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Isotopic constraints on the source of Argentinian loess – with implications for atmospheric circulation and the provenance of Antarctic dust during recent glacial maxima

James Smith [a,*], Derek Vance [b], Rob A. Kemp [a], Corey Archer [b], Phillip Toms [a,1], Matthew King [a], Marcelo Zárate [c,2]

[a] *Centre for Quaternary Research, Department of Geography, Royal Holloway, University of London, Egham TW20 OEX, UK*
[b] *Department of Geology, Royal Holloway, University of London, Egham TW20 OEX, UK*
[c] *CONICET, IANIGLA, CRICYT, CC 330, RA-5500 Mendoza, Argentina*

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Abstract

We present rare-earth element (REE) and Sr–Nd isotopic data for Argentinian loess (28–38°S) with two aims: (1) to examine the source regions of Argentinian loess and the constraints that these put on palaeo-wind directions; (2) to further investigate the source of Antarctic ice-core dust and to test the hypothesis that some of it could be derived from a region to the north of Patagonia – into which the dry, dusty, westerly dominated Patagonian climate expanded during Quaternary glacial maxima. Sr–Nd isotopic data for Argentinian loess from north of 37°S are distinct from Patagonian loess compositions in that they have more radiogenic Sr ($^{87}Sr/^{86}Sr = 0.7059–0.7123$) and less radiogenic Nd ($\varepsilon_{Nd} = -0.8$ to -6.4). REE patterns and Sr–Nd isotopic values are relatively homogeneous for multiple samples taken from single loess sections but show significant differences between sections. In general, there is a northward change from Patagonia-like REE patterns and isotopic values away from volcanogenic signatures and towards those that are more like the continental crust. The latitudinal Nd isotopic pattern is remarkably similar to that for Andean volcanic rocks and suggests derivation of loess from the Andes by more-or-less direct westerly transport. For loess sections in the north, the data imply a contribution from Palaeozoic gneisses to the northwest in the Chilean Altiplano. Sr–Nd data for extra-Patagonian Argentinian loess north of 37°S do not support a significant role for this source region in supplying dust to Antarctica at the last glacial maximum. This conclusion contrasts with previous studies that suggest a significant northward shift in the climatic belts – and in particular the westerlies and the Antarctic Polar Front – during Quaternary glacial maxima. Very systematic relations between the Sr–Nd isotopic composition of loess and their Andean source highlights shifts in the Sr isotopic composition of loess to more radiogenic values and strongly suggests that the slight offset between Patagonian loess and ice-core dust identified by previous workers is due to grain-size differentiation effects.

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[*] Corresponding author. Present address: Department of Geography, University of Durham, Science Laboratories, Durham DH1 3LE, UK. Tel.: +44-191-374-2472; Fax: +44-191-374-2456. *E-mail address:* j.a.smith@durham.ac.uk (J. Smith).

[1] Present address: GEMRU, University of Gloucestershire, Swindon Road, Cheltenham GL50 4AZ, UK.
[2] Present address: CONICET–Universidad Nacional de la Pampa, 6300 Santa Rosa, Argentina.

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1. Introduction

Loess deposits are generally considered to be accumulations of wind-blown dust, most probably from deflation sites on glacial outwash plains or from arid/semi-arid regions (e.g. [1]). Their geochemistry has been the subject of a number of detailed studies in the last two decades. Early work (e.g. [2]) focussed on the question of whether loess could be considered as geochemically representative of the upper continental crust (UCC). More recently, loess deposits have assumed palaeoclimatic importance, with the realisation that loess–palaeosol sequences record changes in continental climate that can be correlated with the detailed history of climate change extracted from deep-sea sediments and polar ice [3–5]. Moreover, the Quaternary history of atmospheric dust loading is recognised as a key feature of the Earth's climate system on glacial–interglacial timescales (e.g. [6]) and is likely to respond to the same processes that control recent loess deposits. Polar ice contains 10–100 times more dust deposited during Quaternary glacial maxima than is accumulating at present [7–9]. Isotopic data [10–12] have identified South America as the most plausible source of elevated Antarctic atmospheric dust contents during glacial maxima, with as little as 10–15% deriving from extra-South American regions (e.g. Australia or South Africa).

The Argentinian loess region (Fig. 1) is the most extensive in the Southern Hemisphere, covering approximately 1.1×10^6 km^2 between 20°S and 40°S [13]. The existing isotopic and geochemical data for the South American loess (apart from one further site in Gallet et al. [23]) is largely limited to Patagonia/Southern Pampas – or an area south of 38°S. Here we present Sr–Nd isotopic data, as well as rare-earth element (REE) patterns, for loess from five locations in this province between latitudes 28°S and 37°S, with the primary aim of expanding the data coverage to include the Argentinian loess province to the north.

More specifically, our aims here are two-fold. Firstly, we seek to investigate the source of Argentinian loess. The identification of sources for loess help in constraining palaeo-wind directions during the major climatic shifts of the Quaternary. Although discussed in several recent publications [14–16,24–26], the source of Argentinian loess remains controversial [14,24]. Sayago [14] suggested that all of it, including loess from mountain areas of northwestern Argentina, was blown from extra-Andean regions of Patagonia by southerly winds. Several other studies, however, based on grain size [24,27], mineralogy [13,18,28], and geochemistry [17–29] have highlighted the possibility of multiple source areas, including the Argentinian continental shelf (sediments derived from Patagonia), the Paraná River basin, the Pampean Hills, the Altiplano–Puna Plateau and/or glaciofluvial deposits of Mendoza, Neuquen and Rio Negro (Fig. 1). Here we use the new isotopic and geochemical data, along with published data for South American loess [10,11,28] and possible source rocks, to shed further light on this issue.

Secondly, we aim to further investigate the possible source regions for dust in East Antarctic ice cores. Grousset et al. [10] first established the isotopic similarity between East Antarctic ice-core dust and loess deposits from northern Patagonia. These authors acknowledged the small size of a putative Patagonian dust source and speculated that the Argentine continental shelf, exposed during the lowered sea level of the glacial maxima, might act as an additional source. However, Basile et al. [11] later showed that the Sr–Nd isotopic compositions of core-top continental shelf sediments were incompatible with such an idea. Basile et al. [11], however, further noted that the movement of the climatic zones during glacial maxima may have created a much larger arid/semi-arid continental region in Argentina, possibly extend-

