discuss loess provenance using isotopic data.

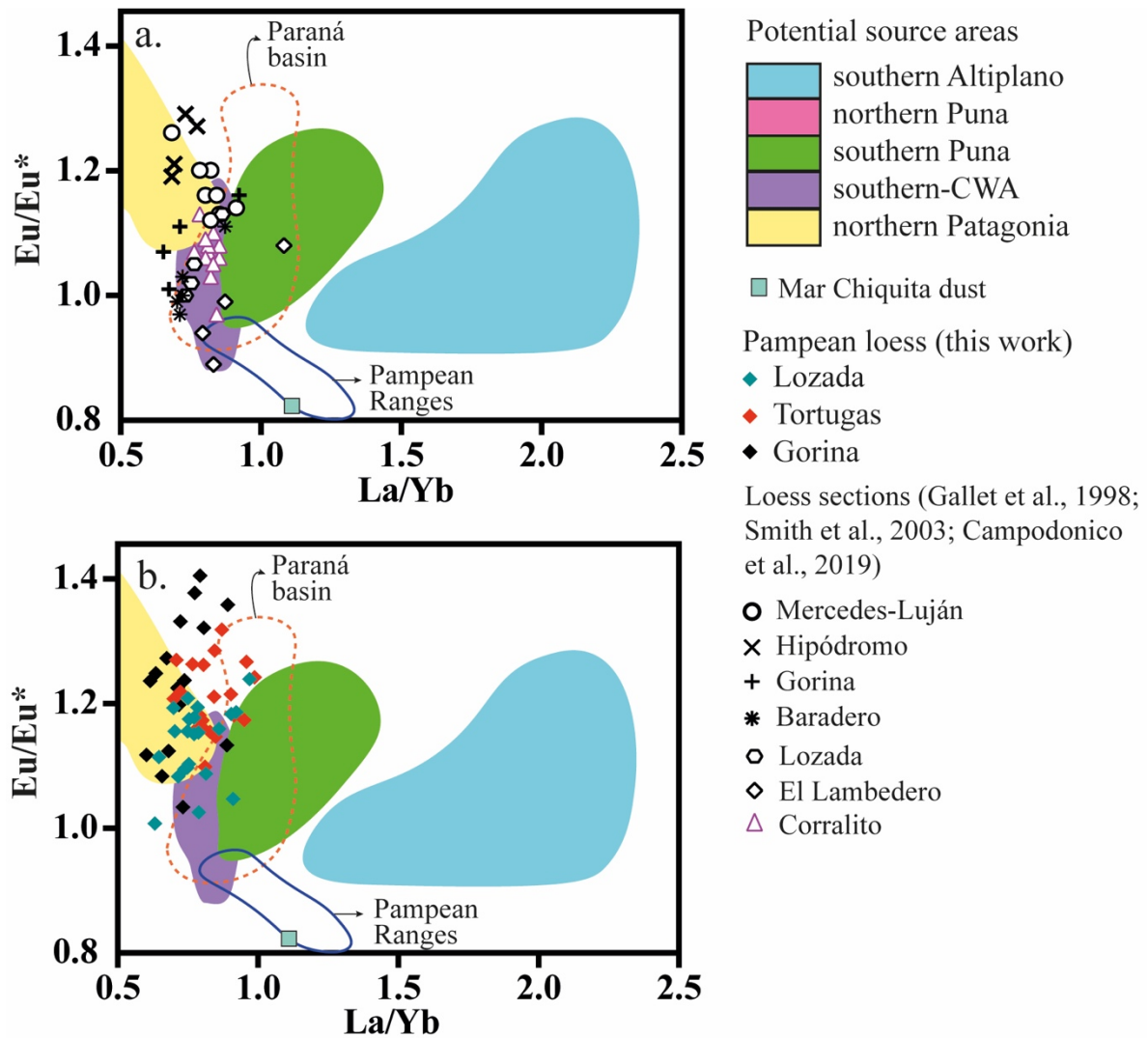


Figure 3. Rare earth element (REE) ratios characterizing dust sources from the Arid Diagonal (colored areas, Gili et al., 2017), the Paraná River basin (Campodonico et al., 2016; Depetris et al., 2003), Mar Chiquita Lake and eastern Pampean Ranges (Torre et al., 2020a). These data are compared with (a) previously published La/Yb vs Eu/Eu* ratios of loess samples (Gallet et al., 1998; Smith et al., 2003; Campodónico et al., 2019) and (b) ratios from the Pampean loess for the Lozada, Tortugas and Gorina sites; white diamonds indicate average values for the three loess sections (Torre et al., 2020a and this work). All samples here are bulk fractions, represented by grain size fraction >75 μm .

378

379 5.2 Sr and Nd isotopes applied to the provenance of the loess

380

381 Previous studies employed Sr and Nd isotopes to analyze the provenance of loess from
 382 different localities distributed in a wider region of the Pampas (Fig. 1b). Our new loess dataset
 383 allow for an update of previous observations made by Smith et al. (2003) and Gaiero (2007)
 384 indicating a general N-S trend for the mean ϵNd values recorded in loess samples from different
 385 latitudes (Fig. 4a). Compared with surface sediments from the PSAs, Nd isotopic data from

386 previous studies and this work show a clear N-S trend composition between a northern
387 Patagonia/S-CWA end-member and a Puna-Altiplano Plateau (PAP) end-member (Fig. 4a-b).
388 That is, the most southerly loess sites (i.e., Hipódromo and Mar del Plata), show a clear
389 Patagonian signal, while the most northerly loess sites (i.e., El Lamedero and La Angostura)
390 show a much more radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ and a less radiogenic ϵNd signature than samples located
391 to the south, both supporting a PAP origin. Interestingly, the loess Nd isotope ratios for samples
392 at intermediate latitudes (i.e., from 31°S to 36°S) do not strictly follow the N-S trend of a more
393 negative (positive) tendency towards the north (south), suggesting that multiple sources seem to
394 explain the loess provenance in the loess belt (Fig. 1b).

395 Combining our new data from all three sites and all grain size fractions, the average isotopic
396 values of 0.7080 ± 0.0114 for $^{87}\text{Sr}/^{86}\text{Sr}$ and -4.45 ± 0.84 for ϵNd are similar to values in loess
397 reported previously: 0.7084 ± 0.0008 for $^{87}\text{Sr}/^{86}\text{Sr}$, and -3.44 ± 1.11 for ϵNd (Smith et al., 2003);
398 and 0.7072 ± 0.0013 for $^{87}\text{Sr}/^{86}\text{Sr}$ and -2.96 ± 1.98 for ϵNd in bulk sediments (Gallet et al., 1998).
399 For the case of the $<5 \mu\text{m}$ samples, average values are higher at 0.7099 ± 0.0010 for $^{87}\text{Sr}/^{86}\text{Sr}$ and
400 -2.02 ± 1.11 for ϵNd (Delmonte et al., 2004) (Fig. 4b and c). Although no age control is reported
401 for most of the previously published isotopic data, these data are very useful for improving loess
402 provenance evaluations. Compared to the published data set, our new data have a small range of
403 Sr and ϵNd isotopic variability (Fig. 4 c-d). The visual analysis of the new data set indicates that
404 multiple sources from SSA have likely contributed to all studied loess sites. At a first glance, the
405 Sr and Nd isotopic data allow us to discard northern Puna and the Paraná basin as significant dust
406 sources for the period under study (Fig. 4b), unlike the REE. Although some rocks from the
407 Pampean Range (e.g., amphibolite) have isotopic compositions similar to northern Patagonian
408 sediments and loess, the average composition from these rocks and the scarce abundance of
409 amphibolites within the Pampean Ranges (e.g., Toselli et al., 1986), do not support a significant
410 role as dust source areas to the Pampean region. Moreover, the low-level atmospheric circulation
411 over this mountainous sector is associated to N-S wind systems, arguing against any efficient
412 mechanism of dust transport to the eastern loess sites (e.g., Tortugas and Gorina). A significant
413 number of the loess samples have intermediate isotopic compositions aligned to a compositional

414 trend defined by the S-CWA/northern Patagonia and southern Altiplano end members (Fig. 4d).
415 Based on Nd isotopic ratios of loess, the role of southern Puna as a source of dust to the Pampas
416 cannot be ruled out considering that many loess samples show similar ϵNd ranges (i.e., -4.9 to -
417 2.8).

