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# Provenance of dust in the Pacific Ocean

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## ABSTRACT

Eolian dust preserved in deep-sea sediment cores provides a valuable indicator of past atmospheric circulation and continental paleoclimate. In order to identify the provenance of eolian dust, Nd and Sr isotopic compositions and Rb, Sr and rare earth element (REE) concentrations have been determined for the silicate fractions of deep-sea sediments from the north and central Pacific Ocean. Different regions of the Pacific Ocean are characterized by distinct air-borne inputs, producing a large range in  $\epsilon_{\text{Nd}}$  (–10 to +1),  $^{87}\text{Sr}/^{86}\text{Sr}$  (0.705–0.721), La/Yb (5–15),  $\text{Eu}_{\text{N}}/\text{Eu}_{\text{N}}^*$  (0.6–1.0) and Sr/Nd (4–33). The average Nd isotopic composition of Pacific deep-sea sediments ( $\epsilon_{\text{Nd}} = -6$ ), is more radiogenic than the average from the Atlantic ( $\epsilon_{\text{Nd}} = -8$ ). In contrast, the average  $^{147}\text{Sm}/^{144}\text{Nd}$  ratio for Pacific sediments (0.114) is identical to that of Atlantic sediments and to that of global average riverine suspended material. The values of  $\epsilon_{\text{Nd}}$  and  $^{147}\text{Sm}/^{144}\text{Nd}$  are positively correlated for the Pacific samples but negatively correlated for Atlantic samples, reflecting a fundamental difference between the dominant components in the end members with radiogenic Nd (island-arc components in the Pacific and LREE-enriched intraplate ocean island components in the Atlantic). Samples from the north central Pacific have distinctive unradiogenic  $\epsilon_{\text{Nd}}$  values of –10,  $^{87}\text{Sr}/^{86}\text{Sr} > 0.715$ , high La/Yb (> 12), and low  $\text{Eu}_{\text{N}}/\text{Eu}_{\text{N}}^*$  (0.6) and Sr/Nd (3–6). These data are virtually identical to the values for loess from Asia and endorse the use of these sediments as indicators of Asian paleoclimate and paleowind directions. Island-arc contributions appear to dominate in the northwest Pacific, resulting in higher  $\epsilon_{\text{Nd}}$  (–1 to +1) and lower  $^{87}\text{Sr}/^{86}\text{Sr}$  ( $\approx 0.705$ ) and La/Yb ( $\approx 5$ ). Sediments from the eastern Pacific tend to have intermediate Sr and Nd isotopic compositions but regionally variable Sr/Nd and REE patterns; they appear to be derived from the west margin of the North and South American continents, rather than from Asia. Our results confirm that dust provenance can be constrained by isotopic and geochemical analyses, which will facilitate reconstructions of past atmospheric circulation and continental paleoclimate.

## 1. Introduction

Eolian dust preserved in deep-sea sediment cores provides a valuable indicator of past atmospheric circulation and continental paleoclimate [1,2]. Early studies of deep-sea surface sediments used variations in the compositions and petrographic characteristics of clay and quartz to deduce provenance and transport mechanisms. Biscaye [3] and Zimmerman [4] investigated clay mineralogy in the Atlantic Ocean and concluded that fluvial processes were the most important

transport mechanism for Atlantic deep-sea sediments. In contrast, Griffin et al. [5] and Rateev et al. [6] studied the mineralogic composition of north Atlantic sediments and concluded that eolian transport by westerlies was dominant. Grousset and Chesselet [7] combined mineralogic, chemical and Sr isotopic data of north Atlantic deep-sea sediments and identified two main transport mechanisms, bottom currents that carry particles from Iceland southward and eolian transport which conveys particles from North America. Although Sr isotopes are powerful tracers in provenance studies, Dasch [8] and Biscaye [9] showed that Sr isotopic ratios, as with mineralogic compositions, can be a function of grain size and therefore less provenance specific than desired. However, the Nd isotopic compositions

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and REE patterns of sediments are thought to be less influenced by the processes of weathering, erosion and sedimentary sorting.

While many Nd isotopic studies have been made of clastic fluvial sediments, carbonates, phosphates, Mn nodules and seawater [10–19], only a few Nd studies of atmospheric particulates have been published [20–22]. The study of continental loess by Taylor and coworkers [22] noted the surprising global uniformity in rare earth element (REE) patterns for loess samples from Europe, China, the U.S.A. and New Zealand, while also noting that loess was isotopically uniform on a regional scale. Goldstein et al. [20] studied particulate contributions to the oceans but included minimal data for the Pacific and the Indian oceans and focussed on samples close to the continents to allow comparison with river inputs. This study also included data for Mount St. Helens ash and the Malan loess from China. Grousset et al. [21] studied geographic variations in Nd isotopic compositions in particulates from the Atlantic Ocean. The data were explained in terms of mixing between eolian dust characterized by unradiogenic Nd derived from locations such as the Saharan desert and volcanic ashes characterized by radiogenic Nd supplied by recent ocean island volcanism. In this paper we report a reconnaissance study of Nd and Sr isotopic and Rb, Sr and REE compositions in the silicate fraction of pelagic sediments from the Pacific Ocean, conducted with a view to evaluating the provenance of the various components.

Dust transport is seasonal, with nearly all of the annual flux occurring in conjunction with one or two large spring storms. Nearly two decades of satellite imagery have allowed atmospheric scientists to track individual storms and determine transport paths, resulting in an unambiguous dataset [23–27]. Thus we know that dust from Asia dominates the entire north Pacific north of the Intertropical Convergence Zone (ITCZ) at an average latitude of about 7°N [1,28,29]. Dust of American provenance is of limited geographic extent in the north Pacific, and generally only occurs beneath the tradewinds in the region south of Mexico and west of Central America [30]. Part of any provenance study is to better document the regional extent of American source materials in the north Pacific.

The important sedimentary process that can confuse or mask the eolian signal is hemipelagic deposition. Hemipelagic deposits are those fine-grained muds deposited within hundreds of kilometers of the continental margin. These muds are derived from winnowing of the continental shelf and upper slope and move offshore within the water column at depths of 2–3 km in a plume or cloud of limited vertical extent. Resulting deposits blanket continental slopes and regions of the seafloor adjacent to continents. The horizontal advection of terrigenous material has been well documented by sediment trap studies and has been shown to be important commonly to 500 km and rarely to as much as 900 km offshore [8,31–33]. Surface and bottom current redistribution of pelagic sediments in the Pacific is significant only at a few but nevertheless well-known localities [1,34–37].

