Supplementary Figure 1.

Geological map of central Taiwan, with the site of the Taiwan Chelungpu-fault Drilling Project (TCDP) and an E-W cross section through the site location[11]. Drilling was carried out near the town of DaKeng in the northern part of the fault system, where a large slip of about 8 m was observed. Two vertical holes, Holes A and B, were drilled about 2 km east of the surface rupture and penetrated the east-dipping Chelungpu fault at about 1.1-1.2 km depth[11,13]. The Chelungpu fault is predominantly developed within and parallel to the bedding of the Pliocene Chinshui shale.

Supplementary Table 1: Chemical compositions of the TCDP core samples

Depth (m)*	FZB1136										FZB1194		
	1134.53 FDZ	1135.88 BrZ	1136.22 GGZ	1136.26 BGZ	1136.31 BGZ	1136.35 BGZ	1136.39 GGZ	1136.43 GGZ	1136.51 BrZ	1136.89 FDZ	1194.68 FDZ	1194.73 BGZ(BM)	1194.75 BGZ
(wt.%)													
SiO_2	66.61	69.63	66.07	68.13	68.08	67.39	67.04	64.99	67.04	65.83	73.65	75.85	69.81
TiO_2	0.99	0.91	1.01	0.96	0.97	0.99	0.89	1.00	0.92	0.97	0.84	0.85	0.96
Al_2O_3	16.17	14.66	16.67	16.13	15.79	16.32	14.79	16.98	15.34	16.24	13.16	12.47	15.32
Fe_2O_3	6.91	6.09	7.19	6.53	6.83	6.67	6.41	7.12	6.73	6.95	5.18	4.51	6.01
MnO	0.09	0.09	0.10	0.02	0.02	0.03	0.14	0.11	0.11	0.08	0.05	0.01	0.04
MgO	2.41	2.11	2.50	2.37	2.52	2.58	2.30	2.63	2.45	2.71	1.77	1.66	2.11
CaO	1.78	1.70	1.29	0.49	0.66	0.82	3.63	1.90	2.51	2.04	0.88	0.45	0.78
Na_2O	1.49	1.53	1.53	1.94	2.03	2.01	1.65	1.54	1.54	1.50	1.56	1.72	1.65
K_2O	3.36	3.15	3.50	3.30	2.97	3.05	3.02	3.59	3.24	3.55	2.78	2.40	3.18
P_2O_5	0.18	0.13	0.14	0.12	0.13	0.15	0.13	0.14	0.13	0.13	0.12	0.09	0.13
(ppm)													
Li	50.4	50.5	62.3	24.1	25.2	24.1	44.2	66.7	65.6	66.6	48.5	21.8	42.0
Rb	116	115	146	126	102	128	120	162	152	155	110	80.8	126
Sr	123	115	124	169	234	187	166	121	120	129	112	162	133
Cs	7.61	7.73	10.9	8.01	5.93	8.37	9.40	13.1	11.9	11.7	7.11	3.13	8.35
Ba	382	391	410	601	558	516	375	438	438	458	403	443	472
La	32.7	36.8	37.5	37.9	37.9	39.9	32.8	35.0	33.9	37.8	33.1	36.2	38.0
Sm	5.67	6.17	6.44	6.42	6.34	6.59	5.85	5.86	5.66	6.78	5.65	6.03	6.56
Pb	21.7	21.1	21.3	21.9	22.9	20.6	19.7	23.7	22.7	26.5	20.7	24.2	25.8
Th	11.4	13.9	14.0	14.0	13.4	14.5	11.8	14.5	14.2	15.7	12.0	12.5	14.2
$^{87}Sr/^{86}Sr$	0.718827	0.719275	0.719400	0.714594	0.713243	0.713966	0.716120	0.720527	0.720645	0.720361	0.719070	0.714001	0.717009
$^{206}Pb/^{204}Pb$	18.5405	18.6073	18.5736	18.5589	18.5759	18.5671	18.5899	18.6007	18.6048	18.6185	18.4745	18.4083	18.5468
$^{207}Pb/^{204}Pb$	15.6533	15.6626	15.6570	15.6542	15.6568	15.6543	15.6578	15.6600	15.6603	15.6625	15.6476	15.6409	15.6552
$^{208}Pb/^{204}Pb$	38.9135	39.1837	38.9504	38.9476	38.9639	38.9472	38.9652	38.9354	38.9391	38.9833	38.8902	38.8477	38.9229
$^{87}Sr/^{86}Sr$ §		0.710215	0.712223	0.711696	0.711504	0.711305	0.711306	0.710863	0.711583	0.711684	0.711120	0.711318	0.712147
$^{206}Pb/^{204}Pb$ §		18.4752	18.5262	18.5539	18.5512	18.5304	18.5098		18.5064	18.5532	18.1355	18.1448	18.3384
$^{207}Pb/^{204}Pb$ §		15.6503	15.6555	15.6561	15.6569	15.6552	15.6518		15.6562	15.6581	15.6190	15.6171	15.6349
$^{208}Pb/^{204}Pb$ §		38.7404	38.8234	38.8519	38.8502	38.8231	38.7991		38.7696	38.8412	38.3185	38.3415	38.5771