418 Notably, the fine fractions of loess from Lozada and Tortugas have similar isotopic
419 compositions, suggesting a common origin (Fig. 4d). This figure also shows that the fine
420 sediments from Tortugas and, to a lesser extent from Lozada, have less (more) radiogenic ϵNd
421 ($^{87}\text{Sr}/^{86}\text{Sr}$) ratios than its coarser fractions. This isotopic difference could be linked to changes of
422 provenance or may be due to a grain size effect. The latter is often observed in fine sediment
423 fractions showing high $^{87}\text{Sr}/^{86}\text{Sr}$ ratios due to higher values in minerals that tend be enriched in
424 fine fractions and weathering effects (Clauer, 1979; Blum et al., 2003). The observed differences
425 in $^{87}\text{Sr}/^{86}\text{Sr}$ ratios between fine and coarse Tortugas and Lozada loess (i.e., average differences of
426 0.0002 and 0.00008 respectively) is smaller than the variation indicated as a consequence of the
427 grain size effect for similar sediment fractions (i.e., difference of ~ 0.0005 between fractions
428 coarser and finer than $50\ \mu\text{m}$) (Feng et al., 2009). Thus, changes in the isotopic ratios of Sr of
429 loess samples could be associated with wind-sorting effects and/or chemical weathering.

430 Figure 4d shows that the Tortugas samples have greater $^{87}\text{Sr}/^{86}\text{Sr}$ offsets between loess
431 fractions compared to the Lozada samples. Accordingly, Tortugas loess has greater relative
432 abundance of the fine ($<44\ \mu\text{m}$) fraction (i.e., average 77%), compared to the same grain size
433 fraction observed in Lozada (i.e., average 56%) (Torre et al., 2020b). Therefore, the Sr isotopic
434 data supports the existence of a grain size effect in the Sr isotopic signal of the loess. This is less
435 apparent in Nd isotopic ratios, which are less affected by sorting effects (Goldstein et al. 1984;
436 Yokoo et al., 2004; Yang et al., 2005). The mean ϵNd difference between coarse and fine fractions
437 is less than 1.1 ϵ unit, in agreement with the observed small differences of $\sim 1\text{--}2\epsilon$ units between
438 clay and silt for grain size fractions in river sediments (e.g. Bayon et al., 2015). In conclusion,
439 although the bimodal grain size distributions in loess samples have been previously suggested to
440 be linked to multiple sources (Torre et al., 2019), the isotopic difference between the finer and

441 coarser fractions is not significant enough to be considered a consequence of variations in
 442 provenance.

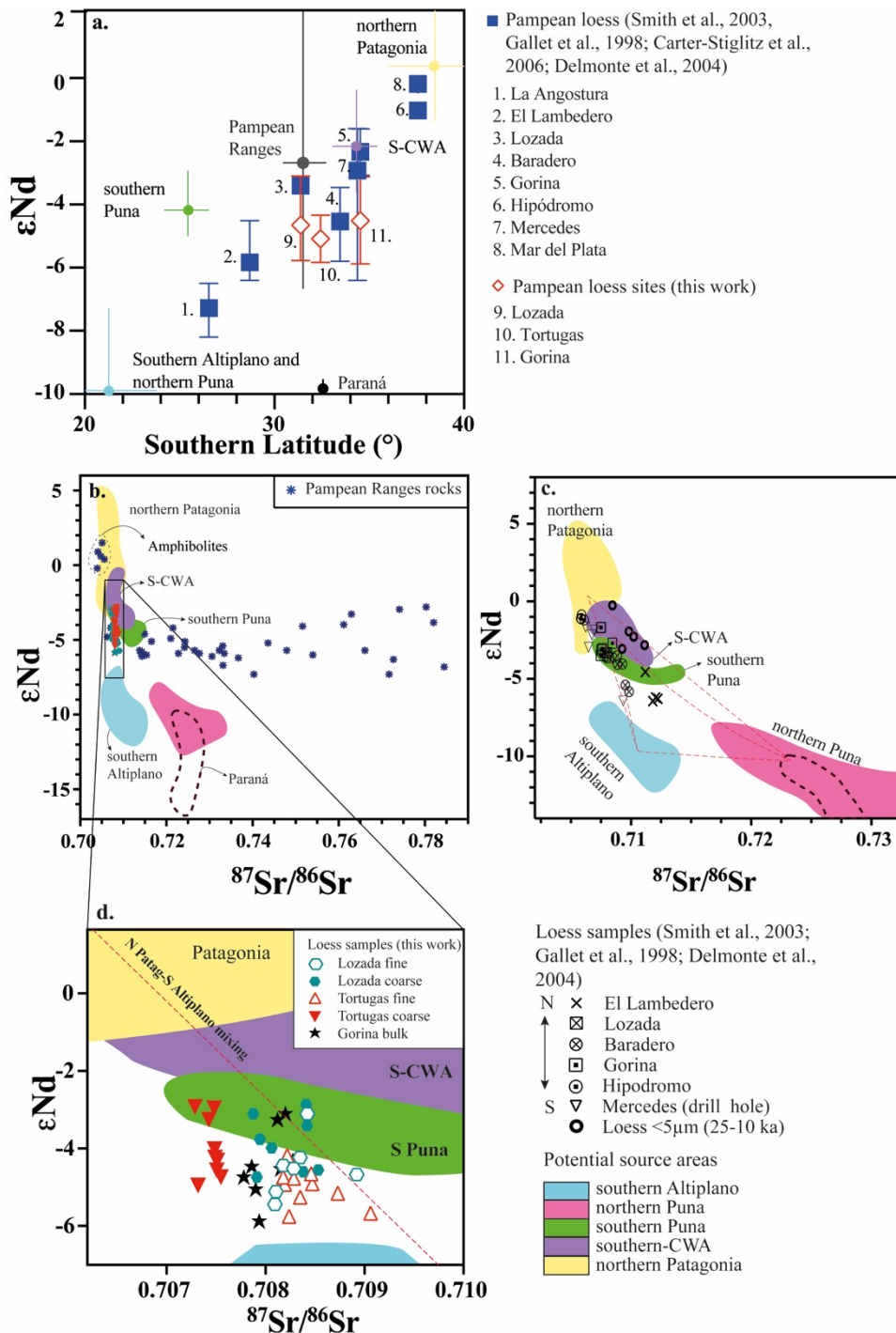


Figure 4. (a) Latitudinal ϵNd variation of Pampean loess. (b-d) Colored areas indicate the isotopic fingerprint of southern South American dust sources from the arid diagonal (Gili et al., 2017), Paraná river sediments (Henry et al., 1996) and outcropping rocks from the Pampean Ranges (Rapela et al., 1998; Pankhurt et al., 1998; Grosse et al., 2002). These potential source areas are compared to: (c) previously published Sr and Nd isotope ratios of Pampean loess (Smith et al., 2003; Gallet et al., 1998; Delmonte et al., 2004), and (b and d) MIS 3-early Holocene Pampean fine ($<44 \mu m$) and coarse ($>44 \mu m$) loess from the Lozada and Tortugas sites, and bulk loess from the Gorina site (this work). Red-dashed lines are mixing lines between potential source areas calculated in Gili et al., (2017).