Atmospheric processes of dust transport are better understood than the seafloor sedimentary record of these processes. Even though Asian dust may reach North America, rapidly accumulating hemipelagic muds from the American continent mask the much more slowly accumulating dust for some unknown hundreds of kilometers west of the coast. Determination of the longitude of the transition from American hemipelagic mud to Asian dust in seafloor surface sediments is a major goal of any provenance study. A complication is that those Pacific rim volcanoes that lie beneath the prevailing westerlies contribute significant amounts of ash to the seafloor in regions immediately downwind. Ash from Japan, the Kuriles and Kamchatka is an important but not yet well quantified component of northwestern Pacific pelagic sediment [38,39]. Aleutian-derived ash is less volumetric than that from Kamchatka, but is important in sediments of the Gulf of Alaska. Products of lower latitude West Pacific volcanic activity are carried farther west by the tradewinds.

Fluxes of dust in the Pacific basin span three orders of magnitude, from over 1000 mg (cm<sup>2</sup> ky)<sup>-1</sup> near the Asian source region to less than 1 mg (cm<sup>2</sup> ky)<sup>-1</sup> in the southern subtropical Pacific. There is a continuous decline in fluxes away from Asia and an order of magnitude dropoff in dust fluxes from north to south across the ITCZ [29]. South of the ITCZ, South America provides

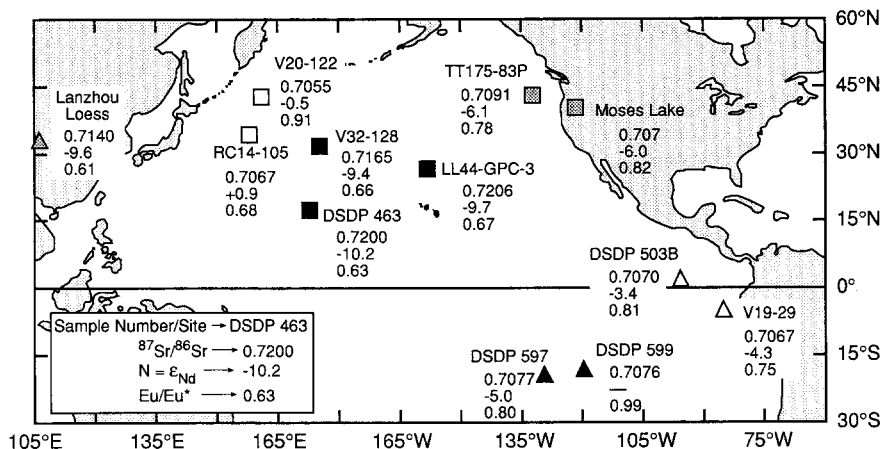


Fig. 1. Map of the Pacific Ocean with Nd isotopic compositions (expressed in  $\epsilon_{Nd}$  units) for silicate fraction of Pacific deep-sea sediments.

a modest amount of dust to the equatorial Pacific [40–42]. Australia, like other large and hyperarid regions such as the core Sahara and parts of the Kalahari and Namib deserts of southern Africa, is essentially deflated [43,44] and contributes no dust to the subtropical Pacific [45,46] and very little to the Indian Ocean [47]. In the southwest Pacific, westerlies may transport some material from Australia and New Zealand to nearby oceanic areas [37,48].

A number of studies [49–52] have attested to the fact that the size of wind-blown grains changes considerably in the first 1000 km of transport and not much beyond this distance. Thus any study of the provenance of dust in oceanic regions needs to include an evaluation of the effects of sorting on the trace element and isotope geochemistry. Since this process is dominant close to the continental source a comparison between continental and oceanic samples is a particularly valuable test.

**2. Sample selection and analytical techniques**

The samples used for this study were collected from ten locations in the northern and central Pacific for various marine sedimentary studies at the University of Michigan over the past years (Fig. 1 and Table 1). Most samples were taken at considerable distances from the continental margin, but two, V19-29 and TT175-83P, are close enough to the Americas so that their mineral component is hemipelagic rather than eolian. All

samples are of Quaternary age, although only those from V20-122, RC14-105, TT175-83P and V32-128 are unambiguously Holocene in age. The samples from V19-29 and DSDP 503B are from the last glacial stage and are about 20 and 32 ky old respectively. LL44-GPC3 is characterized by red clays that have slowly accumulated, so the sample from this core is pre-Holocene. It is unlikely that the cores from DSDP sites 463, 597 and 599 have recovered Holocene sediment at their tops. The analytical results, therefore, can be interpreted only in terms of general Quater-

TABLE 1  
Details of samples

Core	Location	Depth in core/ other information
V20-122	46.3°N, 161.4°E	26–28 cm
RC14-105	39.4°N, 157.3°E	34–35 cm
TT175-83P	46.3°N, 131.7°W	0–3 cm
V19-29	3.6°S, 83.9°W	131–133.5 cm
DSDP 503B	4.1°N, 95.6°W	48–50 cm
V32-128	36.5°N, 177.2°E	10 cm
LL44-GPC3	30.3°N, 157.8°W	73 cm
DSDP 463	21.4°N, 174.7°E	20–24 cm
DSDP 597	18.8°S, 129.8°W	14–18 cm
DSDP 599	19.5°S, 119.9°W	11–14 cm
Moses Lake	eastern Washington State	1.5–2 cm below ground surface after 1980 Mt. St. Helens ash fall
Lanzhou loess	central China	from outcrop in China

nary sediment transport processes rather than specifically Holocene processes.

The mineral component of these samples was isolated by a series of chemical extractions that sequentially removes carbonates using acetic acid, opal using sodium carbonate, and oxides and hydroxides using a strong reducing agent. This procedure is essentially the standard extraction process used for decades by those studying the mineralogy of deep-sea sediments [cf. 53] and is described in detail by Rea and Janecek [54]. This procedure effectively removes all hydrothermal components with the oxides and hydroxides from the sample but does not remove volcanic ash. The mineral residues after the extraction process are presumed to be the eolian (or hemipelagic) component of the pelagic sediments, the operationally defined eolian dust or ODED, and were used for geochemical and isotopic analysis. In any sample of eolian sediment, the clay minerals are finer grained than the silt-sized quartz and feldspar grains that are transported with them, so there is a compositional difference between size fractions. Most mineralogy studies, for analytical

reasons, have concentrated on the  $< 2 \mu\text{m}$  diameter size fraction dominated by clay minerals and thus have under-represented the naturally coarser grains of quartz and feldspar. Different geochemical and isotopic studies of the mineral component of pelagic sediments have studied different size fractions, so studies are only generally comparable. Here we analyze the entire extracted component that is used in eolian studies of paleoclimatology.

