

Data Repository Supplementary Information

Analytical methods: Iron isotope compositions were measured using a Micromass *IsoProbe* on 400 to 300ppb Fe solutions prepared from 25ppm Fe stock solutions. Details of the mass spectrometry measurements are reported in Beard et al. (2003). The precision (1- σ) of measured iron isotope ratios using this method is ± 0.05 ‰ for the $\delta^{56}\text{Fe}$ value and ± 0.07 ‰ for the $\delta^{57}\text{Fe}$ value. This estimate of precision is based on three methods. First, the long-term (1 year) precision of mass spectrometry measurements has been determined by analysis of three ultra-pure Fe standards. These three ultra-pure Fe standards include an Fe solution obtained from High Purity Standards (UW HPS Fe), an iron rod from Johnson Matthey that was dissolved in HCl (UW J-M Fe), and an iron wire purchased from the Institute on Reference Materials and Methods (IRMM-014) that was dissolved in HNO₃. Fifty-two analyses of UW HPS Fe average $\delta^{56}\text{Fe} = 0.49 \pm 0.05$ ‰ and $\delta^{57}\text{Fe} = 0.74 \pm 0.07$ ‰; 47 analyses of UW J-M Fe average $\delta^{56}\text{Fe} = 0.25 \pm 0.05$ ‰ and $\delta^{57}\text{Fe} = 0.39 \pm 0.07$ ‰; and 54 analyses of IRMM-014 average $\delta^{56}\text{Fe} = -0.09 \pm 0.05$ ‰ and $\delta^{57}\text{Fe} = -0.11 \pm 0.07$ ‰. The second method for evaluating the precision of Fe isotope ratios is replicate analysis of individual 25 ppm Fe stock solutions. Of the 136 terrestrial igneous rocks (Beard et al., 2003), aerosols (Table DR1) clastic sedimentary rocks (Tables DR2-DR6), and MOR hydrothermal solutions (Table DR7) that we have analyzed, replicate analyses on 101 different stock solutions have been performed; the average reproducibility is ± 0.05 ‰ for the $\delta^{56}\text{Fe}$ value and ± 0.07 ‰ for the $\delta^{57}\text{Fe}$ value. The third method that we have used to evaluate precision of Fe isotope measurements is to perform complete replicate analyses on 22 different samples, which includes dissolution of different splits from the whole rock powders or aliquots of solutions. The average reproducibility of these complete replicate analyses is ± 0.05 ‰ for the $\delta^{56}\text{Fe}$ value and ± 0.07 ‰ for the $\delta^{57}\text{Fe}$ value. Additionally, we have checked to ensure that our chemical processing of Fe solutions does not produce a bias in measured

Fe isotope compositions by preparing synthetic rock and hydrothermal solutions using the three ultra-pure Fe standards. Iron isotope analysis of these synthetic rock and fluids, following chemical processing, reproduces the Fe isotope composition of the ultra-pure Fe standards. Results of Fe isotope analysis of synthetic hydrothermal solutions are reported in Table DR7.

The precision of an individual Fe isotope measurement is based on the internal counting statistics during the analysis. Our analysis method consists of 30, 10-second on-peak integrations, and the errors associated with each analysis are reported as the standard error of the mean of the 30 measurements. The average internal precision (1-SE) of the 136 samples that we analyzed is ± 0.03 ‰ for the $\#^{56}\text{Fe}$ value and ± 0.02 ‰ for the $\#^{57}\text{Fe}$ value, which significantly underestimates the external error of Fe isotope ratio measurements that we have established. We therefore report 2-SE for individual analyses because it provides a better approximation of the real external (1 σ) precision, which is ± 0.05 ‰ for the $\#^{56}\text{Fe}$ value.

The hydrothermal fluids were collected following the methods of Von Damm et al. (1985). Due to the high concentrations of metals in the hydrothermal solutions, precipitation often occurs within the titanium sampling bottles as they cool to the ambient (2°C) temperature. The solutions were transferred shipboard to an acid cleaned HDPE bottle and acidified with distilled HCl. The Ti sampling bottles were then disassembled and any particles remaining in the bottles were transferred to a 30 ml acid cleaned HDPE bottle using 18.2 megohm water; this fraction is referred to as the "DREGS". In the shore based laboratory, the water samples were filtered in laminar flow benches through 0.4micron Nucleopore filters and collected in new acid cleaned HDPE bottles. This is the

