Sample ID	Sampling site	Temp. (°C)	EC (μS/cm)	pH	He (ppmv)	Ar (vol.%)	N_2 (vol.%)	O_2 (vol.%)	CO_2 (vol.%)	CH_4 (vol.%)	CO_2/N_2	N_2/He	N_2/Ar	He/Ar
20QP-3	Qingping	62.0	1676	6.38	28.0	0.14	5.56	2.27	91.5	0.47	16.5	2101	39.0	0.020
190503-QP	Qingping	62.5	1730	6.35	20.3	0.06	3.34	<0.1	96.1	0.54	28.7	1655	60.4	0.037
21QP-2	Qingping	61.3	1783	6.34	2.30	0.04	2.52	0.56	96.6	0.14	38.3	11,065	68.3	0.006
190504-EL	Eli	55.0	831	6.48	133	1.05	70.6	5.12	21.0	2.15	0.30	5425	67.0	0.013
20TXX-2	Tongxinxiang	34.3	915	6.52	402	0.98	80.5	0.06	18.0	0.39	0.22	2033	81.8	0.041
20TXX-3	Tongxinxiang	34.3	915	6.52	456	0.96	74.5	0.32	23.8	0.41	0.32	1655	77.6	0.048
21TXX-3	Tongxinxiang	34.4	961	6.52	415	0.95	73.1	1.37	24.4	0.12	0.33	1778	76.9	0.044
20LYZ-3	Laoyaozhai	24.0	453	6.98	26.2	0.93	88.1	7.21	3.67	<0.1	0.04	65,251	94.6	0.003
21LYZ-2	Laoyaozhai	25.1	479	7.33	14.0	0.99	89.3	7.51	2.10	<0.1	0.02	70,104	90.5	0.001
20XL-2	Xiling	32.0	489	7.07	34.0	0.95	87.5	8.59	2.91	<0.1	0.03	48,732	91.8	0.004
20XL-3	Xiling	32.0	489	7.07	25.4	0.92	87.7	8.51	2.81	<0.1	0.03	66,516	95.2	0.003
21XL-2	Xiling	31.7	518	7.12	24.0	1.05	92.8	2.86	3.04	0.15	0.03	42,353	88.5	0.002
21DZR-2	Xiling-Daziran	32.4	513	7.05	15.0	1.00	89.4	6.20	3.29	<0.1	0.04	64,904	89.0	0.001
190503-YJ	Yuanjiang	77.8	549	7.71	717	1.34	89.3	0.15	2.93	6.21	0.03	1251	66.7	0.054

Abbreviations: EC, electrical conductivity; Temp., temperature.

Table S1. Chemical compositions (reported in ppmv for helium and vol.% for other gas species) and relative ratios of hydrothermal gases from the Simao block.