We have also analyzed a sample of loess from Lanzhou, China and a soil sample from Moses Lake (a playa) near Mt. St. Helens to compliment existing data for circum-Pacific loess and Mt. St. Helens volcanic products, facilitating the direct comparison between typical continental airborne materials and the oceanic sediments we have analyzed.

Standard ion-exchange procedures were used to separate Sr and Nd from the ODED for isotopic analysis [55]. Rb, Sr and all the REEs were determined by isotope dilution (ID) and were separated from matrix elements using 3 ml of AG50W-X8 cation exchange resin. The REEs

TABLE 2a

Trace element abundances (ppm) on the samples analyzed in this work. (Uncertainties in concentrations of all elements  $< 1\%$ )

Sample	La	Ce	Nd	Sm	Eu	Gd	Dy	Er	Yb	Lu	Rb	Sr
Lanzhou Loess	35.7	71.7	29.7	6.00	1.15	5.34	4.71	2.69	2.50	0.371	83.2	300
RC14-105	18.3	36.2	18.5	4.14	0.947	4.41	4.92	3.37	3.52	0.562	n.d.	185
V20-122	12.1	25.6	12.3	2.79	0.850	2.97	3.36	2.29	2.27	0.359	45.3	243
V32-128	31.2	64.5	24.8	4.43	0.900	3.81	3.86	2.49	2.38	0.369	112	139
DSDP 463	45.1	78.5	35.3	6.24	1.22	5.35	5.32	3.26	3.07	0.476	161	133
LL44-GPC-3	28.9	56.4	21.9	3.74	0.760	3.14	3.39	2.30	2.30	0.362	148	123
Moses Lake	22.9	48.6	23.7	4.96	1.29	4.66	4.05	2.30	2.01	0.310	57.8	358
TT175-83P	21.4	40.5	18.5	3.61	0.857	3.05	3.04	1.96	1.88	0.301	84.9	208
DSDP 503B	15.5	28.6	12.7	2.34	0.578	1.98	1.89	1.20	1.20	0.184	51.5	279
V19-29	10.1	20.1	9.12	1.74	0.409	1.58	1.57	1.07	1.09	0.175	55.8	120
DSDP 597	9.46	19.2	7.68	1.35	0.325	1.10	1.28	0.942	0.988	0.158	65.3	253
DSDP599	9.66	19.0	7.42	1.34	0.412	1.18	1.26	0.842	0.862	0.140	40.1	161

were divided into four fractions (La, Ce and Nd, Sm to Dy, and Er to Lu) using the PTFE-HDEHP method. La was measured with Ta single filaments as  $\text{LaO}^+$  and other REEs were measured with Ta-Re-Ta triple filaments. All measurements were made on a VG Sector thermal ionization multicollector mass spectrometer. Sr and Nd isotopic compositions were acquired using multidynamic analysis. All ID measurements used static analysis. During the course of this study, the average  $^{87}\text{Sr}/^{86}\text{Sr}$  of NIST SRM-987 was  $0.710260 \pm 23$  ( $2\sigma$ ,  $N=34$ ), and the average  $^{143}\text{Nd}/^{144}\text{Nd}$  of the La Jolla Nd standard was  $0.511864 \pm 19$  ( $2\sigma$ ,  $N=18$ ).

### 3. Results and discussion

#### 3.1 The Nd isotopic composition of Pacific dust

The concentrations of Rb, Sr and REEs, together with the Sr and Nd isotopic data, are presented in Table 2. The Nd isotopic composi-

tions of the oceanic samples are highly variable across the Pacific Ocean, with  $\epsilon_{\text{Nd}}$  ranging from  $-10$  to  $+1$  (Table 2b and Fig. 1). The samples near Japan have the highest  $\epsilon_{\text{Nd}}$ , and samples near Central America have relatively high  $\epsilon_{\text{Nd}}$ . The sample from Moses Lake is clearly very different from Mt. St. Helens volcanic products [20,56], but identical to sample TT175-83P from the nearby eastern Pacific. The Nd data for Lanzhou loess are virtually identical to those for Malan loess [20] and Nanking loess [22]. All of these Asian loess samples have  $\epsilon_{\text{Nd}}$  values of  $-10$ . It is notable that all three samples from the central north Pacific contain the least radiogenic Nd, identical to that of loess ( $\epsilon_{\text{Nd}} = -10$ ). These results indicate that substantial quantities of eolian particles are transported across the Pacific from Asia and dominate the silicate fractions of deep-sea sediments over much of the north central Pacific. It is clear that sorting does not significantly affect the Nd isotopic compositions since the Nd isotopic composition of Asian loess is

TABLE 2b  
Sr and Nd isotopic data

Sample	$^{87}\text{Rb}/^{86}\text{Sr}$	$^{87}\text{Sr}/^{86}\text{Sr}^b$	$T_{\text{SrDM}}(\text{Ma})^c$	$^{147}\text{Sm}/^{144}\text{Nd}$	$^{143}\text{Nd}/^{144}\text{Nd}^d$	$\epsilon_{\text{Nd}}^e$	$T_{\text{NdDM}}(\text{Ma})^f$
Lanzhou Loess	0.799	$0.714041 \pm 10$	1075	0.1181	$0.512147 \pm 8$	-9.6	1599
RC14-105	n.d.	$0.706732 \pm 11$	n.d.	0.1355	$0.512683 \pm 11$	0.9	913.4
V20-122	0.537	$0.705495 \pm 10$	472	0.1368	$0.512613 \pm 10$	-0.5	1067
V32-128	2.49	$0.716516 \pm 13$	410	0.1080	$0.512156 \pm 11$	-9.4	1434
DSDP 463	3.48	$0.719954 \pm 10$	362	0.1069	$0.512113 \pm 8$	-10.2	1325
LL44-GPC-3	3.45	$0.720601 \pm 12$	379	0.1035	$0.512141 \pm 10$	-9.7	1396
Moses Lake	0.466	$0.707322 \pm 11$	832	0.1264	$0.512329 \pm 12$	-6.0	1434
TT175-83P	1.18	$0.709140 \pm 13$	432	0.1178	$0.512324 \pm 12$	-6.1	1315
DSDP 503B	0.531	$0.706957 \pm 13$	676	0.1115	$0.512462 \pm 11$	-3.4	1028
V19-29	1.33	$0.706722 \pm 10$	251	0.1153	$0.512420 \pm 11$	-4.3	1133
DSDP 597	0.741	$0.707724 \pm 13$	554	0.1060	$0.512384 \pm 14$	-5.0	937.9
DSDP599	0.719	$0.707619 \pm 17$	566	0.1094	n.d.	n.d.	n.d.

