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Massive crustal carbon mobilization and emission driven by India underthrusting Asia

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The active Himalayan-Tibetan orogen, where India underthrusts into Asia, is an important geological source of carbon dioxide (CO₂) emission into Earth's atmosphere. However, the extent to which Indian underthrusting could stimulate the mobilization of deeply-sourced carbon and its subsequent emission remains unknown. Here, we use a combination of field observations coupled with in-situ CO₂ flux measurements and helium and carbon isotopic data, to study the controls on CO₂ origins and fluxes in a 400-kilometre-long rift transecting northern Himalaya and southern Tibet. High diffuse CO₂ fluxes sustained by pure crustal fluids are confined to rift segments in the northern Himalaya, while toward southern Tibet, CO₂ fluxes become lower but mantle fluid inputs are identified. Such rift-related CO₂ degassing profile suggests metamorphic decarbonation and release of carbon-bearing fluids enhanced by the underthrusting Indian lower crust, agreeing well with Himalayan metamorphism and orogen-parallel lithospheric extension. Deep CO₂ fluxes from extensional tectonics in northern Himalaya and southern Tibet, primarily of crustal origins, are comparable to mantle CO₂ fluxes from global mid-ocean ridges. Our findings demonstrate that geophysical and geo-tectonic responses to continental underthrusting could facilitate massive crustal carbon mobilization and emission, making active collisional orogens globally important carbon sources.

The continental collision between India and Asia built up the Tibetan Plateau¹ that is essential for global carbon cycle and climate changes in the Cenozoic^{2,3}. Over the past few decades, the Tibetan Plateau and its surrounding tectonically uplifted regions have been well documented for their role as atmospheric CO₂ sinks through silicate weathering and organic carbon burial in the geological past^{3,4} and the global warming era at present^{5,6}. Conversely, the Himalayan-Tibetan orogen is also considered as important CO₂ sources due to solid Earth (referring to the mantle and crust) carbon emissions by multi-stage magmatism², regional metamorphism⁷, and enhanced erosion⁸ driven by the India-Asia collision. As a result of ongoing magmatism and metamorphic decarbonation, deep CO₂ emissions are pervasive in modern volcanically and tectonically active regions of the Tibetan Plateau and its surroundings^{9–14}. The co-existence of CO₂ removal from and emission into Earth's atmosphere has boosted a long-lasting debate on the net carbon budget of the Himalayan-Tibetan orogen¹⁵. Particularly, the heterogeneous deep CO₂ emissions at regional to global scales¹⁶

and spatially variable atmospheric CO₂ consumption rates⁴ could cause large uncertainties in whole-Earth carbon cycling models that incorporate the Himalayan-Tibetan orogen as key end-members for the feedbacks between carbon sources and sinks^{17,18}.

With respect to the role of the India-Asia collision zone as a potential deep carbon source, reducing the uncertainties in CO₂ emission rates remains challenging due to the impracticality of carrying out field-based observations in the entire Himalayan-Tibetan orogen, which is the largest one on Earth at present. Nevertheless, clarifying spatial variations in CO₂ origins and fluxes across the India-Asia collisional orogen, as well as the controlling factors, would shed light on the variability of deep CO₂ emissions in response to continental collision dynamics. Northward underthrusting of India beneath Asia has exerted remarkable impacts on lithospheric structure¹⁹, thermal state²⁰, rheological behavior²¹, and active tectonics²² of the Himalayan-Tibetan orogen, where the crustal thickness reaches approximately double that of normal continental crust²³. These

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geophysical and geo-tectonic responses to Indian underthrusting are expected to play a fundamental role in the mobilization of carbon from its source, as well as the uprising and degassing of CO₂-bearing fluids en route to the surface. To elucidate the contribution of Indian continental underthrusting to deep carbon mobilization and emission, it is pivotal to establish a south-to-north CO₂ degassing profile (i.e., following the direction of India-Asia convergence) by linking surface observations of CO₂ emissions (e.g., gas geochemistry and outgassing fluxes) to origins of CO₂-bearing fluids at depth. However, previous studies on CO₂ degassing from active fault zones^{11–13} are limited in space, and an across-orogen profile of the CO₂ origins and fluxes is yet to be constrained. Furthermore, the potential control of India-Asia collision on regional CO₂ emission variabilities, if any, remains untested. To our knowledge, the tectonic CO₂ degassing in the Himalayan-Tibetan orogen has rarely been interpreted from the causal effects of Indian underthrusting that could well reconcile geochemically constrained CO₂ origins and field-based observations of CO₂ fluxes.

Here, we quantitatively constrained the CO₂ origins and fluxes of the Tingri-Tangra Yumco rift (TTYR) in the central part of the India-Asia collision zone, extending approximately 400 km from northern Himalaya to southern Tibet (referring to the Tethyan Himalaya and Lhasa Block, respectively; Fig. 1a). We established a rift-related CO₂ degassing profile along the TTYR (Fig. 1b) based on (i) CO₂ origins constrained by δ¹³C-CO₂, ³He/⁴He, and CO₂/²He of hydrothermal gases, and (ii) field-based measurements of diffuse soil CO₂ fluxes in hydrothermal fields, fault zones, and background areas (“Methods” section). Integrating the analysis of CO₂ origins and fluxes in the TTYR enables us to unravel how Indian underthrusting could facilitate carbon mobilization and emission through extensional tectonics in collisional orogens. By examining the spatial variations in CO₂ origins and fluxes from northern Himalaya to southern Tibet, possible controlling factors associated with Indian underthrusting (e.g., collision-zone geometry, crustal thermal structure, and lithospheric extension) were explored for the rift-related CO₂ degassing profile. Moreover, we

