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A Sm-Nd isotopic study of atmospheric dusts and particulates from major river systems

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$^{143}Nd/^{144}Nd$ ratios, and Sm and Nd abundances, are reported for particulates from major and minor rivers of the Earth, continental sediments, and aeolian dusts collected over the Atlantic, Pacific, and Indian Oceans. Overall, Sm/Nd ratios and Nd isotopic compositions in contemporary continental erosion products vary within the small ranges of $^{147}Sm/^{144}Nd = 0.115 \pm 0.01$ and $^{143}Nd/^{144}Nd = 0.51204 \pm 0.0002$ ($\epsilon_{Nd} = -11.4 \pm 4$). The average period of residence in the continental crust is estimated to be 1.70 ± 0.35 Ga.

These results combined with data from the literature have implications for the age, history, and composition of the sedimentary mass and the continental crust: (1) The average "crustal residence age" of the whole sedimentary mass is about 1.9 Ga. (2) The range of Nd isotope compositions in the continent derived particulate input to the oceans is the same as Atlantic sediments and seawater, but lower than those of the Pacific, demonstrating the importance of Pacific volcanism to Pacific Nd chemistry. (3) The average ratio of Sm/Nd is about 0.19 in the upper continental crust, and has remained so since the early Archean. This precludes the likelihood of major mafic to felsic or felsic to mafic trends in the overall composition of the upper continental crust through Earth history. (4) Sediments appear to be formed primarily by erosion of continental crust having similar Sm/Nd ratios, rather than by mixing of mafic and felsic compositions. (5) The average ratio of $^{143}Nd/^{144}Nd \approx 0.5117$ ($\epsilon_{Nd} \approx -17$) in the upper continental crust, assuming its mean age is about 2 Ga. (6) The uniformity of the Sm-Nd isotopic systematics in river and aeolian particulates primarily reflects efficient recycling of old sediment by sedimentary processes on a short time scale compared to the amount of time the material has resided in the crust.

1. Introduction

Information concerning average chemical and isotopic characteristics of the continental crust has proven to be notoriously difficult to extract from the complex mosaic of terrains that compose the continents. Fortunately, erosion and transport of material on the surface of the Earth reduce some of the chemical diversity through generation of sediments. Goldschmidt's [1] estimate of the average chemical composition of the continents, derived from analyses of the matrix material in glacial deposits, is a classic attempt to exploit the averaging effects of sedimentary processes.

The importance of recycling of crustal material by sedimentary processes is exemplified by the young mean stratigraphic age of the sedimentary mass, compared with the age of the Earth and the continental crust. Estimates of the thicknesses and areas of sediments as a function of stratigraphic age [2,3] show a trend of increasing sedimentary mass with younger stratigraphic ages, so that the half-mass age is probably much less than 600 Ma [4,5]. An important corollary to these observations is that the period of time that the materials that compose a sediment have resided in the continental crust (the "crustal residence age" [6]) on aver-

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age far exceeds the stratigraphic age of the sediment. The mean "crustal residence age" of the rare earth element component of a sediment may be estimated from the Sm-Nd isotope systematics [6,7].

Water and wind are primary media by which surface material is eroded, mixed, and transported. Rivers are by far the most important transport medium [4], with particulate loads predominantly derived from regions of high topography. Winds independently sample large geographical areas, with source regions of particulates determined less by topography than by weather patterns and vegetative cover.

This study reports the Sm-Nd isotopic systematics of materials being deposited by present-day cycles of erosion, including sediments and particulates from the major rivers of the Earth, and atmospheric particulates collected above the Atlantic, Pacific, and Indian Oceans. The primary purposes are: firstly, to ascertain the average "crustal residence age" (t_{CR}) of present-day continental erosion products; secondly, to assess possible systematic geographic variations in the Sm-Nd isotopic characteristics of the particulates; thirdly, to generate a Sm-Nd data base for modern erosion products that can provide the necessary perspective for evaluation of isotopic data on sediments deposited throughout geological history.

2. Sample selection and analytical techniques

Suspended particulates, bank clays, and river bottom sediments have been selected from many of the Earth's major rivers (Fig. 1). Also included are atmospheric dusts collected near sea level in the Atlantic, Pacific, and Indian Oceans; a sample of Mt. St. Helens airfall ash; a late Tertiary to Recent loess which is a source of wind blown dust in the Pacific; and two sediment samples from the Yangtze River basin, from strata which straddle the Precambrian/Cambrian boundary. Size fractions of the sample from the mouths of the Amazon has been analyzed to check for grain-size-dependent isotopic variability. The effect of local geologic variability is examined through inclusion of samples with geographically restricted or geologically exceptional provenances (Hudson, Cam, Columbia, San Francisco Bay, Mt. St. Helens airfall ash).

All samples were washed in ultrapure water prior to dissolution in pressurized PTFE vessels. Sm-Nd isotopic analyses were made at the University of Cambridge using chemical and mass spectrometer techniques described previously [8]. In this paper, model ages calculated from Sm-Nd isotopic measurements are used to estimate the average periods of residence of the rare earth components in the continental crust. Because a sediment sample is likely to have a mixture of Nd components with a variety of provenances, t_{CR} ages represent averages weighted according to the amount of Nd from different sources.

The model t_{CR} ages are assumed to correspond to the time that has elapsed since the sample possessed the same $^{143}Nd/^{144}Nd$ ratio as the mantle which supplied its crustal precursors. This method assumes that the period of time that elapsed between addition of the material to the continental crust, and fractionation to present-day Sm/Nd ratios, was short. If the relevant mantle source was unfractionated relative to a "chondritic uniform reservoir" (CHUR) [9], then model ages based on CHUR evolution, as used by McCulloch and Wasserburg [7], would be appropriate. However, initial $^{143}Nd/^{144}Nd$ ratios from Precambrian terrains suggest that the mantle that has supplied the continental crust has evolved since the earliest Archean with a Sm/Nd ratio which exceeds CHUR [8,10]. Consequently, t_{CHUR} ages are likely to underestimate true crustal residence ages. In this contribution, characteristic ϵ_{Nd} values of a "depleted mantle" (DM) source for the continental crust are assumed to be bounded by the CHUR of Jacobsen and Wasserburg [11] at 4.5 Ga, initial $^{143}Nd/^{144}Nd$ ratios of Archean terrains, and present-day oceanic basalts [12–15]. These conditions can be met by a depleted mantle which evolves linearly over geologic time to a characteristic value today of $\epsilon_{Nd} \approx +10$. Calculated t_{CR} ages in Table 1 are based on a present-day depleted mantle characterized by $^{143}Nd/^{144}Nd = 0.51316$ when normalized to $^{146}Nd/^{144}Nd = 0.7219$. For sediments with Sm/Nd ratios about 0.19 ($f^{Sm/Nd} \approx -0.42$) and $^{143}Nd/^{144}Nd$ ratios about 0.5120 (ϵ_{Nd}

