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Sm–Nd systematics of chondrites

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Abstract

We have studied the $^{147}Sm-^{143}Nd$ and $^{146}Sm-^{142}Nd$ isotopic systems in phosphate fractions and chondrules from six ordinary chondrites and one carbonaceous chondrite, previously dated with Pb–Pb method. $^{147}Sm/^{144}Nd$ ratios vary between 0.182 and 0.191 in phosphates, and between 0.179 and 0.243 in chondrules. The $^{147}Sm-^{143}Nd$ isochron regression through all 34 phosphate and chondrule analyses yields a date of 4588 ± 100 Ma and is in good agreement with more precise Pb–Pb dates of the same chondrites. The initial $^{143}Nd/^{144}Nd$ is 0.50665 ± 0.00014. The same analyses define a $^{146}Sm-^{142}Nd$ isochron with a slope corresponding to $^{146}Sm/^{144}Sm = 0.0075 \pm 0.0027$. Initial $^{142}Nd/^{144}Nd = 1.14160 \pm 0.00011$ corresponds to $\varepsilon^{142}Nd = -2.62 \pm 0.93$. Compilation of the published chondritic whole rock Sm–Nd analyses yields the median $^{147}Sm/^{144}Nd = 0.1964 + 0.0003/-0.0007$, which is our preferred Chondritic Uniform Reservoir (CHUR) value. Using this value and its error limits, we find the present-day CHUR $^{143}Nd/^{144}Nd = 0.512637 + 0.000009/-0.000021$ from the chondritic Sm–Nd isochron that includes all available data for whole rocks, chondrules and phosphates. This value is identical within error with the currently accepted number. An estimate of the bulk earth $^{147}Sm/^{144}Nd = 0.1941 \pm 0.0059$ is obtained from intercept of chondritic $^{146}Sm-^{142}Nd$ isochron with the terrestrial value of $^{142}Nd/^{144}Nd$. This estimate is independent of measured Sm/Nd ratios in chondrites. The same approach was applied to published $^{146}Sm-^{142}Nd$ internal isochrons for differentiated meteorites and yielded similar, although less precise, values. Our data are completely consistent with the currently accepted CHUR parameters and substantiate their use as terrestrial reference values.

Keywords: Chandrites; Sm–Nd; Chur parameter

1. Introduction

The $^{147}Sm-^{144}Nd$ system is an important isotopic tracer of planetary differentiation and crustal growth [1,2], which has been widely used during last 30 years. Accurate interpretation of terrestrial crustal and mantle evolution using Nd isotopes depends on bulk planetary values of Sm/Nd and $^{143}Nd/^{144}Nd$ utilized in the models. Because both Sm and Nd are refractory lithophile elements, it is assumed that the abundance ratio of these elements in chondritic meteorites is similar to the solar system and bulk planetary values [3]. The first high-precision Sm–Nd isotopic study of chondrites [4] established a set of model parameters for the Chondritic Uniform Reservoir (CHUR). These parameters, slightly modified by revision of the oxygen isotope composition used for correction of isotopic ratios measured with NdO^+ ion beam [5], are universally used by the geochemical community. Additional Sm–Nd analyses

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of chondrites of various chemical classes [6–9,53] substantiate these values. It appears therefore that the adopted CHUR Sm–Nd values represent one of the best established and unequivocal basic parameters in modern geochemistry, which hardly needs further investigation.

There are, however, several indications that the Sm–Nd systems of chondrites may be more complex than they seem:

- There are hints of heterogeneity, even if minor, of Sm/Nd in chondrites. These include the difference in Sm/Nd between "light" and "dark" varieties of St. Severin LL6 chondrite [4], and elevated Sm/Nd in the Abee E4 chondrite [6].
- Even greater internal heterogeneity is suggested by Sm/Nd in chondritic phosphates [6,10], which are 3–8% lower than in bulk chondrites. These relatively low Sm/Nd ratios in phosphates imply occurrence of minerals with Sm/Nd ratios higher than the bulk chondritic values.
- Ca–Al-rich refractory inclusions, which are abundant in chondrites of some classes, e.g., CV, tend to have strongly fractionated REE patterns [11,12].

The use of the Sm–Nd system in chondrites as a reference in the studies of planetary evolution is also complicated, as shown by the following observations:

- Although major fractionation of Sm/Nd between the entire solar system, planets and chondrites is unlikely, there is no direct proof that such fractionation did not exist, or that it was negligible.
- On the basis of comparison between terrestrial early Archean, lunar and chondrite Sm–Nd data, it was suggested that the bulk earth and lunar Nd isotopic compositions are probably offset from CHUR by about one ε-unit [13].
- Nd and Hf isotopic ratios in terrestrial crustal and mantle-derived rocks are closely correlated, but the terrestrial Nd–Hf isotopic array is offset from the CHUR value [14–16,53]. There are three possible explanations for this offset: (1) the existence of a "hidden", petrogenetically inactive reservoir in the terrestrial mantle, (2) chondrites may not be representative of the bulk earth in either Lu–Hf or Sm–Nd systems or both, or (3) inaccurate decay constants for ^{176}Lu or ^{147}Sm. The latter two possibilities have to be tested, before the existence of the "hidden reservoir" could be considered.

In this paper, we report new Sm–Nd data for ordinary and carbonaceous chondrites. The approach adopted here differs from the previous studies of Sm–Nd system of chondrites in several ways:

- We have analyzed individual components of meteorites: phosphate minerals and chondrules, rather than whole rocks. This allows us to evaluate the extent of internal fractionation of Sm and Nd. The range of fractionation appears to be sufficiently large for determination of a ^{147}Sm–^{143}Nd isochron age.
- Most of the fractions were also dated using U–Pb and Rb–Sr isotopes. This gives us an independent test of whether the studied phases were formed during the early stages of solar system evolution, and remained a closed system since then.
- For the first time, we have detected the variations of ^{142}Nd/^{144}Nd in chondrites, and determined the initial chondritic ^{146}Sm/^{144}Sm.
- We discuss combined mineral and whole rock Sm–Nd isotope systematics of chondrites and its relationship with the bulk earth Nd isotopic parameters.

2. Studied meteorites and their components

We analyzed phosphate separates and silicate phases (chondrule fragments) from eight ordinary chondrites and one carbonaceous chondrite. The Pb–Pb dates of these chondrites are summarized in Table 1. The Pb–Pb dates of chondrules from seven out of nine meteorites are between 4553 and 4567 Ma, and the Pb–Pb dates of phosphates phases are between 4530 and 4556 Ma. These dates suggest that the studied meteorites have remained undisturbed since at least 4530 Ma, or about 37 Ma. after the formation of the solar system's first solids. Therefore, these meteorites should be suitable for determination of the primary parameters of CHUR. Younger preliminary Pb–Pb dates of chondrules between 4431 and 4489 Ma yielded by two meteorites may be an artifact of common Pb correction

Table 1
Meteorites analyzed in this study and their Pb–Pb ages

Meteorite name	Class	Shock	Chondrule Pb–Pb age (Ma)	2σ	Reference	Phosphate Pb–Pb age (Ma)	2σ	Reference
Richardton	H5	S2	**4562.7**	1.7	[17]	**4550.7**	2.6	[17]
Orlovka	H5		4560.9	4.4	[18]	4555.7	1.9	[19]
Bjurböle	L/LL4	S1	4554.3	3.3	[18]			
Elenovka	L5	S2	4554.2	1.7	[20]	**4535.0**	1.0	[21]
Saratov	L4	S2	4563.6	0.8	[18]	4530.0	6.0	[19]
Krymka	LL3.1	S3	4552.7	10.1	[18]			
Pervomaisky	L6		4431.4	1.5	[18]			
Allende	CV3.2	S1	**4566.8**	1.6	[22]			
Unnamed[a]	L/LL3		**4489**	**43**	[52]			

Chemical class, petrologic type and shock stage are as indicated in [23].
The dates are weighted averages of model $^{207}Pb/^{206}Pb$ dates relative to primordial Pb [24], or Pb–Pb isochron dates.
The isochron dates are highlighted.
[a] This meteorite (not named yet) is described by Herd et al. [52].