"dissolved" fraction. The filters with precipitate were then returned to the original HDPE bottles. This is referred to the "bottle and filter" (B/F) fraction. In the shore based laboratory, the DREGS fraction was evaporated to dryness in laminar flow benches. Both the dregs and B/F fractions were subsequently digested with distilled HNO₃ using teflon Parr microwave digestion vessels. Once the digestions were complete, the B/F and DREGS fractions were each brought to ~10ml total volume with HNO₃. The concentrations in each of the three fractions are measured by flame atomic absorption using NIST-traceable standards, and the values combined based on the volume of the digests and sample to arrive at a total concentration value for the vent fluid sample. In most samples the DREGS fraction appears to be predominately comprised of small pieces of the sulfide chimney that was cored by the bottle snorkels during sample collection, and therefore should not be included in the fluid sample total concentration. For all of the samples reported here, the Fe in the dissolved fraction is >90% of the total Fe for the sample. The iron concentrations, isotopic composition, and relevant masses for each individual sample fraction are reported in Tables DR7 and DR8 and these Fe isotope values are used to calculate the Fe isotope composition of the total solution by mass balance. For those samples with out a corresponding DREGS fraction, the calculated Fe isotope composition is based only on the B/F and dissolved Fe fractions. Because the B/F and DREGS fraction are only a small percentage of the total Fe, the changes in the calculated Fe isotope composition for the bulk fluid samples are not significantly different from those in the dissolved phase. The B/F fraction typically has a lower $\delta^{56}\text{Fe}$ values by 0.3 to 1.1 ‰ as compared to the dissolved Fe fraction, although for two samples the B/F and dissolved Fe fractions have indistinguishable Fe isotope

compositions. The DREGS fraction was analyzed for two samples. For these two samples the DREG fraction has an Fe isotope composition that is analytically indistinguishable from the corresponding dissolved Fe fraction.

References:

- Beard, B.L., Johnson, C.M., Skulan, J., Neelson, K.H., Sun, H., and Cox, L, 2003, Application of Fe isotopes to tracing the geochemical and biochemical cycling of Fe. In Lyons, T, and Zhang, K (eds.) *Isotopic record of microbially mediated processes*, Chemical Geology, in press.
- Ben Othman, D., White, W.M., and Patchett, P.J., 1989, The geochemistry of marine sediments, island arc magma genesis, and crust-mantle recycling. *Earth Planetary Science Letters*, v. 94, p. 1-21.
- Canfield, D.E., 1997, The geochemistry of river particulates from the continental USA: major elements. *Geochimica Cosmochimica Acta*, v. 61, p. 3349-3365.
- Flanagan, F.J., 1976, Descriptions and analyses of eight new USGS rock standards. U.S.G.S. Prof. Paper 840, 192 pp.
- McLennan, S.M., Taylor, S.R., McCulloch, M.T. & Maynard, J.B., 1990, Geochemical and Nd-Sr isotopic composition of deep-sea turbidities: Crustal evolution and plate tectonic associations. *Geochimica Cosmochimica Acta*, v. 54, p. 2015-2050.
- Nance, W.B., and Taylor, S.R., 1976, Rare earth element patterns and crustal evolution—I. Australian post-Archean sedimentary rocks. *Geochimica Cosmochimica Acta*, v. 40, p.1539-1551.
- Plank, T., and Langmuir, C.H., 1998, The chemical composition of subducting sediment: implications for the crust and mantle. *Chemical Geology*, v. 145, p. 325-394.
- Taylor, S.R., McLennan, S.M., and McCulloch, M.T., 1983, Geochemistry of loess, continental crustal composition and crustal model ages. *Geochimica Cosmochimica Acta*, v. 47, p. 1897-1905.
- Von Damm, K.L, Edmond, J.M., Grant, B., Measures, C.I., 1985, Chemistry of submarine hydrothermal solutions at 21°N, East Pacific Rise. *Geochimica Cosmochimica Acta*, v. 49, p. 2197-2220.

Table DR1: Fe Isotope Composition of aerosol samples

Sample Name	Sample Notes	----Analysis 1----		----Analysis 2----		----Analysis 3----		----Mass Spec Average----				----Average of Replicate----			
		$\delta^{56}\text{Fe}$	$\delta^{57}\text{Fe}$	$\delta^{56}\text{Fe}$	$\delta^{57}\text{Fe}$	$\delta^{56}\text{Fe}$	$\delta^{57}\text{Fe}$	$\delta^{56}\text{Fe}$	1-SD	$\delta^{57}\text{Fe}$	1-SD	$\delta^{56}\text{Fe}$	1-SD	$\delta^{57}\text{Fe}$	1-SD
AA-KO-TSP-TM-4	1	0.13±0.05	0.35±0.03	0.10±0.07	0.25±0.05			0.12	0.02	0.30	0.07				
AA-KO-TSP-TM-5	1	0.06±0.05	0.13±0.03	0.07±0.09	0.08±0.04			0.07	0.01	0.11	0.03				
AA-KO-TSP-TM-13	1	-0.11±0.06	-0.07±0.04	0.01±0.09	-0.10±0.05			-0.05	0.09	-0.09	0.02				
AA-KO-TSP-TM-15	1	-0.11±0.06	-0.11±0.03	0.04±0.08	-0.02±0.04	0.13±0.06	0.12±0.04	0.02	0.12	-0.01	0.12				
AA-KO-TSP-TM-21	1	0.10±0.07	0.04±0.03	0.13±0.08	0.13±0.04			0.12	0.02	0.09	0.06				
AA-KO-TSP-TM-23	1	-0.05±0.06	0.02±0.04	0.04±0.06	0.05±0.04			0.00	0.06	0.04	0.02				
AA-KO-TSP-TM-27	1	-0.01±0.06	0.04±0.04	0.11±0.06	0.09±0.03			0.05	0.08	0.07	0.04				
AA-KO-TSP-TM-16	1	0.05±0.07	0.06±0.04	0.08±0.06	0.16±0.03			0.06	0.02	0.11	0.07				
AA-KO-TSP-TM-20	1	-0.02±0.05	0.09±0.04					-0.02	0.05	0.09	0.04				
AA-KO-TSP-TM-2	1	0.02±0.07	0.11±0.04					0.02	0.07	0.11	0.04				
AA-KO-TSP-TM-28	1	-0.07±0.05	-0.01±0.03					-0.07	0.05	-0.01	0.03				
AA-KO-TSP-TM-29	1	-0.09±0.06	-0.08±0.04					-0.09	0.06	-0.08	0.04				
AA-DH-TSP-4	2	-0.17±0.09	-0.16±0.04	-0.14±0.07	-0.15±0.04			-0.15	0.02	-0.16	0.01				
AA-DH-TSP-7	2	-0.10±0.07	-0.13±0.04	-0.10±0.12	-0.05±0.06			-0.10	0.00	-0.09	0.06				
SRM-1649A Aliquot 1	3	-0.12±0.04	-0.13±0.04	-0.03±0.05	-0.04±0.03			-0.07	0.06	-0.09	0.06	-0.09	0.05	-0.10	0.05
SRM-1649A Aliquot 2	3	-0.11±0.05	-0.12±0.03					-0.11	0.05	-0.12	0.03				