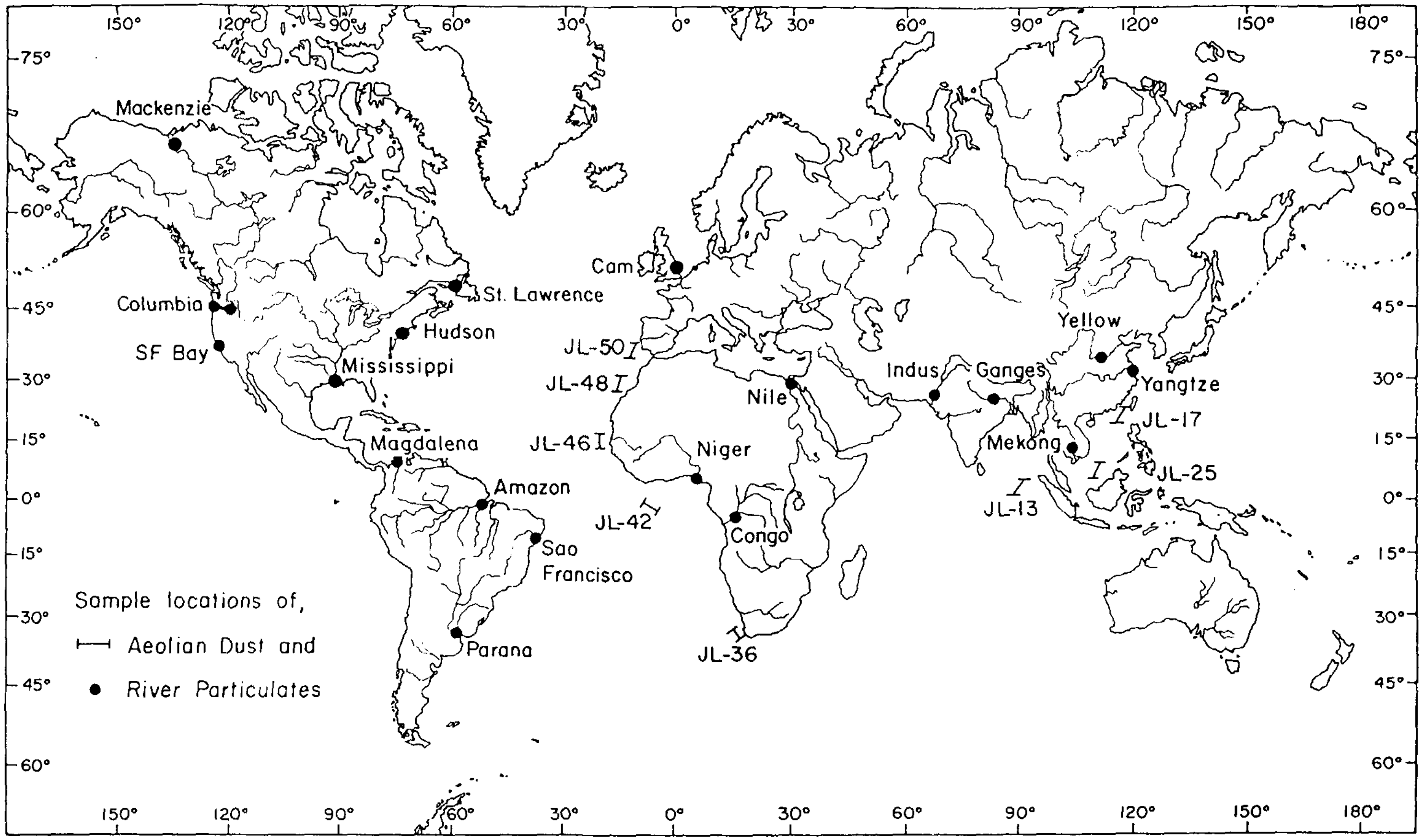


Fig. 1. Map indicating the major drainage systems of the Earth and the locations of river and atmospheric particulates analyzed as part of this study.

TABLE 1

Sm and Nd analytical results

	Sm (ppm)	Nd (ppm)	$^{143}Nd/^{144}Nd$	$^{147}Sm/^{144}Nd$	t_{CR} (Ga)	$\epsilon_{nd}(0)$	$f^{Sm/Nd}$	Sample type
I. Rivers								
Amazon (Brazil)	6.764	35.30	0.512165±16	0.1158	1.54	−9.2	−0.41	1, 5
Cam (England)	4.135	21.40	0.512078±18	0.1168	1.69	−10.9	−0.41	1, 5
Columbia I (U.S.A.)	7.327	30.26	0.512301±18	0.1464	1.93	−6.6	−0.26	1
Columbia II (U.S.A.)	7.691	37.82	0.512414±15	0.1229	1.25	−4.4	−0.37	2
Congo (Congo)	6.339	34.65	0.511813±16	0.1106	1.98	−16.1	−0.44	1, 3
Ganges (India)	6.996	35.83	0.511835±18	0.1180	2.10	−15.7	−0.40	1, 4
(duplicate)	–	–	0.511869±16	–	–	−15.0	–	
Hudson (U.S.A.)	9.213	45.57	0.512057±16	0.1222	1.83	−11.3	−0.38	1, 5
Indus (Pakistan)	5.925	33.75	0.512014±16	0.1061	1.62	−12.2	−0.46	2
Mackenzie (Canada)	3.045	16.09	0.511906±19	0.1144	1.91	−14.3	−0.42	1
Magdalena (Colombia)	5.538	27.49	0.512211±16	0.1218	1.57	−8.3	−0.38	1, 3
Mekong (Cambodia)	7.680	40.62	0.512152±20	0.1143	1.54	−9.5	−0.42	1, 3
(duplicate)	–	–	0.512161±24	–	–	−9.3	–	
Mississippi (U.S.A.)	6.329	32.62	0.512077±18	0.1173	1.70	−10.9	−0.40	1
Niger (Nigeria)	–	–	0.512098±20	–	–	−10.5	–	2
Nile (Egypt)	7.551	34.49	0.512469±18	0.1324	1.29	−3.3	−0.33	2
Parana (Argentina)	7.501	37.10	0.512110±18	0.1222	1.74	−10.3	−0.38	1
St. Lawrence (Canada)	8.982	52.45	0.512366±20	0.1035	1.09	−5.3	−0.47	1, 5
San Francisco Bay (U.S.A.)	4.442	21.30	0.512456±28	0.1261	1.22	−3.6	−0.36	1, 5
São Francisco (Brazil)	5.660	30.95	0.511975±18	0.1105	1.74	−12.9	−0.44	1
Yangtze I (China)	6.148	31.70	0.512087±14	0.1172	1.69	−10.7	−0.40	1, 3
Yangtze II (China)	5.944	31.74	0.512077±22	0.1132	1.63	−10.9	−0.42	2
Yellow (China)	6.302	35.60	0.511994±22	0.1070	1.66	−12.6	−0.46	1, 4
II. Aeolian dusts								
JL-13 (W. Indian)	1.623 (±0.006)	11.93 (±0.02)	0.511931±36	0.0822	1.42	−13.8	−0.58	1
JL-17 (W. Pacific)	7.736	41.62	0.512146±26	0.1124	1.52	−9.6	−0.43	1
JL-25 (W. Pacific)	2.930 (±0.004)	16.42	0.512097±20	0.1079	1.52	−10.6	−0.45	1
JL-36 (Atlantic)	–	–	0.512248±44	–	–	−7.6	–	1
JL-42 (Atlantic)	8.509	45.76	0.512173±18	0.1124	1.48	−9.1	−0.43	1
JL-46 (Atlantic)	6.121	30.67	0.511942±20	0.1206	1.98	−13.6	−0.39	1
JL-48 (Atlantic)	6.683	35.33	0.512016±22	0.1143	1.74	−12.1	−0.42	1
JL-50 (Atlantic)	4.691	21.35	0.512203±22	0.1165	1.49	−8.5	−0.41	1
Mt. St. Helen's ash	2.929	13.69	0.512945±14	0.1320	0.40	+6.0	−0.33	1
III. Other continental sediments								
Malan Loess (China)	5.098	26.07	0.512134±19	0.1182	1.63	−9.8	−0.40	1
Xiling Gorge I (China)	5.891	30.25	0.511937±16	0.1177	1.92	−13.7	−0.40	1 (P€)
Xiling Gorge II (China)	9.931	58.13	0.511866±12	0.1033	1.78	−15.1	−0.47	1 (€)
IV. Amazon mouth								
Size fractions								
bulk	6.764	35.30	0.512165±16	0.1158	1.54	−9.2	−0.41	
coarse ($d > 45$ μm)	3.241	16.71	0.512158±26	0.1172	1.58	−9.4	−0.40	
medium (2 μm $< d < 45$ μm)	6.091	32.13	0.512152±14	0.1146	1.54	−9.5	−0.42	
fine ($d < 2$ μm)	9.332	46.97	0.512145±15	0.1201	1.64	−9.6	−0.39	