(unnamed Antarctic), and/or a result of shock resetting (Pervomaisky).

3. Analytical procedures

Procedures for crushing samples, separating chondrules and mineral fractions, and washing and digesting minerals were described in [17]. For the fractions that were also analyzed for U and Pb, REE were separated from the washes of Pb separation columns. Procedures for Sm and Nd separation and isotopic analyses are similar to those used previously at the Jack Satterly Geochronology Laboratory [25,26]. All fractions were spiked before dissolution with ^{149}Sm–^{150}Nd and, in some cases ^{205}Pb–^{235}U (or ^{205}Pb–^{235}U–^{230}Th) and ^{85}Rb–^{84}Sr mixed tracers. Mutual contamination of spikes, e.g., Sm and Nd introduced with ^{205}Pb–^{235}U and ^{85}Rb–^{84}Sr spikes, was negligible (well below 0.1 pg). Eight procedural blanks measured during this study averaged 2 ± 2 pg for both Sm and Nd, in agreement with our earlier results [26]. Uncertainty in the blank correction is propagated into the errors in $^{147}Sm/^{144}Nd$. This correction slightly increased the errors in $^{147}Sm/^{144}Nd$ for the fractions with less than 1–2 ng of Nd.

Isotopic analyses reported in this study were performed after upgrading our VG-354 mass spectrometer, which included installation of a new data acquisition system (system monitor with built-in digital voltmeters, computer and software) and replacement of an analog Daly detector with an ion counting detector. The analytical protocol set up for the modified mass spectrometer differs in some details from the one used previously. Data acquisition consists of three mass steps using six collectors, and involves measurements of all Nd isotopes as NdO^+, and masses 156, 157, 163 and 165 in each spectrum. The peaks of masses 156 and 157 measured for Ce and Pr interference corrections. The peaks at masses 163 and 165 are used, after appropriate correction for $Nd^{17}O^+$ and $Nd^{18}O^+$, to confirm the absence of Sm in Nd fractions. Small CeO beams were present and were decaying rapidly during analyses. The median $^{140}Ce^{16}O/^{144}Nd^{16}O$ value of 0.00021 (+0.00021/−0.00005,) corresponds to $^{142}Ce/^{142}Nd = 0.000027$, or 23 ppm of $^{142}Nd/^{144}Nd$. The additional uncertainty to $^{142}Nd/^{144}Nd$, introduced by the Ce correction is less than 2 ppm if the isotopic composition of Ce is known with precision of 9 % (a very conservative estimate). The presence of a small Ce signal thus does not compromise precision and accuracy of the $^{142}Nd/^{144}Nd$ ratios. The PrO^+ signal was present at much higher level than Ce (median $^{141}Pr^{16}O/^{144}Nd^{16}O = 0.113 + 0.034/-0.026$), but its impact on precision and accuracy of Nd isotopic ratios is small (only through minor oxygen isotope signal subtraction, which is identical to corrections between Nd isotopes). No Sm beam was detected in any Nd

Table 2
Sm–Nd data and model parameters

No.	Fraction[a]	Fraction weight (mg)	Amount Nd (ng)	Ion yield[b] (%)	[Nd][c] (ppm)	[Sm][d] (ppm)	ε^{150}Sm	2σ	^{147}Sm/^{144}Nd[c]	2σ[c]
1	Richardton Ph	0.740	60.94	1.22	82	25.4			0.1863	0.0006
2	Richardton Ph	1.096	91.97	1.00	84	25.9			0.1866	0.0006
3	Richardton Ph	1.054	108.01	1.20	102	31.9			0.1881	0.0006
4	Richardton Ph	0.261	20.07	3.08	77	23.9			0.1876	0.0006
5	Richardton Ph	2.044	144.83	0.19	71	21.4	2.7	6.4	0.1827	0.0007
6	Orlovka Ph	0.066	5.72	2.67	87	27.5			0.1922	0.0006
7	Orlovka Ph	0.151	13.30	5.57	88	27.8	0.6	6.8	0.1911	0.0006
8	Bjurböle Ph	0.192	25.19	2.52	131	39.8			0.1832	0.0006
9	Bjurböle Ph	0.311	37.54	2.20	121	36.5			0.1830	0.0006
10	Bjurböle Ph	0.230	28.31	1.51	123	37.1	0.7	4.9	0.1821	0.0006
11	Elenovka Ph	0.105	10.43	2.78	99	30.7			0.1867	0.0006
12	Elenovka Ph	0.085	8.88	1.77	104	32.5	4.8	10.0	0.1880	0.0006
13	Elenovka Ph	0.193	19.67	1.00	102	31.6	− 0.4	17.8	0.1872	0.0006
14	Saratov Ph	0.056	2.81	0.60	50	15.5			0.1870	0.0006
15	Richardton Sulf	6.305	0.18	1.72	0.029	0.009			0.1976	0.0020
16	Bjurböle Sulf	0.746	0.09	3.14	0.114	0.035			0.1831	0.0038
17	Richardton LDC	1.441	4.36	1.59	3.03	1.22			0.2429	0.0017
18	Richardton Chond	6.101	2.16	1.28	0.35	0.13			0.2272	0.0012
19	Richardton Chond	6.306	2.17	2.30	0.34	0.13			0.2284	0.0011
20	Richardton Chond	18.561	0.52	1.84	0.028	0.01			0.1787	0.0026
21	Richardton Chond	4.778	2.29	3.83	0.48	0.17			0.2176	0.0008
22	Richardton Chond	19.250	6.95	0.60	0.36	0.14			0.2271	0.0014
23	Richardton Chond	14.290	10.33	0.24	0.72	0.25			0.2049	0.0014
24	Allende Chond	0.636	0.81	3.06	1.27	0.47			0.2262	0.0009
25	Krymka Chond	1.261	1.25	5.45	0.99	0.35			0.2115	0.0007
26	Krymka Chond	1.650	1.00	3.43	0.61	0.20			0.2017	0.0007
27	Krymka Chond	1.295	0.95	2.54	0.74	0.24			0.1963	0.0012
28	Bjurböle Chond	2.203	1.12	1.90	0.51	0.17			0.2048	0.0009
29	Bjurböle Chond	2.054	0.82	3.54	0.40	0.14			0.2199	0.0008
30	Bjurböle Chond	2.437	1.25	0.28	0.51	0.18			0.2135	0.0015
31	Bjurböle Chond	1.016	0.70	1.69	0.69	0.23			0.2041	0.0015
32	Saratov Chond	4.833	2.39	0.85	0.49	0.16			0.2004	0.0010
33	Pervomaisky Chond	2.690	2.38	0.65	0.88	0.29			0.1973	0.0011
34	Unnamed L/LL3	8.270	9.75	0.54	1.18	0.43			0.2207	0.0012
35	Unnamed L/LL3	13.510	35.56	0.15	2.63	0.83			0.1913	0.0020
36	Unnamed L/LL3	36.610	50.35	0.57	1.38	0.45			0.1993	0.0031

[a] Mineral fractions: Ph—Ca phosphate (merrillite and apatite), Sulf—sulfide (mostly troilite), LDC—chondrule fragments picked from low-density fraction ($p < 2.85$), Chond—chondrules and chondrule fragments.