1: Aerosol samples collected during the ACE-ASIA 2000 field season from the Kosan Korea station

2: Aerosol samples collected during the ACE-ASIA 2000 field season from the Dunhuang, China Station

3: SRM-1649A is Urban dust from Washington D.C.; available from National Institute of Standards and Technology

All ACE-ASIA aerosol samples were collected using a Hi-Vol sampler that has an inlet size cut with an aerodynamic diameter in the size range of 50-60 microns

Analysis 1, 2, and 3 refer to different mass spectrometry runs of a sample conducted on different days; the errors are 2-SE from in-run statistics. Mass Spec Average is the average of up to 3 analyses, 1-SD is 1 standard deviation external; if there is only 1 mass spectrometry analysis, the error is 2-SE. Average of Replicate is the average of processing different aliquots of a sample throughout the entire analytical procedure; the best estimate of external reproducibility.

Table DR2: Fe isotope composition of loess

Sample Name	Sample Notes	-----Analysis 1-----		-----Analysis 2-----		-----Analysis 3-----		----Mass Spec Average----			
		$\delta^{56}\text{Fe}$	$\delta^{57}\text{Fe}$	$\delta^{56}\text{Fe}$	$\delta^{57}\text{Fe}$	$\delta^{56}\text{Fe}$	$\delta^{57}\text{Fe}$	$\delta^{56}\text{Fe}$	1-SD	$\delta^{57}\text{Fe}$	1-SD
CY-4a-A	1	0.07 ±0.10	0.19±0.05					0.07	0.10	0.19	0.05
Iowa	1	-0.08 ±0.10	-0.04±0.05					-0.08	0.10	-0.04	0.05
CY-4a-B	1	0.03±0.07	-0.04±0.05	0.11±0.05	0.16±0.03			0.07	0.06	0.06	0.15
CY-4a-C	1	0.04±0.08	0.04±0.06	0.16±0.05	0.24±0.04			0.10	0.08	0.14	0.14
Ksth1-1	2	0.01±0.09	0.11±0.04					0.01	0.09	0.11	0.04
Ksth1-2	2	0.04±0.09	0.11±0.05					0.04	0.09	0.11	0.05
BP-1	3	0.02±0.08	0.07±0.05					0.02	0.08	0.07	0.05
BP-2	3	-0.02±0.06	0.06±0.04					-0.02	0.06	0.06	0.04
BP-3	3	0.10±0.09	0.15±0.04	0.03±0.10	0.06±0.05	0.02±0.08	0.03±0.05	0.05	0.05	0.08	0.06
BP-5	3	0.16±0.08	0.22±0.05	0.07±0.09	0.18±0.05	0.09±0.09	0.19±0.05	0.11	0.05	0.20	0.02

1: Loess samples from Iowa and Kansas, USA; Major- and trace-element and Sr-Nd isotope data reported in Taylor et al. (1983).

2: Loess samples from the Rhine Valley, Germany; Major- and trace-element and Sr-Nd isotope data reported in Taylor et al. (1983).

3: Loess samples from New Zealand; Major- and trace-element and Sr-Nd isotope data reported in Taylor et al. (1983).

Analysis 1, 2, and 3 refer to different mass spectrometry runs of a sample conducted on different days; the errors are 2-SE from in-run statistics. Mass Spec Average is the average of up to 3 analyses; 1-SD is 1 standard deviation external; if there is only one mass spectrometry analysis, the reported error is 2-SE.