Depth (m)*	FZB1194					FZB1243							
	1194.78 BGZ	1194.82 BGZ	1194.86 BGZ	1195.11 GGZ	1195.16 BrZ	1243.30 FDZ	1243.34 GGZ	1243.38 BGZ(BM)	1243.41 BGZ	1243.43 BGZ	1243.47 BGZ	1243.51 GGZ	1243.56 GGZ
(wt.%)													
SiO_2	71.13	70.75	71.43	72.65	72.13	75.00	73.50	73.73	74.06	74.14	75.00	75.96	74.74
TiO_2	0.93	0.94	0.89	0.86	0.90	0.78	0.83	0.90	0.88	0.84	0.80	0.74	0.76
Al_2O_3	14.18	14.23	13.51	13.36	13.48	11.78	12.64	13.94	13.47	12.97	12.34	11.57	12.04
Fe_2O_3	5.88	5.88	5.65	5.45	5.70	4.93	5.35	4.73	4.76	4.89	4.83	4.59	4.93
MnO	0.05	0.06	0.07	0.06	0.05	0.07	0.07	0.01	0.02	0.05	0.05	0.06	0.07
MgO	2.01	2.01	1.95	1.87	1.95	1.65	1.78	1.68	1.69	1.71	1.64	1.58	1.67
CaO	1.07	1.38	1.97	1.27	1.22	1.49	1.39	0.39	0.65	1.11	1.01	1.23	1.46
Na_2O	1.61	1.64	1.59	1.59	1.62	1.60	1.60	1.66	1.67	1.63	1.58	1.55	1.49
K_2O	3.01	2.98	2.83	2.78	2.84	2.58	2.72	2.85	2.69	2.54	2.64	2.63	2.71
P_2O_5	0.12	0.13	0.12	0.11	0.12	0.11	0.12	0.11	0.13	0.12	0.12	0.11	0.12
(ppm)													
Li	46.1	55.5	41.9	54.8	50.1	45.3	42.7	26.5	32.0	33.6	49.4	49.4	49.9
Rb	126	135	114	121	116	99.2	94.6	96.1	91.9	83.5	103	100	101
Sr	123	127	117	116	115	120	125	154	137	148	112	122	114
Cs	8.49	10.1	7.32	8.22	7.71	6.14	6.33	5.54	5.51	4.78	6.92	6.56	6.67
Ba	453	416	437	387	399	401	360	400	390	375	384	408	393
La	37.5	38.3	33.9	34.5	31.3	28.7	30.0	36.7	36.1	35.5	31.8	35.2	31.0
Sm	6.45	6.81	5.91	6.10	5.60	4.98	5.20	6.63	6.20	6.09	5.49	6.08	5.37
Pb	23.9	24.7	21.8	23.0	20.9	19.4	19.7	23.2	20.3	19.7	19.1	18.4	19.0
Th	13.5	14.2	12.5	12.9	11.7	10.6	11.0	13.4	13.1	12.8	16.5	12.9	11.6
$^{87}Sr/^{86}Sr$	0.717810	0.718169	0.717689	0.719108	0.719344	0.718310	0.717226	0.714010	0.714738	0.714400	0.718822	0.718114	0.718171
$^{206}Pb/^{204}Pb$	18.5430	18.5445	18.5388	18.5385	18.5139	18.4906	18.5150	18.4966	18.5068	18.5127	18.5221	18.5084	18.4928
$^{207}Pb/^{204}Pb$	15.6539	15.6539	15.6537	15.6541	15.6518	15.6490	15.6518	15.6507	15.6508	15.6513	15.6528	15.6516	15.6488
$^{208}Pb/^{204}Pb$	38.9360	38.9353	38.9505	38.9099	38.9158	38.9098	38.9732	38.9158	38.9840	38.9816	38.9790	38.9588	38.9169
$^{87}Sr/^{86}Sr$ §	0.712315		0.712587	0.710480	0.711122	0.710409	0.710759	0.710549	0.710771		0.710745	0.709987	0.710910
$^{206}Pb/^{204}Pb$ §	17.9959		17.9181		18.2740	18.1301	17.9483	18.0418	18.0404	18.3650	18.1386		18.2560
$^{207}Pb/^{204}Pb$ §	15.6033		15.5955		15.6296	15.6240	15.5990	15.6065	15.6076	15.6392	15.6177		15.6273
$^{208}Pb/^{204}Pb$ §	38.1164		38.0131		38.4638	38.2359	38.0339	38.1799	38.1785	38.6320	38.3144		38.4695