[b] Number of ions of ^{144}Nd registered by the collectors divided by the number of atoms of ^{144}Nd loaded on the filament.

[c] Sm and Nd concentrations and ^{147}Sm/^{144}Nd ratios are corrected for blank of 2 ± 2 pg for both Sm and Nd. The uncertainty is propagated into the error of ^{147}Sm/^{144}Nd ratio.

[d] Deviation ($\times 10^4$) of ^{150}Sm/^{152}Sm in meteorite fractions from the average value of 0.27612 ± 0.00005 measured with the same procedure in standards and terrestrial rocks.

[e] Nd isotopic ratios are corrected for fractionation relative to the ratio of ^{146}Nd/^{144}Nd = 0.7219, using exponential law and real atomic masses.

[f] The ^{143}Nd/^{144}Nd isotopic ratios are adjusted to ^{143}Nd/^{144}Nd = 0.51186 in the La Jolla standard.

[g] The ε^{143}Nd value at 4.56 ± 0.02 Ga. The error includes uncertainties in ^{147}Sm/^{144}Nd and ^{143}Nd/^{144}Nd ratios and the age.

[h] Present-day ε^{142}Nd relative to ^{142}Nd/^{144}Nd = 1.141901 in the La Jolla standard.

$\frac{^{143}Nd}{^{144}Nd}$ [e,f]	2σ	$\varepsilon^{143}Nd$ [g] (T=4.56)	2σ [g]	$\frac{^{142}Nd}{^{144}Nd}$ [e]	2σ	$\varepsilon^{142}Nd$ [h] (T=0)	2σ	$\frac{^{145}Nd}{^{144}Nd}$ [e]	2σ	$\frac{^{148}Nd}{^{144}Nd}$ [e]	2σ
0.512315	0.000010	− 0.37	0.40	1.141868	0.000019	− 0.29	0.16	0.348409	0.000003	0.241577	0.000005
0.512329	0.000010	− 0.27	0.41	1.141878	0.000021	− 0.20	0.19	0.348403	0.000003	0.241570	0.000005
0.512361	0.000013	− 0.54	0.46	1.141874	0.000043	− 0.24	0.38	0.348411	0.000005	0.241585	0.000010
0.512343	0.000010	− 0.60	0.40	1.141883	0.000023	− 0.16	0.20	0.348406	0.000004	0.241581	0.000007
0.512222	0.000013	− 0.05	0.48	1.141895	0.000042	− 0.05	0.36	0.348398	0.000006	0.241568	0.000011
0.512488	0.000013	− 0.46	0.43	1.141911	0.000033	0.09	0.29	0.348403	0.000007	0.241577	0.000011
0.512473	0.000011	− 0.10	0.42	1.141893	0.000027	− 0.07	0.24	0.348392	0.000007	0.241557	0.000009
0.512217	0.000011	− 0.49	0.41	1.141908	0.000035	0.06	0.31	0.348406	0.000007	0.241581	0.000010
0.512220	0.000012	− 0.27	0.44	1.141872	0.000039	− 0.26	0.34	0.348404	0.000006	0.241569	0.000012
0.512244	0.000010	0.75	0.40	1.141890	0.000024	− 0.09	0.21	0.348403	0.000004	0.241570	0.000007
0.512320	0.000014	− 0.52	0.45	1.141882	0.000040	− 0.17	0.35	0.348408	0.000011	0.241575	0.000016
0.512351	0.000023	− 0.66	0.58	1.141899	0.000066	− 0.02	0.58	0.348390	0.000019	0.241585	0.000026
0.512369	0.000012	0.15	0.44	1.141927	0.000024	0.23	0.21	0.348398	0.000009	0.241564	0.000009
0.512330	0.000022	− 0.49	0.56	1.141901	0.000053	0.00	0.46	0.348381	0.000021	0.241561	0.000024
0.512509	0.000117	− 3.29	2.58	1.141862	0.000313	− 0.34	2.74	0.348402	0.000156	0.241666	0.000130
0.511830	0.000332	− 8.06	6.95	1.140539	0.001005	− 11.93	8.80	0.348241	0.000264	0.242413	0.000318
0.514043	0.000019	− 0.10	1.11	1.141984	0.000053	0.73	0.46	0.348456	0.000011	0.241584	0.000021
0.513563	0.000043	− 0.20	1.14	1.141932	0.000103	0.27	0.90	0.348505	0.000042	0.241541	0.000048
0.513601	0.000025	− 0.16	0.85	1.141957	0.000059	0.49	0.52	0.348543	0.000021	0.241573	0.000028
0.512044	0.000102	− 1.18	2.55	1.141899	0.000223	− 0.02	1.95	0.348877	0.000063	0.241509	0.000105
0.513323	0.000020	0.82	0.62	1.141952	0.000046	0.44	0.40	0.348395	0.000013	0.241567	0.000020
0.513530	0.000022	− 0.79	0.96	1.141915	0.000067	0.12	0.59	0.348484	0.000019	0.241589	0.000027
0.512893	0.000055	− 0.06	1.38	1.141935	0.000147	0.30	1.29	0.348428	0.000043	0.241600	0.000067
0.513559	0.000052	0.32	1.17	1.141954	0.000128	0.46	1.12	0.348343	0.000049	0.241604	0.000076
0.513026	0.000020	− 1.43	0.58	1.141881	0.000060	− 0.17	0.52	0.348415	0.000015	0.241570	0.000020
0.512811	0.000032	0.23	0.77	1.141858	0.000075	− 0.38	0.66	0.348412	0.000032	0.241586	0.000043
0.512694	0.000056	1.15	1.31	1.141866	0.000152	− 0.31	1.33	0.348359	0.000044	0.241527	0.000068
0.512933	0.000048	0.76	1.10	1.141912	0.000110	0.10	0.96	0.348366	0.000042	0.241503	0.000057
0.513371	0.000043	0.36	0.98	1.141849	0.000103	− 0.46	0.90	0.348417	0.000035	0.241578	0.000051
0.513069	0.000134	− 1.72	2.79	1.141919	0.000307	0.16	2.69	0.348330	0.000118	0.241592	0.000167
0.512858	0.000083	− 0.30	1.87	1.141899	0.000275	− 0.02	2.41	0.348380	0.000068	0.241570	0.000108
0.512788	0.000050	0.52	1.17	1.141900	0.000137	− 0.01	1.20	0.348360	0.000043	0.241523	0.000066
0.512621	0.000064	− 0.93	1.41	1.142026	0.000172	1.10	1.50	0.348332	0.000050	0.241599	0.000076
0.513412	0.000021	0.72	0.84	1.142012	0.000065	0.98	0.57	0.348457	0.000016	0.241588	0.000021
0.512474	0.000021	− 0.26	1.26	1.141860	0.000073	− 0.36	0.64	0.348403	0.000017	0.241575	0.000023
0.512755	0.000010	0.55	1.84	1.141938	0.000044	0.32	0.39	0.348420	0.000006	0.241578	0.000018

analysis. Sm isotopic composition was analyzed with a similar procedure but using single mass step. Five SmO^+ masses were measured (163, 165, 166, 168, 170), and NdO^+ interference was monitored on mass 162 ($^{146}Nd^{16}O^+$).