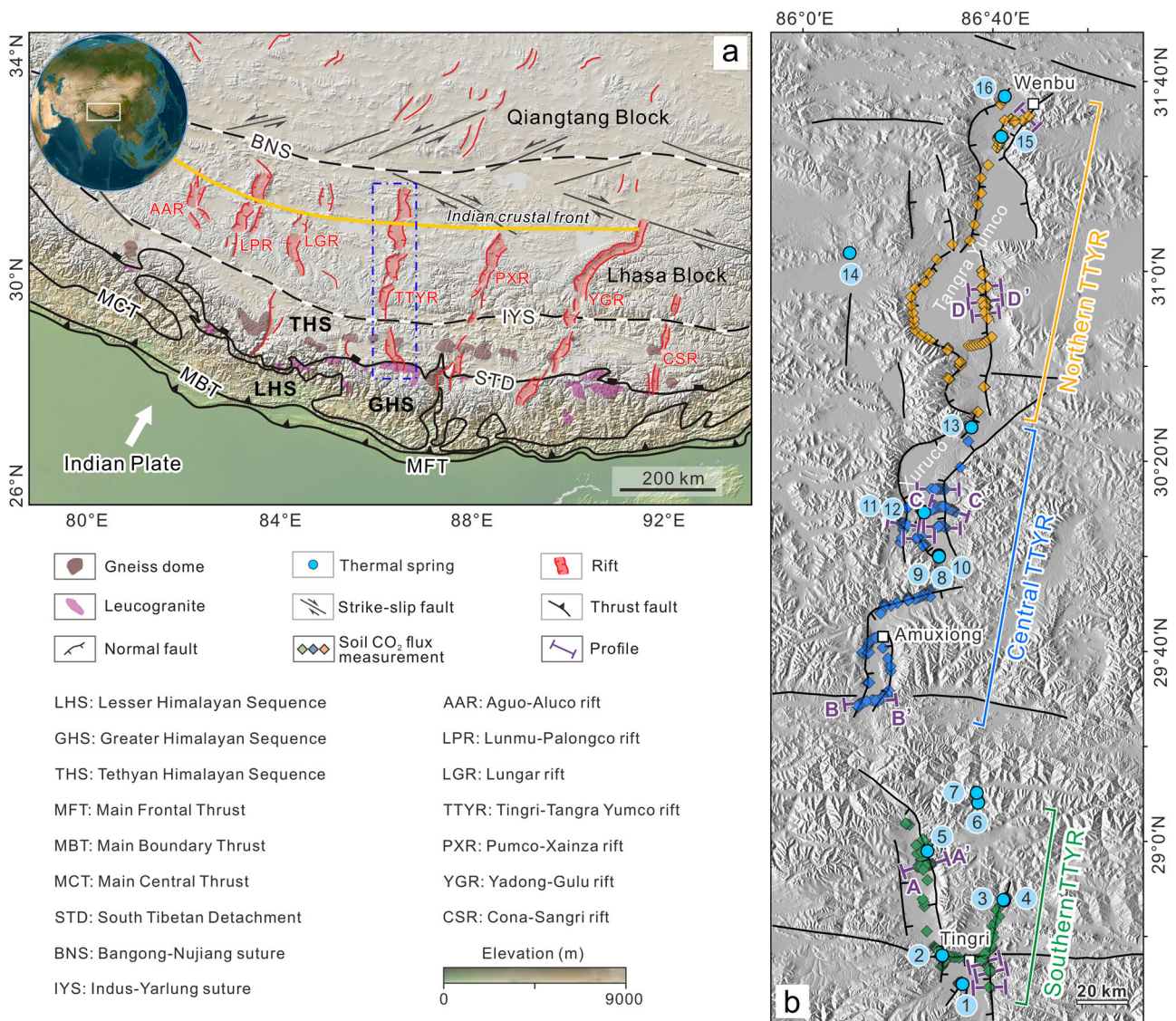


Fig. 1 | Tectonic framework of the Himalayas and southern Tibet, CO₂ flux measurements, and sample distribution in the study area. a Map of the Himalayan-Tibetan orogen showing major tectonic boundaries and extensional rift systems. Base map was created with GeoMapApp (www.geomapp.org)/CC BY, and the hemisphere inset was generated using ArcGIS Earth software. Localities of the Himalayan domes and leucogranites are after ref. 51. The dash-dotted blue box shows the location of the TTYR. The yellow line represents the seismically inferred

northern extent of the Indian crustal front¹⁹. **b** Distribution of CO₂ flux measurements and thermal springs in the southern, central, and north segments of the TTYR. Profiles AA', BB', CC', and DD' represent soil CO₂ measurements across active fault zones, as shown in Supplementary Fig. 8. The base map is from a 30-meter SRTM digital elevation model (www.earthexplorer.usgs.gov). Numbers represent hydrothermal fields from which bubbling gas samples were collected for geochemical analysis, with carbon and helium isotope data shown in Fig. 3a, b.

estimated the deep CO₂ output of extensional tectonics (i.e., rifts and normal faults) in the northern Himalaya and southern Tibet based on carbon isotope mass balance model and flux extrapolation (“Methods” section and Supplementary Information). Taking into account the CO₂ origins and fluxes across the India-Asia collisional orogen, we suggest that the geophysical and geo-tectonic responses to Indian underthrusting are the first-order controls on mobilization and emission of massive crustal carbon, making the Himalayan-Tibetan orogen a globally important carbon source.