$^{143}Nd/^{144}Nd$ ratios are normalized to $^{146}Nd/^{144}Nd = 0.7219$. Reported errors are 2σ on the mean.
Concentrations are determined to a precision of better than 0.1%.
The $\epsilon_{Nd}(0)$ and f notations describe deviations of $^{143}Nd/^{144}Nd$ (in parts per 10^4) and Sm/Nd from a CHUR reservoir respectively. t_{CR} is the crustal residence age calculated relative to depleted mantle ($\epsilon_{Nd}^{4.5} = 0$; $^{143}Nd/^{144}Nd$ (today) = 0.51316), which has evolved linearly over geologic time. Present-day CHUR parameters are $^{147}Sm/^{144}Nd = 0.1966$, $^{143}Nd/^{144}Nd = 0.512638$ [11]. Amazon size fractions were kindly supplied by R.B. Stallard (Princeton University). Sample types: 1 = bulk sample; 2 = < 60 μm size fraction; 3 = suspended material; 4 = bank sediment; 5 = bottom sediment. Details about samples may be obtained from the authors.

≈ -12.5), these t_{CR} model ages are approximately 0.5 Ga greater than t_{CHUR} ages, and 0.2 Ga greater than "t_{DM}" ages of DePaolo [16].

3. Results

Sm and Nd concentrations, $^{143}Nd/^{144}Nd$ ratios, and crustal residence ages (t_{CR}) are listed in Table 1. $^{147}Sm/^{144}Nd$ ratios of all samples analyzed are less than CHUR (= 0.1966) [11], and there is a total range of $-0.58 < f^{Sm/Nd} < -0.25$. Excluding the Mt. St. Helens airfall ash data, which are typical of arc volcanism, the total variation of $^{143}Nd/^{144}Nd$ ratios is 0.5118–0.5125 ($-16 < \epsilon < -3$). The highest values are lower than oceanic basalts, the lowest are the same as Proterozoic and Phanerozoic continental sediments [6,7]. Fig. 2a, b shows that despite a large range of Sm and Nd concentrations (Table 1) the bulk of the data for both river and wind-borne particulates varies within a small range of $^{147}Sm/^{144}Nd = 0.115 \pm 0.01$ ($f = -0.42 \pm 0.06$) and $^{143}Nd/^{144}Nd = 0.51205 \pm 0.0002$ ($\epsilon = -11.5 \pm 4$). These ranges appear to be a general averaged feature of the portion of the continental crust that is being eroded today. Wind and water, utilizing independent means to sample the Earth's surface, give indistinguishable results. The $^{147}Sm/^{144}Nd$ ratios are similar to those found in many continental rock types (Table 2), and likely represent the average Sm/Nd (≈ 0.19) of the upper continental crust. For the river samples, the overall uniformity of the data set is in spite of the variety of sample types analyzed (size fractions, bulk samples, suspended particulates, bank clays, bottom sediments).

3.1. *Atypical river sediments and particulates*

Only the samples from the Columbia and Nile Rivers, St. Lawrence Estuary, and San Francisco Bay (Table 1) are outside the ranges typical of large-scale erosional products. The sediment load of these rivers accounts for less than 3% of the total load of rivers included in this study (Table 3). The St. Lawrence Estuary and San Francisco Bay samples have $^{147}Sm/^{144}Nd$ ratios close to typical upper crust, but have high $^{143}Nd/^{144}Nd$ ratios,

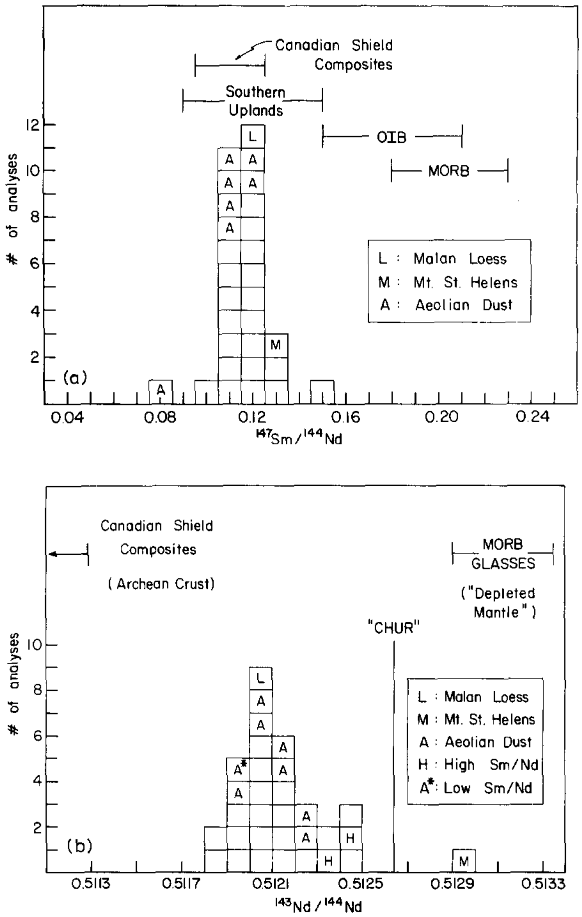


Fig. 2. (a) Histogram of $^{147}Sm/^{144}Nd$ ratios in atmospheric particulates (marked *A*), Asian loess (marked *L*), Mt. St. Helens airfall ash (marked *M*) and river particulates (all others) listed in Table 1. Atmospheric dusts and the loess have the same Sm/Nd ratios as the bulk of the river particulates. Results are compared with mid-ocean ridge basalts [13,15], ocean island basalts [12,14], Southern Uplands shales and greywackes [6], and Canadian Shield composites [37]. (b) Histogram of $^{143}Nd/^{144}Nd$ ratios. The loess and atmospheric dusts are the same as the bulk of the river particulates. Two river samples with high $^{143}Nd/^{144}Nd$ ratios also have high Sm/Nd ratios. Nd isotope ratios in these samples are higher than "Archean upper crust" [7], and lower than the "chondritic uniform reservoir" [11], and "depleted mantle" [13,15].