Sample ID	$^{4}He/^{20}Ne$	Measured $^{3}He/^{4}He$ (R/Ra, 1σ)	Corrected $^{3}He/^{4}He$ (R/Ra, 1σ) [a]	Helium inventory (%) [b]			MORB Mantle He (%) [c]	^{4}He in spring water (cm^{3} STP g^{-1} H_2O) [d]	^{4}He in mantle fluids (cm^{3} STP g^{-1} H_2O) [d]	Fluid flow rate (mm yr^{-1}) [d]	Total ^{3}He flux (atoms m^{-2} s^{-1}) [d]	Total ^{4}He flux (atoms m^{-2} s^{-1}) [d]	Mantle ^{4}He flux (10^{10} atoms m^{-2} s^{-1})	Crustal ^{4}He flux (10^{10} atoms m^{-2} s^{-1})
				M	C	A								
20QP-3	4.69	5.16 ± 0.06	5.41 ± 0.06	63.6	29.7	6.70	67.5	2.21×10^{-7}	1.40×10^{-7}	315	2.22×10^{8}	2.98×10^{13}	1890	1086
190503-QP	41.9	5.79 ± 0.06	5.83 ± 0.06	72.3	27.0	0.70	72.8	1.69×10^{-7}	1.16×10^{-7}	477	2.77×10^{8}	3.45×10^{13}	2360	1086
21QP-2*	28.0	5.72 ± 0.09	5.78 ± 0.09	71.3	27.6	1.10	72.1	1.90×10^{-8}	1.29×10^{-8}	4162	2.70×10^{8}	3.38×10^{13}	2295	1086
190504-EL	14.2	5.11 ± 0.05	5.20 ± 0.05	63.5	34.3	2.20	64.9	1.08×10^{-6}	6.58×10^{-7}	60	2.00×10^{8}	2.79×10^{13}	1703	1086
20TXX-2	16.3	6.64 ± 0.07	6.75 ± 0.07	82.7	15.4	1.90	84.4	3.11×10^{-6}	2.46×10^{-6}	56	6.88×10^{8}	7.37×10^{13}	5854	1520
20TXX-3	20.1	6.82 ± 0.07	6.92 ± 0.07	85.1	13.4	1.50	86.4	3.53×10^{-6}	2.87×10^{-6}	54	7.78×10^{8}	8.14×10^{13}	6622	1520
21TXX-3	27.0	7.17 ± 0.11	7.24 ± 0.11	89.5	9.40	1.10	90.5	3.23×10^{-6}	2.75×10^{-6}	74	1.03×10^{9}	1.03×10^{14}	8742	1520
20LYZ-3*	0.54	2.96 ± 0.04	4.80 ± 0.06	29.6	11.5	58.9	–	8.59×10^{-8}	–	–	–	–	–	–
21LYZ-2	2.90	2.58 ± 0.03	2.73 ± 0.03	30.7	58.4	10.9	34.0	9.50×10^{-8}	3.04×10^{-8}	550	8.44×10^{7}	2.23×10^{13}	715	1520
20XL-2*	0.55	0.49 ± 0.01	0.03 ± 0.001	>0	>42.5	<57.5	–	1.22×10^{-7}	–	–	–	–	–	–
20XL-3*	0.54	0.49 ± 0.01	0.01 ± 0.001	>0	>41.6	<58.4	–	8.90×10^{-8}	–	–	–	–	–	–
21XL-2	3.00	0.40 ± 0.01	0.34 ± 0.01	3.50	85.9	10.6	4.05	1.65×10^{-7}	6.30×10^{-9}	223	7.49×10^{6}	1.58×10^{13}	60	1520
21DZR-2	3.20	0.40 ± 0.01	0.35 ± 0.01	3.60	86.5	9.90	4.12	1.05×10^{-7}	4.06×10^{-9}	354	7.63×10^{6}	1.58×10^{13}	61	1520
190503-YJ	57.1	0.050 ± 0.002	0.045 ± 0.002	0.30	99.2	0.50	0.32	6.77×10^{-6}	2.81×10^{-8}	7.2	1.30×10^{6}	2.08×10^{13}	8.7	2075
Crust	1000	–	0.02	–	–	–	–	–	–	–	–	–	–	–
Mantle	1000	–	8.00	–	–	–	–	–	–	–	–	–	–	–

[*] Uprising fluid flow rate of Sample 21QP-2 is not considered due to low helium content that is suspicious in causing considerable uncertainty of calculated flow rate of uprising fluids (Table S1); Samples 20LYZ-3, 20XL-2, and 20XL-3 are ruled out for calculation and discussion because of their low $^{4}He/^{20}Ne$ ratios and possible high degrees of air contamination.

[a] Measured $^{3}He/^{4}He$ ratios (R/Ra) are corrected for air contamination, assuming that (i) all ^{20}Ne is derived from air-saturated water, and/or (ii) the N_2/Ar ratio represents the mixture between air and air-saturated water where applicable, both following the case of a groundwater recharge temperature of 15 °C (see details in the Supplementary Methods).

[b] Total helium inventory is calculated from the mixing of the MORB mantle (M), crust (C), and air (A) end-members. Samples 20XL-2 and 20XL-3 do not yield valid results of total helium inventory due to their low $^{4}He/^{20}Ne$ ratios that may be fractionated during gas purification processes before measurements; nevertheless, the corresponding estimates are shown for comparison.