Results and discussion

Carbon origins and secondary hydrothermal processes

The origin and evolution of CO₂-bearing fluids in hydrothermal systems are controlled by a series of physico-chemical processes^{24–28}, such as partial melting of the mantle or crustal source rocks, metamorphic decarbonation reactions, gas-water-rock interaction, calcite precipitation, and multi-component mixing. Particularly, the intricate secondary hydrothermal processes, which could result in He-CO₂ elemental and isotopic fractionation as reflected by δ¹³C-CO₂ and CO₂/³He data^{24,29}, indicate the need for caution when attempting to trace the origins of CO₂-bearing fluids using He-CO₂ systematics. For example, the CO₂/³He ratios of the TTYR samples vary from 8.7 × 10⁸ to 9.6 × 10¹² and appear to decrease from south to north (Supplementary Fig. 1). However, it would be misleading to infer a south-to-north transition from mantle carbon sources to crustal carbon sources simply by comparing CO₂/³He of samples with reference values of the crustal and mantle end-members^{30,31}. Instead, hydrothermal degassing is more likely to result in preferential loss of He relative to CO₂ from the residual fluids (Supplementary Fig. 2). It should be noted that the relative lower CO₂/³He and negative δ¹³C-CO₂ values of several samples were likely modified by calcite precipitation and/or CO₂ partial dissolution in groundwater (Fig. 2), as supported by the presence of travertine deposits around the spring mouths and the higher solubility of CO₂ relative to He in the groundwater²⁴ (Fig. 2).

To further evaluate the influence of secondary hydrothermal processes, we modeled the trends of (i) calcite precipitation at 125 °C and 192 °C

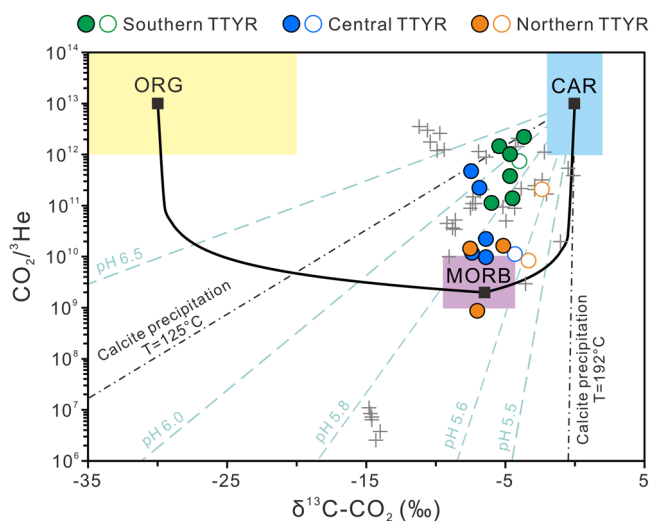


Fig. 2 | Plot of CO₂/³He versus δ¹³C-CO₂ for hydrothermal gases. The mixing lines between mantle (MORB), organic sediment (ORG), and carbonate (CAR) end-members are shown for comparison. Filled and open symbols represent data in this study and literature data, respectively. Data from the Yadong-Gulu rift (YGR) are shown by gray crosses. Black dotted lines represent predicted calcite fractionation model trends for CO₂ loss by calcite precipitation at 125 °C and 192 °C (after ref. 26). Light blue dashed lines show the predicted trends for gas dissolution over a range of pH values at 50 °C (average temperature of the studied thermal springs) (after ref. 25). Assuming a starting carbonate-like δ¹³C value is 0.3‰ for metamorphic CO₂ and CO₂/³He = 1 × 10¹³ in the predicted model lines for calcite precipitation and partial gas dissolution (after ref. 44).

following Barry et al.²⁶ and (ii) CO₂ dissolution during gas-water interaction over a range of pH values (5.5–6.5) at a fixed temperature of ~50 °C (i.e., average sample temperature measured in the field) following Gilfillan et al.²⁵ (Fig. 2). The selection of temperature for calcite precipitation modeling is supported by reservoir temperature (average = 146 °C; ref. 32) of the TTYR thermal springs. Considering that the Himalayan-Tibetan orogen is characterized by a remarkably thick crust (thickness up to 70 km¹⁹) and abundant carbon-bearing metamorphic rocks and sedimentary rocks^{12,33}, the carbonate/metamorphic end-member was chosen as the starting point for calcite fractionation and CO₂ dissolution models (Fig. 2). This is consistent with the He-C isotopic data and mixing calculations that show predominant origins of the CO₂-bearing fluids from a crustal carbonate/metamorphic end-member (see details below). The modeled lines agree well with the dataset and the hydrothermal gases may have been affected by calcite precipitation and/or CO₂ dissolution to different degrees (Fig. 2). Notably, it should be clarified that the role of secondary processes deduced from the modeling is inevitably simplified for the complex migration paths of hydrothermal fluids through the thick crust of the Himalayan-Tibetan orogen.

Although secondary hydrothermal processes occurred and may have modified chemical and isotopic compositions of the CO₂-bearing fluids²⁴ (i.e., masking their true source), we note that most samples are within the envelope area defined by the MORB, CAR, and ORG end-members (Fig. 2), which represents the expected results of the ternary mixing model proposed by Sano and Marty (ref. 31). This may suggest limited impacts of secondary hydrothermal processes on the chemical and isotopic composition of the CO₂-bearing fluids (i.e., limited He-CO₂ fractionation) for most samples, and this possibility requires further assessment. Based on the above assumption, we calculated the proportions of different carbon source components (i.e., mantle, carbonate rocks, and organic sediments) following a ternary mixing model³¹ (Fig. 2) as expressed by the equations below:

$$\begin{aligned} (^{13}\text{C}/^{12}\text{C})_{\text{Obs}} &= f_{\text{MORB}} \times (^{13}\text{C}/^{12}\text{C})_{\text{MORB}} + f_{\text{CAR}} \times (^{13}\text{C}/^{12}\text{C})_{\text{CAR}} \\ &+ f_{\text{ORG}} \times (^{13}\text{C}/^{12}\text{C})_{\text{ORG}} \end{aligned} \quad (1)$$

$$\begin{aligned} 1/(^{12}\text{C}/^3\text{He})_{\text{Obs}} &= f_{\text{MORB}}/(^{12}\text{C}/^3\text{He})_{\text{MORB}} + f_{\text{CAR}}/(^{12}\text{C}/^3\text{He})_{\text{CAR}} \\ &+ f_{\text{ORG}}/(^{12}\text{C}/^3\text{He})_{\text{ORG}} \end{aligned} \quad (2)$$