Table DR3: Fe isotope composition of soil samples

Sample Name	Sample Notes	----Analysis 1----		----Analysis 2----		----Mass Spec Average----				----Average of Replicate----			
		$\delta^{56}\text{Fe}$	$\delta^{57}\text{Fe}$	$\delta^{56}\text{Fe}$	$\delta^{57}\text{Fe}$	$\delta^{56}\text{Fe}$	1-SD	$\delta^{57}\text{Fe}$	1-SD	$\delta^{56}\text{Fe}$	1-SD	$\delta^{57}\text{Fe}$	1-SD
Asian Mineral Dust	1	0.05±0.04	0.01±0.03			0.05	0.04	0.01	0.03				
ZBT-IM012215	2	0.11±0.07	0.22±0.04			0.11	0.07	0.22	0.04				
Kosan	3	0.02±0.05	0.03±0.03	-0.01±0.05	0.12±0.03	0.00	0.02	0.08	0.06				
URC-IM012213	4	0.09±0.05	0.14±0.04			0.09	0.05	0.14	0.04				
DNH5-IM012216	5	-0.03±0.05	-0.04±0.04			-0.03	0.05	-0.04	0.04				
Xinjiang	6	0.00±0.04	-0.01±0.02			0.00	0.04	-0.01	0.02				
ZBT1-IM012214	7	0.14±0.05	0.22±0.03			0.14	0.05	0.22	0.03				
Xian, Sanxi	8	0.06±0.05	0.14±0.03			0.06	0.05	0.14	0.03				
SRM-2709 Aliquot 1	9	-0.04±0.04	-0.03±0.03	-0.03±0.06	0.02±0.03	-0.03	0.01	0.00	0.03	-0.05	0.04	-0.04	0.06
SRM-2709 Aliquot 2	9	-0.10±0.05	-0.10±0.04			-0.10	0.05	-0.10	0.04				

1-8: Soil sample collected during ACE-ASIA 2000 field season at various field sites in China and Korea, sieved to 62 microns to have a comparable size to aerosols.

9: Soil sample from a field in the San Joaquin Valley, CA; available from the National Institute of Standards and Technology

Analysis 1, and 2 refer to different mass spectrometry runs of a sample conducted on different days; the errors are 2-SE from in-run statistics. Mass Spec Average is the average of up to 2 analyses, 1-SD is 1 standard deviation external; if there is only 1 mass spectrometry analysis, the error is 2-SE. Average of Replicate is the average of processing different aliquots of a sample throughout the entire analytical procedure; the best estimate of external reproducibility.

Table DR4: Fe isotope composition of modern marine sediments

Sample Name	Sample Notes	----Analysis 1----		----Analysis 2----		----Analysis 3----		----Mass Spec Average----				----Average of Replicate----			
		$\delta^{56}\text{Fe}$	$\delta^{57}\text{Fe}$	$\delta^{56}\text{Fe}$	$\delta^{57}\text{Fe}$	$\delta^{56}\text{Fe}$	$\delta^{57}\text{Fe}$	$\delta^{56}\text{Fe}$	1-SD	$\delta^{57}\text{Fe}$	1-SD	$\delta^{56}\text{Fe}$	1-SD	$\delta^{57}\text{Fe}$	1-SD
MAG-1	1	0.01±0.07	0.09±0.04	-0.09±0.06	-0.02±0.04			-0.04	0.07	0.04	0.08				
183-10A4	2	0.09±0.05	0.18±0.05					0.09	0.05	0.18	0.05				
183-10A5	3	-0.10±0.14	-0.07±0.15	-0.04±0.04	-0.05±0.03			-0.07	0.04	-0.06	0.01				
GS7605-55	4	0.21±0.08	0.26±0.07	0.13±0.06	0.28±0.04			0.17	0.06	0.27	0.02				
RC14-67	5	0.08±0.05	0.16±0.04					0.08	0.05	0.16	0.04				
V33-75	6	0.07±0.05	0.19±0.04					0.07	0.05	0.19	0.04				
V14-55	7	-0.07±0.05	0.00±0.02					-0.07	0.05	0.00	0.02				
CA26-S Aliquot 1	8	0.09±0.06	0.17±0.04					0.09	0.06	0.17	0.04	0.07	0.02	0.08	0.13
CA26-S Aliquot 2	8	0.06±0.06	-0.01±0.03					0.06	0.06	-0.01	0.03				
BA18-M	9	0.06±0.06	0.14±0.05					0.06	0.06	0.14	0.05				
CC10-M	10	0.05±0.05	0.18±0.04					0.05	0.05	0.18	0.04				
TE7-M	11	0.14±0.05	0.18±0.04					0.14	0.05	0.18	0.04				
TE2-M Aliquot 1	12	0.02±0.05	0.06±0.03	0.10±0.09	0.03±0.05	0.06±0.06	0.04±0.03	0.06	0.04	0.04	0.02	0.06	0.06	0.08	0.12
TE2-M Aliquot 2	12	0.11±0.06	0.26±0.04	-0.03±0.07	-0.07±0.04			0.04	0.10	0.10	0.24				
TE2-M Aliquot 3	12	0.10±0.05	0.15±0.03					0.10	0.05	0.15	0.03				
CA28-M	13	0.00±0.05	0.12±0.04					0.00	0.05	0.12	0.04				
FA-36-M	14	-0.01±0.05	0.04±0.03					-0.01	0.05	0.04	0.03				