444 5.3. *Pb isotope ratios applied to the provenance of loess*

445

446 In contrast to the Sr and Nd isotopic pair, the grain size dependence of Pb isotopes is not
447 well-documented, showing inconsistent results (e.g., Pettke et al., 2000; Unruh, 2001; Grousset
448 and Biscaye, 2005; Feng et al., 2010). Gili et al. (2016) showed that the differences between Pb
449 isotope ratios for the fine (<5 μm) and coarse (<63 μm) grain size fractions are small for most of
450 the PSAs from SSA and surface Pampean loess samples. The small differences observed between
451 the Pb isotopic compositions of the fine and coarse fractions of sediments from PSAs means that
452 sorting by size allows for attempts to relate the Pb isotope data in loess to the different PSAs. In
453 general, the Pb isotopic compositions of modern sediments in central and eastern SSA is
454 suspected to be anthropogenically contaminated by industrial emissions and leaded petrol
455 (Vallelonga et al., 2010). Thus, Pb isotopes in surface sediments from potential dust sources
456 representing Mar Chiquita Lake, and the Paraná basin and rivers draining the Pampean Ranges,
457 cannot be used for loess provenance analyses. These areas in any case have been found to be
458 insignificant sources of dust to the Pampean loess. Therefore, the following discussion involves
459 only Pb isotopic data of PSAs whose surface sediments are not suspected of anthropogenic
460 contamination.

461 Previously published Pb isotopes from loess samples showed compositions similar to S-
462 CWA and southern Puna, indicating a latitudinal trend from more to less radiogenic values
463 southward (Fig. 5a-b) (Gili et al., 2016). Data also indicate that fine-grained samples are less
464 radiogenic compared to the coarse fraction (<63 μm) and bulk material (i.e., Gili et al., 2016;
465 Vallelonga et al., 2020), suggesting a grain size dependence on Pb isotopes.

466 Gili et al (2016), demonstrated that the differences between Pb isotope ratios for the fine and
467 coarse grain size fractions are small for most of the SSA samples. Based on this conclusion, we
468 focus the analysis of the Pb isotopic ratios in the fine loess fractions (i.e., <44 μm) (i.e., Lozada
469 and Tortugas), and the bulk fraction from the Gorina site. In term of $^{206}\text{Pb}/^{207}\text{Pb}$ and $^{206}\text{Pb}/^{204}\text{Pb}$,
470 Figure 5c-f shows the existence of a gradient from more radiogenic (Lozada) to less radiogenic
471 (Gorina) isotope ratios. Data from Tortugas show intermediate values, partially overlapping with

472 values from the two other sections. The whole dataset, including data from Gili et al. (2016) and
 473 Vallelonga et al. (2010) (Fig. 5a-b, Fig. 5d-f), define a linear trend along the compositional fields
 474 represented by S-CWA and southern Puna. The slope value of the linear array defined by loess
 475 samples (i.e., 0.0406 ± 0.0140) is too low to signify an age (Pb isotope systematics require a slope
 476 greater than 0.046). The fact that $^{207}\text{Pb}/^{204}\text{Pb}$ vs $^{206}\text{Pb}/^{204}\text{Pb}$ ratios from the Pampean sediments
 477 cannot be interpreted as an isochron reinforces the idea of the supply of aeolian sediments from
 478 more than one source area.

479 Figure 5 suggests that Pampean loess results from mixing of S-CWA and southern Puna.
 480 This observation contrasts with the interpretation based on Sr and Nd isotopes, which indicate a
 481 subordinate origin from the southern Altiplano (Fig. 4b-c). This inconsistency may be explained
 482 by the spatial distribution of Pb isotopes, which might indicate that the sedimentary Pb isotopic

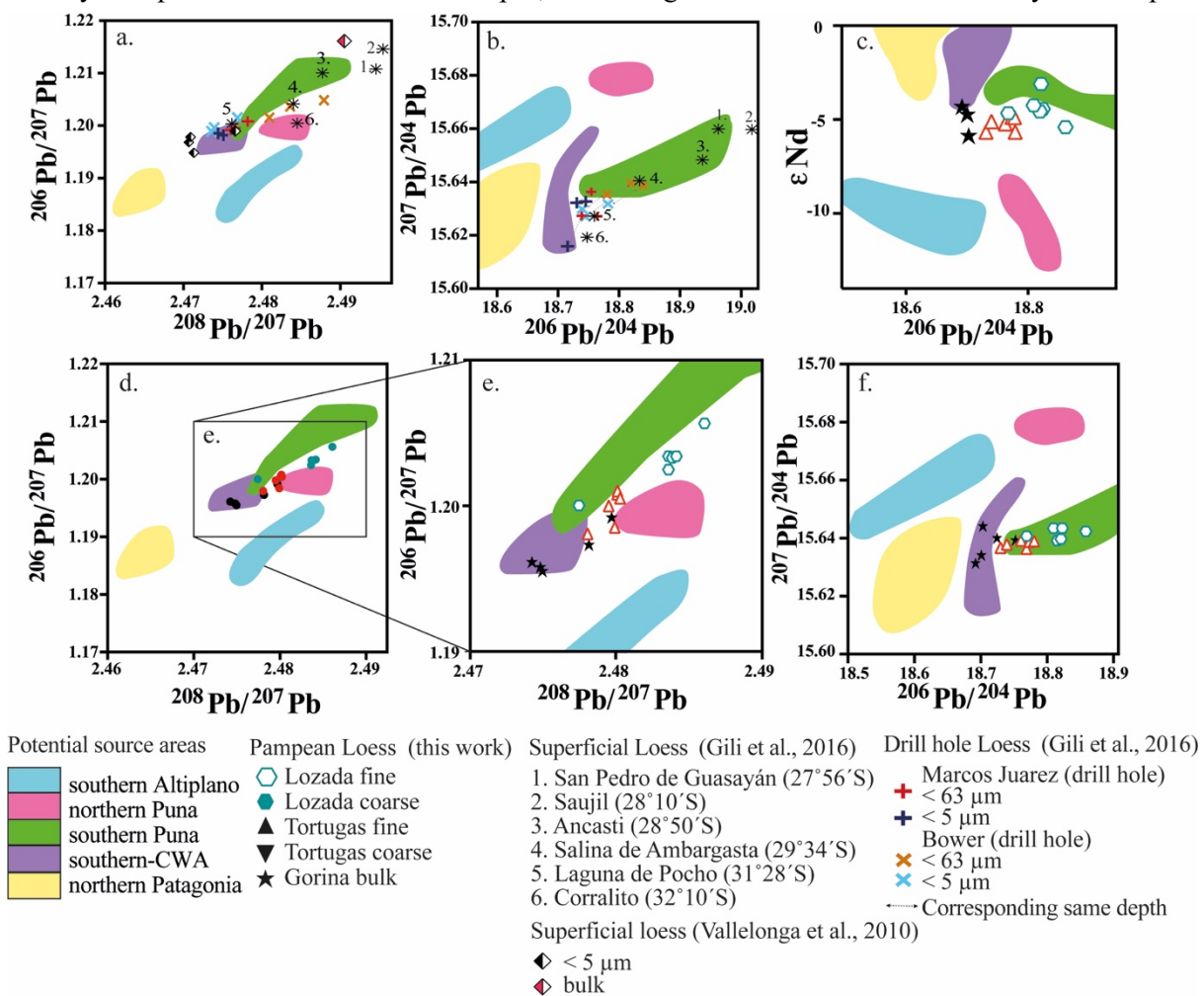
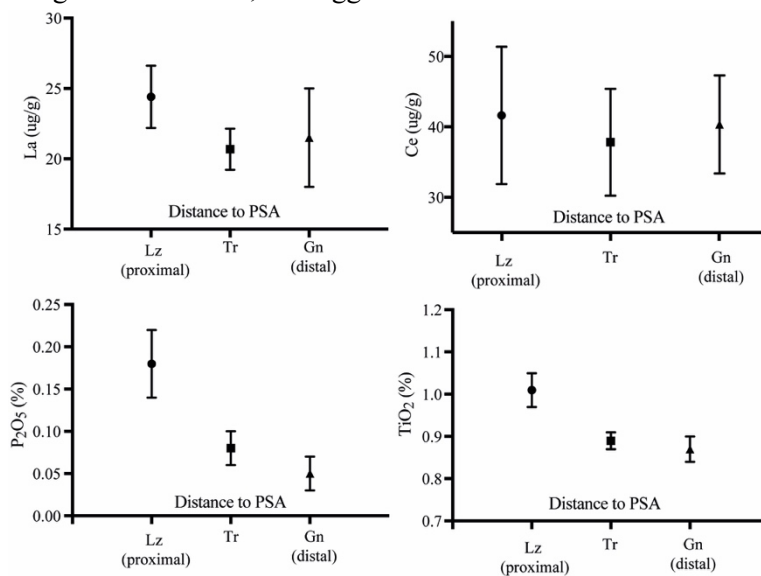