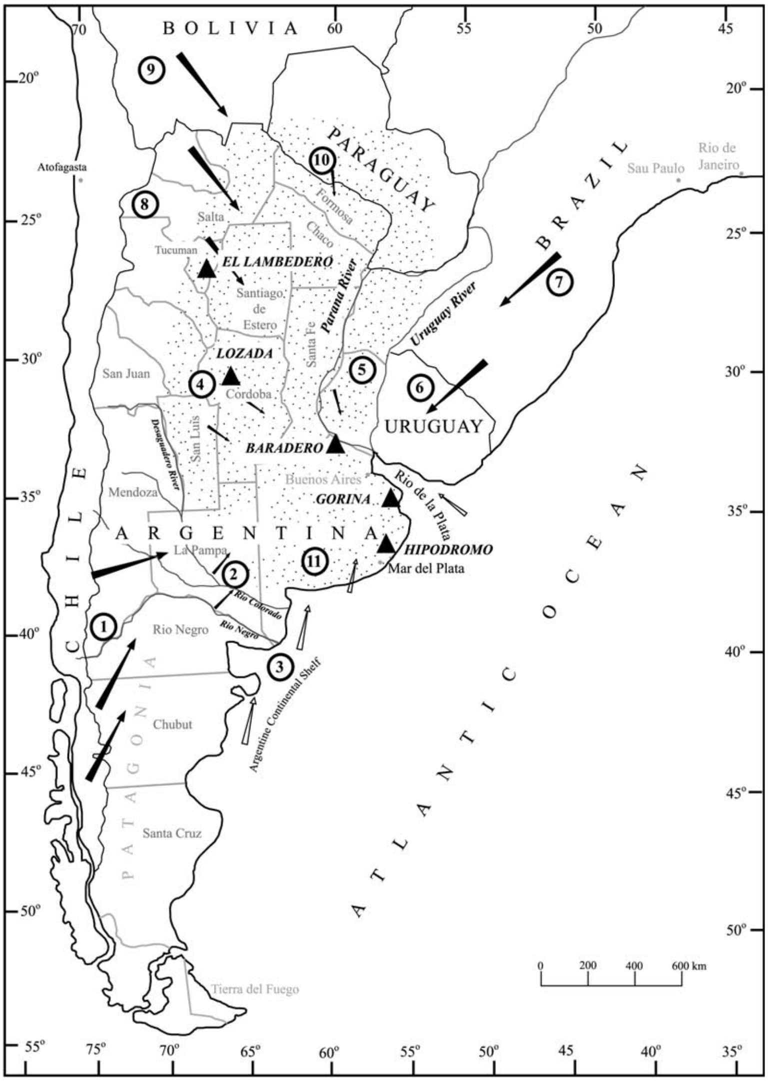

Fig. 1. Sample sites (triangles), potential source areas (numbered circles) and likely transport directions (arrows). The distribution of loess (based on [13]) is also shown (dot-dashed line): (1) Patagonia and extra Andean regions [14,15]; (2) Rio Negro and Colorado [16]; (3) Argentine continental shelf [16]; (4) sediments derived from the Cordoba and San Luis hills (Sierra Pampeanas/Pampean Hills) [17,18]; (5) Paraná/Uruguay River basin [18]; (6) Palaeozoic/basaltic rocks, Uruguay [18]; (7) Brazilian Shield [19]; (8) NW Argentina – Antofalla/Antofagasta ranges/Cerro Galan region/Palaeozoic rocks of Central Andes [20,21]; (9) Bolivian Andes and Altiplano–Puna Plateau [22]; (10) Mega-fans of Paraguay [20]; (11) Tandillia Range, Buenos Aires [18].

ing as far north as 20°S, which could have acted as an additional source of dust with similar isotopic characteristics as Patagonia. Only one site in this region (near Buenos Aires, 33°S; [23]) has previously been studied isotopically. Here we expand the available dataset considerably in order to test the hypothesis that East Antarctic ice-core dust could have derived from the Argentinian loess province.

2. Sample locations and analytical procedures

Loess samples were collected from sections

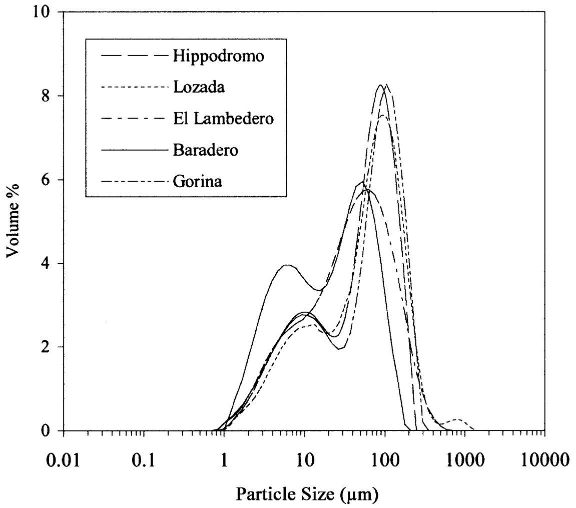

Fig. 2. Average grain-size distributions for loess samples. All samples have a distinct bimodal pattern with a minimum at ca. 20 μm.

originally excavated for a regional study of loess–palaeosol sequences. Thus, the five sites sampled span a wide geographical area (Fig. 1) extending from Tucumán (El Lambedero; E1–E4) through Córdoba (Lozada; L1–L4) and northern Buenos Aires province (Baradero; B1–B5/Gorina; G1–G4) to southern Buenos Aires province (Hipódromo; H1–H4). Samples were taken from loess units in reverse stratigraphic order (i.e. E1 is shallower and therefore younger than E2).

Particle-size characteristics were determined using a Malvern Laser (2000L) granulometer. Samples were pre-treated with 20% hydrogen peroxide to remove organic matter and dispersed using 5% sodium hexametaphosphate. The size results are displayed in Fig. 2, where it can be seen that the distribution is bimodal with a minimum at around 20 μm. Malvern granulometers are thought to bias towards the fine fraction in micaceous sediments. However, our purpose here was simply to obtain a rough estimate of the grain-size distributions in order to decide on a suitable cut-off point for a coarse and fine fraction for separate isotopic analyses on one sample (see below).

All 23 bulk sediment samples were measured for REE, ^{87}Sr/^{86}Sr and ^{143}Nd/^{144}Nd. One Gorina sample was separated into < 20 μm and > 20 μm fractions based on the grain-size distributions in Fig. 2, in order to allow the investigation of the variation in ^{87}Sr/^{86}Sr ratios and ^{143}Nd/^{144}Nd ratios in different size fractions of the same sample. These fractions were separated by gravity settling. The suspended < 20 μm were decanted and the sequences repeated as necessary.

REE concentrations on the bulk sediment fractions were determined using techniques described elsewhere [30]. Analyses were performed on a Perkin Elmer Scex Elan 5000 inductively coupled plasma mass spectrometry (ICP-MS). Replicate analyses of BCR-1 USGS standard rock show agreement with the reference values to within about 5%.