* Depth for the upper surface of sample.

§ Isotope ratio of H_2O-soluble component.

Supplementary Methods

Calculation of the concentration of a trace element in the gouge after the interaction with fluid

Assuming chemical equilibrium, the solid/fluid bulk distribution coefficient (*D*) of a trace element is defined by

$$D = \frac{C_s}{C_f} \quad (1)$$

where C_s and C_f are the element concentrations in the solid (gouge) and fluid, respectively.

Our model 1 assumes interaction of solid with a single batch of fluid. Letting *R* to be the fluid/solid-mass ratio, mass balance requires

$$C_s + R \cdot C_f = C_{s0} + R \cdot C_{f0} \quad (2)$$

where C_{s0} and C_{f0} are the element concentrations in the solid and fluid, respectively, prior to solid-fluid interaction. From equations (1) and (2), we obtain the equation for calculating the trace element concentration of solid after the interaction with fluid:

$$C_s = D \cdot \frac{C_{s0} + R \cdot C_{f0}}{R + D} \quad (3\text{-}1)$$

In model 2, we assume interaction of solid with multiple batches of fluids, each with a small, constant fluid/solid-mass ratio, *r*. Each of the batches of fluid, which originally had the element concentration C_{f0}, is equilibrated with the solid, and then removed from the system. In this case, the element concentration in the solid which interacted with the fluid *i* times can be calculated by the following equation:

$$C_{si} = D \cdot \frac{C_{si-1} + r \cdot C_{f0}}{r + D} \quad (3\text{-}2)$$

and total *R* is given as the sum of the *r* values for each of the batches of fluid, *i·r*. In this study, we assumed a very small *r* value of 0.01.

The model 1 and model 2 represent the both extremes of the mode of solid-fluid interaction, and trace elements with low *D* values are more effectively extracted from the solid in model 2 than in model 1 (see Fig. 3).

Parameters used in the calculations

D values

Solid/fluid bulk distribution coefficient (*D*) values at 250, 300, and 350°C were

estimated based on two sets of hydrothermal experimental data.

You *et al.*[19] carried out their experiments at 25-350°C and 80 MPa using hemipelagic mudstones from the decollement zone in the Nankai Trough (ODP site 808) and synthetic NaCl-CaCl$_2$ solutions of composition similar to the in situ pore water (initial water/rock ratio = 3). The sediments they used consisted mainly of quartz, plagioclase, carbonate, illite, smectite, kaolinite, and chlorite, and their total organic carbon content was <0.4%[31]. After the experiments, quartz, plagioclase, illite (including celadonite), chlorite (clinochlore), and possibly maghemite were identified in the residual solids.