<sup>a</sup> Reported errors for isotopic ratios are  $2\sigma$  of the mean; <sup>b</sup> data normalized to  $^{86}\text{Sr}/^{88}\text{Sr} = 0.1194$ ; <sup>c</sup> calculated using present-day depleted mantle values of  $^{87}\text{Sr}/^{86}\text{Sr} = 0.702$  and  $^{87}\text{Rb}/^{86}\text{Sr} = 0.02$ ; <sup>d</sup> data normalized to  $^{144}\text{Nd}/^{146}\text{Nd} = 0.7219$ ; <sup>e</sup> deviations in parts in  $10^4$  from the present-day bulk earth value of  $^{143}\text{Nd}/^{144}\text{Nd} = 0.512638$ ; <sup>f</sup> calculated using present-day depleted mantle values of  $^{143}\text{Nd}/^{144}\text{Nd} = 0.513151$  and  $^{147}\text{Sm}/^{144}\text{Nd} = 0.21362$

uniform and that of the central north Pacific is also uniform and identical to that of Asian loess.

The  $\epsilon_{Nd}$  values and Sm-Nd crustal residence ages [57] of the samples are plotted against  $^{147}\text{Sm}/^{144}\text{Nd}$  in Fig. 2, together with results for eolian particles from other oceans [20,21], continental loess and Mt. St. Helens products. The  $^{147}\text{Sm}/^{144}\text{Nd}$  ratios of Pacific samples range from 0.104 to 0.137, with a mean of 0.114. With the exception of one sample with an extremely high value, the ratios for Atlantic samples [21] have an almost identical range (0.095–0.135) and an iden-

tical mean (0.114). These averages are also identical to average exposed continental crust, estimated at 0.114 by Goldstein and Jacobsen [58] on the basis of ratios in river water dissolved and suspended material, and the mean of  $0.116 \pm 0.004$  for loess samples reported by Taylor et al. [22]. Therefore in this respect the oceanic samples appear to be representative of exposed continental crust and equivalent to continental loess.

The Atlantic samples have  $\epsilon_{Nd}$  values that are, on average, more negative than those of Pacific samples. In contrast to the Pacific  $\epsilon_{Nd}$  values, which are never less radiogenic than Asian loess, only two Atlantic samples have  $\epsilon_{Nd}$  values that are significantly *more positive* than Asian loess. This reflects the contributions from older cratons surrounding the Atlantic Ocean. When the isotopic data are compared with the Sm/Nd ratios, distinct features emerge for each ocean. The  $\epsilon_{Nd}$  of the samples from the Pacific Ocean are positively correlated with  $^{147}\text{Sm}/^{144}\text{Nd}$  ratios, in contrast to the negative trend for the Atlantic samples (Fig. 2a). Similarly, the Atlantic crustal residence ages increase with Sm/Nd, in contrast to the modest negative trend displayed by the Pacific data (Fig. 2b). The crustal residence ages of loess samples from around the world do not correlate positively or negatively with the Sm/Nd ratios, which are relatively constant [22]. As such, the Nd isotopic data for the components sampled in the deep-sea sediments cannot be explained simply in terms of mixtures of continental loess with different crustal residence ages. Other components with very different REE compositions to that of loess are involved. The most likely sources of these components are young volcanoes, contributing either direct inputs of volcanogenic ash or erosional products.

The distinct features of each ocean can be explained by considering the different end-member components in the two oceans. As Grousset et al. [21] pointed out, ocean islands such as the Azores and Iceland supply ash and erosional products with radiogenic Nd and low  $^{147}\text{Sm}/^{144}\text{Nd}$  ratios to the Atlantic Ocean. In the case of the Pacific samples, volcanic products from island arcs are an important end member. The young volcanic products with radiogenic Nd have mixed with circum-Pacific crustal materials that are younger on average than circum-Atlantic crust.

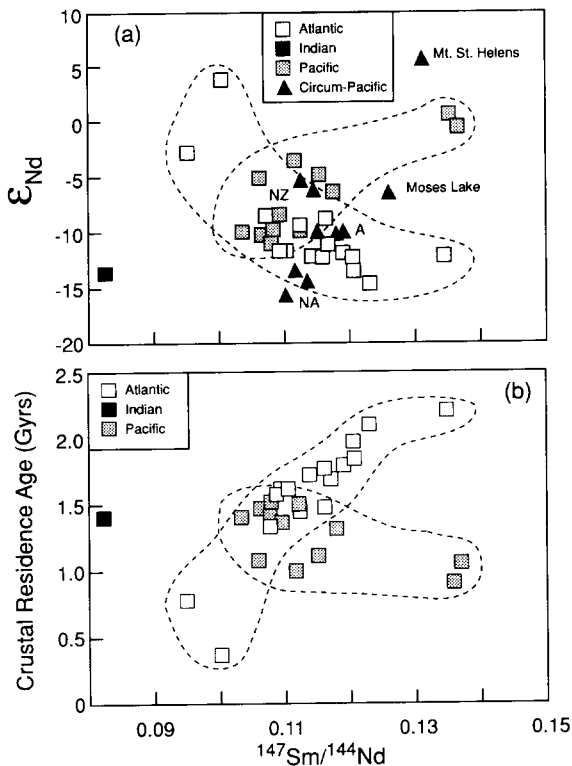


Fig. 2. Comparison between the Sm-Nd isotopic data for particulates from the Pacific (mainly this study), Atlantic [20,21] and Indian [20] Ocean. Excluding one sample from the Atlantic with an anomalously high Sm/Nd ratio [21] the average Sm/Nd ratios of Atlantic and Pacific particulates are very similar. However, the relationships with Nd isotopic composition and crustal residence age are distinct, implicating different components in the two oceans. The single datum for the Indian Ocean plots outside the fields for both the Atlantic and Pacific Ocean. The data for loess [20,22,this study] display regional differences in Nd isotopic composition but show no relationship between Nd isotopic composition and Sm/Nd, which is relatively uniform. *A* = Asian loess; *NZ* = New Zealand loess; *NA* = North American loess.