- 1: USGS rock standard; marine mud from the Gulf of Maine; sample description and compositional data in Flanagan (1976).
2: Turbidite clay from DSDP 183 Aleutian Islands; sample description and compositional data reported in Plank and Langmuir (1998).
3: Turbidite sand from DSDP 183 Aleutian Islands; sample description and compositional data reported in Plank and Langmuir (1998).
4: Terrigenous sediment from the Antilles Islands; sample description and compositional data reported in Ben Othman et al. (1989).
5: Volcanogenic sediment from Indian Ocean Sunda arc; sample description and compositional data reported in Ben Othman et al. (1989).
6: Terrigenous sediment from Indian Ocean Sunda arc; sample description and compositional data reported in Ben Othman et al. (1989).
7: Siliceous and calcareous ooze with clay from Atlantic Ocean Sandwich arc; sample description and compositional data reported in Ben Othman et al. (1989).
8: Turbidite sand layer from Middle America arc; sample description and compositional data reported in McLennan et al. (1990).
9: Turbidite mud layer from south China basin; sample description and compositional data reported in McLennan et al. (1990).
10: Turbidite mud layer from Ganges cone; sample description and compositional data reported in McLennan et al. (1990).
11: Turbidite mud layer from Angola basin; sample description and compositional data reported in McLennan et al. (1990).
12: Turbidite mud layer from Newfoundland basin; sample description and compositional data reported in McLennan et al. (1990).
13: Turbidite mud layer from Peru-Chile trench; sample description and compositional data reported in McLennan et al. (1990).
14: Turbidite mud layer from east Japan Basin; sample description and compositional data reported in McLennan et al. (1990).

Analysis 1, 2, and 3 refer to different mass spectrometry runs of a sample conducted on different days; the errors are 2-SE from in-run statistics. Mass Spec Average is the average of up to 3 analyses, 1-SD is 1 standard deviation external; if there is only 1 mass spectrometry analysis, the error is 2-SE. Average of Replicate is the average of processing different aliquots of a sample throughout the entire analytical procedure; the best estimate of external reproducibility.

Table DR5: Fe isotope composition of suspended load from rivers

Sample Name	Sample Notes	----Analysis 1----		----Analysis 2----		----Analysis 3----		----Mass Spec Average----				----Average of Replicate----			
		^{56}Fe	^{57}Fe	^{56}Fe	^{57}Fe	^{56}Fe	^{57}Fe	^{56}Fe	1-SD	^{57}Fe	1-SD	^{56}Fe	1-SD	^{57}Fe	1-SD
Connecticut	1	0.03±0.09	-0.03±0.05	-0.03±0.04	-0.05±0.04			0.00	0.04	-0.04	0.01				
Ohio River	1	0.02±0.08	0.04±0.04	0.00±0.05	-0.01±0.04			0.01	0.02	0.02	0.04	0.02	0.02	0.02	0.03
Aliquot A															
Ohio River	1	0.04±0.09	0.03±0.05					0.04	0.09	0.03	0.05				
Aliquot B															
Wabash	1	-0.05±0.09	0.00±0.04	-0.04±0.04	-0.04±0.03			-0.05	0.01	-0.02	0.03				
Illinois	1	0.02±0.07	-0.02±0.05	-0.07±0.06	-0.02±0.03	-0.03±0.06	-0.06±0.04	-0.03	0.05	-0.03	0.02				
Mississippi	1	-0.06±0.09	-0.06±0.04	-0.11±0.05	-0.10±0.03			-0.08	0.04	-0.08	0.02				
Venice Surf															
Mississippi	1	-0.02±0.06	-0.05±0.03	-0.05±0.05	-0.06±0.04			-0.03	0.02	-0.05	0.01				
Venice, Bottom															
Colorado, CO	1	-0.05±0.08	-0.05±0.04	0.06±0.05	0.01±0.04			0.00	0.08	-0.02	0.04				
Red River	1	0.00±0.09	0.07±0.03	0.00±0.04	0.00±0.04			0.00	0.00	0.03	0.05				
Brazos River	1	-0.04±0.08	0.03±0.04	0.00±0.06	0.05±0.03			-0.02	0.03	0.04	0.02				
Missouri R.	1	-0.14±0.10	-0.21±0.04	-0.12±0.11	-0.15±0.05			-0.13	0.01	-0.18	0.04				
Colorado	1	0.12±0.14	0.20±0.12	0.10±0.07	0.14±0.04			0.11	0.01	0.17	0.04				
River, TX															
Pecos River	1	0.04±0.03	0.08±0.03					0.04	0.03	0.08	0.03				
Rio Grande, TX	1	0.00±0.09	0.03±0.05					0.00	0.09	0.03	0.05	-0.02	0.03	0.03	0.00
Aliquot A															
Rio Grande, TX	1	-0.04±0.07	0.02±0.04					-0.04	0.07	0.02	0.04				
Aliquot B															

1: Sample description and major element data reported in Canfield (1997).

Analysis 1, 2, and 3 refer to different mass spectrometry runs of a sample conducted on different days; the errors are 2-SE from in-run statistics. Mass Spec Average is the average of up to 3 analyses, 1-SD is 1 standard deviation external; if there is only 1 mass spectrometry analysis, the error is 2-SE. Average of Replicate is the average of processing different aliquots of a sample throughout the entire analytical procedure; the best estimate of external reproducibility.