[c] Mantle helium proportions are calculated assuming a binary mixing model between the MORB mantle and crust based on air-corrected $^{3}He/^{4}He$ ratios.

[d] Details about the calculation of helium concentrations, uprising fluid flow rates, and total outgassing fluxes of ^{3}He and ^{4}He are given in Text S2.

Table S2. Helium isotopic compositions, total helium inventory of hydrothermal gases, and the calculated helium fluxes from the Simao block.

Site No.	Site name	Latitude (N)	Longitude (E)	Elevation (m)	Temp. (°C)	Salinity (‰)	D_{trans} (km)	U (ppm)	Th (ppm)
1	QP	23°19'05.0"	101°26'05.1"	1183	61.3–62.5	0.92–0.98	15	2.70	10.5
2	EL	23°26'49.5"	101°24'57.3"	1130	53.9	0.46	15	2.70	10.5
3	TXX	22°57'18.6"	101°05'07.2"	1238	34.3–34.4	0.50–0.53	21	2.70	10.5
4	LYZ	23°02'24.8"	101°05'49.0"	1372	24.0–25.1	0.25–0.26	21	2.70	10.5
5	DSJ	22°48'37.8"	101°16'29.2"	1007	43.5	0.28	21	2.70	10.5
6	XL	23°02'34.9"	101°00'15.7"	1377	31.7–32.4	0.27–0.28	21	2.70	10.5
7	XS	23°12'30.0"	101°00'09.2"	1104	48	0.28	21	2.70	10.5
8	YJ	23°31'03.0"	101°53'54.0"	958	76.0–93.0	0.30	35	2.16	8.82

Abbreviations: QP, Qingping; EL, Eli; TXX, Tongxinxiang; LYZ, Laoyaozhai; DSJ, Dashujiao; XL, Xiling; XS, Xisa; YJ, Yuanjiang. The salinity of spring water is calculated using in situ measured electrical conductivity data (Supplementary Table 1). The distance of fluid transport from source to the surface (i.e., H_{crust}) is estimated based on the depths of high conductivity zones beneath Ning'er and Tongguan Quaternary volcanoes (Cheng et al., 2019). An inclined fluid uprising scenario is assumed for helium transport at No. 3–7 hot springs, corresponding with a transport distance of approximately 21 km from a 15-km-depth magma chamber. For the Ailao Shan-Red River shear zone (i.e., Site No. 8), the total crustal thicknesses of ~35 km (Wang et al., 2017) are taken as the distance of fluid transport. The recommended U and Th contents are cited from Rudnick and Gao (2014), assuming (i) upper continental crustal values for hot spring in the Quaternary volcanic field (i.e., No. 1–7), and (ii) averaged continental crustal values for the Yuanjiang hot spring in the non-volcanic region based on relative contribution proportions of 0.66:0.28:0.06 for radiogenic ^{4}He production in the upper, middle, and lower continental crust (Ballentine and Burnard, 2002; Rudnick and Gao, 2014).

Table S3. Reference parameter values used in calculation of helium fluxes.

Region	Active period (year of historical eruption)	Air-corrected $^3He/^4He$ (R/Ra)	References
Changbaishan volcanic field	Holocene (AD 1903)	6.33	Gao et al. (2006)
Tengchong volcanic field	Holocene (AD 1609)	5.92	Zhao et al. (2012)
Wudalianchi volcanic field	Holocene (AD 1719–1721)	3.18	Zhao et al. (2019)
Qiongbei volcanic field	Holocene	1.27	Xu et al. (2012)
Xianshuihe fault	Holocene	3.79	Zhou et al. (2015)
Litang fault	Holocene	1.03	Tang et al. (2017)
Karakoram fault	Holocene	2.24	Klemperer et al. (2013)
Tan-Lu fault	Holocene	1.87	Shangguan et al. (1998)
Gulu-Yadong rift	Holocene	1.02	Zhang et al. (2017)
Active faults in Yanqing, Beijing	Holocene	2.52	Lu et al. (2021)
Active faults in Zhangzhou, Fujian	Holocene	1.39	Tian et al. (2021)

Only the highest air-corrected $^3He/^4He$ ratio is compiled for the individual region to show the maximum degree of mantle helium degassing through hydrothermal systems.