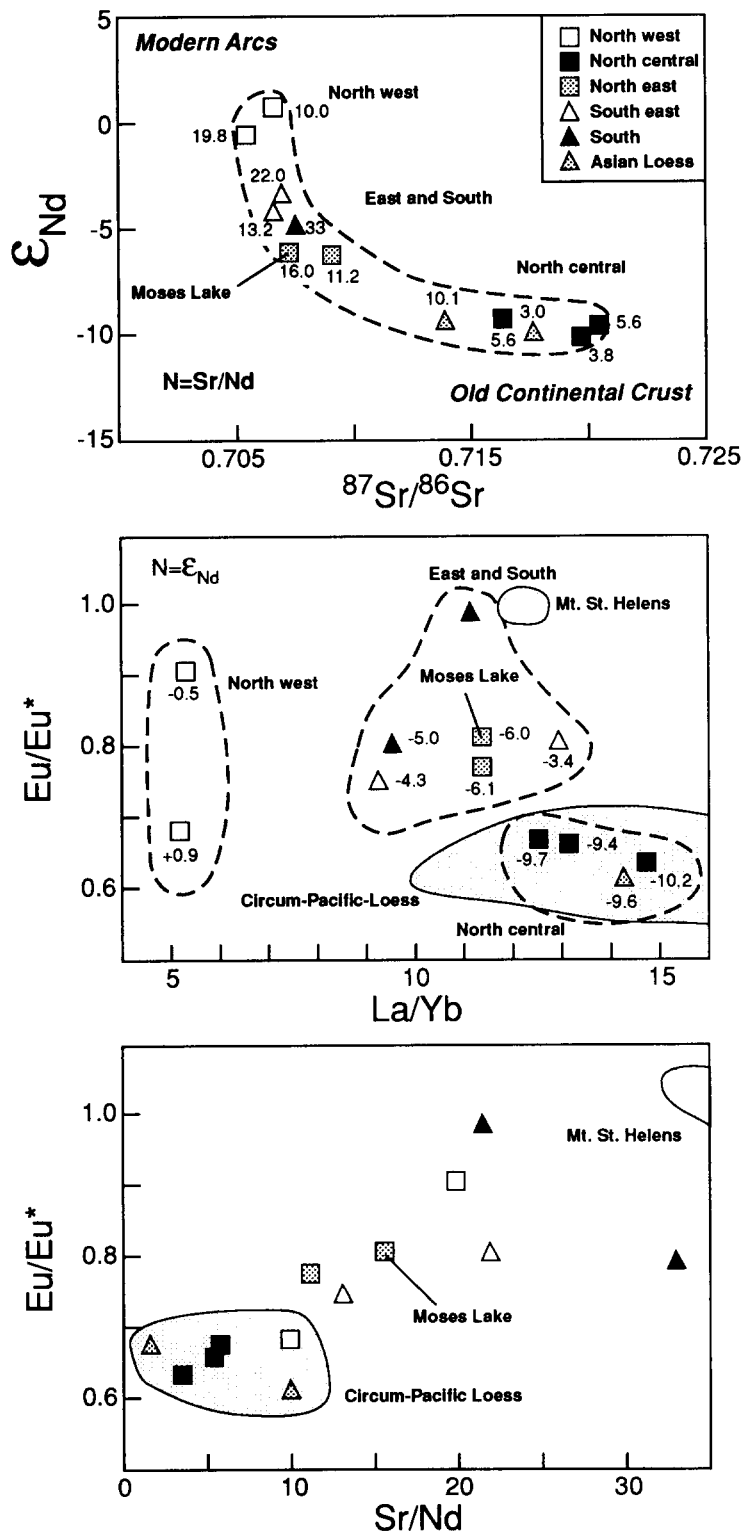


Fig. 3. Plots of Nd vs. Sr isotopic compositions and chondrite-normalized Eu anomaly vs. La/Yb and Sr/Nd ratios for the silicate fraction of Pacific deep-sea sediments.

While ocean island basalts are extremely LREE enriched with low Sm/Nd, island-arc volcanic products have relatively flat REE patterns, comparable to those found in some of the Pacific dust samples. Hence the opposite trends shown in Fig. 2 are to be expected.

It is well established that the Nd isotopic composition of seawater is distinct in different oceans [14,17,59,60]. Piegras and Wasserburg [17] attributed the less radiogenic Nd found in Atlantic seawater (average  $\epsilon_{Nd} = -12$ ) compared to Pacific seawater (average  $\epsilon_{Nd} = -8$ ) to an older average age of the continents that drain into the Atlantic. The average  $\epsilon_{Nd}$  for the Pacific sediment samples studied here is  $-6$ , whereas the average for the equivalent samples from the Atlantic is  $-10$ , mimicking the differences in the isotopic composition of seawater. There is no correspondence between the regional variations in the Nd isotopic compositions of the oceans, as indicated by data for seawater, metalliferous sediments and ferromanganese encrustations [15,18], and the pattern indicated for the eolian particles here (Fig. 1). If eolian particles supply significant Nd to seawater by leaching on the seafloor, the local residence time of Nd must be about 300–600 yrs. This would be sufficiently long to allow seawater to mix and dilute or eliminate regional variations, but short enough to prevent homogenization of the Nd isotopic composition of the world's oceans.

### 3.2 Combined Nd and Sr isotopic compositions of Pacific dust

The combined Nd and Sr isotopic compositions of Pacific dust can be described as approximating a hyperbolic mixing curve between an arc-like end member ( $^{87}\text{Sr}/^{86}\text{Sr} \approx 0.7055$ ,  $\epsilon_{Nd} \geq +1$ ) and a loess component ( $^{87}\text{Sr}/^{86}\text{Sr} \geq 0.714$ ,  $\epsilon_{Nd} \approx -10$ ) similar to old cratonic crust (Fig. 3). The curvature of the data array would suggest that the arc-like end member has higher Sr/Nd. This is broadly confirmed by the ratios measured for each sample (Fig. 3). While there are distinctions between the northwestern Pacific, north central Pacific and south and eastern Pacific, further geographic discrimination is not apparent (Figs. 1 and 3, *top*). The integrity of the Nd-Sr isotopic relationships as shown in Fig. 3 is evi-

dence that the Sr isotopic compositions have not been greatly affected by sorting or alteration. However, there is evidence that sorting may have affected the samples to some degree. Although, the north central Pacific samples have similar Sr isotopic characteristics to that of loess from Asia, the data for both the Asian loess and the north central Pacific samples exhibit greater variability in Sr than in Nd isotopic compositions (Fig. 3). Proximal sorting might be responsible for some of the variability in Sr isotopic compositions in the Asian loess and north central Pacific samples.

The northwestern Pacific most clearly contains a volcanogenic component, probably from Japan or the Kuriles–Kamchatka arc. The  $\epsilon_{Nd}$  for volcanic rocks from Japan [61] vary from  $+8$  to  $+2$ , significantly higher than the values for the northwestern Pacific. Assuming arc volcanism in Japan supplies ash with  $\epsilon_{Nd}$  of  $+6$  and that the ash mixed with airborne particles derived from the continental crust with  $\epsilon_{Nd}$  of  $-10$ , the two samples with high  $\epsilon_{Nd}$ , RC14-105 and V20-122, collected near Japan contain 20–30% of older, continental, eolian-transported Nd. Younger crustal materials in the North American continent with higher  $\epsilon_{Nd}$  compared to the north central Pacific could contribute to the Nd budget of the south and eastern Pacific as hemipelagic and eolian particles, resulting in higher  $\epsilon_{Nd}$ . For example, major portions of the Cordilleran circum-Pacific batholiths commonly have low  $^{87}\text{Sr}/^{86}\text{Sr}$  ( $< 0.705$ ) and high  $\epsilon_{Nd}$  ( $> -2$ ) [62–64]. The mixing of such components with older materials from near the continental margin could explain the observed isotopic characteristics of the south and eastern Pacific samples.