For isotopic analysis, approximately 100 mg of loess was initially treated with HCl to dissolve carbonate, followed by digestion in HF+HNO_3. Post-depositional processes may cause carbonate precipitation in, or removal from, loess sections. Such a process potentially compromises the use of Sr isotopic compositions to examine source–loess relationships. However, carbonate contents of these loess samples are low (Table 1) and there is no relationship between the amount of carbonate and the Sr isotopic composition of the samples (see below and Table 1). The HCl leachates were retained with the rest of the sample for isotopic analysis. Sr and Nd were separated for isotopic analysis using procedures that have been reported elsewhere [31]. The techniques used for isotopic analysis of Sr by thermal ionisation mass spectrometry and of Nd by multi-collector magnetic-sector ICP-MS have also been described previously [32,33]. The measured ^{87}Sr/^{86}Sr and ^{143}Nd/^{144}Nd were corrected for mass fractionation by normalising to $^{86}\text{Sr}/^{88}\text{Sr} = 0.1194$ and $^{146}\text{Nd}/^{144}\text{Nd} = 0.7219$, respectively. Precision was monitored using multiple analyses of standards whose reproducibility is given in a footnote to Table 1. Nd data are reported in units of ε_{Nd}, the deviation of a sample ^{143}Nd/^{144}Nd from the present-day chondritic reservoir value (CHUR) (0.512638) in parts per 10^4, as follows:

$$\varepsilon_{\text{Nd}} = \left[\frac{(^{143}\text{Nd}/^{144}\text{Nd})}{(^{143}\text{Nd}/^{144}\text{Nd})_{\text{CHUR}}} - 1\right] \times 10^4$$

3. Results

3.1. *Effects of carbonate content and grain size on $^{87}Sr/^{86}Sr$ ratio*

Carbonate has high Sr concentrations that may have a distinct Sr isotopic signature, so that post-depositional mobility of carbonate potentially compromises the use of Sr isotopes to examine the sources of loess. The carbonate contents of these samples (Table 1) are low and, though minor variations in their Sr isotopic composition might be attributable to variations in $CaCO_3$ content, there is no relationship between the latter and either the Sr concentration or the Sr isotopic ratio. The critical point is that any differences in Sr isotopic ratios within sites that might be attributable to $CaCO_3$ content are smaller than the differences between the sites. For example, the $CaCO_3$ contents of samples B1 and B3 encompass almost the entire range seen in the entire sample set yet their Sr isotopic compositions are similar and the difference between these two samples is certainly smaller than the differences between sites. The same goes for samples G2 and G4 and, to an even greater degree, for samples E2 and E4.

Variations in the Sr and Nd isotopic compositions of sediments in different grain-size fractions have been reported in several studies (e.g. [34–38]) but the results have not been consistent. Of all these studies, those of Dasch [34], Goldstein et al. [35], and Derry and France-Lanord [38] are the most relevant to the grain sizes encountered

Table 1
Sr–Nd isotopic data for Argentinian loess samples

Site[a]	Sample	$CaCO_3$[b] (wt%)	[Sr] (ppm)	$^{87}Sr/^{86}Sr$[c]	$^{143}Nd/^{144}Nd$	ε_{Nd}	$^{147}Sm/^{144}Nd$[d]	T_{DM}
Hipódromo	H1	–	–	0.705964	0.512581	−1.07	0.1231	0.96
Hipódromo	H2	1.2	310	0.705986	0.512594	−0.81	0.1372	1.11
Hipódromo	H3	0	371	0.705914	0.512583	−1.04	0.1370	1.13
Hipódromo	H4	0.1	347	0.706207	0.512577	−1.14	0.1319	1.07
Gorina	G1B	2.6	–	0.707624	0.512478	−3.08	0.1192	1.08
Gorina	G1C	2.5	–	0.707557	0.512460	−3.43	–	–
Gorina	G1F	2.1	–	0.708551	0.512500	−2.65	–	–
Gorina	G2	0.2	222	0.707701	0.512482	−3.00	0.1313	1.23
Gorina	G3	2.3	521	0.707617	0.512554	−1.61	0.1409	1.25
Gorina	G4	1.1	232	0.707592	0.512552	−1.64	0.1265	1.05
Baradero	B2	–	–	0.709927	0.512339	−5.79	0.1225	1.35
Baradero	B1	0.2	209	0.709612	0.512361	−5.37	0.1283	1.41
Baradero	B3	3.6	216	0.708987	0.512430	−4.02	0.1277	1.27
Baradero	B4	2.8	203	0.708950	0.512459	−3.46	0.1234	1.17
Baradero	B5	2.7	191	0.709387	0.512432	−3.99	0.1309	1.32
Lozada	L1	–	–	0.708089	0.512458	−3.47	0.1221	1.15
Lozada	L2	3.1	320	0.708270	0.512452	−3.59	0.1321	1.30
Lozada	L3	0.2	315	0.708135	0.512469	−3.26	0.1317	1.26
Lozada	L4	0.6	522	0.707904	0.512468	−3.27	0.1313	1.26
El Lambedero	E1	–	–	0.711894	0.512308	−6.40	0.1154	1.30
El Lambedero	E2	3.6	227	0.711274	0.512405	−4.51	0.1292	1.34
El Lambedero	E3	–	289	0.712331	0.512316	−6.25	0.1258	1.44
El Lambedero	E4	0.3	264	0.712134	0.512320	−6.17	0.1329	1.56

[a] Locations of sample sites as follows: Hipódromo – 37.57°S, 57.38°W; Gorina – 34.54°S, 58.01°W; Baradero – 33.47°S, 59.30°W; Lozada – 31.39°S, 64.64°W; El Lambedero – 28.70°S, 66.06°W.

[b] $CaCO_3$ concentrations measured using a calcitimeter [44].

[c] Precision is dominated by external reproducibility. For the period of these analyses, NIST SRM987 Sr gave $^{87}Sr/^{86}Sr = 0.710247 \pm 0.000011$ (two standard deviations of the mean, $n = 17$). The La Jolla Nd standard gave 0.511856 ± 0.000007 (two standard deviations of the mean). This is equivalent to an uncertainty on ε_{Nd} of 0.14.

[d] Derived from Nd and Sm concentrations measured by ICP-MS. Sr concentrations also measured by ICP-MS.

in this study. Goldstein et al. [35] found no difference in Nd isotopic composition between grain sizes ranging from <2 μm to >45 μm, while Derry and France-Lanord [38] observed differences of around 1–2 ε units between grain-size fractions <2 μm and <50 μm. Clearly, at these small grain sizes, differences in $\varepsilon_{\mathrm{Nd}}$ between size fractions are not very significant. The situation for Sr is more complicated. Dasch [34] compared the $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$ ratios of two grain-size fractions and the bulk material from the same sample and concluded that $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$ ratios may increase with decreasing grain size, due to the dominance of Rb-rich micas in the clay fraction. Derry and France-Lanord [38], on the other hand, found that the <50 μm fraction in sediments from the Bay of Bengal consistently contained more radiogenic Sr than the <2 μm fraction. However, there is some doubt as to whether the less radiogenic Sr in the fine fraction represents a true source characteristic or whether it resulted from alteration by pore waters in a submarine setting.