Figure 5. Lead isotope ratios for Pampean loess compared to potential dust source areas in southern South America (Gili et al., 2016).

483 budget is influenced by mineral sorting, thus obscuring dust provenance interpretations. In fact,
 484 the decreasing concentration of elements typically associated with heavy minerals such as zircons,
 485 monazites and allanites (i.e., P, La, Ce, Ti) with distance from the source (Fig. 6) supports the
 486 idea that the Pb isotopic signature is influenced by mineral sorting processes during aeolian
 487 transport. This is because heavy minerals tend to be enriched in the coarse sedimentary fraction
 488 (Garçon et al., 2014). While Gorina has systematically lower average concentrations compared
 489 to the Lozada data, similar concentrations of La and Ce are observed compared to the Tortugas
 490 loess record (Fig. 6). The similar concentrations between Tortugas and Gorina is also reflected in
 491 overlapping Pb isotope ratios between these two sites. Such a heavy mineral effect has already
 492 been noted for aeolian (Sun and Zhou, 2010; Bird et al., 2020) and fluvial transport processes
 493 (Garçon et al., 2013). In this sense, many coarse and dense minerals such as zircons, monazites,
 494 and allanites have more radiogenic Pb isotope ratios and are deposited closer to the source areas,
 495 whereas long-distance deposits are preferentially enriched in platy phyllosilicates and other slow-
 496 settling minerals. Thus, we suggest that the observed inverse correlation between Pampean loess



505
 Figure 6. Average concentrations for each Pampean loess sampling site (Lz, $n = 6$; Tr and Gn, $n = 5$) of selected trace and major elements associated with heavy minerals. Error bars are 1σ based on the measured samples from each site.

Pb ratios and the distance to the sources might be caused in part by mineral sorting during aeolian transport. Future mineralogical studies in loess sites would help to assess the importance of Pb isotopes as a sediment provenance tool.

507

508 *5.4. Modern Pampean dust provenance for deciphering loess origin*

509

510 Based on Sr-Nd isotopic fingerprinting, earlier studies showed that modern dust collected in
511 the central Pampas (e.g., Marcos Juárez station, located 25 km W of the Tortugas site) (Fig. 1) is
512 sourced from the southern Altiplano, and from S-CWA and/or southern Puna (Gaiero et al., 2013;
513 Gili et al., 2017) (Fig. 7). These modern dust samples from the Marcos Juárez monitoring station
514 (white diamonds in Fig. 7) have a 12-day mean sampling interval (Cosentino et al., 2020). Four
515 of these samples have signatures consistent with a dust source from S-CWA while two other
516 samples have clear southern Altiplano signatures. Interestingly, three samples indicate mixed
517 signatures explained by S-CWA/southern Puna and southern Altiplano, similar to the isotopic
518 compositions showed by loess samples from the three Pampean sections (grey area in Fig. 7). The
519 average isotopic composition of modern dust (n = 12, black diamond, Fig. 7) has a similar ϵ_{Nd}
520 value to the Pampean loess, suggesting that similar sources (mainly S-CWA, southern Puna and
521 southern Altiplano) were active from MIS3 to modern times. On the other hand, the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio
522 of the Pampean loess is lower than modern dust. The analyzed modern dust samples have almost
523 half the grain size mode (Torre et al., 2020) compared to the fine fraction of loess, suggesting an
524 important particle size control over the Sr isotope ratios of these samples.

525 The isotopic compositions of modern dust samples show that despite the short 12-day mean
526 sampling period, these samples summarize the isotopic compositions of multiple PSAs, attesting
527 to the extremely high temporal variability of dust sources deposited on the Pampean plain. Thus,
528 the geochemical signature of each loess sample is an integration of the contribution of different
529 dust sources (e.g., S-CWA, southern Puna and/or southern Altiplano) over time scales of hundreds
530 to thousands of years. Based on the time resolution, each loess sample integrates time periods
531 between 100-150 years at Lozada and Tortugas, and over ~600 years at Gorina (Torre et al.,
532 2019).

533 After experimenting with the mixing of Sr-Nd composition of modern dust samples, we
 534 found that two extreme Marcos Juárez dust end members (e.g., two samples with clear northern
 535 Patagonia/S-CWA compositions and two samples with clear southern Altiplano compositions)
 536 permit us to obtain a mixing line that represents most of the compositions of the fine-grained
 537 Pampean loess (see calculation in Supplementary Material, S3) (Fig. 7). Moreover, a similar
 538 mixing experiment, combined with isotopic data from a dust sample collected in Buenos Aires
 539 province, supports the interpretation that northern Puna was insignificant as a dust source (Fig.
 540 7). Furthermore, the isotopic mixing line based on the modern Marcos Juárez dust also helps to
 541 explain the composition of the coarse loess fraction. Notably, coarser sediments have an increased
 542 proportion of more radiogenic Nd sources (e.g., S-CWA and northern Patagonia), while the finer
 543 sediments are slightly closer to less radiogenic Nd sources (e.g., southern Altiplano). This mixing
 544 exercise ignores the role played by the southern Puna as a contributor of dust to the Pampas. This