indicating they are primarily composed of relatively young continental crust. These may reflect significant input of material of Grenville age or younger into the St. Lawrence, and from the Sierra Nevada [17] into San Francisco Bay. High

TABLE 2

Sm/Nd in different rock types

	$^{147}Sm/^{144}Nd$ *	$f^{Sm/Nd}$
River and aeolian particulates [a]	0.115	−0.42
North American shales (Pz) [b]	0.105	−0.47
Southern Uplands shales and greywackes (Early Pz) [c]	0.122	−0.38
Delradian slates and schists (Prot-Pz) [d]	0.103	−0.48
Pilbara shales (Archean) [e]	0.116	−0.41
Canadian Shield composites (Archean and Proterozoic) [f]	0.106	−0.46
Isua grey gneisses and metasediments (Archean) [g]	0.105	−0.47
Felsic granulites [h]	0.104	−0.47
Granites [i]	0.113	−0.43
Mid-ocean ridge basalt glasses [j]	0.200	+0.02
Columbia river basalts [k]	0.143	−0.27
South Sandwich basalts [l]	0.205	+0.04
Aleutian basalts [m]	0.157	−0.20
Andean andesites [n]	0.120	−0.39
Marine manganese nodules [o]	0.144	−0.27
Siliceous ooze [p]	0.150	−0.24
Seawater [q]	0.122	−0.38
"CHUR" (bulk Earth) [r]	0.1966	0.00

[a] Average of river and aeolian particulates in Table 1; [b] North American shales [7]; [c] average of 16 SU sediments, Scotland [6]; [d] average of 5 Delradian metasediments, Scotland [6]; [e] average of 12 Pilbara metasediments, Australia [36]; [f] best estimate of Canadian Shield from table 4 of Shaw et al. [37]; [g] average of 5 Isua grey gneisses and metasediments, Greenland [8]; [h] average of 48 felsic granulites [8,38–40]; [i] average of 100 granite analyses [17,41–44]; [j] average of 17 MORB glasses [13,15]; [k] average of 24 CRB [18]; [l] average of 7 SS basalts [45,46]; [m] average of 7 Aleutian basalts [47]; [n] average of 12 andesites, Ecuador and Chile [48]; [o] average of 29 MMN [30,49]; [p] average of 7 siliceous oozes [49]; [q] average of 17 seawater samples [31,50]; [r] $^{147}Sm/^{144}Nd$ of "CHUR" [11] if $^{146}Nd/^{144}Nd = 0.7219$.

* $^{147}Sm/^{144}Nd$ are estimated for some samples by the relationship: $^{147}Sm/^{144}Nd = 0.605 \times Sm/Nd$.

$^{147}Sm/^{144}Nd$ ratios (0.13–0.15) in one of the Columbia River samples and the Nile indicate the presence of rare earth components less fractionated than typical upper crust, possibly derived, respectively, from Columbia River [18] and East African volcanism. That the Nile data reflect input from East African volcanism rather than Saharan sediment is implied by the coincidence of results for atmospheric dusts with Saharan sources (JL-46, JL-48) with the bulk of the river and atmospheric particulate data.

Application of these data requires the assumption that the samples are representative of the sediment load of the rivers. Evidence that this assumption is (in general) valid is provided by coincidence of $^{143}Nd/^{144}Nd$ ratios in a Yangtze suspended particulate and the < 60 μm fraction of a sand sample, and in different size fractions of an Amazon sediment (Table 1).

3.2. *Loess and Yangtze basin sediments*

The Precambrian/Cambrian boundary sediments from the Yangtze basin have Nd isotopic compositions nearly the same as sediments and particulates from the Yangtze mouth (Table 1). These results suggest that the sediments are typical of the material sampled by the Yangtze River, and that they were probably close to the global average when they were deposited ca. 600 Ma. The Malan Loess has Sm/Nd and $^{143}Nd/^{144}Nd$ ratios close to other loess from North America and Eurasia [7,19], and river and atmospheric particulates.

3.3. *Amazon River size fractions*

Coarse, medium, and fine fractions have a three-fold variation of Nd abundances (17–47 ppm) but the same $^{143}Nd/^{144}Nd$ ratios ($-9.6 < \epsilon_{Nd} < -9.3$). $^{147}Sm/^{144}Nd$ ratios exhibit a range from 0.115 to 0.120. Homogenization of Nd may have occurred through cation exchange between particulates and river water during transport, or perhaps less likely through solution-precipitation reactions. Possibly the solid particles are well mixed by the river during transport, but Sm and Nd are differentially dissolved from different size fractions. In any case it can be anticipated that the $^{143}Nd/^{144}Nd$ ratio of Amazon River water is the same as the particulates.

These results show that low-temperature processes fractionate the rare earth elements a small but measureable degree. The implication that isotopes approach equilibrium during fluvial transport may be a unique reflection of the enormous size of the Amazon, and/or its high water/particulate ratio (Table 3). Studies of size fractions

TABLE 3

Vital statistics of rivers in this study

River	Estimated annual suspended sediment load ($\times 10^{-14}$ g/a)	Estimated annual discharge (km^3/a)	Area of drainage basin (10^3 km^3)
Amazon	3.6 [a]	5720 [a]	6915 [c]
	9.0 [b]		
Columbia	0.09 [b]	229 [a]	669 [c]
Congo	0.65 [a]	1250 [a]	3800 [c]
Ganges	14.5 [a]	371 [a]	1060 [c]
Hudson	0.01 [b]	12 [b]	20 [b]
Indus	1.0 [b]	175 [a]	960 [c]
	4.4 [a]		
Mackenzie	1.0 [b]	306 [b]	1800 [c]
Magdalena	2.2 [b]	237 [b]	0.24 [b]
Mekong	1.6 [b]	350 [a]	810 [c]
Mississippi	2.1 [b]	362 [a]	3220 [c]
	3.1 [b]		
Niger	0.4 [b]	192 [b]	2090 [c]
	0.04 [a]		
Nile [g]	1.1 [a]	89 [a]	2870 [c]
Parana	0.92 [b]	470 [a]	2304 [a]
St.Lawrence	0.04 [b]	447 [a]	1290 [c]
São Francisco	0.06 [b]	97 [b]	600 [c]
Yangtze	4.78 [b]	688 [a]	1800 [c]
Yellow	18.8 [a]	47.4 [a]	745 [c]
	10.8 [b]		
Total	50 [d]	11,000 [e]	31,000
Total Earth	135 [b]	36,000 [a]	127,000 [c,f]
	225 [a]		

(If large differences exist between estimates in different references, more than one is given in the table.)

[a] Holeman [51].

[b] Milliman and Meade [34].

[c] Holland [52].

[d] Uses estimates of Milliman and Meade [34] where available.

[e] Uses estimates of Holeman [51] where available.

[f] Excludes Australia and Antarctica.

[g] Pre-Aswan dam.

from other river systems, and a better understanding of the extent to which rare earth elements reside in lattices versus surface sites in clay minerals, are required before these results can be generalized.

4. Discussion

4.1. Homogeneity of Nd isotope ratios in continental erosion products

The river systems in this study drain about 25% of the exposed continental crust, excluding Australia and Antarctica, and account for some 25–35% of the particulate flux and 30% of the water discharge to the oceans (Table 3). This coverage is augmented by the aeolian dusts. The overall variation of $^{143}Nd/^{144}Nd$ ratios is small (Fig. 2b) despite representation of diverse portions of the continental crust.