In this study, one sample (G1 from Gorina) was chosen to investigate the effects of grain size on isotopic values (Table 1 – samples G1B (bulk), G1F (fine) and G1C (coarse)). For the Gorina data it can be seen that: (1) the bulk sediment isotopic value is between that for the coarse and fine fractions; and (2) the difference between the bulk isotopic value and that for the coarse fraction is of the same order as that between different bulk samples from the same site. One of our principal aims is to constrain the source of the South American loess so that the bulk sample is the most appropriate one for us to use. For this reason the bulk sediment fraction was chosen for the rest of the analyses. For the purpose of investigating the source of the ice-core dust, however, it must be borne in mind that transport-related modification of the Sr isotopic ratios may occur. We show later (Fig. 7 below) that, in contrast to the case for Nd isotopes, there is good evidence for transport-related differentiation of Sr isotopes, so that finer material that is transported further has higher Sr isotopic ratios.

3.2. Rare earth elements

REE data for the five sites analysed are given in Table 2 and are shown as chondrite-normalised

Table 2
REE concentrations (ppm) of Argentinian loess as determined by ICP-MS

Site[a]	Sample	La	Ce	Pr	Nd	Sm	Eu	Gd	Dy	Ho	Er	Yb	Lu	$(La/Yb)_N$	Eu/Eu*
Hipódromo	H1	23.4	48.3	4.8	24.7	5.03	1.24	4.06	4.24	0.93	2.48	2.35	0.39	6.66	0.84
Hipódromo	H2	20.71	43.16	5.36	20.93	4.75	1.03	3.35	3.23	0.68	1.85	2.21	0.35	6.27	0.79
Hipódromo	H3	23.28	46.03	5.77	22.51	5.10	1.14	3.51	3.31	0.66	1.99	2.21	0.35	7.04	0.83
Hipódromo	H4	19.98	40.28	5.04	20.85	4.55	1.00	3.41	3.36	0.67	1.87	2.14	0.34	6.24	0.78
Gorina	G1B	29.1	59.7	5.9	28.3	5.58	1.22	4.34	4.41	0.93	2.45	2.32	0.40	8.39	0.76
Gorina	G2	23.20	47.88	5.90	22.75	4.94	1.01	3.69	3.70	0.71	2.04	2.41	0.38	6.44	0.73
Gorina	G3	22.05	48.60	5.89	23.17	5.40	0.99	3.91	3.63	0.72	2.01	2.40	0.37	6.14	0.66
Gorina	G4	18.82	47.39	5.57	22.12	4.63	0.91	3.46	3.29	0.65	1.81	2.13	0.35	5.91	0.70
Baradero	B2	31.2	65.9	6.5	30.7	6.22	1.31	4.92	5.01	1.04	2.91	2.63	0.46	7.93	0.73
Baradero	B1	25.57	52.48	6.24	25.12	5.33	0.98	4.01	3.98	0.80	2.24	2.61	0.38	6.55	0.65
Baradero	B3	26.70	53.31	6.78	27.60	5.83	1.04	4.32	4.12	0.80	2.36	2.75	0.39	6.49	0.64
Baradero	B4	28.87	53.44	7.17	29.74	6.07	1.15	4.86	4.68	0.92	2.59	3.03	0.45	6.37	0.65
Baradero	B5	27.02	56.28	7.01	27.02	5.85	1.13	4.52	4.20	0.82	2.35	2.76	0.41	6.55	0.68
Lozada	L1	30.1	62.0	6.1	29.5	5.96	1.28	4.77	4.84	1.00	2.76	2.58	0.43	7.80	0.74
Lozada	L2	25.99	53.96	6.63	25.67	5.61	1.02	4.12	3.84	0.78	2.16	2.62	0.40	6.63	0.65
Lozada	L3	25.24	53.55	6.65	25.61	5.58	1.03	4.00	3.84	0.74	2.18	2.46	0.39	6.86	0.67
Lozada	L4	24.95	51.88	6.49	25.09	5.45	1.04	3.94	3.81	0.75	2.16	2.42	0.40	6.89	0.69
El Lambedero	E1	37.5	81.7	8.0	37.0	7.06	1.44	5.53	5.18	1.05	2.81	2.55	0.41	9.83	0.71
El Lambedero	E2	36.27	69.97	9.32	36.12	7.72	1.26	5.73	5.35	1.05	2.96	3.21	0.50	7.56	0.58
El Lambedero	E3	33.42	68.84	8.26	31.09	6.47	1.20	5.05	4.58	0.90	2.58	2.81	0.45	7.95	0.65
El Lambedero	E4	29.95	61.52	7.44	27.06	5.95	1.05	4.60	4.35	0.84	2.41	2.77	0.45	7.23	0.62

[a] Locations of sample sites given in Table 1.

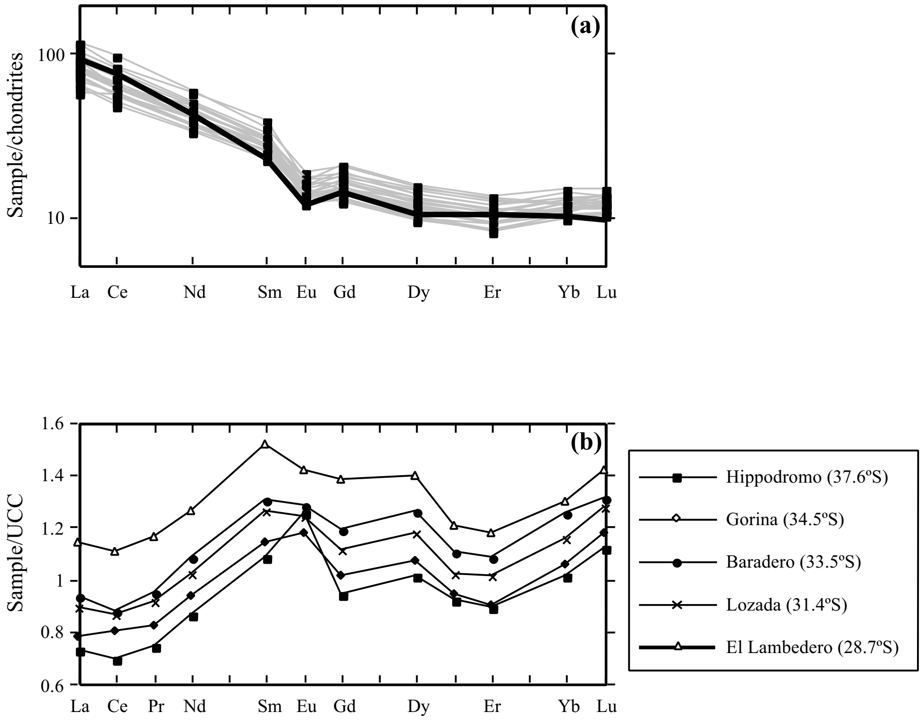

Fig. 3. (a) Chondrite-normalised (chondrite values from [39]) REE plots for all Argentinian loess samples analysed (black squares and grey lines). Also plotted for comparison (thick black line) are the UCC values of [40]. (b) Loess REE concentrations normalised to the UCC values of [40]. There are small but significant differences both between samples and UCC and between samples in the suite analysed here – see text for further discussion.

patterns in Fig. 3a. Generally, all samples are characterised by light REE (LREE) enriched patterns, relatively flat heavy REE (HREE) trends and negative europium anomalies. All samples are very similar to the average UCC (bold line in Fig. 3a) pattern of [40]. Furthermore, the patterns are generally consistent with loess deposits from other regions of the world [3–5,23].