$$f_{\text{MORB}} + f_{\text{CAR}} + f_{\text{ORG}} = 1 \quad (3)$$

where subscripts Obs, MORB, CAR, and ORG refer to the observed value of the sample, mantle, carbonate rocks, and organic sediments, respectively. Reference values for the MORB, CAR, and ORG end-members are as those in Sano and Marty (ref. 31), which are: δ¹³C = -6.5 ± 2.5‰ and CO₂/³He = 2 × 10⁹ for MORB-type mantle, δ¹³C = 0 ± 2‰ and CO₂/³He = 10¹³ for CAR, and δ¹³C = -30 ± 10‰ and CO₂/³He = 10¹³ for ORG.

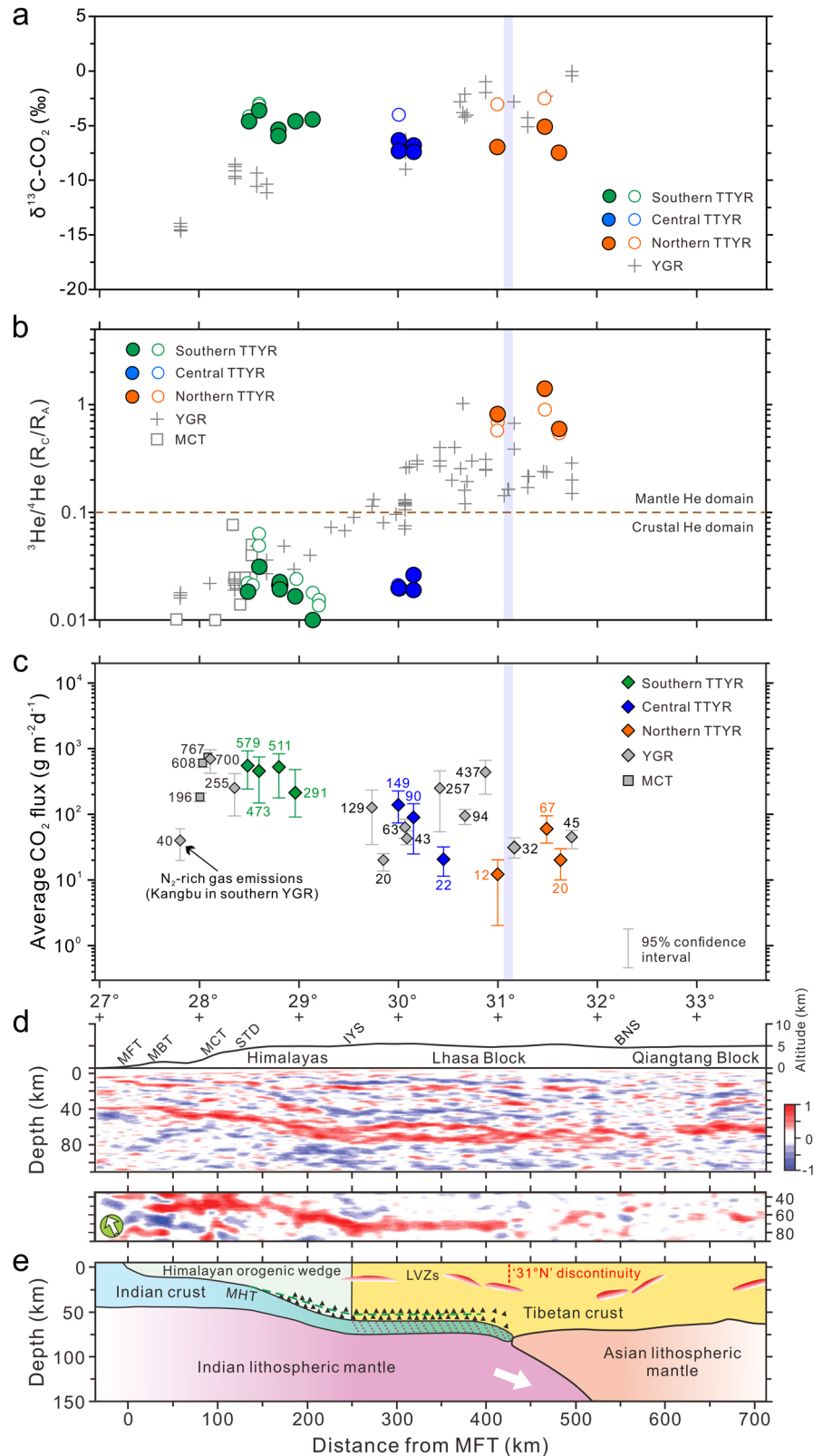
The results show that three samples (21CZX1-1, 21CZX2-3, and 21CZX3-2 from the Chazi hydrothermal field) may have been remarkably affected by secondary processes because of their low CO₂/³He ratios and abnormally high proportion of mantle carbon inputs (Supplementary Data 1); the latter is at odds with the pure crustal fluid sources evidenced by He isotope data (see details below). It is evident that the crustal carbon components are dominant in the entire TTYR; and if not considering the Chazi samples, the southern and central TTYR would be characterized by >99% crustal carbon contributions, while remarkable mantle carbon inputs (12–14%) are found in the northern TTYR (Supplementary Data 1). We speculate a pure crustal (metamorphic) carbon source for the southern and central TTYR and a crust-dominated (but showing clear mantle inputs) carbon source for the northern TTYR. Notably, the relative proportion of mantle carbon and crustal metamorphic carbon in the deeply-sourced carbon end-member is challenging to be quantitatively constrained due to the difficulty in distinguishing between metamorphic carbon and shallow carbon (e.g., carbonate dissolution and organic matter

decomposition) solely based on the ternary mixing model (Fig. 2). In addition, considering the fraction of the original carbon removed by secondary processes during the upward migration of deeply-sourced CO₂-bearing fluids, it is plausible that our observations of deep CO₂ fluxes at the surface only represent part of the original amount of carbon (e.g., CO₂) below the study area.