### 3.3 Deducing provenance from combined isotopic and trace element data

While the Nd and Sr isotopic compositions are capable of resolving dust provenance in a general sense, the potential for more precise connections between dust and source would appear limited, given that the entire eastern Pacific appears to have intermediate Sr and Nd isotopic compositions (Fig. 3). However, combining the isotopic and geochemical compositions of dust provides a more powerful and specific discriminant in assigning source regions. REE patterns (Fig. 4) for



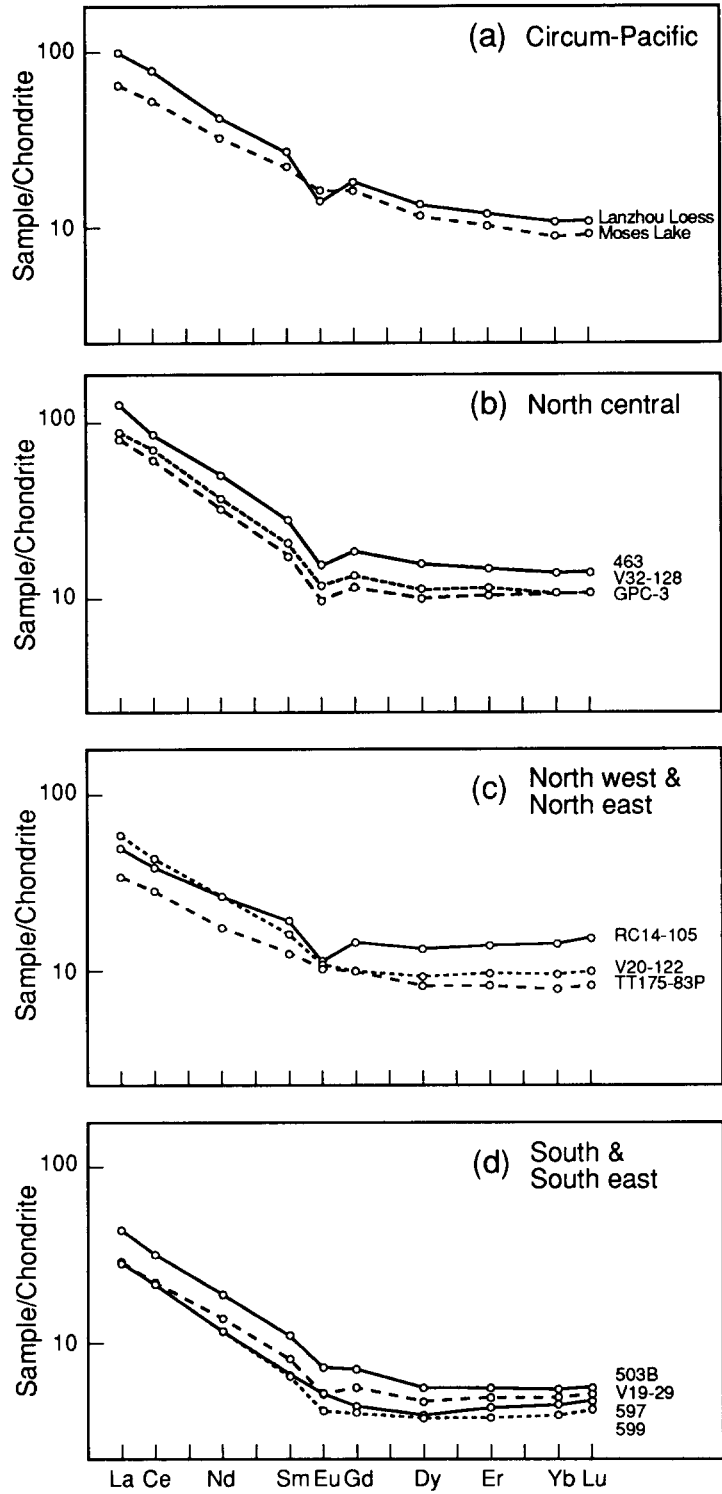


Fig. 4. Plots of chondrite-normalized REE compositions for silicate fractions of Pacific deep-sea sediments. The REE abundances of the Leedeey chondrite [70] were used for normalization.

the Pacific samples are similar to typical crustal materials (e.g., loess), being enriched in LREEs, with a slight negative Eu anomaly and with relatively flat HREE. Regional variations in Nd and Sr isotopic composition are paralleled by variations in Eu anomaly and La/Yb ratio (Figs. 1 and 3). The north central Pacific has a steep pattern (Fig. 4 b) ( $\text{La}/\text{Yb} = 14$ ) with significant Eu depletions typical of post-Archean shales ( $\text{Eu}_N/\text{Eu}_N^* = 0.65$ ), while the south and east Pacific is characterized by a less negative Eu anomaly and more moderate La/Yb (Fig. 4d). The northwestern Pacific group has a distinctive, relatively flat REE pattern (Fig. 4c). The loess sample plots close to the north central Pacific data (Fig. 3, *middle*), confirming the isotopic evidence that eolian particles derived from the Asian continent dominate the deep-sea sediments in this region. This observation endorses the use of the REE compositions of these deep-sea sediments as indicators of provenance. Other circum-Pacific loess samples from Asia, North America and New Zealand also plot with the north central Pacific data. However, only the Asian samples have similar Nd isotopic composition. This plot further highlights the fact that samples from other portions of the Pacific contain non-loess components. There is no Pacific sample with a loess REE pattern that has a Nd isotopic composition like that of loess from New Zealand or the U.S. The only major loess component comes from Asia.