James *et al.*[20] carried out similar experiments at 50-350°C and 40 MPa using hemipelagic sediments from the Escanaba Trough (ODP site 1037) and synthetic solutions with fluid composition comparable to the fluids thought to recharge sediment- and basalt-dominated hydrothermal systems at mid-ocean ridges (initial water/rock ratio = 4). The sediments they used consisted of 29% quartz, 24% feldspars (20% plagioclase, 4% K-feldspar), 6% clay minerals (illite and chlorite), 5% heavy minerals, and 34% lithic fragments of largely volcanic origin. The total inorganic and total organic carbon contents were as low as 0.4% and 0.6%, respectively.

In these experiments, the solution samples for chemical analysis were intermittently extracted from the reaction cell at controlled pressure and temperature during heating and cooling episodes. The empirical D value at each temperature can be estimated as

$$D = \frac{C_{s\,\text{exp}}}{C_{f\,\text{exp}}} = \frac{m_{s0\,\text{exp}}C_{s0\,\text{exp}} + m_{f0\,\text{exp}}C_{f0\,\text{exp}} - m_{f\,\text{exp}}C_{f\,\text{exp}}}{m_{s\,\text{exp}}C_{f\,\text{exp}}} \quad (4)$$

where $m_{s0\,\text{exp}}$, $m_{f0\,\text{exp}}$, $m_{s\,\text{exp}}$ and $m_{f\,\text{exp}}$ are the masses of the starting sediment, starting fluid, reacted sediment, and reacted fluid, respectively, and $C_{s0\,\text{exp}}$, $C_{f0\,\text{exp}}$, $C_{s\,\text{exp}}$ and $C_{f\,\text{exp}}$ are the element concentrations in the starting sediment, starting fluid, reacted sediment, and reacted fluid, respectively. The D values were calculated using the experimental data from the heating episode by assuming $m_{s\,\text{exp}} = m_{s0\,\text{exp}}$, and corrected for the loss of fluid and elements due to repeated extraction of the sample solution.

In both experiments, temperature was increased step wise to 350°C in increment of 25-50°C. Total holding times at 250, 300, and 350°C were 72 hours for all temperatures in the experiment by You *et al.*[19] and 99, 119 and 47 hours, respectively, in the experiment by James *et al.*[20]. In the latter experiment at 250°C, the compositions of two fluid samples recovered after holding times of 48 hours and 99 hours were within analytical error, suggesting that near steady-state conditions were attained. You and Gieskes[32] performed another experiment using the same conditions as You *et al.*[19], but they added [6]Li- and ^{18}O-enriched spikes to the starting solution. Despite the contrasting Li contents in the starting solutions (0 µM and 8400 µM), at 350°C these

two experiments yielded nearly identical D_{Li} values (1.2 and 1.3) and D_{Sr} values (24 and 27), which is consistent with the attainment of trace element equilibrium between the fluid and the sediment.

It has been suggested that organic materials in sediment may affect the mobility of alkaline and alkali earth metals[33]. Although organic carbon content (at <0.6 %) was low in the TCDP sediments and in the sediments used by You *et al.*[19] and James *et al.*[20], and therefore unlikely to control the mobility of elements investigated here, the effect of the organic materials can be evaluated on the basis of the data of a hydrothermal experiment by Thornton and Seyfried[34]. They carried out their experiments at 350°C and 50 MPa for a total holding time of 2639 hours, using a diatomaceous ooze (a nearly equal mixture of pelagic clay and biogenic silica) from the Guaymas Basin and original sediment pore fluid (initial water/rock ratio = 3). The sediment they used consisted of biogenic silica, quartz, K-feldspar, plagioclase, and minor amounts of smectite and barite, and contained 3.99% total organic carbon. The D_{Sr} and D_{Ba} values estimated from their experiment were 24 and 9.4, respectively, and are comparable to those estimated from the data of You *et al.*[19] (D_{Sr} = 24 and D_{Ba} = 13) and James *et al.*[20] (D_{Sr} = 20 and D_{Ba} = 15). Unfortunately, Li and Rb contents of initial sediment are not available in the data of Thornton and Seyfried[34]. However, a Li/Rb ratio of 1.9 observed for the sample solution at 350°C, which is likely to be controlled by equilibrium between clay minerals and fluid, is comparable to the value of 2.4 observed by You *et al.*[19] and James *et al.*[20]. Thus, it is conceivable that our estimates of D were not affected by the presence of organic materials in the sediment.