Rift-related hydrothermal degassing profile from northern Himalaya to southern Tibet

Following the Indian motion to the north, we compared the δ¹³C-CO₂ and ³He/⁴He values of hydrothermal gases (Figs. 3a and 3b) and average soil CO₂ fluxes of hydrothermal fields (Fig. 3c) relative to the distance from the Main Frontal Thrust (MFT), which represents the southernmost boundary

Fig. 3 | Carbon and helium isotopes of hydrothermal gas samples, average soil CO₂ fluxes, geophysically imaged collision-zone structure, and geological profile across the Himalayas and southern Tibet. Abbreviations are as in Fig. 1. Filled and open symbols in Panels a and b represent data in this study and literature, respectively. **a** Plot of δ¹³C-CO₂ values versus latitude of sampling sites for hydrothermal gases. **b** Plot of ³He/⁴He (R_C/R_A) versus latitude of sampling sites for hydrothermal gases. Samples from the YGR^{11,12,35,38} and MCT (see ref. 38, and references therein) are shown for comparison (see compiled data in Supplementary Data 2). **c** Average soil CO₂ flux (g m⁻² d⁻¹) versus latitude of the hydrothermal fields. The compiled soil CO₂ flux data of hydrothermal fields in the Himalayas and southern Tibet are summarized in Supplementary Table 1. Note that the results of soil CO₂ fluxes in fault zone profiles and background areas investigated in this study are not shown in Panel c. **d** Receiver function image¹⁹ showing seismic velocity profile across the Himalayas and southern Tibet. Panel d refers to Panel e and provides seismic evidence for collision-zone structure from crustal to mantle depths. Note that there are no corresponding relationships between Panel d and Panels a, b, and c. **e**, Geological interpretation of the India-Asia collision-zone structure (modified from ref. 19). Main Himalayan Thrust (MHT) represents the lower boundary of the Himalayan orogenic wedge. The dashed purple lines show eclogitization of the horizontal portion of the underthrusting Indian lower crust. A broad layer of LVZs (low-velocity zones) in the Himalayas and southern Tibet is shown by the areas of dashed green lines and black triangles above the underthrusting India. Shallow LVZs are scattered in the upper crust. The light blue band in Panels a–c represents the Indian crustal front constrained by the 31°N discontinuity¹⁹.



recording the India-Asia collision. This comparison defines a rift-related hydrothermal degassing profile in terms of C-He isotopes and diffuse CO₂ emissions from northern Himalaya to southern Tibet. Unlike the relatively wide $\delta^{13}\text{C-CO}_2$ range (-14.8 to +0.2‰; refs. 11,34,35) observed in the Yadong-Gulu rift (YGR; Fig. 1a), the TTYR samples have a much narrower range of $\delta^{13}\text{C-CO}_2$ values (-7.5 to -2.6‰; Fig. 3a). Spatially, there are almost no variations in $\delta^{13}\text{C-CO}_2$ among southern, central, and northern segments of the TTYR, which differs from a northward-increasing trend in $\delta^{13}\text{C-CO}_2$ along the YGR¹². The $\delta^{13}\text{C-CO}_2$ data could be influenced by multiple factors that may contribute to the origin and transport of CO₂-bearing fluids, such as mantle carbon inputs, crustal lithology (e.g., limestone vs. meta-sedimentary rocks), addition of biogenic carbon into the hydrothermal fluids, and solubility-controlled phase separation (e.g., hydrothermal degassing and calcite precipitation). As for the majority of the TTYR samples that show limited influence by He-CO₂ elemental and isotopic fractionation (Fig. 2), we find that the southern and central TTYR are characterized by a pure crustal carbon source (mantle carbon <1%), while mantle carbon inputs (12–14%) are remarkable in the northern TTYR. This is consistent with the relative proportions of the crustal and mantle fluids calculated from He isotope systematics of the hydrothermal gases (Supplementary Data 1), as detailed below.

Since (i) CO₂ is believed to be a primary carrier for He migration through the crust³⁰ and (ii) He is chemically inert and is a powerful tracer for quantitatively constraining the inputs of mantle and crustal fluids, the origins of CO₂-bearing fluids can be interpreted from air-corrected ³He/⁴He data (R_C/R_A , where $R_A = \text{air } ^3\text{He}/^4\text{He} = 1.39 \times 10^{-6}$). We note that a first-order feature of the rift-related hydrothermal degassing profile is the contrasting He source components among different rift segments (Fig. 3b). Specifically, a pure crustal domain of He sources dominates the southern and central TTYR (³He/⁴He = 0.01–0.06 R_A , corresponding to null or <1% mantle He inputs), whereas in the northern TTYR, the proportion of mantle fluids in the CO₂-bearing fluids could reach ~7–17% [³He/⁴He = 0.55–1.41 R_A , well over the canonical crustal value of 0.02 R_A (ref. 36)]. The along-rift ³He/⁴He variations in the TTYR are consistent with previously identified patterns of ³He/⁴He distribution in the Himalayas and the hinterland Tibetan Plateau^{37–39} (Fig. 3b; see also Supplementary Fig. 3). Moreover, to the south of the Himalayan crest, large amounts of crustal fluids (³He/⁴He = 0.003–0.076 R_A ; refs. 38,40) have also been observed in the Main Central Thrust (MCT), suggesting a pure crustal fluid source for hydrothermal systems in the Himalayan fold-and-thrust belt. In stark contrast, the near N-S striking extensional rifts that transect the northern Himalaya and southern Tibet (generally located to the north of the Himalayan crest; Fig. 1a) show a prominent transition in origins of CO₂-bearing fluids from a pure crustal domain in the south to a domain characterized by discernible northward-increasing inputs of mantle fluids (e.g., up to 7–17% in the northern TTYR; Fig. 3b). We suggest that such transition in fluid origins constrained by C-He elemental and isotopic systematics (Figs. 2 and 3b) may point to the impact of continental collision-related properties of the Himalayan-Tibetan orogen, such as crust-mantle structure¹⁹ and crustal thermal state²⁰, on the mobilization of carbon in its sources from northern Himalaya to southern Tibet.