Data were processed off-line, and were corrected for minor oxygen isotopes using the ratios $^{18}O/^{16}O = 0.00214 \pm 0.00002$ and $^{17}O/^{16}O = 0.000397 \pm 0.000002$ [25]. Nd isotopic ratios are corrected for fractionation relative to $^{146}Nd/^{144}Nd = 0.7219$, using an exponential law and real atomic masses. Twenty-seven 0.5–10 ng loads of the La Jolla standard run during this study yielded the following weighted average isotopic ratios: $^{142}Nd/^{144}Nd = 1.141901 \pm 0.000008$, $^{143}Nd/^{144}Nd = 0.511873 \pm 0.000004$, $^{145}Nd/^{144}Nd = 0.348403 \pm 0.000003$, $^{148}Nd^{144}Nd = 0.241577 \pm 0.000005$, $^{150}Nd/^{144}Nd = 0.236469 \pm 0.000005$. Weighted average values of nonradiogenic Nd isotopic ratios measured in chondrites: $^{145}Nd/^{144}Nd = 0.348406 \pm 0.000006$ and $^{148}Nd^{144}Nd = 0.241573 \pm 0.000003$, agree with the La Jolla standard values. Ion yields of the La Jolla runs vary between 1.0% and 20.2% (median 3.22 +3.26/− 0.82); isotopic analyses of Nd separated from the chondrites gave ion yields of 0.15–5.6% (median 1.77 + 0.59/ − 0.50), marginally lower than the standard runs.

Isochrons, weighted means and medians and their uncertainties were calculated using Isoplot/Ex version 2.49 [27]. All isochron, weighted mean and median errors have 95% confidence intervals, unless indicated otherwise.

4. Results

4.1. Sm isotopic composition and possible neutron capture effects

One of the Sm isotopes—^{149}Sm—has large effective cross-section for (n,γ) reaction on thermal neutrons [54]. In meteorites that were exposed to cosmic rays (mainly high-energy protons that interact with meteoroid surfaces and produce thermal neutrons) for millions of years, ^{149}Sm is consumed and the equal amount of ^{150}Sm atoms are produced. Thermal neutron fluences determined from Sm and Gd isotopic shifts in chondrites and some other stony meteorites vary mostly between 10^{15}–10^{16} n cm^{-2} ([55,56] and references therein), with corresponding shifts in $\varepsilon^{149}Sm$ (deviation in parts per 10^4 of the $^{149}Sm/^{152}Sm$ ratio in the sample from the $^{149}Sm/^{152}Sm$ ratio in a material that did not experience neutron irradiation) about − 1 to − 7. Higher neutron fluxes and shifts in $\varepsilon^{149}Sm$ up to − 32 were found in aubrites (enstatite achondrites [57]).

If meteorites analyzed in this study have accumulated large neutron irradiation doses, then decrease in natural abundance of ^{149}Sm could compromise determination of Sm/Nd ratios using ^{149}Sm tracer. Since irradiation-related shifts in abundances of ^{149}Sm and ^{150}Sm are correlated ($\varepsilon^{149}Sm/\varepsilon^{150}Sm = -0.534$), the possible isotopic shifts in natural abundance of ^{149}Sm can be estimated from $\varepsilon^{150}Sm$. The procedure for Sm isotopic analysis used here was not designed for complete separation of Nd and high-precision measurement of ^{150}Sm; however, five phosphate fractions from four meteorites (Richardton, Orlovka, Bjurböle and Elenovka) yielded Sm analyses with low Nd background and sufficiently precise $\varepsilon^{150}Sm$ for evaluation of neutron capture effects (Table 2). The $\varepsilon^{149}Sm$ values corresponding to these $\varepsilon^{150}Sm$ values are between 0 and − 2.5 ε-units (possibly up to − 5 ε-units considering the errors), in agreement with the previous Sm isotopic studies of chondrites [55,56]. These possible shifts are insignificant compared to the shifts in $^{149}Sm/^{152}Sm$ due to spiking (between 30% and 500% in most samples); therefore, no corrections for neutron capture effects were applied.

4.2. Sm–Nd systems

The Sm–Nd analytical results are presented in Table 2 and in Figs. 1 and 2. Sm and Nd concentrations in the phosphates are about 100–200 times higher than the abundances in CI chondrites [28]. Sm and Nd concentrations in chondrules are broadly similar to the CI values. Chondrules from unequilibrated (petrologic type 3) chondrites Allende, Krymka and the unnamed have slightly higher Nd concentrations of 0.61–2.63 ppm, than chondrules from equilibrated ordinary chondrites, mostly 0.3–0.5 ppm. This difference may be explained by migration of REE from chondrules and matrix to growing merrillite crystals during chondritic metamorphism. Two fractions of Richardton chondrule fragments show anomalous concentrations of REE.

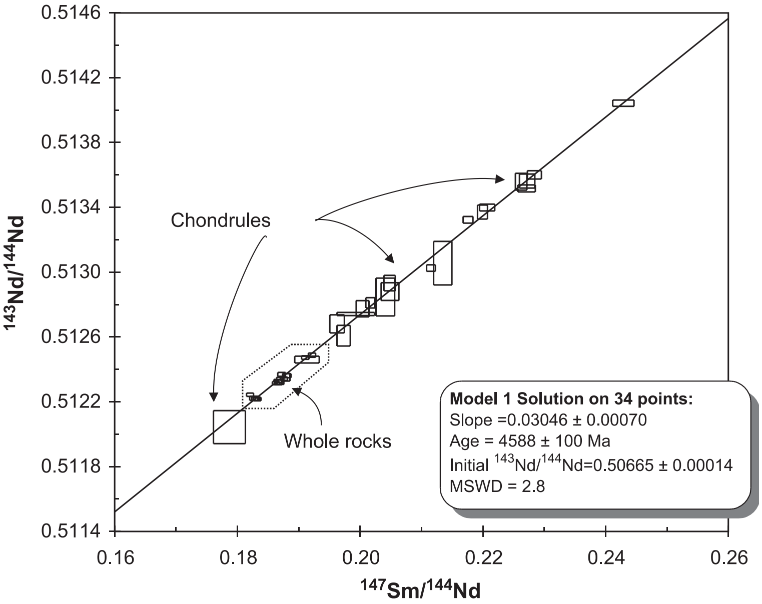

Fig. 1. $^{147}Sm-^{143}Nd$ isochron plot for chondritic phosphates and chondrules analyzed at the Jack Satterly Geochronology Lab, Toronto. Data point error bars are 2σ in all figures.

The chondrule population (#17, Table 2) picked from the low-density fraction (bromoform floats, $\rho < 2.85$), which consists of small, presumably plagioclase-rich, chondrules and their fragments, has Nd concentration of 3 ppm, higher than all the other chondrule fractions. The Sm/Nd of this fraction is also higher than of the other chondrules. The fraction consisting of light-colored, olivine-rich chondrule fragments (#20,

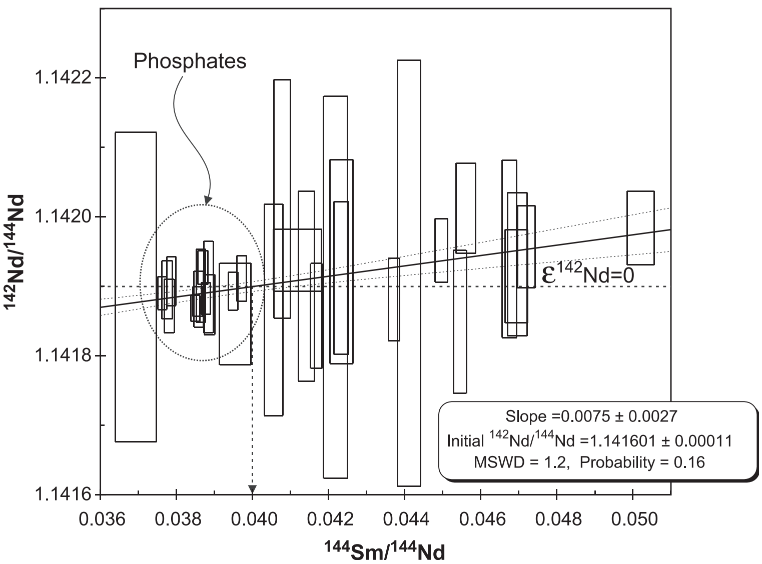

Fig. 2. $^{146}Sm-^{142}Nd$ isochron plot for chondritic phosphates and chondrules analyzed at the Jack Satterly Geochronology Lab. Horizontal dashed line corresponds to $\varepsilon^{142}Nd = 0$. Vertical dashed arrow points to the $^{144}Sm/^{144}Nd$ value corresponding to $\varepsilon^{142}Nd = 0$.