In detail, however, there are subtle differences both between these samples as a group and other continental loess deposits as well as between individual sites within the sample set. Generally, there is a relationship between geographical location and LREE content (Fig. 3b) – with samples from the most southerly site (Hipódromo) having LREE contents below those for UCC and the most northerly site (El Lambedero) containing more LREE than UCC. These geographical trends also extend to the magnitude of the europium anomaly (Fig. 4, left-hand panel) and, to a lesser extent, to the degree of LREE enrichment as measured by $(La/Yb)_N$ (Fig. 4, right-hand panel). The small differences between the sample suite as a whole and other continental loess samples presumably reflect the greater influence of young volcanic source regions (Andes/Patagonia/Paraná) on South American loess, while the subtle differences between sites reflect the varying importance of this provenance relative to contributions from old continental crustal material.

One of the primary aims of this paper is to investigate the source of dust to Antarctica during glacial periods. Fig. 4, however, shows that the range in the Eu anomaly and $(La/Yb)_N$ ratio in this ice-core dust almost covers the entire range in the loess samples studied here. As a result we have to turn to the isotopic compositions of the loess samples to address this issue.

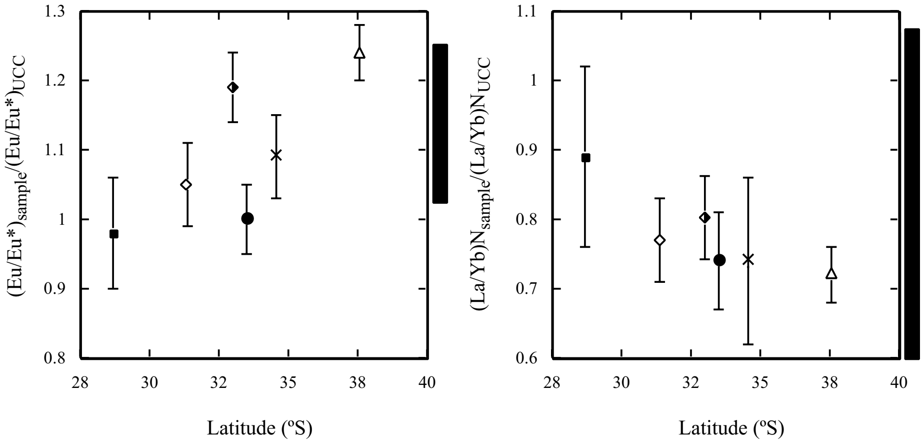

Fig. 4. Europium anomaly (Eu/Eu*) and $(La/Yb)_N$ of Argentinian loess, normalised to those for UCC, as a function of latitude. The data points represent the means for each location and the error bars one standard deviation. The half-filled diamond represents data for Buenos Aires loess from [23]. Other symbols as in Fig. 3. The most northerly samples have values identical to UCC but there is a progressive southward increase in Eu/Eu* accompanied by a less distinct decrease in $(La/Yb)_N$. The black bars at the right hand edge of each plot give the range for Antarctic ice-core dust (from Basile et al. [11] and I. Basile, personal communication).

3.3. Nd and Sr isotopes

The results of $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ isotopic analyses are given in Table 1 and graphically represented in Fig. 5. Samples are characterised by a broad range in $^{87}Sr/^{86}Sr$ (0.7059–0.7123) values and restricted $^{143}Nd/^{144}Nd$ ratios (0.5123–0.5126), but do show significant geographical variability (e.g. Hipódromo vs. El Lambedero). Individual sites, on the other hand, show less isotopic variability, implying uniform source regions throughout the period of deposition investigated (ca. 10–150 ka). The geographic pattern is consistent with results obtained from REE analysis, with the more northerly samples exhibiting the least radiogenic Nd and most radiogenic Sr values (e.g. El Lambedero), implying again a greater importance for a differentiated continental crustal provenance. In contrast, more southerly sites (e.g. Hipódromo) have the least radiogenic Sr and most radiogenic Nd, reflecting the greater contribution of a young volcanic source region. These differences are further reflected in the depleted mantle model ages (Table 1), which generally increase northwards from 1–1.1 Ga for Hipódromo to 1.3–1.6 Ga for El Lambedero.

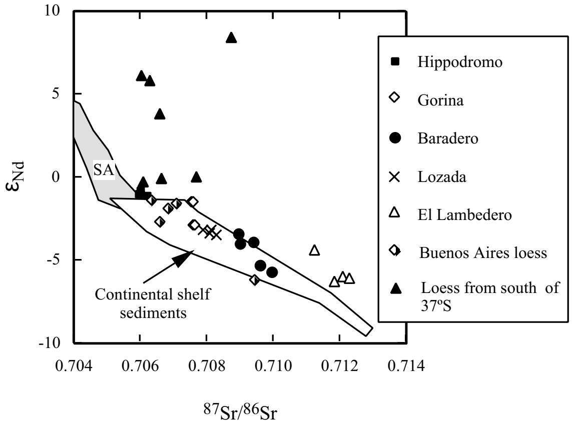

Fig. 5. Sr–Nd isotopic data for Argentinian loess compared to literature data for other South American loess and sediments and to data for southern Andean/Patagonian volcanics (field SA). Data for loess from south of 37°S (solid triangles; [10,11]) are distinct from loess from north of 37°S (all other data; this study and [23]). Data for South American and Falkland continental shelf sediments from [10,11]. Data for southern Andean/Patagonian lavas (34–54°S) from [41] and [42].

4. Discussion

4.1. Comparisons with other South American loess and sediment samples

It is generally accepted that Argentinian loess is characterised by a large proportion of volcanic minerals [29] and that the great majority of such minerals are derived from andesitic and basaltic rocks [13] of the southern Andes of Mendoza and Neuquen (Fig. 1). In detail, however, the origin of Pampean loess is still rather controversial, with some studies suggesting a single Andean/Patagonian source (e.g. [14]) while other data suggest that, in the central and northern part of Argentina especially, other sources might have contributed [20–22,29,45]. In the former view, although the primary transport of material down the Pampean plains would be via southwesterly winds, fluvial systems such as the Desaguadero River basin in Mendoza [46] and the Negro and Colorado rivers [16] would have supplied large amounts of fine-grained sediment to the Pampas of central Argentina. During glacial times these fine-grained clastic sediments would have then been driven northwards by southerly and southwesterly winds [16,24,47]. The general SW–NE decrease in grain size (e.g. [15]) lends support to the idea of a single Patagonian source, though grain size might be controlled by previous fluvial grain-size zonation [16] rather than exclusively aeolian granulometric differentiation.