Figure 3c illustrates the variations in average soil CO₂ fluxes of hydrothermal fields from the Himalayas to southern Tibet (Supplementary Table 1). Clearly, the southern TTYR exhibits higher average soil CO₂ fluxes (291–579 g m⁻² d⁻¹) than observed in the central and northern TTYR (12–149 g m⁻² d⁻¹) (Supplementary Fig. 4). Except for one hydrothermal field dominated by N₂-rich gas emissions (i.e., Kangbu in southern YGR¹²; Fig. 3c), the available data of soil CO₂ fluxes show that the Himalayas are currently experiencing vigorous degassing of CO₂-bearing fluids [e.g., 196–767 g m⁻² d⁻¹ in the MCT¹⁰, 255–700 g m⁻² d⁻¹ in southern YGR¹², and 291–579 g m⁻² d⁻¹ in the TTYR (this study)], as expected in the global-scale tectonic CO₂ degassing model¹⁶. To the north of the Indus-Yarlung suture (Fig. 1a), most hydrothermal fields ($n = 13$) in the TTYR and YGR have lower average soil CO₂ fluxes (12–149 g m⁻² d⁻¹) than observed in the Himalayas (Fig. 3c). Two localities showing high soil CO₂ fluxes (253 and

437 g m⁻² d⁻¹, respectively) in the northern YGR¹¹ may result from localized high influx of CO₂-rich fluids. If only considering hydrothermal fields with CO₂-rich gas emissions, the average diffuse soil CO₂ fluxes would generally exhibit a good correlation with latitude [$R^2 = 0.528$, $p < 0.001$ ($n = 22$); Supplementary Fig. 5], suggesting spatially variable controls on CO₂ degassing from the Himalayas to southern Tibet.

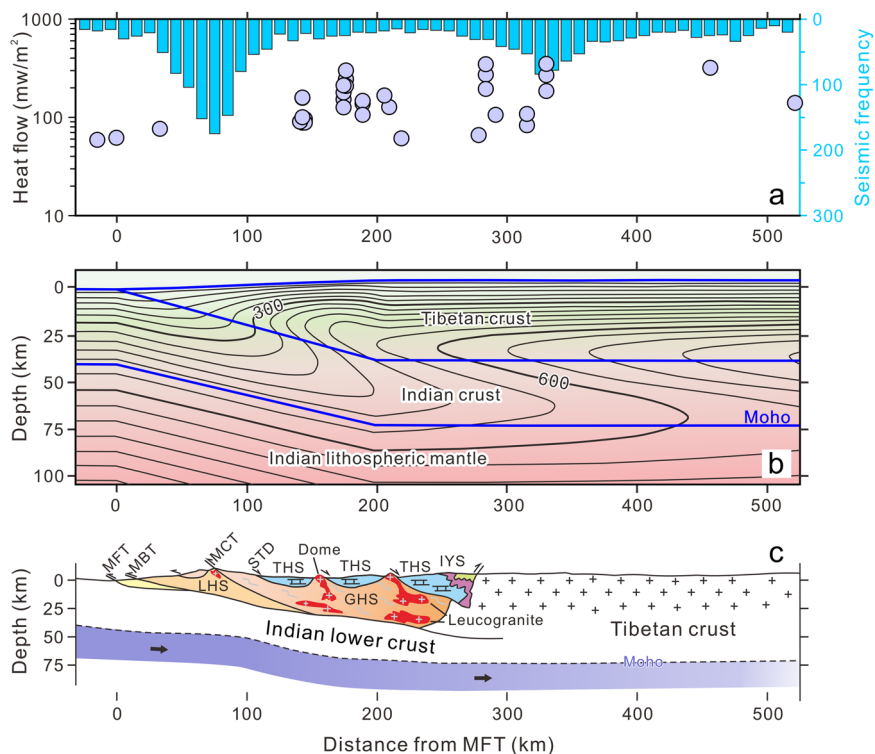
Taking CO₂ origins and fluxes together, we find that high CO₂ fluxes are sustained by pure crustal fluids in the Himalayan orogenic wedge, a wedge-shaped portion above the underthrusting Indian lower crust (Fig. 3d, e), while relatively lower CO₂ fluxes dominate rift segments in southern Tibet where mantle carbon inputs are discernible (Fig. 3b, c). Notably, tectonic CO₂ emissions from the Himalayan-Tibetan orogen are generally dominated by crustal decarbonation. Furthermore, it is striking that the geophysically imaged geometry of the orogenic crust (Fig. 3d, e; ref. 19) corresponds well with variations in CO₂ origins and fluxes from the Himalayas to southern Tibet. As observed in the TTYR, pure crustal carbon sources are generally identified in regions above the underthrusting Indian lower crust, although it remains uncertain whether mantle carbon inputs have occurred in the gap zone between the central and northern TTYR [i.e., the rift segment approximately between 30°N and 31°N to the south of the Indian crustal front that is defined by the so-called “31°N discontinuity” (Fig. 3b); ref. 19]. But in the YGR, mantle fluid signals (³He/⁴He > 0.1 R_A) were identified as far as ~110 km southward from the surface projection of the Indian crustal front^{34,37,38}, suggesting that mantle carbon inputs to hydrothermal systems may vary among the near N-S striking extensional rifts. A possible explanation for such rift-dependent carbon mobilization and emission mode could be the orogen-parallel changes in morphology (e.g., subduction angle) and collision-zone dynamics (e.g., tearing and/or break-off) of the subducting Indian plate^{38,39,41}, which have the potential to determine the distribution, amounts, and rates of mantle carbon supply to the extensional rifts in southern Tibet.

