|  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Sample ID | Sampling site | Temp. (oC) | EC (μS/cm) | pH | He  (ppmv) | Ar  (vol.%) | N2  (vol.%) | O2  (vol.%) | CO2  (vol.%) | CH4  (vol.%) | CO2/N2 | N2/He | N2/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 | 4He/20Ne | Measured  3He/4He  (R/Ra, 1σ) | Corrected  3He/4He  (R/Ra, 1σ) a | Helium inventory (%) b | | | MORB Mantle  He (%) c | 4He in spring water (cm3 STP g−1 H2O) d | 4He in mantle fluids (cm3 STP g−1 H2O) d | Fluid flow rate (mm yr−1) d | Total 3He flux (atoms m−2 s−1) d | Total 4He flux (atoms m−2 s−1) d | Mantle 4He flux (1010 atoms m−2 s−1) | Crustal 4He flux (1010 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 × 108 | 2.98 × 1013 | 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 × 108 | 3.45 × 1013 | 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 × 108 | 3.38 × 1013 | 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 × 108 | 2.79 × 1013 | 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 × 108 | 7.37 × 1013 | 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 × 108 | 8.14 × 1013 | 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 × 109 | 1.03 × 1014 | 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 × 107 | 2.23 × 1013 | 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 × 106 | 1.58 × 1013 | 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 × 106 | 1.58 × 1013 | 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 × 106 | 2.08 × 1013 | 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 4He/20Ne ratios and possible high degrees of air contamination.

a Measured 3He/4He ratios (R/Ra) are corrected for air contamination, assuming that (i) all 20Ne is derived from air-saturated water, and/or (ii) the N2/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 4He/20Ne 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 3He/4He ratios.

d Details about the calculation of helium concentrations, uprising fluid flow rates, and total outgassing fluxes of 3He and 4He 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. (oC) | 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 4He 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  (1010 atoms m−2 s−1) | Crustal 4He flux  (1010 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.