Regions of geologically recent volcanism are by and large outside the basins of major rivers. The rivers in this study with Quaternary arc volcanism in the drainage basins are the Columbia River and San Francisco Bay; the ones with potentially large inputs from continental flood basalts are the Columbia and Parana Rivers. The Columbia River and San Francisco Bay show effects attributable to prominent local geologic features; the Parana flood basalts must be a minor sediment source for the Parana River.

The homogeneity of Nd isotope compositions might be attributed to either (a) thorough mixing of isotopically heterogeneous material by contemporary erosional cycles, or (b) overall sampling of crust with similar Nd isotope ratios. A definitive answer must await intensive study of a single river system, however, available data support the latter explanation. Small rivers such as the Cam and Hudson, and the continental sediments in Table 1, and other Phanerozoic continental sediments [6,7,19,20] have $^{143}Nd/^{144}Nd$ ratios close to the aeolian dusts and particulates from large rivers of this study. This uniformity likely reflects mixing of sediments through a number of erosional cycles. The observed trend of increasing sedimentary mass with young depositional age [4,5] is better explained by recycling of older sediments to form new sediments than by increase of the total mass of sediments toward the present day [2]. Sediments are the rock type most likely to be destroyed by sedimentary processes simply because they are the most abundant surficial rock type, and they are disaggregated by wind and water more easily than crystalline rocks. The primary source of sediments is probably cannibalized older sediments.

Present-day contributions from regions with Sm-Nd systematics substantially different from contemporary erosion products are probably small. Large cratonic areas, such as the Canadian Shield [7], are not significant sources of eroded material. For volcanic arcs there appears to be a time lag between appearance and incorporation into major drainage systems.

4.2. *Sediments and the composition of the continental crust*

Sm-Nd isotopic investigations of metamorphosed continental rocks (e.g. [21,22]) have provided the general impression that on scales larger than that of local mineral equilibria, metamorphic processes have small effects on Sm-Nd systematics. The degree of fractionation of Sm/Nd ratios by sedimentary processes is insufficient to disturb the overall Sm-Nd systematics in a river sediment (section 3.2). In the continental crust, magmatic processes fractionate Sm and Nd to the degree that it affects the Sm-Nd isotopic systematics of a rock. In general, Sm/Nd ratios decrease with increasingly silicic bulk compositions.

Fig. 3 and Table 2 show that Sm/Nd ratios in different rock types vary with composition. Geological provinces dominated by basaltic magmatism (island arcs, continental flood basalts, mid-ocean ridges, oceanic islands) are regions on the earth's surface with characteristic Sm/Nd ratios higher than river and aeolian particulates. Because Sm/Nd ratios broadly correlate with the composition of a rock, they can indicate the compositional provenance of a sediment. Aeolian dusts and particulates from major rivers sample large geographical areas, and the observed uniformity in their Sm/Nd ratios implies that they represent an estimate of the average Sm/Nd (≈ 0.19) of the upper crust.

$^{143}Nd/^{144}Nd$ ratios reflect both the Sm/Nd ratio and time. Sediments, metasediments, and composites of variable "stratigraphic" age, which represent mixtures of large regions of the upper crust, have variable $^{143}Nd/^{144}Nd$ ratios reflecting their age. In general these have $^{147}Sm/^{144}Nd$ ratios in the range of 0.10–0.13, lower than those found in basalts, but similar to river and aeolian particulates. The decay of ^{147}Sm has had small effect on the $^{147}Sm/^{144}Nd$ ratios of the older rocks in Fig. 3. The constancy of Sm/Nd ratios in the products of large-scale surficial sampling throughout geologic history precludes the likelihood that the composition of the upper continental crust has undergone significant mafic to felsic secular trends, as suggested by many investigators [2,23,24]. In detail the literature data on sediments, metasediments, and composites (Table 2) are consistent with a small increase in average Sm/Nd ratios from $^{147}Sm/^{144}Nd \approx 0.105$ in the early Archean to about 0.115 today. The increase is consistent with addition of material to the crust from a mantle which has been progressively depleted in lithophile elements through time. The magnitude of the increase is not large enough to imply a pronounced change in composition.