Table 2) has anomalously low Nd concentration of 0.028 ppm, and the lowest Sm/Nd ratio among all chondrule fractions. Sulfides from Richardton and Bjurböle have very low REE contents, and yielded small amounts of Nd (0.09–0.18 ng), insufficient for precise analysis. The sulfide fractions are therefore not considered in the discussion of isotope systematics.

Sm/Nd ratios in phosphates, from 0.182–0.191, cover the same range as in the previously published chondritic phosphate data [6,10]. The silicates show a spread of Sm/Nd from 0.179 to 0.243, wider than the previously studied whole chondrites and phosphates.

The ^{147}Sm–^{143}Nd isochron regression through all 34 phosphate and chondrule analyses (Fig. 1) yields a date of 4588 ± 100 Ma and initial ^{143}Nd/^{144}Nd = 0.50665 ± 0.00014. This date is rather imprecise, but it is consistent with more precise Pb–Pb dates of the same chondrites (Table 1), with exception of the preliminary Pb isotopic dates for the shocked chondrite Pervomaisky, and the date for the unnamed L/LL3 chondrite based on rather unradiogenic Pb isotopic data. Our data do not show any signs of disturbance at 2–2.5 Ga, suggested by the Sm–Nd chondrule data of Krestina et al. [58–60]. Internal isochron regression through five phosphate and seven chondrule fractions from Richardton yielded the date of 4624 ± 120 Ma and initial ^{143}Nd/^{144}Nd = 0.50660 ± 0.00016 (MSWD = 1.8).

The same 34 phosphate and chondrule analyses define a ^{146}Sm–^{142}Nd isochron (Fig. 2), with the slope corresponding to ^{146}Sm/^{144}Sm = 0.0075 ± 0.0027. Initial ^{142}Nd/^{144}Nd = 1.14160 ± 0.00011 corresponds to ε^{142}Nd = -2.62 ± 0.93. Because the Sm–Nd isochron relationships of chondrite components were established shortly after chondrite formation, ^{146}Sm/^{144}Sm and ε^{142}Nd closely approximate initial chondritic values.

5. Discussion

5.1. Comparison with previous chondritic Sm–Nd results and with accepted CHUR parameters

The new phosphate and chondrule data can be considered together with published whole rock and phosphate analyses. In order to compare the data, we need to recalculate them to a consistent scheme of normalization. For renormalization, we used an exponential fractionation law as expressed in [29], real atomic masses, and the accepted ^{146}Nd/^{144}Nd = 0.7219. We therefore expect that all the previous data should be compatible with our data, unless there are some additional analytical biases. The Caltech data [4,6] are renormalized using ^{146}Nd/^{144}Nd=0.724134 [5]. The more recent Caltech data reported in [8] are renormalized using ^{146}Nd/^{144}Nd = 0.724137. The Scripps data for chondritic phosphates [10], which were originally normalized to the ^{148}Nd/^{144}Nd ratio, are renormalized to ^{146}Nd/^{144}Nd = 0.721878 reported in [30]. The Berkeley data [7] were first converted to the Caltech system using their reported ^{146}Nd/^{142}Nd (0.636130 in [7] vs. 0.636151 in [5]), and then normalized the same as the early Caltech data (the equivalent Berkeley ^{146}Nd/^{144}Nd = 0.724122). All these renormalized data are presented in the Supplementary Table 1. The data reported by the University of Arizona group [53] are obtained using the same normalization scheme as used in this paper, and are included in the Supplementary Table 1 without additional correction. We have to note that renormalization of published data is not quite straightforward, because various isotopic ratios, fractionation laws, oxygen isotope correction and spike subtraction schemes were involved in the primary data normalization at various labs. Therefore, some residual biases between the data from different labs may be expected. Since all the Nd isotopic data included in our compilation are either reported relative to the accepted ^{143}Nd/^{144}Nd = 0.51186 for the La Jolla Nd standard, or the original papers report the La Jolla standard results very close to this value, we expect the residual biases to be small.

In Fig. 3, all mineral and whole rock data are plotted together. The combined isochron yields the date of 4547 ± 110 Ma and initial ^{143}Nd/^{144}Nd = 0.50671 ± 0.00015. These values are similar to the values obtained from phosphate and chondrule data alone.

In order to reveal potential fine-scale differences between the data sets, we plot the ^{147}Sm–^{143}Nd data as deviations from the accepted CHUR values. We use the present-day CHUR ^{143}Nd/^{144}Nd = 0.512646, which is derived from the value of 0.511847 in the Caltech normalization system [5,6] through the normalization procedure accepted in this study. This value is 0.14 ε-units higher than ^{143}Nd/^{144}Nd = 0.512638, derived

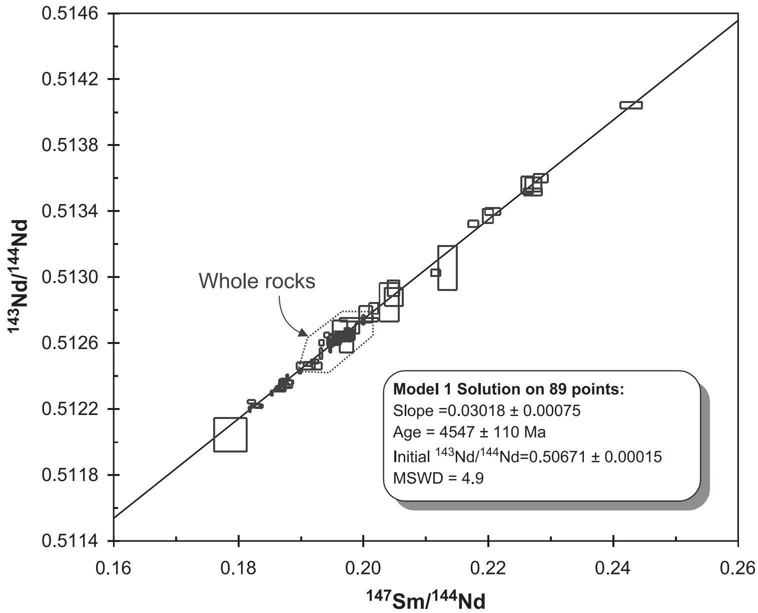

Fig. 3. ^{147}Sm–^{143}Nd isochron plot for chondritic phosphates, chondrules and whole rocks ([4,6–8,10,53] and this study).