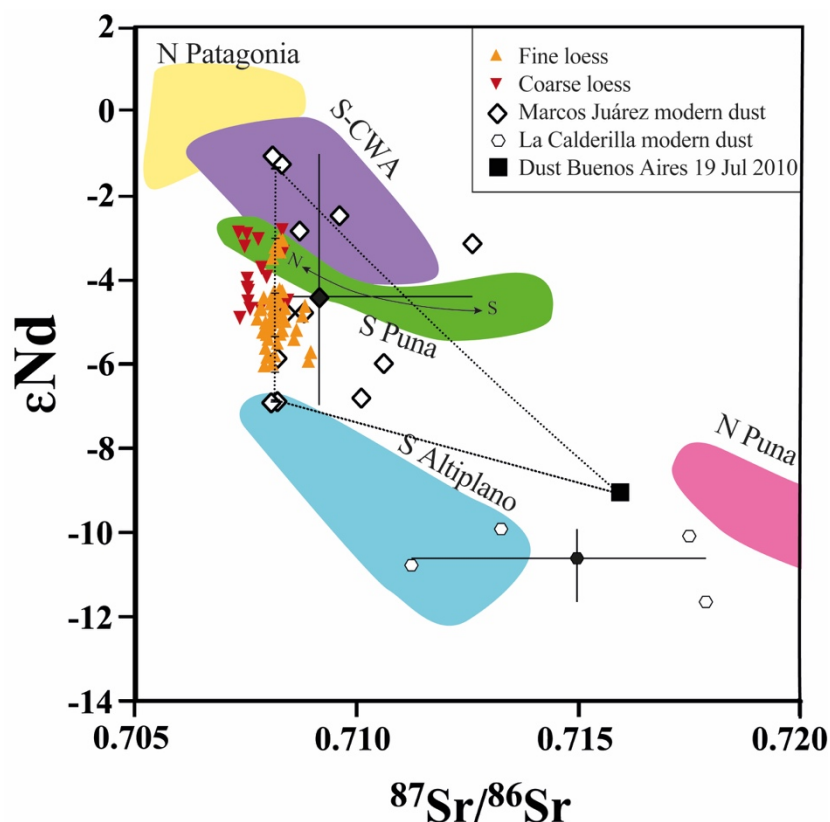


Figure 7. Nd-Sr isotope ratios of modern dust collected at the Marcos Juárez (diamonds) and La Calderilla (hexagons) monitoring stations (Gaiero et al., 2013; Gili et al., 2017). Black symbols represent the average composition and the range for each dust monitoring station. Dotted lines represent the calculated mixing curves between two end members from Marcos Juárez station, and between La Calderilla station. Data from La Calderilla represent the isotopic composition of dusts near their PAP source (Gili et al., 2017). Triangles indicate the Sr-Nd isotopic compositions of fine and coarse loess (this work).

PSA has a clear N-S isotopic gradient (i.e., southernmost Puna has lower Nd and higher Sr isotope ratios than the northern sector of southern Puna as illustrated in Fig. 7) and hence, a possible contribution from the northern sector of southern Puna may be obscured as its composition lies within the mixing line closer to

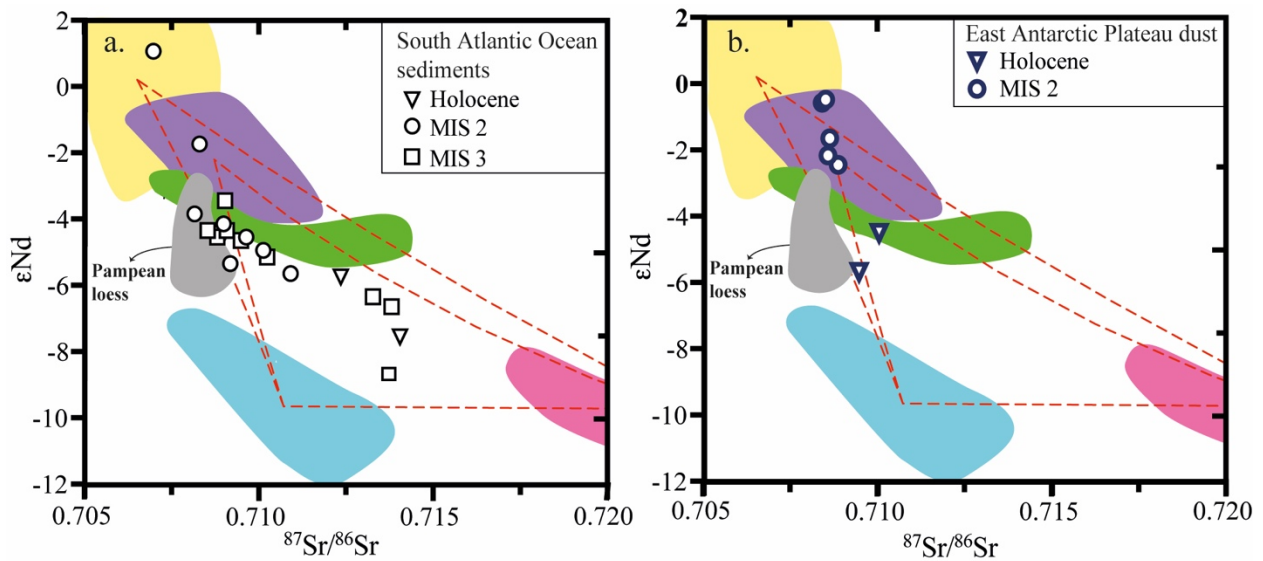
560 the S-CWA/northern Patagonia end member.

561

562 *5.5. Short- and long-range deposition of southern South American dust*

563

564 Along with the Pampean loess, the isotopic compositions of MIS3-Holocene dust collected
565 in marine sediment cores from the Southern Atlantic Ocean and in ice cores of the eastern
566 Antarctic Plateau are consistent with the main provenance coming from SSA (Fig. 8) (Gili et al.,
567 2017, Noble et al., 2012; Walter et al., 2000; Delmonte et al., 2004, 2007, 2010). Among these
568 dust archives, the Pampean loess represents the proximal record of SSA dust (Li et al., 2008;
569 Albani et al., 2012), while South Atlantic Ocean and East Antarctic Plateau sedimentary deposits
570 respectively characterize the middle- and long-range dust transport from SSA. The restricted
571 isotopic variation shown by loess samples during MIS 3-early Holocene contrasts with the large
572 range of isotopic compositions of the medium-to-long range dust deposited during the same
573 period. Their increased range of Sr-Nd isotopes values are well-constrained by the isotopic
574 variability observed for the PSAs from SSA, indicating that these sources could have been
575 important dust suppliers. Furthermore, Sr-Nd isotopes indicate that there could have been sources
576 that supplied dust to more distal environments that are not recorded in the Pampean loess (e.g.,
577 northern Puna, southern Patagonia, Tierra del Fuego, southern Africa, New Zealand) (Sugden et
578 al., 2009; Noble et al., 2013; Koffman et al., 2021). The isotopic mismatch between Pampean
579 loess and the more distal archives may suggest that Pampean loess was not a significant dust
580 source to the distal environments during the past glacial-interglacial cycle as previously suggested
581 (Delmonte et al., 2010). This agrees with stratigraphic studies indicating no erosional hiatuses in
582 the Pampean loess record between MIS 3 and the early Holocene, meaning that deflation over the
583 Pampas was not widespread during this period (Torre et al. 2019, Zárata et al., 2009; Kemp et al.,
584 2004, 2006).