In the north, the loessic mineral assemblage has been used to suggest a source in the Puna Altiplano and/or the Bolivian Andes [21]. At a regional scale, Iriondo [24] suggested that the source areas of the Chaco loess region, which is in the north, were located in the Andes Cordillera, including the Altiplano. In this model, northern winds deflated the material to the south, forming the loess cover. In contrast, Sayago et al. [15] have argued that because loess deposits in this area generally mantle the eastern sides of the main bedrock ridges, this must indicate winds from the south and southeast. The latter interpretation disagrees with observational data as well as present-day atmospheric circulation, which show a dominance of northerly winds during January [20]. Furthermore, geological evidence of Late Pleistocene palaeo-wind direction shows wind scoured ignimbrite ridges aligned in a NW–SE orientation from about 18 to 28°S [48]. On a more local scale, mineralogical studies of the clay, silt and sand fractions in the eastern and northeastern parts of the Santa Fe province indicate that the surficial loessic deposits, including alluvial material, originated in the Paraná River basin [29,49].

In general terms, the loess samples analysed in this study display nearly volcanogenic isotope values similar to those reported by [10,11] and [23]. Significant differences do exist, however. Fig. 5 compares isotopic values from this study with: (1) loess from the southern pampas and the Patagonian plateau (i.e. south of 37°S; [10,11]); (2) Buenos Aires loess [23]; and (3) Argentine continental shelf sediments (including one datum for Rio de la Plata mouth sediments; [11]).

It is immediately clear from this plot that there are significant differences between the Patagonia/Southern Pampa loess and loess samples from further north. The Nd isotopic signatures of the southern samples are very similar to volcanic rocks of the southern Andes and Patagonia, which crop out extensively between 34 and 54°S both in the Andes themselves and across the eastern foothills [41–43]. A Patagonian/southern Andean source for this loess is consistent with previous mineralogical and isotopic investigations, which have shown that the loess of the Pampas is composed primarily of pyroclastic material, and displays an isotopic signature which is dominated by volcanogenic material [10,11,20,23]. As noted by previous authors, the slight offset between the more Sr-radiogenic Patagonian loess samples and a potential Patagonian/southern Andean source is probably due either to the admixture of a small amount of material from another source or to a transport/grain-size differentiation effect [10].

Not surprisingly, our most southerly site, Hipódromo (southern Buenos Aires), is the one that is closest isotopically to the loess of the southern Pampas and Patagonia. Previous data [16] have suggested that dust was transported to the Hipódromo area from the lower reaches of the Rio Colorado and Negro, which both had their source in the then active ice cap of the Patagonia Andes (south of 38°S) and the alpine glaciers of the south central Andes (north of 38°S). Our results, therefore, support previous studies, which indicate a source rock that originated in Patagonia, subsequently reworked by fluvial and aeolian processes.

Fig. 5 also clearly shows that the rest of our sites, as well as those studied by Gallet et al. [23], are isotopically distinct from those of the southern Pampas/Patagonia and, therefore, cannot have been derived entirely from Patagonia (cf. [14]). Loess, isotopically, becomes progressively more unlike Patagonian/southern Andean basalts northward in a way that is reminiscent of changes in REE concentrations and patterns (Fig. 4). For example, loess from El Lambedero (Tucumán province) has much more radiogenic $^{87}Sr/^{86}Sr$ and less radiogenic $^{143}Nd/^{144}Nd$ than other samples.

4.2. Comparisons with other potential source areas

The isotopic data presented here place strong constraints on the source of South American loess. Fig. 5 shows isotopic data for South American loess in comparison to most of the potential source areas. A notable feature of the published dataset for Andean volcanics is the latitudinal gradients in Sr and Nd isotopic ratios. In general, southern Andean volcanic rocks have values that are close to those for the mantle, with little evidence for assimilation of continental crust. However, there is a progressive change northward so that the southern Andes occupy a separate field in Sr–Nd isotopic space (Fig. 6). The south central Andean field largely overlaps that for the Paraná basalts, though the latter is shifted towards slightly more radiogenic Sr. Finally, Uruguayan rhyolites and Palaeozoic granites from the Chilean Andes have very heterogeneous Sr and Nd isotopic values but are, overall, much more radiogenic in Sr and less radiogenic in Nd than Andean volcanics.

Clearly, the loess measured here and those of [23] do not lie in the field for Patagonian/southern Andean volcanics and Fig. 6 indicates that this source alone cannot explain the Sr–Nd data. The loess data, in fact, fall largely in the field for the south central Andes, though some samples have slightly more radiogenic Sr isotopic values that are more compatible with the Paraná field (cf. [18]). The fact that all the loess fall in or close

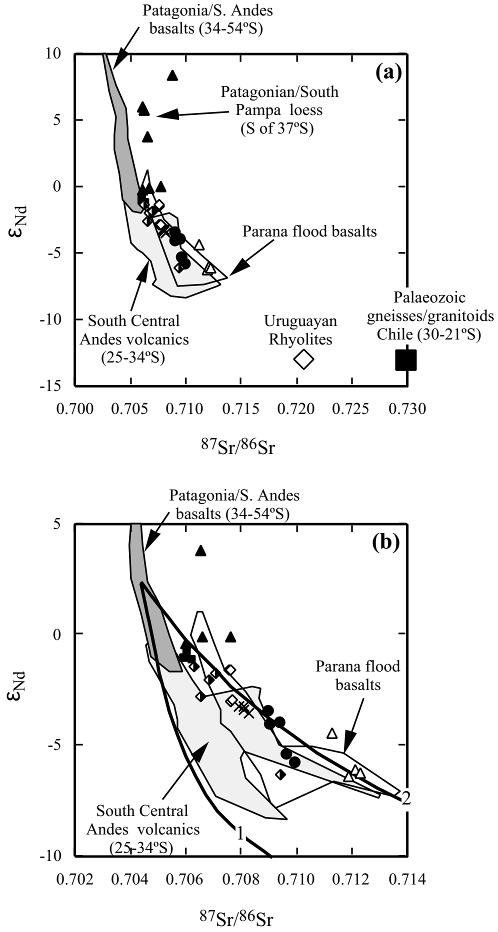

Fig. 6. (a) Data for South American loess (data sources and symbols from Figs. 3–5) compared to possible source areas. Data for possible source areas from: Patagonian/southern Andes volcanics (34–54°S) from [41,42,50]; south central Andes (25–34°S) from [42,51,52]); Paraná flood basalts from [53]; Uruguayan rhyolites (*average* plotted as open diamond) from [54]; Chilean (30–21°S) Palaeozoic gneisses and granitoids from [55] (*average* plotted as filled square). (b) Enlarged view of part of panel a. Heavy solid lines denote mixing between an average Patagonian/Andean volcanic and (1) an average Uruguayan rhyolite; (2) an average Palaeozoic granitoids/gneiss. These mixing hyperbolae were calculated using the measured Sr–Nd concentration in the source rocks [54,55].