from the same starting value by power law, nominal mass normalization [5]. The ε^{143}Nd (4.56 Ga) values are plotted in Fig. 4 show that there are, indeed, systematic shifts between the data sets. The greatest difference of 0.96 ± 0.30 is observed between the weighted average values of more recent Caltech data (-0.60 ± 0.22 [8]), and the Berkeley data ($+0.36 \pm 0.20$ [7]). There may be also smaller biases, not exceeding 0.5 ε-units, between the other data sets. The weighted average ε^{143}Nd (4.56 Ga) values are -0.23 ± 0.18 for the Toronto data, 0.06 ± 0.12 for the Scripps phosphate analyses [10], -0.28 ± 0.13 for the 1980–1984 Caltech whole rock analyses [4,6], and 0.10 ± 0.27 for the 2003 Arizona data [53]. The observed differences are not caused by using an inappropriate reference age, because all the data points, especially whole rocks and phosphates, have ^{147}Sm/^{144}Nd ratios close enough to the accepted CHUR value to make ε^{143}Nd(T) rather insensitive to the choice of T. It is also unlikely that the differences are caused by analytical bias in ^{147}Sm/^{144}Nd ratios, because all the labs involved in Sm–Nd studies routinely measure these ratios with precision and accuracy of 0.05–0.2% (larger errors in the analyses of very small samples are produced by blank correction), and their mixed Sm–Nd spike calibrations are tested against the widely distributed normal solution [5]. The cause of these interlaboratory biases in ε^{143}Nd(T) is unknown. We realize that it would be the best to assure complete analytical consistency between all the data, but at this point we have no reason to exclude any particular data set on analytical grounds.

The combined chondrite Sm–Nd data set allows us to reevaluate the CHUR parameters. Following the approach of [4,31], we seek to define the present day rather than initial parameters. The first step is choosing the representative ^{147}Sm/^{144}Nd value. As it was pointed out by [4], there is no unique way of selecting a unique solar system (CHUR) value from a data set of variable chondritic ^{147}Sm/^{144}Nd. ^{147}Sm/^{144}Nd in all analyzed bulk chondrites are summarized in a histogram plot in Fig. 5. The median value of $0.1964 + 0.0003/-0.0007$ is chosen as the representative CHUR value. We use the median because it is insensitive to outliers and does not require assumptions about the shape of the distribution. Using this value and its error limits, we calculate from the all-data isochron line slope of 0.03018 and y-intercept of 0.50671 (Fig. 3) the present-day CHUR ^{143}Nd/^{144}Nd $= 0.512637 + 0.000009/-0.000021$.

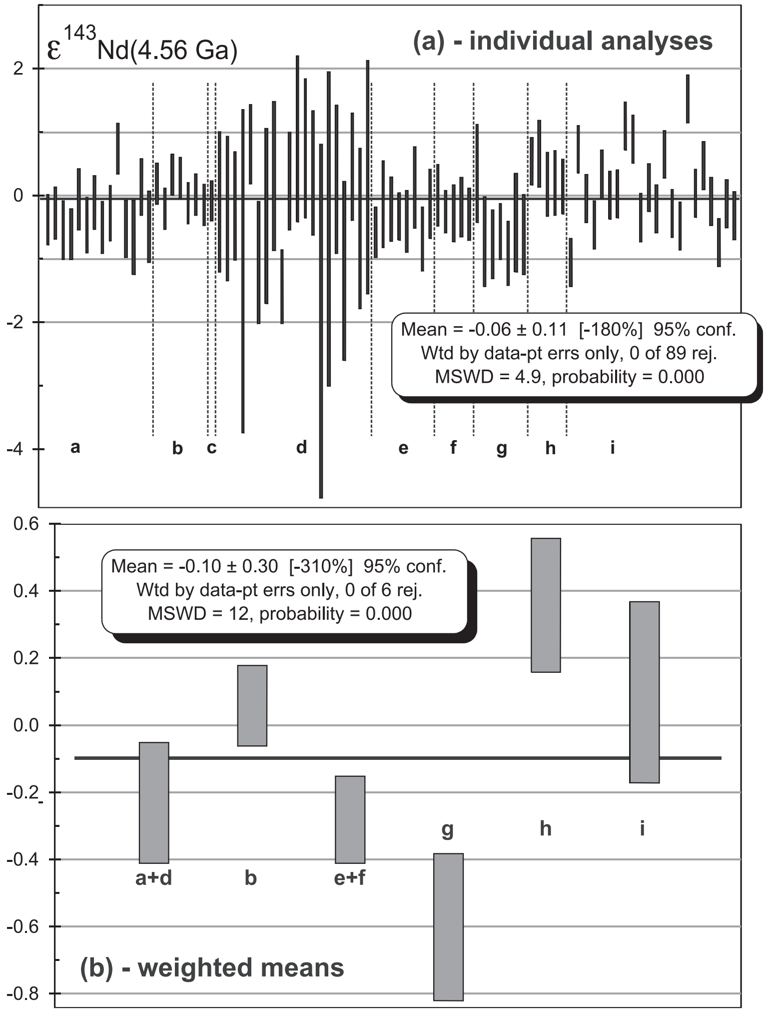

Fig. 4. ε^{143}Nd values at 4.56 Ga for chondritic phosphates, chondrules and whole rocks ([4,6–8,10,53] and this study), calculated relative to the CHUR parameters ^{147}Sm/^{144}Nd = 0.1966 and ^{143}Nd/^{144}Nd = 0.512646 (see text for details): (a) individual analyses, (b) weighted means. Data sets: (a) phosphates, this study; (b) phosphates, Brannon et al. [10]; (c) phosphate, Jacobsen and Wasserburg [4]; (d) chondrules, this study; (e) whole rocks, Jacobsen and Wasserburg [4]; (f) whole rocks, Jacobsen and Wasserburg [6]; (g) whole rocks, Prinzhofer et al. [8]; (h) whole rocks, DePaolo et al. [7]; (i) whole rocks, Patchett et al. [53].

These CHUR parameters agree exceptionally well with the currently accepted numbers.

The CHUR ^{147}Sm/^{144}Nd and ^{143}Nd/^{144}Nd values calculated here are rather precise; however, they do not necessarily exactly match solar, bulk earth, or even average chondrite values. The precise relationship between chondritic and solar abundances of Sm and Nd is complicated by the fact that only CI chondrites show abundance patterns of refractory lithophile elements similar to the solar pattern, whereas the other chondrites deviate from the solar pattern [28,61–63]. The only CI chondrite (Orgueil) studied for Sm–Nd isotopic systematics yielded ^{147}Sm/^{144}Nd = 0.1962 [8], in excellent agreement with the median value for all chondrites.