Because La, Sm and Pb data are not available in James *et al.*[20], the D_{La}, D_{Sm} and D_{Pb} values estimated on the basis of experimental results of You *et al.*[19] were used for both calculations shown in Fig 2.

C_{s0} and C_{f0} values

The initial gouge concentrations (C_{s0}) we used were the same as the reference values for FZB1136 used in Fig. 2, that is, the averages of the sample data from 1136.22 and 1136.51 m. The initial fluid concentration (C_{f0}) values for Li, Rb, Sr, Cs and Ba were estimated from the compositions of pore waters of 49-67°C (n = 8) collected from neritic sediment in the High Island Field, offshore Texas, at depths of about 1800-2100m[22]. Concentrations of these elements in the pore waters correlate well with Na contents, and we used the values at a Na concentration of 11 000 ppm for C_{f0}. For La, Sm, and Pb, the C_{f0} values were taken from the fluid compositions obtained by hydrothermal experiments at 25 and 63°C[19]. The $^{87}Sr/^{86}Sr$ and $^{206}Pb/^{204}Pb$ ratios of initial gouge and initial fluid we used were the average values of the samples from 1136.22 and 1136.51 m and the lowest values of the H_2O-soluble components in

FZB1136 (the 1135.88m sample), respectively.

	D (You *et al.*)			*D* (James *et al.*)			C_{s0}	C_{f0}
	250°C	300°C	350°C	250°C	300°C	350°C	(ppm)	(ppm)
Li	11	3.7	1.2	15	8.4	2.8	63.9	0.44
Rb	290	120	13	81	67	25	149	0.085
Sr	17	24	24	16	17	20	122	21
Cs	80	42	9.1	92	60	26	11.4	0.024
Ba	17	54	13	20	17	14	424	30
La	41000	47000	26000				35.7	0.0002
Sm*	41000	47000	26000				6.05	0.0002
Pb	1900	350	110000				22.0	0.005

							Initial gouge	Initial fluid
$^{87}Sr/^{86}Sr$							0.72002	0.7102
$^{206}Pb/^{204}Pb$							18.589	18.48

*D and C_{f0} of Sm were assumed to be the same as those of La.

Supplementary references

31. Berner, U. & Koch, J. in *Proceedings of the Ocean Drilling Program, Scientific Results, Vol. 131* (eds., Hill, I. A., Taira, A., Firth, J. V., *et al.*) 379-385 (Ocean Drilling Program, College Station, Texas, 1993).

32. You, C.-F. & Gieskes, J. M. Hydrothermal alteration of hemi-pelagic sediments: experimental evaluation of geochemical process in shallow subduction zones. *Appl. Geochem.* **16**, 1055-1066 (2001).

33. Clauer, N., Chaudhuri, S., Lewan, M. D. & Toulkeridis, T. Effect of thermal maturation on the K–Ar, Rb–Sr and REE systematics of an organic-rich New Albany Shale as determined by hydrous pyrolysis. *Chem. Geol.* **234**, 169-177 (2006).

34. Thornton, E. & Seyfried, W. E., Jr. Reactivity of organic-rich sediment in seawater at 350°C, 500 bars: experimental and theoretical constraints and implications for the Guaymas Basin hydrothermal system. *Geochim. Cosmochim. Acta* **51**, 1997-2010 (1987).