Table DR6: Fe isotope composition of Proterozoic to Phanerozoic Shales

Sample Name	Sample Notes	----Analysis 1----		----Analysis 2----		----Analysis 3----		----Mass Spec Average----				----Average of Replicate----			
		$\delta^{56}\text{Fe}$	$\delta^{57}\text{Fe}$	$\delta^{56}\text{Fe}$	$\delta^{57}\text{Fe}$	$\delta^{56}\text{Fe}$	$\delta^{57}\text{Fe}$	$\delta^{56}\text{Fe}$	1-SD	$\delta^{57}\text{Fe}$	1-SD	$\delta^{56}\text{Fe}$	1-SD	$\delta^{57}\text{Fe}$	1-SD
SDC-1	1	0.03±0.11	0.06±0.05					0.03	0.11	0.06	0.05				
SGR-1	2	-0.25±0.07	-0.33±0.04	-0.46±0.04	-0.68±0.03			-0.36	0.15	-0.50	0.25				
M11 Aliquot 1	3	0.17±0.09	0.17±0.04	0.22±0.05	0.30±0.03			0.19	0.03	0.23	0.09	0.15	0.06	0.23	0.06
M11 Aliquot 2	3	0.08±0.08	0.18±0.04	0.13±0.11	0.25±0.05			0.11	0.04	0.22	0.05				
M12	3	-0.01±0.05	0.04±0.02					-0.01	0.05	0.04	0.02				
M14	3	0.06±0.07	-0.02±0.05	0.11±0.05	0.17±0.03			0.09	0.04	0.07	0.13				
M15	3	0.05±0.05	0.11±0.04	-0.09±0.05	-0.04±0.04			-0.02	0.10	0.03	0.11				
PL1	4	0.01±0.08	-0.02±0.05					0.01	0.08	-0.02	0.05				
PL6	4	0.00±0.07	-0.03±0.05					0.00	0.07	-0.03	0.05				
PW4	5	-0.03±0.08	-0.03±0.05	0.02±0.09	0.09±0.05			-0.01	0.04	0.03	0.08				
PW5	5	0.03±0.06	0.01±0.06					0.03	0.06	0.01	0.06				
PW7	5	0.21±0.06	0.20±0.05	0.21±0.05	0.23±0.05			0.21	0.00	0.22	0.02				
SC1	6	-0.04±0.10	-0.11±0.06					-0.04	0.10	-0.11	0.06				
SC2	6	-0.02±0.06	0.02±0.04					-0.02	0.06	0.02	0.04				
SC4	6	-0.04±0.04	-0.02±0.03					-0.04	0.04	-0.02	0.03				
SC5	6	0.02±0.07	0.02±0.04					0.02	0.07	0.02	0.04				
SC7	6	0.01±0.09	0.02±0.07					0.01	0.09	0.02	0.07				
AO6	7	0.04±0.08	0.09±0.04	0.02±0.08	0.03±0.04			0.03	0.02	0.06	0.04				
AO7 Aliquot 1	7	-0.34±0.10	-0.46±0.05	-0.40±0.08	-0.52±0.04	-0.30±0.09	-0.38±0.04	-0.35	0.05	-0.45	0.07	-0.28	0.10	-0.43	0.06
AO7 Aliquot 2	7	-0.16±0.09	-0.36±0.05	-0.22±0.06	-0.41±0.03			-0.19	0.05	-0.39	0.03				
AO9	7	-0.01±0.10	0.06±0.05					-0.01	0.10	0.06	0.05				
AO10	7	0.08±0.08	0.12±0.05	0.14±0.09	0.26±0.04	0.06±0.09	0.19±0.04	0.10	0.04	0.19	0.07				
AO12 Aliquot 1	7	0.44±0.08	0.69±0.04	0.49±0.10	0.69±0.06	0.34±0.04	0.55±0.03	0.42	0.08	0.64	0.08	0.39	0.07	0.60	0.11
AO12 Aliquot 2	7	0.37±0.07	0.64±0.04	0.31±0.10	0.43±0.05			0.34	0.04	0.53	0.15				

1: USGS rock standard mica schist; a metasedimentary rock from the Sykesville formation Washington, D.C.; Late Precambrian to Cambrian depositional age; see Flanagan (1976).

2: USGS rock standard Shale of the Green River; from the Green River basin, Wyoming; Tertiary depositional age; see Flanagan (1976) for details.

3: Shale from the Mt Isa group, Australia; Middle Proterozoic depositional age; sample description and compositional data in Nance and Taylor (1976).

4: Shale from the Canning basin, Australia; Permian to Carboniferous depositional ages; sample description and compositional data in Nance and Taylor (1976).

5: Shale from Perth Basin, Australia; Triassic depositional age; sample description and compositional data in Nance and Taylor (1976).

6: Shale from State Circle Shale Formation, Australia; Silurian depositional age; sample description and compositional data in Nance and Taylor (1976).

7: Shale from the Amadeus Basin, Australia; Cambrian to Upper Proterozoic depositional ages; sample description and compositional data in Nance and Taylor (1976).

Analysis 1, 2, and 3 refer to different mass spectrometry runs of a sample conducted on different days; the errors are 2-SE from in-run statistics. Mass Spec Average is the average of up to 3 analyses, 1-SD is 1 standard deviation external; if there is only 1 mass spectrometry analysis, the error is 2-SE. Average of Replicate is the average of processing different aliquots of a sample throughout the entire analytical procedure; the best estimate of external reproducibility.