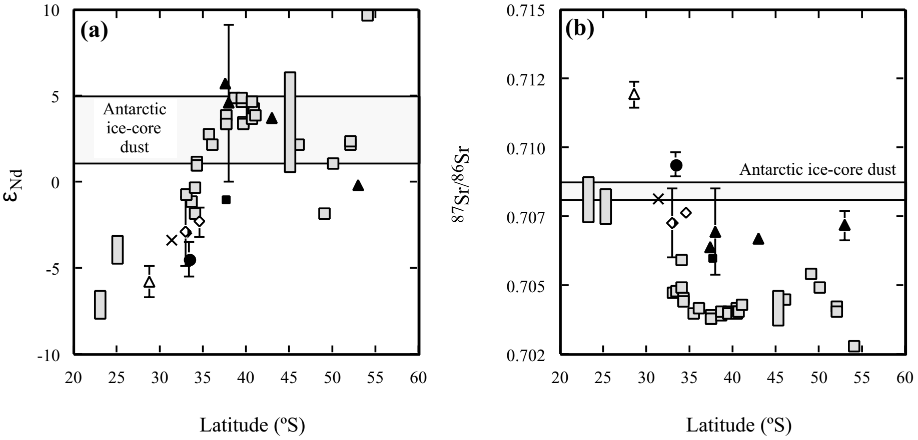

Fig. 7. Plot of (a) ε_{Nd} and (b) $^{87}Sr/^{86}Sr$ versus latitude for South American loess (averages and one standard deviation where more than one datum available, symbols as in Fig. 3, data sources for Buenos Aires and Patagonian loess as for Fig. 5) and Andean volcanics (heavy stippled boxes). Data sources for Andean volcanics as for Fig. 6. Light stippled bands give data for Antarctic ice-core dust (see text for discussion).

to the field for the south central Andes is strongly suggestive that they were derived from such a source. However, there remains the possibility that they are the result of mixing between a source with unradiogenic Sr and radiogenic Nd (e.g. Patagonia) and one with more radiogenic Sr and unradiogenic Nd – the two obvious candidates for the latter being Uruguayan rhyolites and the Palaeozoic gneisses/granitoids of the Chilean Andes. Fig. 6b shows the trends that would be produced by such mixing (see caption to Fig. 6 for details). Taken at face value, these mixing lines allow mixing of Patagonia with a source similar to a granitic Palaeozoic end-member but appear to preclude mixing with a source similar to Uruguayan rhyolites. However, given the possibility of shifts in the Sr isotopic ratio of the loess as a result of grain-size effects, a contribution from these rhyolites is still theoretically possible. However, there are two further observations that argue against such a scenario.

Fig. 7 shows ε_{Nd} and $^{87}Sr/^{86}Sr$ for South American loess plotted against latitude. Superimposed on this plot are the values for Andean volcanics. The striking observation from the Nd plot (Fig. 7a) is that the data for loess follow the trend for the volcanic rocks almost perfectly. The ε_{Nd} data for both loess and Andean volcanics rise from values around −8 at 20–25°S to a peak of around +5 at 40°S before declining again to around 0 at the southern extremity of the continent – through there is one outlier in the volcanic database of $\varepsilon_{Nd} = +10$ at 55°S. The faithfulness with which the loess replicates the bedrock values directly west of them is strong, albeit circumstantial, evidence for the loess being derived predominantly from the Andes by westerly transport processes – be they rivers, winds or some combination of both. If such a close match were to be produced by mixing of two end-members it would be wholly coincidental.

It is also worth noting here that, while the Sr isotopic data for loess show a pattern of variation with latitude that is similar to that for Andean volcanics, the loess data are shifted very significantly (>0.001) towards more radiogenic values. This feature, in combination with the very good match for Nd isotopes, again highlights the issue of grain-size differentiation and transport and its effect on Sr isotopic ratios of dust versus the source of that dust (e.g. [34,56], this study). This problem is particularly clearly highlighted here and requires caution in the interpretation of Sr isotopic data for dust in terms of a source region (cf. [10,56]).

The Nd data in Fig. 7 do not preclude some involvement of the Paraná basalts in the source of Argentinian loess because the Sr–Nd isotopic characteristics of the Paraná basalts are very similar to parts of the south central Andes. Neither, however, do they require such a contribution. This is further illustrated in Fig. 8, where the Sr isotopic composition of loess is plotted against the inverse of the Sr concentration and compared with values for the important sources. Mixing trends on this plot will be linear. The plot again clearly shows the importance of the south central Andes in producing the isotopic characteristics of the loess but, again, a role for the Paraná basalts is neither precluded nor required. However, the observation of key importance here is that the El Lambedero samples trend away from the south central Andes/Paraná field towards the field for Palaeozoic gneisses and granitoids in the Chilean Andes (30–21°S) and clearly suggest a contribution from this source to the El Lambedero site.

It is therefore apparent that a single Patagonian/southern Andean dust source for South American loess is much too simplistic. Our isotopic data are wholly consistent with the conclusion of Iriondo and Kröhling [46] and Morrás [29] that multiple sources are involved in the origin of the Pampean aeolian record, with a great majority of sediment originating in the Desaguadero River and the Colorado River basins, being ultimately derived from the Andes.

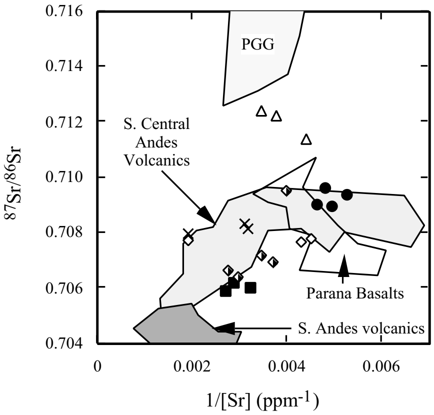

Fig. 8. 1/[Sr] versus $^{87}Sr/^{86}Sr$ for Argentinian loess studied here and the samples of [23]. Published data for potential source areas from references cited in caption to Fig. 6. The data for Palaeozoic gneisses and granitoids (PGG) are very heterogeneous and this field extends to higher 1/[Sr] and very high $^{87}Sr/^{86}Sr$. Many of the loess samples fall close to the data for volcanics from the south central Andes but the El Lambedero data require a contribution from a source similar to Palaeozoic gneisses and granitoids outcropping at 21–30°S in the Chilean Andes.