Potential source areas from southern South America

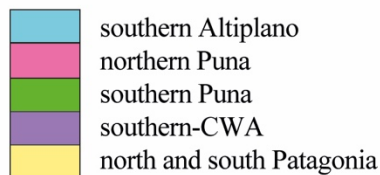


Figure 8. Sr-Nd isotopic signature of MIS 3 to Holocene Pampean loess deposits (grey area), together with (a) dust from marine sediment cores in the South Atlantic Ocean (Noble et al., 2012; Walter et al., 2000), and (b) dust from ice cores in the East Antarctic Plateau (Delmonte et al., 2004, 2007, 2008).

585

586 5.6. Dust provenance unmixing

587

588 Although the visual inspections of Figures 3-5 add critical observations to the understanding
 589 of loess provenance, the quantitative apportionment of each source, as well as the potential to
 590 identify the lesser prevalent dust sources, require additional techniques such as unmixing models.
 591 We used the SIMMR unmixing model (Parnell et al., 2020) with PSAs of SSA. To constrain the
 592 model we discarded, based on the discussions presented in sections 5.1 and 5.2, the northern Puna,
 593 Paraná River, Pampean Ranges and Mar Chiquita Lake PSAs, which were singled out as not
 594 significant dust sources to the Pampean region.

595 As it was observed that there is a possible grain-size and mineralogical dependence of Sr and
 596 Pb isotopic ratios in loess samples, here we contrasted SIMMR model results using only Nd
 597 isotope data with results obtained combining Nd and Sr data. Interestingly, the results with and

598 without Sr isotope data were very similar. On
 599 the other hand, the model output obtained
 600 adding Pb isotopes is consistently different
 601 compared to the one using only Nd or the Nd-
 602 Sr isotopic pair. This is probably linked to the
 603 mineralogical sorting effect that we found in
 604 our data that could affect Pb isotopic system
 605 (section 5.3).

606 The statistical unmixing was performed
 607 on the fine loess samples from Lozada and
 608 Tortugas, and on bulk loess samples from the
 609 Gorina site. The unmixing model supports the
 610 existence of multiple dust sources to the
 611 Pampean loess (Fig. 9), indicating that none
 612 of the considered PSAs of SSA are negligible
 613 contributors of dust to the Pampas during
 614 MIS 3 to early Holocene (i.e., all of them
 615 show inputs greater than 10%) (Table S3)
 616 (see Supplementary Material, section S4). For
 617 the three loess sites, distal sources associated
 618 with the STJ circulation (i.e., southern
 619 Altiplano + southern Puna) represent the
 620 main dust suppliers (an average input of 55%)
 621 to Pampean loess deposited between MIS 3 and
 622 early Holocene. In turn, the model indicates
 623 that sources associated to the SWW circulation
 624 (i.e., S-CWA and northern Patagonia) have

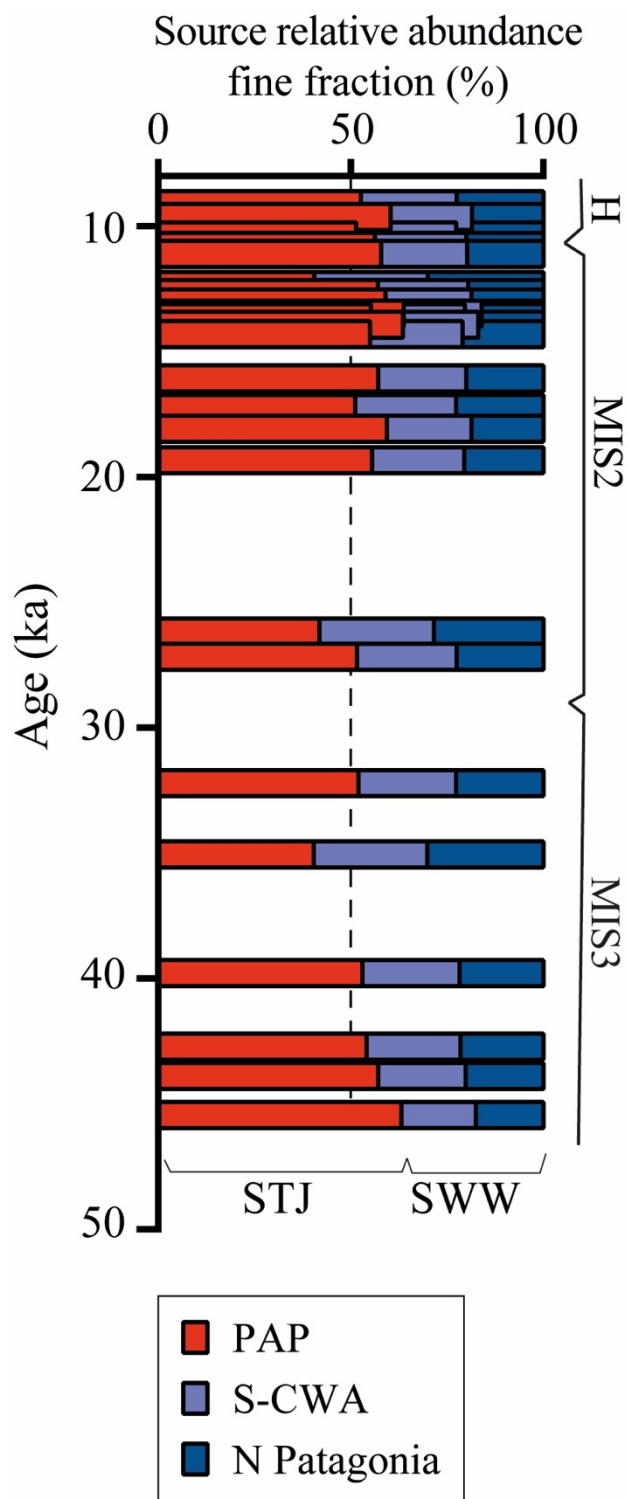


Figure 9. Analyses of the relative inputs of dust to the Pampean loess belt determined by the SIMMR unmixing model. The model was constrained with data of the fine fraction (<44 μm) from Lozada and Tortugas, and bulk loess from Gorina. The reddish colors indicate the proportion of dust supplied from sources interacting with the subtropical jet stream (STJ) (i.e., Puna-Altiplano Plateau, PAP). The purple colors indicate the proportion of dust supplied from sources interacting with the southern westerly winds (SWW) (i.e., southern central-west Argentina and northern Patagonia).

625 supplied an average of 45% of the dust reaching the studied Pampean sites.