Table DR7: Fe isotope composition of MOR Hydrothermal fluids (Dissolved, B/F, and Dregs fractions)

Sample Name	Sample Notes	----Analysis 1----		----Analysis 2----		----Analysis 3----		----Mass Spec Average----				----Average of Replicate----			
		$t^{56}\text{Fe}$	$t^{57}\text{Fe}$	$t^{56}\text{Fe}$	$t^{57}\text{Fe}$	$t^{56}\text{Fe}$	$t^{57}\text{Fe}$	$t^{56}\text{Fe}$	1-SD	$t^{57}\text{Fe}$	1-SD	$t^{56}\text{Fe}$	1-SD	$t^{57}\text{Fe}$	1-SD
<i>Pacific Ocean MOR Hydrothermal Fluids</i>															
2360-1C Diss.	1	-0.29±0.07	-0.42±0.04	-0.38±0.07	-0.53±0.03			-0.33	0.06	-0.47	0.08				
2360-1C B/F		-0.50±0.06	-0.81±0.04	-0.55±0.04	-0.81±0.04			-0.52	0.04	-0.81	0.01				
2735-4 Diss. Aliquot 1	2	-0.46±0.08	-0.58±0.04	-0.53±0.05	-0.72±0.03			-0.49	0.05	-0.65	0.10	-0.49	0.04	-0.18	0.05
2735-4 Diss Aliquot 2	2	-0.47±0.05	-0.65±0.04					-0.47	0.05	-0.65	0.04				
2735-4 B/F Aliquot 1		-1.54±0.07	-2.32±0.04					-1.54	0.05	-2.32	0.04	-1.61	0.06	-2.38	0.06
2735-4 B/F Aliquot 2		-1.67±0.06	-2.44±0.04	-1.61±0.04	-2.38±0.03			-1.64	0.04	-2.41	0.04				
2735-4 DREGS		-0.54±0.05	-0.79±0.04	-0.59±0.05	-0.79±0.04			-0.57	0.03	-0.79	0.01				
3030-11 Diss. Aliquot 1	3	-0.55±0.09	-0.83±0.05	-0.56±0.06	-0.82±0.03			-0.56	0.01	-0.82	0.01	-0.54	0.03	-0.28	0.03
3030-11 Diss. Aliquot 2	3	-0.50±0.05	-0.79±0.03					-0.50	0.05	-0.79	0.03				
3030-11 B/F Aliquot 1		-0.70±0.07	-0.95±0.03	-0.68±0.05	-0.95±0.03			-0.69	0.02	-0.95	0.01	-0.73	0.05	-1.00	0.07
3030-11 B/F Aliquot 2		-0.80±0.05	-1.10±0.03	-0.72±0.07	-1.00±0.04			-0.76	0.06	-1.05	0.07				
3030-11 DREGS		-0.69±0.05	-1.00±0.03	-0.57±0.05	-0.94±0.03			-0.63	0.08	-0.97	0.04				
3290-13 Diss.	4	-0.21±0.08	-0.43±0.04	-0.31±0.07	-0.50±0.04			-0.26	0.07	-0.46	0.05				
3290-13 B/F		-0.31±0.06	-0.42±0.03	-0.31±0.04	-0.43±0.03			-0.31	0.01	-0.42	0.01				
3306-12C Diss Aliquot 1	5	-0.32±0.09	-0.53±0.04	-0.32±0.09	-0.50±0.04	-0.26±0.09	-0.46±0.04	-0.30	0.03	-0.50	0.04	-0.31	0.04	-0.20	0.01
3306-12C Diss Aliquot 2	5	-0.36±0.08	-0.60±0.04	-0.28±0.09	-0.43±0.05			-0.32	0.06	-0.51	0.12				
3306-12C B/F		-0.50±0.06	-0.81±0.04	-0.55±0.04	-0.81±0.02			-0.52	0.04	-0.81	0.01				
<i>Atlantic Ocean MOR Hydrothermal Fluids</i>															
3114-11 Diss Aliquot 1	6	-0.47±0.07	-0.71±0.04	-0.51±0.06	-0.67±0.03			-0.49	0.03	-0.69	0.03	-0.45	0.05	-0.19	0.05
3114-11 Diss Aliquot 2	6	-0.39±0.06	-0.53±0.04	-0.44±0.06	-0.60±0.03			-0.42	0.04	-0.56	0.05				
3133-10 Diss	7	-0.23±0.07	-0.34±0.04	-0.19±0.05	-0.29±0.03			-0.21	0.03	-0.32	0.04				
3133-10 B/F		-0.49±0.09	-0.59±0.04					-0.49	0.09	-0.59	0.04				
<i>Synthetic Hydrothermal Test Solutions</i>															
Black Smoker 1A test	8	0.17±0.09	0.31±0.04	0.22±0.12	0.46±0.06			0.19	0.04	0.38	0.11	0.22	0.05	0.41	0.08
Black Smoker 1B test	8	0.22±0.10	0.40±0.05	0.28±0.10	0.47±0.04			0.25	0.04	0.44	0.05				
Black Smoker 2A	9	0.28±0.06	0.39±0.03					0.28	0.06	0.39	0.03	0.28	0.06	0.48	0.11
Black Smoker 2B	9	0.27±0.06	0.50±0.04	0.20±0.14	0.36±0.06			0.24	0.05	0.43	0.10				
Black Smoker 2C	9	0.30±0.17	0.52±0.08	0.37±0.08	0.62±0.04			0.33	0.05	0.57	0.07				
Black Smoker 3A test	10	0.33±0.11	0.59±0.06	0.38±0.08	0.42±0.04			0.36	0.04	0.50	0.12	0.28	0.09	0.43	0.12
Black Smoker 3B test	10	0.18±0.10	0.41±0.05	0.22±0.09	0.29±0.05			0.20	0.03	0.35	0.08				
Black Smoker 4A test	11	0.29±0.08	0.57±0.06	0.35±0.08	0.68±0.05			0.32	0.04	0.63	0.08	0.33	0.03	0.64	0.06
Black Smoker 4B test	11	0.33±0.07	0.67±0.04					0.33	0.07	0.67	0.04				