The internal variations of ^{147}Sm/^{144}Nd observed in chondrules and chondritic minerals in this study suggest that the variations among bulk chondrite samples

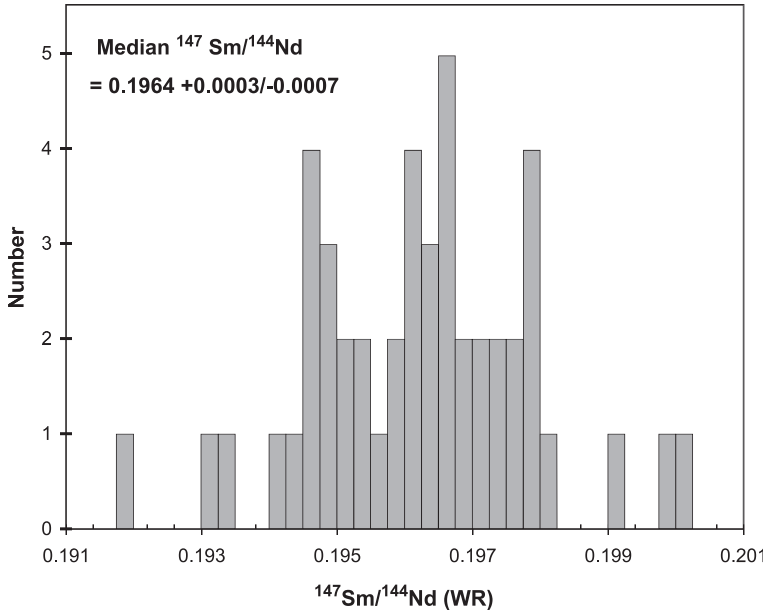

Fig. 5. Histogram showing $^{147}Sm/^{144}Nd$ ratios in all analyzed chondrite whole rocks [4,6–8,53].

may be caused by variable proportions of chondrules, matrix, and secondary minerals. More representative data can be thus obtained from large chondrite samples. However, since chondrites are not direct and unmodified condensates from the solar nebula but are mixtures of components of various origins and thermal histories, the sample size alone is not sufficient to assure that the observed Sm/Nd represents either solar or average chondritic values. We suggest that the future studies should examine the correlation between chondritic Sm/Nd ratio (as well as Lu/Hf, and other elemental ratios of interest for isotope geochemistry) and chondrite composition and primary mineralogy (i.e., chemical group), metamorphic conditions (i.e., petrologic type), and sample size.

The relationship between the CHUR and the bulk silicate Earth Sm–Nd systematics is perhaps even more complex. In the following section, we propose a new way for evaluation of the bulk terrestrial Sm/Nd ratio.

5.2. $^{146}Sm–^{142}Nd$ systematics of chondrites and bulk terrestrial Sm/Nd ratio

Bulk earth Sm/Nd can be estimated from the intercept between a meteorite primary $^{146}Sm–^{142}Nd$ isochron and the line corresponding to the modern average terrestrial value (i.e., $\varepsilon^{142}Nd = 0$), as shown in Fig. 2. This estimate does not depend on measured Sm/Nd in chondrites, but it does depend on two conditions: homogeneous initial $^{146}Sm/^{144}Sm$ in the solar system, and early accretion of the earth over a period of time much shorter than the half-life of ^{146}Sm.

The first condition of homogeneous solar system $^{146}Sm/^{144}Sm$ can be checked by plotting together two parameters of $^{146}Sm–^{142}Nd$ isochron regressions for various meteorites: $^{146}Sm/^{144}Sm$ (isochron slope) and initial $\varepsilon^{142}Nd$ (isochron y-intercept). This approach has been used previously to verify the homogeneity of initial $^{146}Sm/^{144}Sm$ in differentiated meteorites [8,32]. The $^{146}Sm–^{142}Nd$ isochron results based on published data for differentiated meteorites and chondrite data reported here (Table 3) show close correlation (Fig. 6), which reflects isochron rotation with decay of ^{146}Sm. If the initial distribution of ^{146}Sm was heterogeneous, the data would have been scattered.

Homogeneity in the initial distribution of ^{146}Sm can also be tested by plotting the $^{146}Sm/^{144}Sm$ ratios (isochron slopes) against absolute ages of the mete-

Table 3
Summary of $^{146}Sm-^{142}Nd$ isochron regressions[a]

No.	Meteorite	Class	MSWD	Points	$\frac{^{146}Sm}{^{144}Sm}$	Error	$\varepsilon^{142}Nd$ initial	Error	$^{147}Sm/^{144}Nd$ at $\varepsilon^{142}Nd = 0$	Error[a]	Reference
1	Angra dos Reis	Angrite	0.61	7	0.0047	0.0023	− 1.36	0.67	0.1610	0.0195	[33]
2	Moama[c]	Eucrite	1.70	3	0.0041	0.0013	− 1.62	0.58	0.2195	0.0293	[6]
3	Angra dos Reis	Angrite	0.80	3	0.0117	0.0032	− 4.17	1.00	0.1976	0.0122	[6]
4	Bholdhati	Howardite	1.13	4	0.0032	0.0016	− 0.82	0.62	0.1415	0.0927	[34]
5	LEW-86010	Angrite	0.15	5	0.0071	0.0017	− 2.58	0.61	0.2024	0.0122	[35]
6	LEW-86010	Angrite	4.6	12	0.0084	0.0024	− 2.93	0.94	0.1902	0.0171	[36]
7	Ibitira	Eucrite	5.5	6	0.0092	0.0042	− 3.00	1.60	0.1780	0.0317	[8]
8	Acapulco	Acapulcoite	0.80	6	0.0070	0.0019	− 2.21	0.74	0.1756	0.0195	[8]
9	Morristown[d]	Mesosiderite	0.85	4	0.0075	0.0015	− 2.13	0.54	0.1561	0.0244	[8]
10	VacaMuerta p16	Mesosiderite	18	7	0.0069	0.0052	− 2.10	2.40	0.0878	0.1268	[32]
11	VacaMuerta p12	Mesosiderite	0.60	6	0.0063	0.0010	− 0.95	1.20	0.0878	0.0878	[32]
12	VacaMuerta p5	Mesosiderite	5.5	6	0.0050	0.0031	− 1.70	1.20	0.1854	0.0585	[32]
13	Mt Padbury	Mesosiderite	1.90	6	0.0058	0.0016	− 1.99	0.61	0.1878	0.0244	[32]
14	Caldera	Eucrite	0.77	4	0.0074	0.0015	− 3.01	0.55	0.1537	0.0195	[37]
15	Caddo	IAB iron	3.3	4	0.0090	0.0050	− 2.89	1.30	0.1756	0.0927	[38]
16	Piplia Kalan	Eucrite	3.2	4	0.0046	0.0056	− 1.80	2.30	0.2146	0.0488	[39]
17	Chond Px + Ph	Chondrites	1.20	34	0.0075	0.0027	− 2.62	0.93	0.1941	0.0059	This
18	Chond WR + min	Chondrites	1.60	21	0.0071	0.0033	− 2.50	1.20	0.1976	0.0073	[4,6,8,53], this
19	Richardton	H5 chondrite	0.46	12	0.0088	0.0032	− 3.16	1.10	0.1995	0.0078	This
	Weighted Mean—all differentiated meteorites				0.0062	0.0010	− 2.12	0.47	0.1857	0.0117	
	Weighted Mean—LEW-86010 and Acapulco				0.0074	0.0011	− 2.53	0.42	0.1937	0.0088	

[a] Isochron regressions are performed using Isoplot—Ex version 2.49. Errors are 95% confidence intervals, and include appropriate error magnification for isochrons with excessive scatter of the data points. In order to minimize normalization-related biases, the data are regressed as $\varepsilon^{142}Nd$ (reported in the original publication, or calculated from sample and standard $^{142}Nd/^{144}Nd$ ratios) vs. $^{146}Sm/^{144}Nd$.

[b] The errors are estimated from the intersection of the $\varepsilon^{142}Nd$ line with the isochron error envelopes. These error limits are in many cases asymmetric. For simplicity, we have accepted the larger of the two error limits for each isochron.

[c] Two pyroxene analyses averaged.