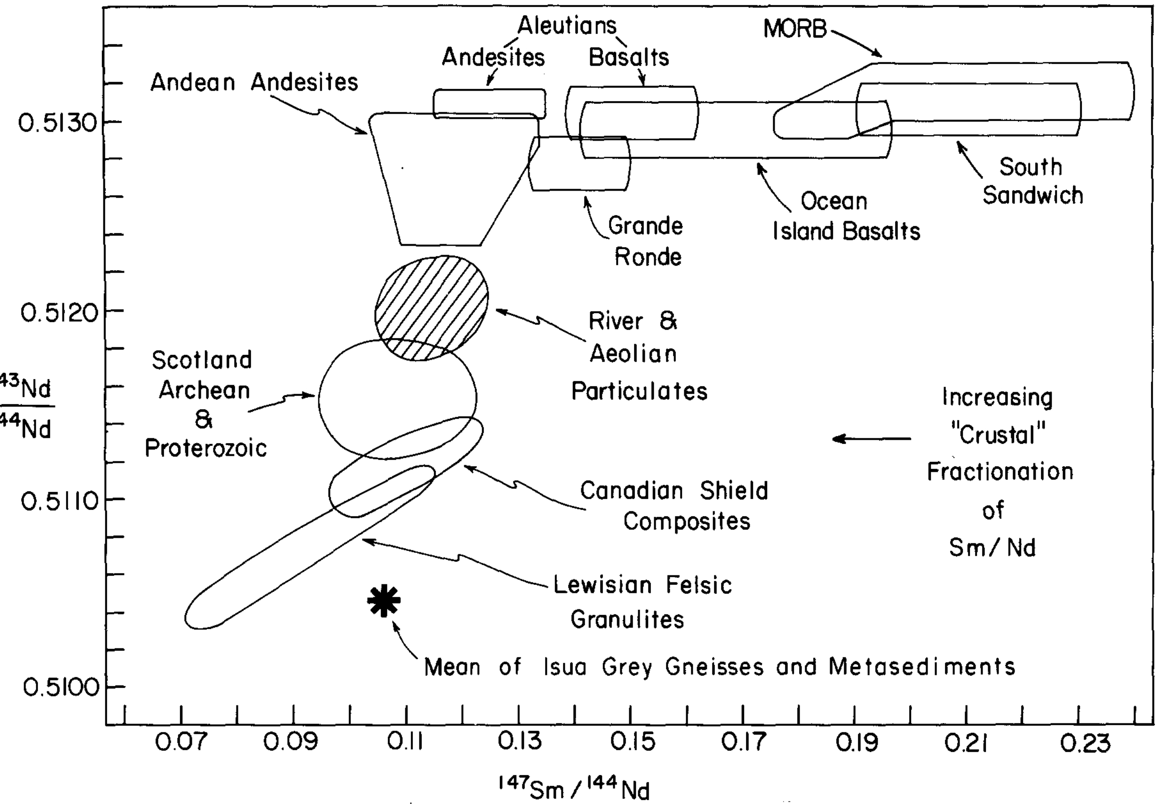


Fig. 3. Comparison of the fields of $^{147}Sm/^{144}Nd$ and $^{143}Nd/^{144}Nd$ for different rock types. Variations in Sm/Nd primarily reflect magmatic processes, while $^{143}Nd/^{144}Nd$ ratios reflect both Sm/Nd ratios and time. Recent river and aeolian particulates have nearly the same Sm/Nd ratios as Isua metasediments and grey gneisses, Canadian Shield composites, and Scotland Archean and Proterozoic sediments, metasediments, and gneisses. This near constancy implies the absence of secular trends in the composition of the continental crust through geologic time. Mixing on this diagram is linear. Sm/Nd ratios in sediments require that addition of basaltic rocks with high Sm/Nd ratios compared to the sedimentary mass be balanced by addition of rocks having correspondingly low Sm/Nd ratios. Although Lewisian felsic granulites seem to be a possible low Sm/Nd component, average Sm/Nd ratios in granulites and granites appear to be close to those of modern sediments (Table 2). If a highly fractionated (low Sm/Nd) reservoir does not exist, the upper continental crust must have been formed by addition of new material with similar Sm/Nd ratios, and rocks of basaltic composition can not be, and (since the earliest Archean) could not have been volumetrically important components. See text for discussion. Data are from the following sources: Isua [8], Lewisian [39], Canadian Shield [7], Scotland [6], river and aeolian dusts (this study), Andes [48], Aleutians [47], Grande Ronde [18], mid-ocean ridge basalts [12–16], ocean island basalts [12,14], South Sandwich [45,46].

The constancy of Sm/Nd ratios through geologic time in the upper continental crust requires that its bulk composition since 3.8 Ga has been, on average, more silicic than basalt. Obviously, continental flood basalts cannot meet this requirement, however neither can island arc extrusives. Karig and Kay [25] have estimated the average composition of volcanics at convergent plate margins to be 53–54% SiO_2. Fractionation on the way to the surface and contamination by older upper crust assures that the composition of primary melts are more mafic than average arc lava. Andean arcs appear to be the only environments in which large amounts of young material of typical composition (low Sm/Nd, high $^{143}Nd/^{144}Nd$) are being added to the upper continental crust in the present day (Fig. 3).

4.3. *The compositional diversity of the upper crust*

A sediment can be considered to be a mechanical mixture of its sources, with respect to Sm and

Nd. River and aeolian particulates are derived from sources that have Sm/Nd ratios lower than basalts, but the same as sediments and metasediments in the geologic record, and Canadian Shield composites (Fig. 3). Two possible means by which these compositions can be generated are (a) mixing of basalts with material having Sm/Nd ratios lower than sediments, or (b) mixing of materials that have similar Sm/Nd ratios. The latter explanation minimizes the volumetric importance of compositional diversity in the upper continental crust, and appears to be the mechanism which best explains the Sm/Nd ratios of sediments.

If basalts were a volumetrically large component of the upper continental crust, the Sm/Nd ratios of sediments require that a large reservoir of material with Sm/Nd ratios lower than sediments also exists. The component plotted in Fig. 3 which can mix with basalts to form the intermediate compositions of sediments are the Lewisian granulites. Although the Sm/Nd ratios of felsic granulites are highly variable, they appear to have, on average, the same Sm/Nd ratios as sediments. The same appears to be true for granites (Table 2). The absence of a large upper crustal reservoir characterized by Sm/Nd ratios lower than clastic sediments indicates that material with $^{147}Sm/^{144}Nd > 0.13$, such as basalts, have constituted since 3.8 Ga a volumetrically minor portion of the exposed continental crust.

4.4. *Implications for the marine chemistry of Nd*

The isotopic composition of Nd has proved valuable in studies of rare earth provenance in the oceans. O'Nions et al. [27] showed that the continents are the major source of Nd in manganese nodules and metaliferrous sediments. Subsequently, Goldstein and O'Nions [28,29] and Piepgras et al. [30] found that ferromanganese deposits exhibit small ranges of $^{143}Nd/^{144}Nd$ ratios within each ocean basin, but that the values in the Pacific are higher than in the Indian, whose mean is higher than that of the Atlantic (Fig. 4a). Nd isotope compositions in seawater from the Atlantic and the Pacific are the same as ferromanganese deposits from those oceans [31]. The results suggest that the residence time of Nd in the oceans is longer than the intra-basin mixing times, but shorter than the mixing time of the whole ocean. Higher $^{143}Nd/^{144}Nd$ ratios in the Pacific were attributed to derivation of Nd from younger sources in the Pacific than the Atlantic, either from the (relatively) young continental crust surrounding the Pacific or circum-Pacific magmatism [28–31].

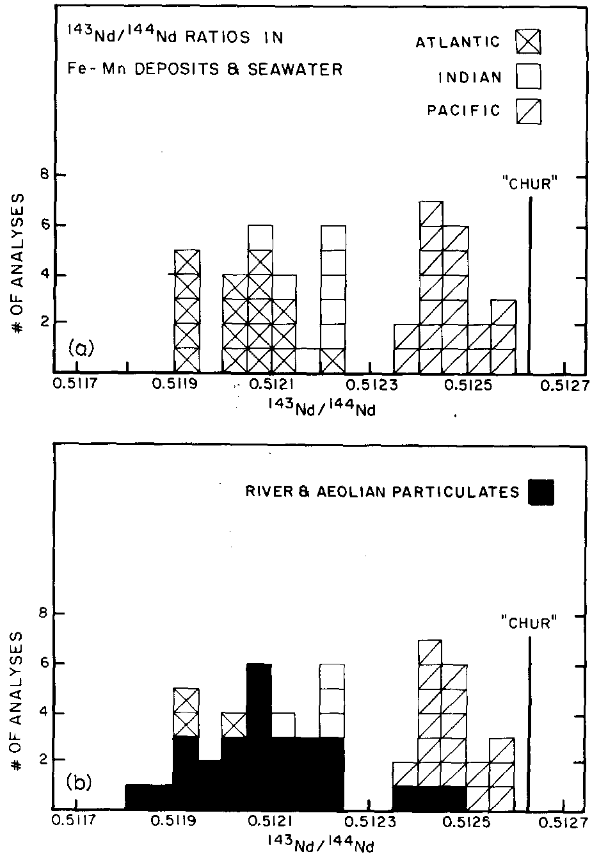


Fig. 4. (a) Histogram showing $^{143}Nd/^{144}Nd$ ratios in ferromanganese deposits and seawater, distinguished by ocean basin. $^{143}Nd/^{144}Nd$ ratios are less than "CHUR" [11], an estimate of the "bulk Earth" composition, which shows that the primary source of rare earth elements in marine authigenic sediments and seawater is the continental crust. Data from references 27, 29, 30. (b) Overlay of the data on river and aeolian particulates from Table 1 on the marine data. Marine Nd in the Atlantic and Indian Oceans has the same isotope ratios as the continental flux. The Pacific marine data require a component derived from Pacific volcanism.

The results of this study (Table 1) demonstrate that the material flux from the continents to the oceans is characterized by $\epsilon_{Nd} = -11.4 \pm 4$. This range appears to be independent of geography and transport medium. The latter consideration is important because wind-derived particulates may be the primary source of pelagic sediments [32,33]. The Nd isotopic composition of river water is likely to be the same as the particulates (section 3.3).