Carbon mobilization driven by Indian underthrusting

An intriguing feature of the rift-related CO₂ degassing profile lies in the crustal portion above the underthrusting Indian lower crust, which is bounded by the MFT to the south and the Indian crustal front to the north (Fig. 3e). Intensive mobilization of carbon is expected to occur at crustal levels beneath the Himalayan-Tibetan orogen, sustaining high CO₂ fluxes observed at the surface, particularly in the Himalayan orogenic wedge (Fig. 3c, e). This may suggest the fundamental role of Indian underthrusting in mobilization of crustal carbon from its sources. To address this possibility, we integrated plausible constraints from geophysics and geo-tectonics of the collision-zone crust that have close affinities with the underthrusting Indian continent (Fig. 4). Considering the absence of volcanic activities in southern Tibet for the past ca. 8 million years¹, and the non-volcanic nature of the Himalayan orogenic wedge over even longer timescales⁴², the partial melting and intruded magmas as the reservoirs to feed CO₂ degassing are unlikely scenarios. Instead, the mobilization of carbon through metamorphic decarbonation has been widely invoked as a plausible mechanism for CO₂ origins in field-based and experimental studies on active orogenic degassing^{9,12,33,43,44}. In this section, the controlling factors for metamorphic decarbonation are considered to explain the high CO₂ fluxes observed in the Himalayan orogenic wedge.

A primary feature of young and mature collisional orogenic systems, such as the Cenozoic Himalaya, is the evolution of crustal thermal structure during continental collision processes^{45,46}. Currently, the active extensional rifts (mostly initiated at 13 ± 3 Ma⁴²) in the northern Himalaya and southern Tibet have high surface heat flows (up to 100–300 mW m⁻² as shown in Fig. 4a; refs. 21,47), consistent with the predictions in thermal structure model of continental collisional orogens⁴⁶. In addition, thermal modeling results of the Indian underthrusting beneath Asia have suggested temperatures of ≤ 600 °C for the crust of the Himalayan orogenic wedge (Fig. 4b; ref. 20). Under such a crustal thermal structure, metamorphic decarbonation reactions could occur roughly within crustal depths of 10–25 km (>300 °C; ref. 48). Notably, the simulated geotherms become shallower to

Fig. 4 | Surface heat flows, earthquakes, thermal structure, and geological evidence for the massive crustal carbon mobilization and emission in the Himalayan orogenic wedge. **a** Surface heat flows and seismic frequency versus distance from MFT across the Himalayas and southern Tibet. Filled circles represent measured surface heat flows from the global heat flow database: update 2023 (www.ihfc-iugg.org). Earthquake events are from USGS. **b** Model of crustal thermal structure across the Himalayas and southern Tibet³⁰. Contours are at 50 °C intervals. Blue lines denote the boundaries between the Tibetan crust, Indian crust, and the mantle based on seismic images of lithospheric structure¹⁹. **c** Geological profile across the Himalayan-Tibetan orogen showing outcropping of domes and leucogranites in the Himalayan orogenic wedge, especially the northern Himalaya (modified from ref. 51). **c**. Abbreviations are as in Fig. 1.



the surface in the regions approximately 100–200 km north of the MFT (Fig. 4b), which agrees well with the observed high CO₂ fluxes in the MCT and southern segments of the TTYR and YGR (Fig. 3c). We, therefore, suggest that metamorphic decarbonation reactions at crustal levels could be enhanced by warming (i.e., elevated geotherms^{20,21}) of the Himalayan orogenic wedge during India-Asia continental collision (i.e., the underthrusting of India beneath Asia).

In accretionary wedges of collisional orogens, a thermally weakened infrastructure is likely to generate lateral crustal flows toward the orogenic foreland⁴⁵. This is a widely accepted model for the Himalayas where geophysically imaged anomalies of low electric resistivity⁴⁹ and low seismic velocity¹⁹ are plausible mid-crustal channel flow that migrates toward the Himalayan erosional front^{23,50}. The channel flow probably represents hot and ductile materials (e.g., anatexic melts involved in the formation of Himalayan leucogranites⁵¹) that could promote crustal CO₂ production by heating the overlying carbon-bearing rocks (e.g., carbonate and calc-silicate rocks). Possible geological evidence for such heating-related metamorphic decarbonation could be the presence of the Himalayan leucogranites and gneiss domes in southern segments of the extensional rifts^{42,50} (Fig. 4c), including the southern TTYR and southern YGR (Fig. 1a). Notably, geological evidence for metamorphism was not observed in the southern Tibet where CO₂ fluxes are low, suggesting that the Himalayan metamorphism is critical for crustal carbon mobilization. Besides the heating effects of mid-crustal channel flow, the infiltration of aqueous fluids could remarkably increase the efficiency of metamorphic decarbonation⁵², which is a plausible scenario for the Himalayan orogenic wedge due to dehydration of the Indian lower crust above the inclined portion of the Main Himalayan Thrust (MHT; ref. 21).