[d] Four low-precision analyses not included in regression, as suggested in the original publication.

orites obtained with $^{147}Sm-^{144}Nd$ or another extant isotopic chronometer, for example U–Pb (e.g., Fig 6 in [32]). If the initial distribution of ^{146}Sm was homogeneous, the absolute ages are accurate, and the $^{146}Sm-^{142}Nd$ system as well as the isotopic system of the absolute chronometer were closed simultaneously, then the data would plot along the decay curve of ^{146}Sm, which starts at the initial $^{146}Sm/^{144}Sm$ at the time of formation of the solar system. However, we have to withhold from using this approach, because of the major uncertainties in absolute age determination of differentiated meteorites with complex igneous and metamorphic history [8,32,40].

The second condition is early and fast accretion of the earth. If the earth material separated from the solar nebula during first 30–50 Ma of accretion, then the bulk earth $^{146}Sm-^{142}Nd$ value would plot on the primary isochron, irrespective of whether the Sm/Nd ratio of the earth is chondritic or not. The timing and the rate of accretion are hard to test directly, but there is extensive indirect evidence for fast terrestrial and other planetary core formation, based on $^{182}Hf-^{182}W$ isotopic system [41,42]. These studies establish the upper time limit on the accretion of the earth. Other short-lived isotopic systems [43] and dynamic accretion models [44] also suggest that the accretion is fast compared to the ^{146}Sm decay.

There is, therefore, strong evidence that both of the conditions involved in determination of bulk terrestrial Sm/Nd ratio from $^{146}Sm-^{142}Nd$ systematics are satisfied. We further assume that the standards used in Nd isotopic analyses, probably originating from REE deposits, represent bulk earth $^{142}Nd/^{144}Nd$. This as-

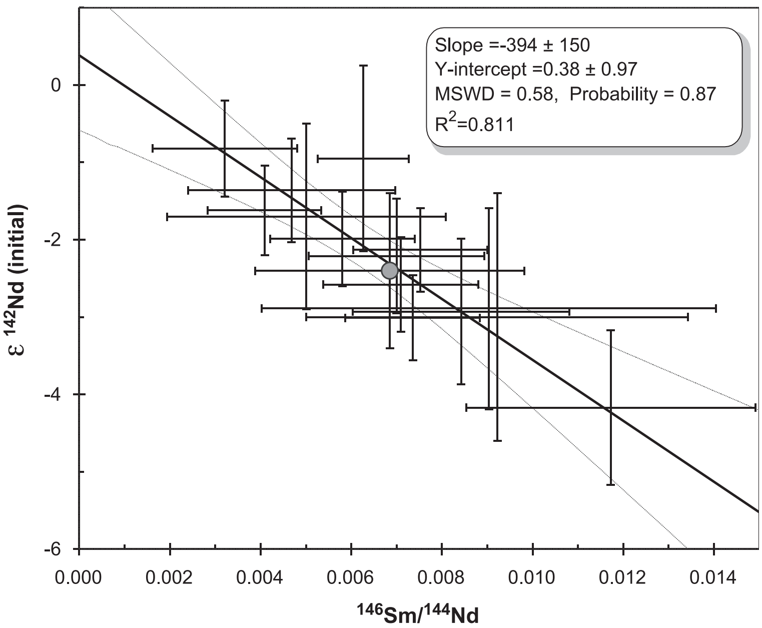

Fig. 6. $^{146}Sm/^{144}Sm$ (isochron slope) vs. initial $\varepsilon^{142}Nd$ (isochron *y*-intercept), obtained from $^{146}Sm-^{142}Nd$ isochron regressions for various meteorites. The chondrite isochron value is marked with a circle. The curves outline the 95% confidence error envelope. For the data, their sources, and additional information, see Table 3.

sumption is likely to be valid because terrestrial $^{142}Nd/^{144}Nd$ appears to be homogeneous, with the exception of small (less than 0.15–0.33 ε-units) variations in some early Archean rocks [45–50].

$^{147}Sm/^{144}Nd$ calculated from intercepts of meteoritic $^{146}Sm-^{142}Nd$ isochrons with the $\varepsilon^{142}Nd = 0$ (terrestrial $^{142}Nd/^{144}Nd$) line are summarized in Table 3. The isochron of chondrules and chondritic phosphates gives the most precise terrestrial $^{147}Sm/^{144}Nd = 0.1941 \pm 0.0059$. Similar values, with a weighted average of 0.1937 ± 0.0088, are obtained from internal isochrons for primitive achondrites, which are thought to have relatively simple history: angrite LEW 86010 [35,36] and Acapulco [8] (the data for Angra dos Reis are not included in this estimate because of unresolved conflict between two published data sets [6,33]). The data set for all differentiated meteorites is more scattered and yields a less precise weighted average of 0.1857 ± 0.0117, which is also consistent with the chondrite and primitive achondrite estimates. All these estimates overlap with the range of $^{147}Sm/^{144}Nd$ ratios measured in chondrites and agree very well with the CHUR best estimate of 0.1964.

The estimates of bulk terrestrial Sm/Nd from currently available meteorite $^{146}Sm-^{142}Nd$ isochrons are not sufficiently precise to be used in the reference Sm–Nd data set for terrestrial applications in place of the CHUR parameters. However, with new generation high precision mass spectrometers (e.g., Finnigan Triton TI), maximizing the sensitivity of Nd isotopic analysis, and careful selection and preparation of sufficiently large samples of chondrule fragments, it may be possible to considerably improve precision of chondritic Sm–Nd isochrons. This would allow direct determination of the bulk terrestrial Sm–Nd isotopic parameters with similar or better precision than the uncertainty in CHUR. Consistency (or discrepancy) between CHUR and bulk earth Sm–Nd parameters may allow detection of a "hidden reservoir" in the earth mantle [14–16,51] and evaluation of its size.

6. Conclusions

(1) The range of variations of Sm/Nd ratios among chondritic phosphates and chondrules greatly exceeds the variations among whole chondrites.

These variations were sufficient to define a $^{147}Sm-^{143}Nd$ isochron with precision of about ± 100 my. Although this precision is not high enough to study the timing of chondrite formation and metamorphism, the agreement of the $^{147}Sm-^{143}Nd$ isochron date with more precise Pb–Pb ages confirms closed system behavior of Sm–Nd in chondrites.
(2) For the first time, we directly determined the initial abundance of ^{146}Sm in chondrites, corresponding to $^{146}Sm/^{144}Sm = 0.0075 \pm 0.0027$, from a $^{146}Sm-^{142}Nd$ isochron defined by the same chondrule and phosphate fractions.
(3) From a compilation of published Sm–Nd analyses of bulk chondrites, and a $^{147}Sm-^{143}Nd$ isochron that combines whole rock, phosphate and chondrule analyses converted to one normalization system, we reevaluated the Chondritic Uniform Reservoir parameters. Our preferred values are $^{147}Sm/^{144}Nd = 0.1964 + 0.0003/-0.0007$ (the median of published bulk chondrite analyses with 95% confidence interval), and present-day $^{143}Nd/^{144}Nd = 0.512637 + 0.000009/-0.000021$.
(4) We propose a new approach to determine the bulk terrestrial Sm/Nd independent of measured Sm/Nd ratios in chondrites, from the intercept of the chondritic $^{146}Sm-^{142}Nd$ isochron with the terrestrial value of $^{142}Nd/^{144}Nd$. The chondrite isochron yields the bulk earth $^{147}Sm/^{144}Nd = 0.1941 \pm 0.0059$. Published $^{146}Sm-^{142}Nd$ internal isochrons for differentiated meteorites yield similar, although less precise, values. With improved precision of future Sm–Nd analyses of chondrites, it may be possible to determine bulk earth Sm–Nd isotopic parameters with similar or better precision than the uncertainty in CHUR.
(5) Our data are entirely consistent with the accepted CHUR parameters and substantiate their use as terrestrial reference values.

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