626

627 *5.7. Major dust fluxes to the Pampas under El Niño-like conditions*

628

629 Figure 10 contrasts loess mass accumulation rates (MARs), loess provenance model outputs,
630 lake levels reconstructions for Mar Chiquita Lake and lakes from the PAP area (e.g., proxies for
631 humidity conditions over the Pampas and PAP respectively), and dust accumulation rates
632 recorded at the South Atlantic Ocean and East Antarctic Plateau for the late Pleistocene to the
633 early Holocene. Loess mass accumulation rates (MARs) varied substantially on millennial
634 timescales (Fig. 10a) as a result of changes in dust availability at the sources, the intensity and/or
635 position of the SWW or STJ, and/or dust settling/removing mechanisms (Torre et al., 2019). The
636 three loess sections indicate that periods with increased MARs of the fine-grained fraction of dust
637 are generally associated to increased dust contributions from PAP (e.g., up to ~64%) (Fig. 10d).
638 This feature is clearly observed along the loess records in four different time windows; i.e., 43-
639 41 ka BP; 20-18 ka BP (late MIS 2); 14.6-12.6 ka BP (Antarctic Cold Reversal); and 11.4-8.9 ka
640 BP (early Holocene) (Fig. 10, shaded areas). Interestingly, the periods showing high dust fluxes
641 over the Pampas are preceded by high lake level phases in the Altiplano (Fig. 10e-f) (Baker et al.,
642 2001; Sylvestre et al., 1999; Fornari et al., 2001; Fritz et al., 2004; Placzek et al., 2006). Moreover,
643 there is a positive trend between the relative magnitudes of preceding high-lake level events with
644 increased MARs in the Pampean loess. For example, the most prominent humid Tauca phase (Fig.
645 10e) preceded the highest dust input recorded at Tortugas, which was mainly derived from the
646 PAP region (~64%, Fig. 10d). In turn, the less intense Coipasa humid phase preceded the PAP
647 source input of 60% observed at Lozada. Through these observations, we propose that high MARs
648 observed in the Pampas are associated to preceding humid phases in the PAP region. Humid
649 phases over the region caused significant accumulation of sediments on the lake shores, thus
650 inducing a greater dust activity during the subsequent arid climatic phase.

651 If we assume that similar sources supplied dust both proximal and distal environments, it is
652 likely that the relatively wet conditions in the Pampas from 13.5 ka until 8.5 ka BP (González et

653 al., 1994; Prado and Alberdi, 1999; Piovano et al., 2009; Cuña-Rodríguez et al., 2020) (Fig. 10b)
654 enhanced wet dust deposition over proximal sectors (e.g., Pampas), thus reducing SSA-derived
655 dust fluxes to more distal environments (compare Fig. 10a with Fig. 10g-h). This is also supported
656 by high MARs detected in the distal loess site (Gorina) before 40 ka BP, which appears to be
657 associated to a wet period, as indicated by soil development in NW Argentina (Zech et al.,
658 2009a,b).

659 Although the Pampean loess has a persistent isotopic fingerprint consistent with a mixed
660 provenance, the variability of this signature allows us to determine fluctuations in the contribution
661 of these sources. The activation of the Puna+southern Altiplano dust sources occurs at times of
662 increased MARs on loess deposits, suggesting the existence of a mechanism of enhanced deflation
663 over the sources and/or increased wet removal of atmospheric dust downwind in the Pampean
664 lowlands. This is compatible with the dominance of El Niño-like conditions in South America
665 during the last glacial-interglacial cycle. In this sense, negative present-day rainfall anomalies are
666 observed over the PAP during El Niño events, which are consistent with a stronger-than-average
667 STJ over the Altiplano that hampers the advection of moisture from the eastern lowlands
668 (Garreaud and Aceituno, 2007), enhancing deflation over the PAP region (e.g., Gaiero et al.,
669 2013). In contrast, upper-level circulation anomalies tend to occur during La Niña episodes
670 (Garreaud and Aceituno, 2007), enhancing moisture transport towards the Altiplano and favoring
671 wetter-than-average conditions and lower dust emissions. El Niño Southern Oscillation (ENSO)-
672 related rainfall anomalies at a global scale show that El Niño episodes are typically associated
673 with anomalously wet conditions in the central-eastern portion of SSA (Ropelewski and Halpert,
674 1987), which in turn could have promoted increased scavenging of PAP-derived atmospheric dust
675 over the Pampas (Torre et al., 2019).

676 There is no scientific consensus about past variability of ENSO conditions during the late
677 Pleistocene and early Holocene. Nevertheless, from 38.2 to 8.4 ka BP strong El Niño-like
678 manifestations have been inferred from studies of alluvial sequences in the coastal region of
679 southern Peru (Keefer et al., 1998; 2003; Ortlieb et al., 2003). Other studies have concluded that
680 these records probably reflect local short-lived rainfall events, rather than through El Niño

681 episodes (Vargas et al., 2006). Moreover, reconstructions of temperature variability in the Pacific
682 Ocean have indicated reduced ENSO conditions during the LGM compared to the Holocene (Ford
683 et al., 2015). At the same time, some model simulations indicate intensified ENSO variability for
684 this glacial period (e.g., Timmerman et al., 2004; Otto-Blesiner et al., 2003). Nonetheless, at
685 millennial timescales, results from geological records and models still suggest persistent ENSO
686 variability throughout the entire last glacial-interglacial cycle, and weaker frequencies and
687 amplitudes of major El Niño-like events may have characterized the latest Pleistocene and the
688 early Holocene compared to the late Holocene (Rodbell et al., 1999; Moy et al., 2002; Tudhope
689 et al., 2001, Clement et al., 1999). Future studies of younger loess deposits spreading towards the
690 mid-late Holocene could help in the identification of the influence of the ENSO variability on
691 aeolian deposits between Late Pleistocene and Holocene times.

692

693 *5.8. Lower dust input to the Pampas under equatorward-shifted SWW during the LGM and ACR*

694

695 Many uncertainties still exist about the paleoclimatic conditions prevailing over SSA's dust
696 PSAs during the LGM. This period is barely observed in the Gorina profile, with only two isotopic
697 data points available for dust provenance analysis. The LGM is of great interest for the
698 understanding of paleo-atmospheric circulation and the interaction between dust and climate
699 because of the sharp increase of dust fluxes recorded in the South Atlantic Ocean and East
700 Antarctic Plateau (e.g., Lambert et al., 2012) (Fig. 10g-h). In spite of geomorphological evidence
701 (Zárate and Tripaldi, 2012; Tripaldi et al., 2011) and Earth system simulations (e.g., Albani et al.,
702 2012) that support an important role for the S-CWA as a dust supplier during the LGM to the
703 South Atlantic Ocean and East Antarctic Plateau (Fig. 8) (Gili et al., 2017), a similar dust flux
704 increase is not observed during the LGM as recorded at the Gorina site (Torre et al., 2019).
705 However, the unmixing model demonstrates a clear provenance change at the beginning of MIS
706 2, indicating increased inputs from S-CWA+northern Patagonia, representing more than 60% of
707 the dust input to the Gorina site (Fig. 10c). We hypothesize that the lower dust deposition rates
708 recorded over the continent between 47-43 ka BP, 40-22 ka BP, 17-14 ka BP and 12.3-11.4 ka

709 BP reflect reduced close-to-source dust wet deposition due to drier conditions at the Pampas
710 (Prieto, 2000; Behling, 2002; Iriarte, 2006; Behling and Pillar, 2007; Cuña-Rodríguez et al.,
711 2020). In turn, higher dust deposition rates at distal environments were probably facilitated by a
712 reduced hemispheric hydrological cycle that permitted longer lifetimes of atmospheric dust
713 particles (e.g., Lambert et al., 2008; Markle et al., 2018).

714 Reduced dust deposition at the Pampas with increased relative input from S-CWA+northern
715 Patagonia (i.e., ~62%) is also observed during a short time-lapse at the end of the ACR (Figs.
716 10a,d) . The activation of S-CWA+northern Patagonia sources during colder periods (i.e., LGM
717 and the ACR) was probably powered by an overall strengthening of Zonda winds induced by
718 more vigorous SWW, while they were shifted northward, closer to the equator (e.g., Gili et al.,
719 2017). Future studies of Pampean loess at higher temporal resolution during the LGM will help
720 to better understand dust dynamics during this important period in Earth's history.