$^{143}Nd/^{144}Nd$ ratios of Atlantic ferromanganese deposits and pelagic sediments [29] and seawater [31] are within the range of the continental input (Fig. 4b). The higher values in the Pacific are both outside this range, and higher than the values of the Columbia River and San Francisco Bay (Table 1). The only likely sources of radiogenic Nd which can satisfy mass balance requirements are volcanic ash emissions and exchange or dissolution of Nd by hydrothermal circulation of seawater at mid-ocean ridges. The appearance of the effect in the Pacific likely reflects relatively high sea-floor spreading rates in that ocean.

Goldstein and O'Nions [29] found that Pacific pelagic clays have variable $^{143}Nd/^{144}Nd$ ratios, with values in western Pacific clays that are lower than in proximal manganese nodules. These results suggested that manganese nodules, and by inference, Pacific seawater, contain a component of Nd which has not been derived from the source of pelagic clays. If the average input from the continents is characterized by $^{143}Nd/^{144}Nd \approx 0.51205$, and the input from Pacific magmatism is ca. 0.5131, then about 25% of the Nd in western Pacific pelagic clays and 40% in Pacific ferromanganese deposits and seawater is derived from circum-Pacific magmatism. Intermediate $^{143}Nd/^{144}Nd$ ratios in the Indian Ocean are likely to arise from mixing of Pacific seawater with the continental input and Atlantic seawater.

The ratio of Sm/Nd is higher in marine sediments than in continental erosion products and seawater (Table 2), and in contrast to $^{143}Nd/^{144}Nd$ ratios, shows no inter-basinal variations. Authigenic sediments in the Pacific could be considered simple mixtures of continental and ocean volcanic components, however, Atlantic sediments are incompatible with this notion. The higher Sm/Nd ratios in marine precipitates relative to seawater suggest that Sm is preferentially incorporated into authigenic phases. The behavior of the rare earth elements during aqueous-particulate interactions remains one of the outstanding problems of marine Sm-Nd geochemistry.

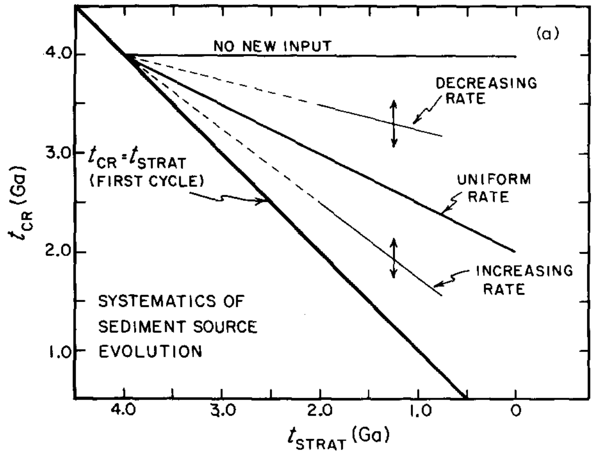
4.5. The age and evolution of the continental crust

The rivers studied (Table 1) include ten of the largest 15 rivers on Earth in estimated sediment load [34]. These account for 95% of the sediment load represented by the data set, and have a mean crustal residence age (t_{CR}) of 1.70 ± 0.35 Ga. The mean of all river and aeolian particulate data is 1.62 Ga. A mean age in this range (1.6–1.7 Ga) is also obtained by weighting the data according to the sediment load of the rivers. We regard this as the mean crustal residence age of continent that is subject to erosion and deposition in the present day.

The overall uniformity of the Sm-Nd systematics of contemporary continental erosion products justifies the use of sediments to evaluate the history of the continental crust. It must be assumed that sediments in the geologic record have crustal residence ages that reflect average eroding continent. One requirement since 3.8 Ga for sediments to be typical of the upper continental crust is that they have $^{147}Sm/^{144}Nd$ ratios in the range of 0.10–0.13.

The stratigraphic age distribution of the sedimentary mass appears to follow an exponential decay function with a half-mass age of 500 Ma [2,4]. Based on this relationship and Sm-Nd isotopic data on sediments in the geologic record [6,8,19,20], the mean t_{CR} age of the entire sedimentary mass is about 1.9 Ga. The small mass of Archean sediments has little effect on the overall mean age.

The t_{CR} ages of sediments, metasediments, and composites are compared to their "stratigraphic" ages (t_{STRAT}) in Fig. 5b. Samples from the Archean have $t_{CR} \approx t_{STRAT}$. Those with t_{STRAT} ages less than 2.0 Ga have t_{CR} ages substantially greater than the t_{STRAT}. Crustal residence ages of Phanerozoic sediments are not very different from those being deposited today. For those metasediments and composites whose plotted t_{STRAT} ages are metamorphic ages, actual depositional ages



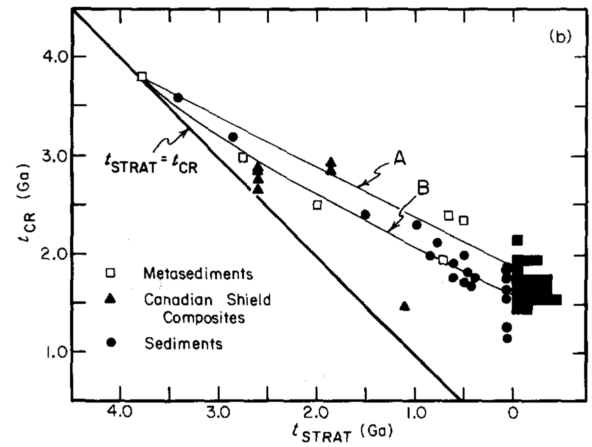


Fig. 5. Stratigraphic vs. crustal residence ages for sediments, metasediments, and composites. (a) Diagram illustrating the systematics of the evolution of the source of sediments. t_{CR} is the "crustal residence" age and t_{STRAT} is the stratigraphic age. Sediments that are deposited soon after emplacement of their source in the crust, without mixing of older crustal material, would fall close to the line of equal t_{CR} and t_{STRAT}. These are described as "first cycle" sediments. For everything else the slopes as well as the positions are important. A horizontal trend implies that no new material is added to the source of sediments. A uniform input rate of new material would result in a trend with a slope of -0.5. The lines on the diagram illustrate the trends expected from no growth and uniform growth since 4.0 Ga. An increasing input rate would imply dominance by new material and result in a trend with a steeper slope than implied by a uniform rate. Conversely, a decreasing input rate would result in a trend with a shallower slope. (b) Comparison of t_{CR} and t_{STRAT} for Archean to Recent sediments, metasediments, and composites. "Stratigraphic" ages of metasediments and composites are metamorphic ages. The sediment data represent the Sm-Nd systematics of the source of sediments, rather than the bulk sedimentary mass or the continental crust. Line *A* represents evolution of a sedimentary mass which has grown since 3.8 Ga at a uniform rate. Line *B* represents the evolution of the source of sediments for the conditions that the half-mass stratigraphic age of sediments is always 500 Ma and this age distribution is the result of erosion and re-deposition of old sediments (and pre-existing continental crust under certain conditions—see text). Data are from this study and references 6, 7, 8, 19. Data from strata of the same age in O'Nions et al. [6] are averaged. The stratigraphic age of the Fig Tree Shale has been adjusted to 3.4 Ga (J. Barton, personal communication).

must be older, and the use of metamorphic ages increases the appearance of scatter in the data. Systematics of the t_{CR} vs. t_{STRAT} diagram are illustrated in Fig. 5a. The concordance of t_{CR} and t_{STRAT} ages in the Archean can be the result of local derivation of sediments from newly formed continent without significant input from pre-existing continent, as might be the case if the Archean Earth were covered with microcontinents separated from one another, or substantial mixing of older sediment with young material, with domination of the source of sediments by younger input. In post-Archean times the source of sediments has been dominated by pre-existing continental crust. Although values of estimated t_{CR} ages are model dependent, these same observations hold if crustal residence age calculations are based on models of crustal source evolution other than the one used in this study (e.g. [7,16]).