Further geographical distinctions can be resolved using Sr/Nd ratios (Fig. 3, *bottom*). In general terms the Sr/Nd ratios of the samples increase with increasing  $\text{Eu}_N/\text{Eu}_N^*$ , as is predictable from feldspar fractionation in the continental crust. Many circum-Pacific volcanic arc components such as the dacites of Mt. St. Helens [56] plot with  $\text{Sr}/\text{Nd} \approx 35$  and  $\text{Eu}_N/\text{Eu}_N^* \approx 1$  at one end of the trend defined by the Pacific data. Loess samples plot at the other end with much lower values for both parameters. The north central Pacific samples have Sr/Nd and  $\text{Eu}_N/\text{Eu}_N^*$  that are unresolvable from circum-Pacific loess. This implies that both Sr/Nd and  $\text{Eu}_N/\text{Eu}_N^*$  are also largely unaffected by sorting during transport to the oceans. The northwestern Pacific samples show no special distinction on this diagram, but nevertheless can be resolved by their high  $\epsilon_{\text{Nd}}$  and low La/Yb (Fig. 3). The south and east

Pacific samples show important geographic distinctions. Those from close to Central America are displaced to higher Sr/Nd ratios than those from close to North America (southeast and northeast respectively in Fig. 3). The samples from the south Pacific also have high Sr/Nd and only a small Eu anomaly, implicating sources that have undergone little feldspar fractionation. These regional variations imply that more precise estimates of provenance may be elucidated with a combined isotopic and trace element approach.

### 3.4 Trace element and model age fractionation—*natural and artifact*

Certain features of the data are consistent with some trace element fractionation either naturally, during weathering, transport, sedimentation and early diagenesis, or during sample preparation. For example, although the north central Pacific samples have Sr and Nd isotopic compositions as well as La/Yb, Sr/Nd and  $\text{Eu}_N/\text{Eu}_N^*$  that are identical to their known provenance, Asian loess, there is some evidence of minor fractionation of the MREEs. This is evident in Fig. 4 but is more effectively highlighted with a plot of  $\text{La}_N/\text{Sm}_N$  vs.  $\text{Gd}_N/\text{Yb}_N$  (Fig. 5a). All the Pacific samples have  $\text{Gd}_N/\text{Yb}_N$  less than the upper crust average of 1.4 whereas most of the loess samples and the Moses Lake sample have higher values. Even more strikingly, the north central Pacific samples have  $\text{La}_N/\text{Sm}_N$  values that are higher than any of the loess samples. These features are difficult to explain unless there has been minor fractionation of the MREEs. Arc-like components would plot with low  $\text{La}_N/\text{Sm}_N$  and  $\text{Gd}_N/\text{Yb}_N$ . Intraplate OIB sources such as Hawaii would have high  $\text{La}_N/\text{Sm}_N$  and  $\text{Gd}_N/\text{Yb}_N$ . Loess has moderate  $\text{La}_N/\text{Sm}_N$  and  $\text{Gd}_N/\text{Yb}_N$ . There is no obvious component that could generate the high  $\text{La}_N/\text{Sm}_N$  and low  $\text{Gd}_N/\text{Yb}_N$  of the north central Pacific samples. Furthermore it is difficult to see how an additional component could have this type of effect and yet apparently not change the isotopic compositions observed. If there has been some loss of MREEs during the extraction process it appears not to have changed Sr/Nd or  $\text{Eu}_N/\text{Eu}_N^*$  (Fig. 3, *middle*). Therefore, loss of carbonate is not likely to be important since the Sr/Nd ratios are very

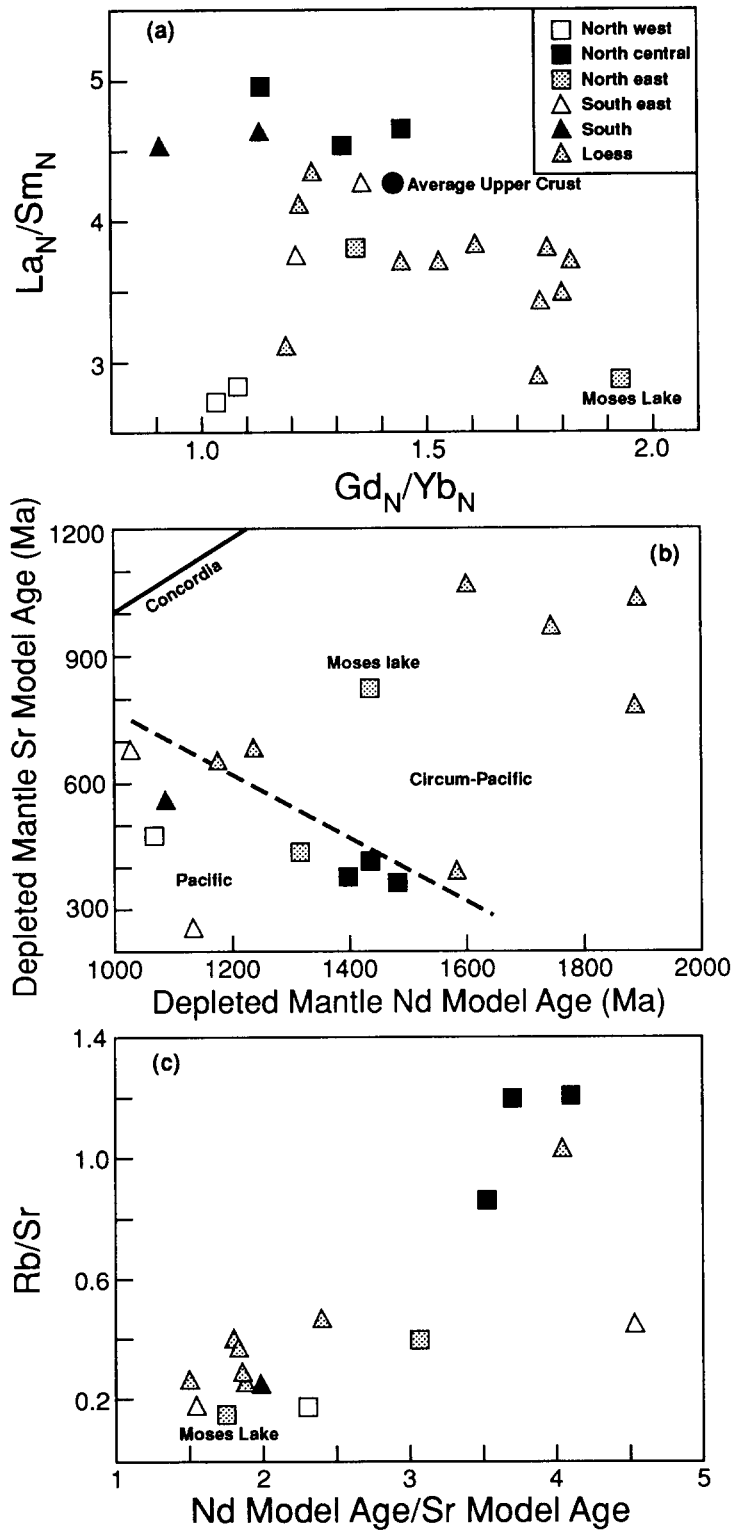


Fig. 5. (a) Plot of  $La_N/Sm_N$  vs.  $Gd_N/Yb_N$  showing MREE loss in Pacific samples relative to loess. (b) Nd model age vs. Sr model age showing the younger Sr model ages observed in both Pacific and loess samples and (c) the ratio of Nd to Sr model age plotted against Rb/Sr.

high ( $> 100$ ) [12]. Similarly, metalliferous oxides are unlikely to be relevant since they have low Sr/Nd ratios ( $< 0.1$ ) [14,15,17,19]. Apatite is known to preferentially incorporate significant amounts of MREEs and has Sr/Nd ratios of about 1–10 [13,65,66]. Certain forms of low-temperature apatite are soluble in acetic acid [65–67]. Hence, it is possible that this portion of the oceanic sample was partially lost during sample preparation, although we are engaging in further research to ascertain whether this is indeed the case.