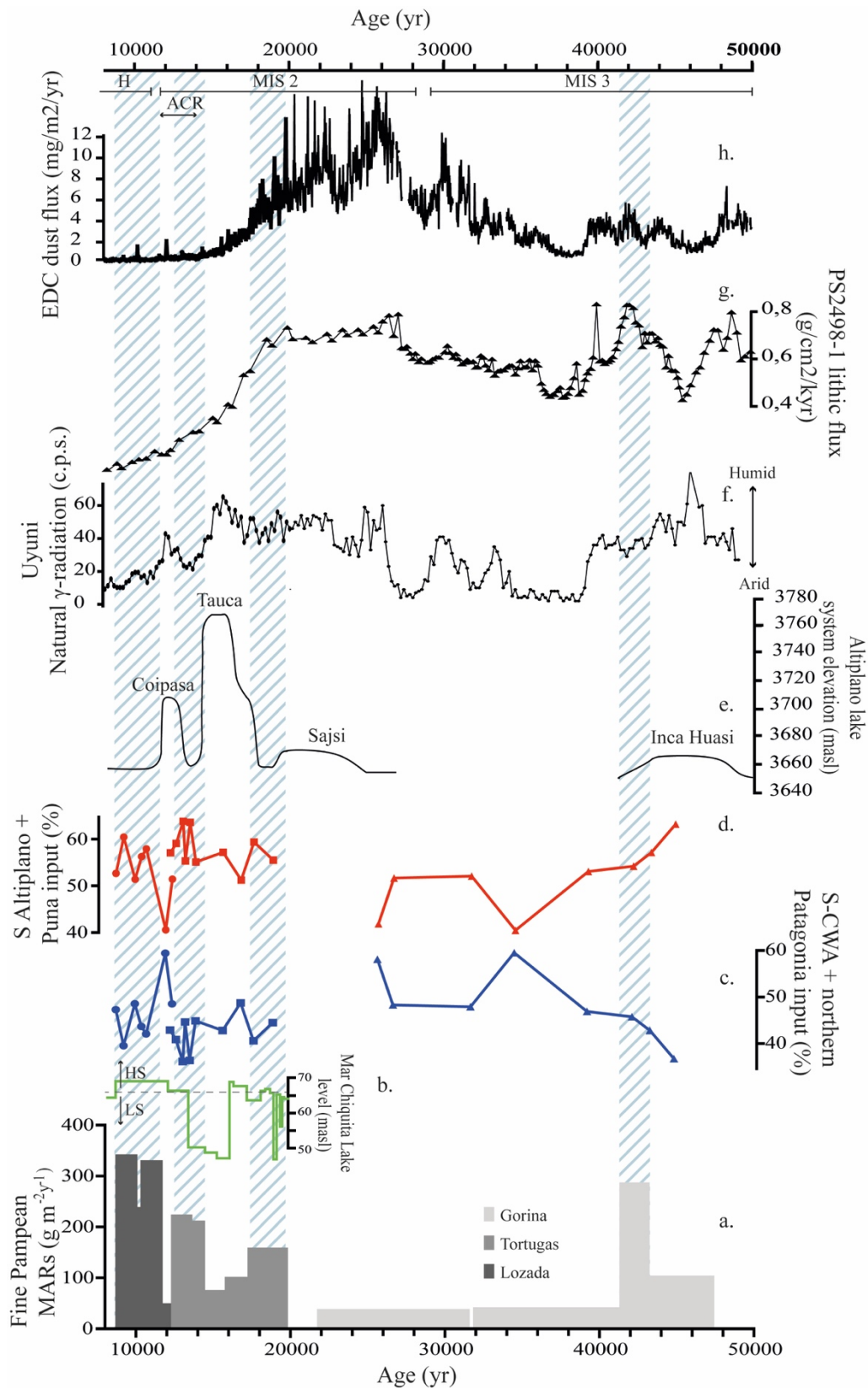


Figure 10. (a) Temporal variability of the mass accumulation rate (MAR) calculated for the fine dust fraction deposited on three Pampean loess sites. (b) Mar Chiquita Lake level (Cuña-Rodríguez et al., 2020). Contribution of dust derived from (c) south-central western Argentina (S-CWA) + northern Patagonia; and from (d) southern Altiplano + southern Puna the fine fraction of Pampean loess. (e,f) Paleo-lake levels in the Altiplano (Placzek et al., 2013; Baker et al., 2001). (g) Lithic fluxes at the South Atlantic Ocean (Anderson et al., 2014), and (h) dust fluxes at EPICA Dome C (EDC), East Antarctica (Lambert et al., 2012). Shaded areas denote times of high MAR.

722 **5. Final remarks**

723

724 The use of geochemical and unmixing model tools for understanding the provenance of the
725 Pampean loess underlines the paleoclimatic importance of aeolian deposits for improving
726 knowledge of past dust cycles. In SSA the loess belt is located at a relatively long distance from
727 the Andean dust sources, and we have observed that mineral sorting effects on Pb isotopes
728 obscures loess provenance interpretations. However, Pb isotopes combined with Sr-Nd isotopes
729 allow us to deduce that multiple sources have supplied dust to the Pampean loess belt between
730 MIS 3 and the early Holocene. Although REE are less diagnostic compared to radiogenic isotopes,
731 they are complementary tools for loess provenance studies.

732 We observed that modern dust collected at the core of the Pampas has a similar geochemical
733 composition to Pampean loess, suggesting that modern PSAs were also active during MIS 3 and
734 the early Holocene, and that modest changes in the geochemical signature of loess respond to the
735 activation/deactivation of the different dust sources. The data on modern dust and loess point to
736 the northern Patagonia+S-CWA and southern Puna+southern Altiplano as the main dust suppliers
737 to the Pampean loess. Our data excluded the northern Puna as an important Pampean loess dust
738 supplier during the late Pleistocene. In order to improve the understanding on the behavior of the
739 subtropical jet stream latitudinal movement, the absence of a northern Puna isotopic signature in
740 loess samples along with the possible role play by the southern Puna should be further
741 investigated.

742 The geochemical fingerprint of Pampean loess does not explain the entirety of the isotopic
743 composition of dust in distal paleo-records located at the South Atlantic Ocean and the East
744 Antarctic Plateau. This suggests that during the studied period the Pampean loess was not an
745 important dust supplier to distal depositional environments and/or that there were active sources
746 that not detected at the Pampean loess.

747 Revisiting geochemical data from previous loess studies along with new data from this study
748 has permitted us to discard some PSAs located close to the Pampean loess (e.g., Mar Chiquita
749 Lake, Paraná River, Pampean Range) as significant dust contributors to this region. Although the

750 Pampean loess record suggests that multiple SSA dust sources have been active during the past,
751 the dominance of the S-CWA and the northern Patagonia signature on dust recovered from distal
752 environments (e.g., Antarctica) point to the existence of a higher transport efficiency of the SWW
753 during the last glacial period. During colder periods (e.g., the LGM and ACR), the activation of
754 S-CWA/northern Patagonia sources could have been powered by the strengthening of the foehn
755 Zonda wind, induced by a more vigorous SWW. We suggest that higher dust deposition rates at
756 distal environments were promoted by a reduced hemispheric hydrological cycle that permitted
757 longer airborne residence times of atmospheric dust particles and reduced close-to-source
758 deposition.

759 Finally, the elevated dust deposition rates observed during some periods in the Pampas is
760 interpreted as associated with the increased frequency/intensity of El Niño-like conditions. This
761 allowed for sediment deflation from a dryer Puna-Altiplano region and deposition over the more
762 humid Pampas.

763

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772

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