At the present day, the prevailing wind direction along the eastern side of the Andes, between 27 and 37°S, is from the south. To the east, in the Pampas, northerly and northeasterly winds prevail throughout the year. However, the northward displacement of the subtropical high-pressure belt in winter means that westerly winds dominate during the winter in the southern Pampas. In the summer, easterly to southeasterly winds are characteristic in the northern Pampas [57]. The close correspondence between the latitudinal changes in ε_{Nd} for volcanics and loess does not permit much of a southerly component to the transport direction. However, it has been suggested (e.g. [58]) that there was a significant northward movement in the Polar Front in the Southern Hemisphere during recent glacial maxima leading to more intense and frequent westerly winds, that now blow over Patagonia, in the entire Pampean region under consideration here [14,57–61]. The data presented here support that suggestion and, in this case, transport of dust eastward from the Andes into the Argentinian pampas seems straightforward to achieve.

It is also well known [62] that strong winds from the Altiplano blow down-slope at the present day in a south-southeastward direction. It is likely that such winds have existed throughout the Quaternary, augmented during glacial times by katabatic winds generated by the mountain ice caps [20]. Delivery of dust to the plains to the south and southeast of the Altiplano would have involved the subtropical jet stream, which crosses the Andes at about 25° [20]. Geological evidence of these winds during the Late Pleistocene period includes impressive wind-scoured plateau surfaces (e.g. Cerro Galan) and sand dunes, both of which show a southerly wind direction [48]. These data support the suggestion made here, that loess at El Lambedero contains material derived from Palaeozoic granitoids and gneisses in the Chilean Andes at 21–30°S.

4.3. Implications for dust found in East Antarctica and atmospheric circulation during the Last Glacial Maximum (LGM)

It is now well established from ice-core studies that the concentration of dust in the Antarctic atmosphere increased by a factor of 10–100 during Quaternary glacial maxima (e.g. [9]). However, the reasons for this increase are still unclear, with all attempts to produce it by atmospheric general circulation models (AGCMs) ending in failure (e.g. [6,63,64]). Early work (e.g. [64]) suggested that the transport efficiency of the atmosphere during glacial maxima was not much higher than the present-day and that any increase in Southern Hemisphere atmospheric dust loading had to arise from an expansion of the source areas. Subsequent work [6], however, using model-calculated soil moisture contents to identify source areas for dust (and which did predict expansions in source areas), similarly failed to produce the large increases in atmospheric dust content implied by ice-core data. Even the most recent AGCMs [63] do not simulate the increase in Antarctic atmospheric dust. It was noted by Grousset et al. [10] that an essential observational constraint on these models was the identification of the continental source of the dust and these authors used Sr–Nd isotopic analyses of ice-core dust and loess from around the Southern Hemisphere to show that such a source had to lie in Patagonia.

Though the isotopic data of Grousset et al. [10] and Basile et al. [11] are compelling in the identification of Patagonia as the source of ice-core dust, there is a residual concern over the small size of such a source area. Grousset et al. [10] noted that the Argentinian continental shelf, exposed during the lowered sea level of glacial maxima, could double the size of a Patagonian dust source but Basile et al. [11] showed that the shelf sediments had Sr–Nd isotopic characteristics that were incompatible with this possibility.

Transport of dust to Antarctica is likely to be achieved by deflation from loess sites by westerly winds, followed by entrainment in the circumpolar vortex, the cyclonic circulation cell centred on Antarctica [11,61]. At the present day, this westerly dominated Patagonian climate extends between 40 and 56°S. However, there is ample evidence that this westerly belt migrated northward during the LGM, perhaps as far as 30°S, bringing a dry, windy climate to an extended region to the north of Patagonia [14,61,65,66]. Moreover, the circumpolar vortex is controlled by the location of the Antarctic Polar Front, which itself depends on sea-surface temperatures and sea-ice distribution in the Southern Ocean, and is likely to have moved northward during Quaternary glacial maxima [58]. A further possibility, then, is that dust deposited in Antarctica during the LGM was derived from a much expanded region of South America beyond and to the north of Patagonia.

This possibility is explored in Fig. 7a, which, in addition to the Nd isotopic data for loess and Andean volcanics, shows (horizontal stippled band) the existing data for ice-core dust from East Antarctica [10,11]. It is clear from this figure that the loess measured here from latitudes north of about 37°S cannot be a significant contributor to Antarctic dust. In fact, the dust can only come from Patagonia/Southern Pampas (i.e. south of 37°S), with Nd isotopic characteristics as originally identified by the analyses of Grousset et al. [10]. The Sr isotopic data in Fig. 7b would, at face value, seem to be incompatible with the Nd data and to suggest that Antarctic dust could be derived from South American loess at 30–35°S. However, a grain-size differentiation effect is well established for Sr, and though the direction of such a shift is still not certain ([38], this study) it is highly likely that the incompatibility between Figs. 7a,b is caused by grain size. The data presented here for one sample suggest that these loess contain more radiogenic Sr in their fine fractions. More importantly, perhaps, the data in Fig. 7 strongly suggest that the Sr isotopic signature of the loess has been shifted towards more radiogenic values during transport from the loess source regions. In that case, there is a strong likelihood of further changes in the Sr isotopic composition during transport of the dust to Antarctica that would make the Nd and Sr data mutually compatible. In fact, this diagram suggests very strongly that the slight offset in Sr isotopic composition between Patagonian/Southern Pampas loess and ice-core dust is wholly due to a transport/grain-size effect rather than necessarily requiring the admixture of small proportions of African/Australian [11] or Pampean [61] material to ice-core dust (cf. [11]). Verification of this idea, however, would require further analysis of South American loess at grain sizes equivalent to those recovered from the ice cores themselves (e.g. [67]).

5. Summary

On the basis of this geochemical and isotopic study of Argentinean loess, we can make the following conclusions:

1. A combined isotopic (Sr–Nd) and geochemical (REE) approach provides a powerful tool with which to trace loess sources at a reasonably detailed level. Sr–Nd isotope and REE values do not vary significantly between samples at individual sites, suggesting relatively uniform source regions for the loess, at least through the Late Pleistocene.
2. Argentinian loess from latitudes north of about 37°S has very different isotopic characteristics from Patagonian and Southern Pampean aeolian sediments.
3. A single Patagonian source for Argentinian loess is precluded by our data. Loess in South America has Nd isotopic characteristics very similar to Andean/Patagonian volcanics directly west of the sites studied, an observation that is wholly consistent with westerly transport processes. However, the El Lambedero site in the north has a significant component of material from old crustal rocks to the northwest in the Chilean Altiplano. A small contribution to some loess samples from the Paraná Basalt province is not excluded by our data.
4. Loess from north of about 35–37°S cannot contribute significantly to dust in Antarctic ice cores. Patagonian loess is the only possible source of this dust yet identified. Sr isotopic data presented here show latitudinal patterns that are similar to those for Nd but with a significant offset to more radiogenic values that suggests changes in the signature during transport. As suggested by Basile et al.[11], this is the most likely explanation for the slight offset in Sr isotopic values between Patagonian loess and ice-core dust.

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