This REE fractionation extends to a shift in Sm/Nd ratio and Nd model age. The three north central Pacific samples have  $^{147}\text{Sm}/^{144}\text{Nd}$  about 10% lower than the three Asian loess samples. This translates into a roughly 10% decrease in Nd model age. The Nd and Sr model ages are compared in Fig. 5b. It can be seen that despite the lowering of Nd model ages, the Sr model ages are even lower. This is not an artifact induced during preparation of the oceanic samples. Taylor et al. [22] noted the same feature in loess (Fig. 5b) and ascribed it to Rb enrichment during weathering. The ratio of Nd model age to Sr model age is correlated with Rb/Sr (Fig. 5c). The data for Nanking loess [22] and the north central Pacific samples are virtually identical. However, the sample of Lanzhou loess (Table 2) plots with the other loess data. Goldstein [68] noted that this difference between Nd and Sr model age was true of the sedimentary mass as a whole and explained it in terms of preferential recycling of radiogenic Sr into the mantle. Goldstein argued that radiogenic Sr was lost from the continents to seawater and thence to the mantle by hydrothermal exchange at ridges. He also pointed out that the Rb/Sr ratio of the continents might be increased by carbonate subduction. Both processes would lead to a decrease in Sr model age of the sedimentary mass. Another mechanism for decreasing Sr model age is feldspar fractionation in relatively young crustal systems. This will produce terranes that are biased toward younger Sr model ages and generate correlated depletions in Sr/Nd and  $\text{Eu}_\text{N}/\text{Eu}_\text{N}^*$ , as exemplified by Fig. 3.

Weathering is anticipated to induce preferential Sr loss [58,68]. Regardless of whether this takes place with Sr equilibration it will leave a residue with higher Rb/Sr and younger model

age. Mineral sorting will further enhance the spread in Sr model age. If distal samples are enriched in (finer) clays they will be Rb rich and hence define younger Sr model ages. All three north central Pacific samples have higher Rb concentrations than loess. If the age of the last major Rb/Sr mineral fractionation is young (such as in young igneous complexes, recently uplifted terranes or recent weathering), the weathering products, if sorted mineralogically, will define the average Rb-Sr mineral age of these processes, provided Sr is not equilibrated during weathering. The data for Asian loess and north central Pacific samples display a correlation between Sr isotopic composition and Rb/Sr (not shown), the slope of which corresponds to an apparent age of the order of 100 Ma, consistent with the importance of Mesozoic and Cenozoic igneous and tectonic activity in southeast Asia. Hence, at least a part of the bias to younger Sr model age appears to be due to mineralogic sorting rather than Rb gain or Sr loss caused by mobility during weathering.

#### 4. Conclusions regarding provenance

From these preliminary data we can draw some immediate conclusions regarding provenance. First, the two samples collected in the northwestern Pacific have the highest  $\epsilon_{\text{Nd}}$  and lowest La/Yb ratios of all the samples, implying a significant component of arc volcanic products. This is supported by the high abundances (6 and 17%) of smectite in these two samples [69], which indicate contributions from mafic volcanic sources. The age of these core samples is ca. 6000 yrs, which is coincident with the violent eruption of Kikai volcanoes in the Ryukyu arc, one of the largest eruptions in Japan during the Holocene, ejecting about  $100 \text{ km}^3$  of Akahoya volcanic ash. Another possible source of the sediment is the weathered surface of the Japanese islands. Goldstein and Jacobsen [58] reported that the suspended materials of four rivers in Japan have  $^{87}\text{Sr}/^{86}\text{Sr}$ ,  $^{147}\text{Sm}/^{144}\text{Nd}$  and  $\epsilon_{\text{Nd}}$  of 0.707–0.708, 0.10–0.14 and  $-3.3$  to  $+0.5$  respectively, similar to the values of the two deep-sea sediments studied here. Second, broad reaches of the north central Pacific are characterized by eolian sediments that

almost perfectly mimic the geochemical signature of the Asian loess deposits. The suggestion that North American dust supplies significant Nd to seawater in this region of the Pacific Ocean [18] is clearly in error. Third, nearer the Americas, core TT175-83P, raised from the western flank of the Juan de Fuca Ridge, is dominated by sediments that show distinct affinity with the Cascades of North America, as sampled at Moses Lake near Mt. St. Helens. The Nd isotopic compositions and trace element compositions are quite distinct from those of the loess derived from Asia and interior North America (Kansas and Iowa) as reported here and by Taylor et al. [22]. This is entirely consistent with the dominance of hemipelagic deposition from the nearby continental margin. Fourth, the sample from core DSDP Site 503B is characterized by a mineral component more like that of northwestern South America, as seen in hemipelagic core V19-29, than like that of western Pacific sources. This confirms prior interpretations regarding the nature of the ITCZ as an eolian transport barrier and the downwind extent of the area influenced by South American dust sources [40–42]. In the south Pacific, the sample from core DSDP Site 597 displays an isotopic signature and REE pattern consistent with a significant contribution from young arc material, implicating South America as the ultimate source of the silicate mineral component at this location. The nearby Site 599, somewhat closer to South America and the East Pacific Rise, is characterized by similar trace element composition and  $^{87}\text{Sr}/^{86}\text{Sr}$  (Fig. 1). The lack of a Eu anomaly, the low La/Yb and the high Sr/Nd all indicate that the source of this sediment is not Asian loess, nor New Zealand loess [22], and must be derived from a different source, probably the South American arcs.

Our results confirm that isotopic compositions combined with trace element data can distinguish the provenance of the mineral component of deep-sea sediments. Additional combined chemical and isotopic analyses are clearly needed to define the regional variations more fully. However, the demonstrated ability to distinguish provenance in general, and the relatively undiluted and unaltered loess signal preserved in central Pacific regions in particular, indicates that this approach will be useful in downcore studies

to determine past changes in dust provenance and hence paleoclimate.

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