1: Hydrothermal solution from East Pacific Rise (EPR) 9-10°N A Vent sampled 1991; 2: Hydrothermal solution from EPR 9-10°N Bio 9 vent sampled 1994; 3: Hydrothermal solution from EPR 9-10°N Bio 9 vent sampled 1995; 4: Hydrothermal vapor from EPR 21.5°S East Pacific Rise Brandon Vent vapor sampled 1998; 5: Hydrothermal brine from EPR 21.5°S Brandon Vent sampled 1998; 6: Hydrothermal solution from mid Atlantic Ridge (MAR) Lucky Strike, 2608 vent sampled 1997; 7: Hydrothermal solution from MAR, Logatchev, Irina vent, ultramafic hosted, sampled 1997; 8: Synthetic test solution with Cl, Si, Na, K, Ca, Fe, and Mg contents (45, 5, 40, 1, 2, 2, 5 mmol/kg, respectively) to match an average low Cl vent solution; the UW J-M Fe standard was used for Fe; 9: Synthetic test solution with Cl, Si, Na, K, Ca, Fe, and Mg contents (850, 18, 700, 40, 45, 5, 5 mmol/kg, respectively) to match an average vent brine solution; the UW J-M Fe standard was used for Fe; 10: Synthetic test solution with Cl, Si, Na, K, Ca, Fe, and Mg contents (500, 18, 400, 20, 30, 5, 5 mmol/kg, respectively) to match an average vent solution; the UW J-M Fe standard was used for Fe; 11: Synthetic test solution with Cl, Si, Na, K, Ca, Fe, and Mg contents (500, 5, 500, 10, 10, 0.2, 50 mmol/kg, respectively) to match hydrothermal fluid modified seawater; the UW J-M Fe standard was used for Fe

Analysis 1, 2, and 3 refer to different mass spectrometry runs of a sample conducted on different days; the errors are 2-SE from in-run statistics. Mass Spec Average is the average of up to 3 analyses, 1-SD is 1 standard deviation external; if there is only 1 mass spectrometry analysis, the error is 2-SE. Average of Replicate is the average of processing different aliquots of a sample throughout the entire analytical procedure; the best estimate of external reproducibility.

Table DR8: Measured Fe concentration, mass of solution and isotope composition of different hydrothermal fluid fractions and the mass-balance calculated Fe isotope composition of the total solution.

Sample ID	3114-11	3133-10	3290-13	3306-12C	2360-1C	2735-4	3030-11
Vent ID	8.2	Logatchev	Ba.2	Be.5	Aa.1	B9.4	B9.8
Mg (AA) mmoles/kg	1.62	4.82	3	7	7.907	4.08	1.82
Mass Filtered	621.5	720.05	645.36	615.83	384.07	611.42	184.4
Mass B/F	9.95	10.4955	10.8676	11.5588	10.1114	10.2879	9.836
Mass Dregs	9.3732	No dregs	No dregs	No dregs	No dregs	10.3269	9.9891
Dissolve Fe σ moles/kg	915.8	2356	8754	10468	1430	1200	5859
Digest B/F Fe σ moles/kg	32.03	1079	3200	1448	1660	1050	1670
Digest-Dregs Fe σ moles/kg	52.61	No dregs	No dregs	No dregs	No dregs	1870	865.5
Totals-All Splits Fe σ moles/kg	917	---	---	---	---	1243	5959
Totals Dissolved and B/F σ moles/kg	916	2372	8808	10495	1474	1218	5948
$\delta^{56}\text{Fe}$ Dissolved fraction	-0.45	-0.21	-0.26	-0.31	-0.33	-0.49	-0.54
$\delta^{56}\text{Fe}$ B/F fraction	Not analyzed	-0.49	-0.31	-0.52	-0.52	-1.61	-0.73
$\delta^{56}\text{Fe}$ Dregs fraction	Not analyzed	No dregs	No dregs	No dregs	No dregs	-0.57	-0.63
Calculated $\delta^{56}\text{Fe}$ Dissolved and B/F	---	-0.21	-0.26	-0.31	-0.34	-0.51	-0.54