Table S4. Compiled $^3He/^4He$ data of representative active/Quaternary volcanic fields and active fault zones in mainland China.

Study area	Tectonic setting	Mantle ^4He flux (10^{10} atoms m^{-2} s^{-1})	Crustal ^4He flux (10^{10} atoms m^{-2} s^{-1})	References
Mt. Minakami	volcanically active region	24135	5865	Wakita et al. (1978)
Lake Nyos	volcanically active region	19583	10417	Sano et al. (1990)
Lake Van	volcanically active region	21.4524	3.5476	Kipfer et al. (2002)
Lake Taupo	volcanically active region	0.621	2.379	Torgersen (1983, 2010)
Lake Nemrut	volcanically active region	515.86	84.14	Kipfer et al. (2002)
Crater Lake	volcanically active region	45.36	9.64	Kipfer et al. (2002)
Lake Mashu	volcanically active region	72.03	19.97	Kipfer et al. (2002)
Lac Pavin	volcanically active region	44.97	15.03	Kipfer et al. (2002)
Laacher See	volcanically active region	617.7	382.3	Kipfer et al. (2002)
Lake Baikal	tectonically active region	3.47	17.53	Kipfer et al. (2002)
Teggau Lake	tectonically active region	6.76	443.24	Torgersen and Clarke (1978), Torgersen (2010)
Green Lake	tectonically active region	0.34	810.66	Torgersen (2010)
Futagawa fault	tectonically active region	116	470	Sano et al. (2016), Kim et al. (2020)
Futagawa fault	tectonically active region	134	472	Sano et al. (2016), Kim et al. (2020)
Futagawa fault	tectonically active region	505	463	Sano et al. (2016), Kim et al. (2020)
Futagawa fault	tectonically active region	81.2	461	Sano et al. (2016), Kim et al. (2020)
Futagawa fault	tectonically active region	116	462	Sano et al. (2016), Kim et al. (2020)
Futagawa fault	tectonically active region	35.0	461	Sano et al. (2016), Kim et al. (2020)
Futagawa fault	tectonically active region	37.7	459	Sano et al. (2016), Kim et al. (2020)
Heunghae fault	tectonically active region	34.5	322	Kim et al. (2020)
Heunghae fault	tectonically active region	72.0	155	Kim et al. (2020)
Heunghae fault	tectonically active region	98.8	276	Kim et al. (2020)
Heunghae fault	tectonically active region	135	292	Kim et al. (2020)
Heunghae fault	tectonically active region	216	287	Kim et al. (2020)
Heunghae fault	tectonically active region	271	283	Kim et al. (2020)
Heunghae fault	tectonically active region	153	301	Kim et al. (2020)
Heunghae fault	tectonically active region	57.2	277	Kim et al. (2020)
Great Artesian Basin	sedimentary basin	0.008	3.092	Torgersen and Clarke (1985)
Great Hungarian Plain	sedimentary basin	0.028	0.172	Stute et al. (1992)
Great Hungarian Plain	sedimentary basin	0.666	7.334	Martel et al. (1989)
Paris Basin	sedimentary basin	0.0073	0.9927	Marty et al. (1993), Pinti and Marty (1995), Pinti et al. (1997), Castro et al. (1998a, 1998b)
Molasse Basin	sedimentary basin	0.0008	0.1992	Andrews et al. (1985)
Carrizo Aquifer, TX	sedimentary basin	0.0075	0.9925	Castro et al. (2000)
Black Sea	sedimentary basin	0.0001	1.2999	Top and Clarke (1981)
San Juan Basin, NM	sedimentary basin	0.008	0.792	Castro et al. (2000)
Eastern Paris Basin	sedimentary basin	0.100	9.900	Ballentine et al. (2002)

Table S5. Compiled helium fluxes data of representative volcanically and tectonically active regions and sedimentary basins worldwide.