If there is a quantitative relationship between the amount of new continental crust that is made and the amount of new material that is added to the sedimentary mass, then the trends of the data in Fig. 5 must in some way be related to the evolution of the continental crust. The relative crustal residence ages of the source of sediments (the quantity that is reflected by the t_{CR} age of a sediment), the whole sedimentary mass, and the continental crust, are determined by the processes that are operative in the formation and destruction of sediments. In the general case any of these three reservoirs may be older or younger than the others. A detailed discussion is beyond the scope of this contribution, and one example can suffice: if there is a linear relationship between the amounts of new material that are added to the continental crust and sedimentary mass, then the mean ages of the continental crust and the sedimentary mass are the same. This holds if sediments are recycled to the continental crust (for example, through granitization or high-grade metamorphism), as long as sediment-to-crust recycling is balanced, with respect to age and mass, by erosion of the crystalline crust to form sediments.

The concordance of t_{CR} and t_{STRAT} ages for the Archean, and the discordance thereafter, may be interpreted, as by Allègre and Rousseau [20], to require increasing rates of continental growth through the Archean, and decreasing rates in post-Archean times. This would be the case if the evolution of the source of sediments directly reflects that of the continental crust, and continental growth has been continuous. However, recycling of old sediments to form new sediments (sediment-sediment recycling), and erosion of previously existing crystalline continental crust, are processes that cause the evolution of the source of sediments to diverge from that of the whole continental crust.

Fig. 5b shows that the Sm-Nd data from sediments, metasediments, and composites are compatible with uniform growth of the entire sedimentary mass since 3.8 Ga. Line *A* represents the mean age of the sedimentary mass through time for uniform growth (and the continental crust, for the condition that there is a linear relationship between addition of new material to the continental crust and the sedimentary mass). The trajectory of line *B*, representing the evolution of the source of sediments, assumes that sediments are formed by mixing newly formed continental crust with pre-existing continent. The conditions are that the stratigraphic mass-age distribution of sediments throughout Earth history is described by exponential decay with a half-mass age of 500 Ma, and that this has been a result of sediment-sediment recycling, and conservation, with respect to mass and age, of sediment-to-crust recycling and erosion of old crystalline crust. Under these conditions, in the early Archean, the mass of sediment from newly formed crust would be large compared to the total sedimentary mass, and would dominate the system. As the sedimentary mass grows, new continental crust would become less important.

It must be emphasized that the model in Fig. 5b is presented to illustrate the important effects of sediment-sediment recycling, and not to advocate uniform growth of the sedimentary mass. A number of continental growth models can be compatible with the Sm-Nd data on sediments. In general, the older the mean age of the continental crust, the greater the required amount of recycling of sediment out of the interactive crystalline crust-sedimentary mass system. Two possible reservoirs for such recycling are the craton and the mantle. Models of continental growth that call for a near constancy of continental mass from the Archean [35] can be compatible if accompanied by initially very fast but constantly decreasing rates of continent-to-mantle recycling.

4.6. Nd isotopic compositions of crustal reservoirs

Fig. 6 is a Sm-Nd isotopic variation diagram with superimposed model iso-age lines. This diagram can be used to estimate bulk compositions or ages of different portions of the crust if two of the three variables of $^{147}Sm/^{144}Nd$, $^{143}Nd/^{144}Nd$, and

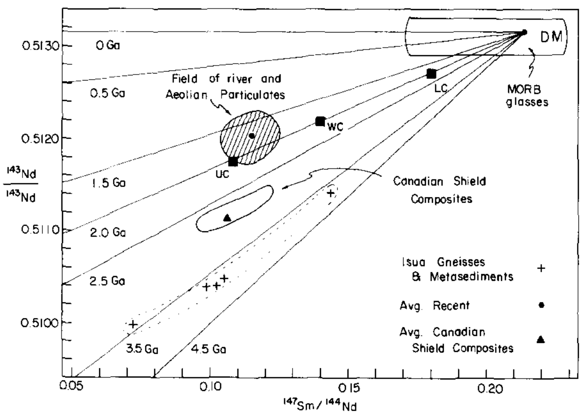


Fig. 6. Comparison of $^{147}Sm/^{144}Nd$ and $^{143}Nd/^{144}Nd$ ratios and lines of equal model age based on a depleted mantle (DM) which has evolved in a single stage from CHUR [11] at 4.6 Ga to $\epsilon_{Nd} \approx +10$ today. Plotted are the fields of most river and aeolian particulates (Table 1), MORB glasses [13,15], data on Isua grey gneisses and metasediments [8], and Archean and Proterozoic Canadian shield composites [7]. Overall, Isua and the composites have Sm/Nd ratios similar to the river and aeolian particulates, and correspond to the Taylor-Shaw upper crustal (*UC*) estimates [37,53]. Using these estimates and those of the Taylor-McLennan whole crust (*WC*) and lower crust (*LC*) [54] the mean $^{143}Nd/^{144}Nd$ ratios of these reservoirs are estimated to be ca. 0.5118, 0.5122, and 0.5127 ($\epsilon_{Nd} = -17, -9, +1$) respectively, if the mean age of these reservoirs is about 2.0 Ga.

t_{CR} are known. For example, the Sm/Nd ratio of the upper continental crust is strongly constrained by the constancy of this ratio since the Archean. If the average age of the upper continental crust is ca. 2.0 Ga, close to that of the sedimentary mass, then its average $^{143}Nd/^{144}Nd$ ratio is about 0.5118 ($\epsilon_{Nd} \approx -17$). The mean $^{143}Nd/^{144}Nd$ ratio of the whole continental crust and the lower continental crust can be estimated if the ratio of Sm/Nd in these reservoirs and their average ages are known.

5. Concluding remarks

The uniformity of Nd isotopic compositions and Sm/Nd ratios in contemporary products of erosion appear to be largely independent of lithological and tectonic diversity within the source regions for aeolian dusts and the major rivers of the Earth. The homogeneity probably results from efficient mixing which has accompanied repeated cycles of erosion and sedimentation. Sm-Nd isotope analyses of sediments preserved in the geologic record provide a powerful tool which can be used to trace the evolution of the sedimentary mass and the continental crust. The identification of the relatively homogeneous character of the continental input into the oceans has important implications for the marine geochemistry of Nd.

Sm-Nd isotopic analyses are likely to be useful not only for regional studies, but also for investigations of local provenance where the fine structure is important. An indication of the average age and composition of a small area of the crust may be provided by analyses of river particulates. Additional analyses of modern and ancient sediments should help to clarify some of the complexities of the evolution of the continents.

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