Carbon emission facilitated by extensional tectonics

A global-scale carbon emission model¹⁶ reveals that the Tibetan Plateau and its surroundings have a high probability of CO₂ degassing related to extensional tectonics, which is in comparable magnitude with the East African rift system that was suggested to have CO₂ outgassing flux of $71 \pm 33 \text{ Mt yr}^{-1}$ (ref. 53). Tectonically, the control of Indian underthrusting on deep CO₂ emissions from extensional rifts such as the TTYR could be

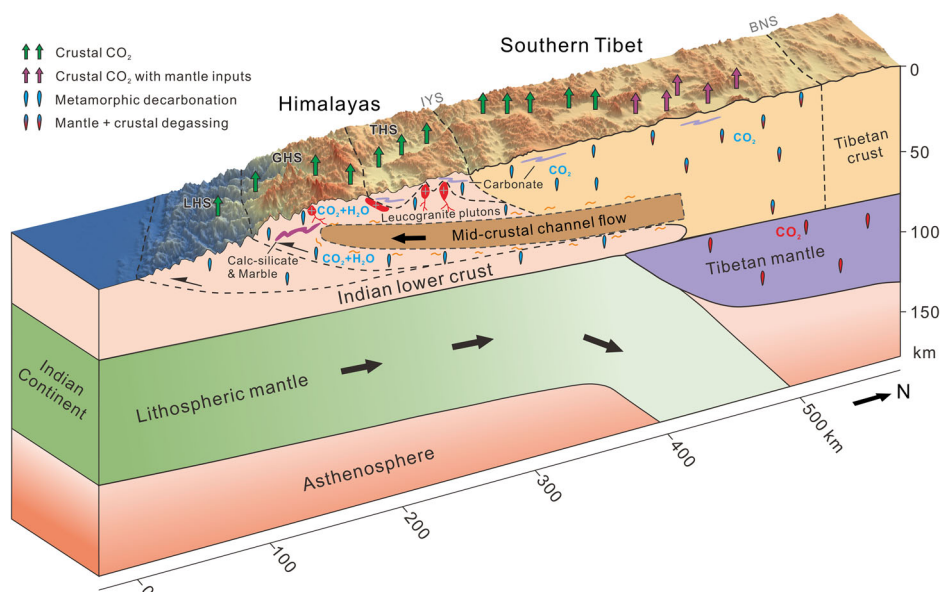
interpreted in models that the eclogitized Indian lower crust may have served as a dynamic trigger for the orogen-parallel lithospheric extension in the Himalayas and southern Tibet^{22,54,55}. In the extensional regimes of rifts and normal faults²², the CO₂-bearing fluids could migrate efficiently to the surface and thus sustain high CO₂ fluxes¹⁶. Due to the ongoing continental collision, earthquake events could promote the formation of highly permeable fracture networks in the brittle upper crust and facilitate efficient uprising and degassing of the CO₂-bearing fluids^{10,56}. As shown in Fig. 4a, the spatial variations in the frequency of earthquake events, which mostly occurred in the Himalayan orogenic wedge, may suggest more favorable pathways for the upward migration of CO₂-bearing fluids in the Himalayas than in southern Tibet. Thermodynamic modeling results show that the fluid immiscibility in metamorphic C–O–H–salt fluids is crucial for driving the upward CO₂ transport from the deep crust of active orogens³³. This process is particularly facilitated by earthquake-induced brittle fracturing of the crust. Overall, regional/contact metamorphism, crustal thermal state (e.g., geothermal gradients), and active structures (e.g., extensional tectonics) involved in source-to-surface transport of the mobilized crustal carbon through the extensional rift systems have close affinities with the underthrusting India, which could well reconcile geochemically constrained CO₂ origins and field-based observations of CO₂ fluxes.

Himalayan-Tibetan orogen as important carbon sources

We established the potential links between surface observations of active CO₂ degassing (i.e., origins and fluxes) and deep metamorphic decarbonation in the Himalayan-Tibetan orogen (Fig. 5). The combined geophysical and geo-tectonic responses to the Indian underthrusting, such as the collisional metamorphism that characterizes the Himalayan orogenic wedge^{23,50}, could stimulate massive crustal carbon mobilization. By efficient outgassing through rifts and normal faults^{11,12}, as well as in the Himalayan fold-and-thrust belt^{9,43}, the CO₂-rich fluids produced by metamorphic decarbonation reactions at crustal levels³³ represent remobilization of carbon that has been stored in crustal rocks over geological timescales.

Our estimation yielded a total CO₂ flux of $17.4 \pm 4.1 \text{ Mt yr}^{-1}$ for the TTYR, and by subtracting shallow carbon contributions, we obtained a deep CO₂ flux of $2.97 \pm 1.01 \text{ Mt yr}^{-1}$ (Supplementary Table 2). Notably, our

Fig. 5 | Conceptual model showing massive crustal carbon mobilization and emission driven by the underthrusting of India beneath Asia. Abbreviations are as in Fig. 1. The CO₂ emissions in southern and central TTYR are derived from metamorphic decarbonation at lower crustal and upper crustal depths, while those in the northern TTYR are supplied mainly by crustal fluids but show contributions from mantle fluids (up to 7–17%). The migration of mid-crustal channel flow toward the Himalayan erosional front is shown, which could be partially molten, serve as the heat source for the overlying carbon-bearing rocks (e.g., calc-silicate and marble) in the upper crust, and mobilize massive CO₂ from the crustal sources. The Himalayan gneiss domes and leucogranite plutons are shown as the geological evidence for the extension and melting of the Himalayan orogenic crust. The presence of CO₂ + H₂O fluids is expected for dehydration of the Indian lower crust above the inclined portion of the Main Himalayan Thrust²¹, which is likely to facilitate the metamorphic decarbonation reactions.



calculation methods only take into account the deep carbon fraction, which could yield conservative and relatively unbiased estimates of deep CO₂ fluxes for the Himalayan-Tibetan orogen. Further extrapolation of our estimates from the TTYR to seven major near N-S striking rifts and other smaller-scale normal faults in the northern Himalaya and southern Tibet gave rise to a total deep CO₂ output of $37 \pm 21 \text{ Mt yr}^{-1}$ (Methods and Supplementary Information). This estimate is comparable to mantle CO₂ fluxes from global mid-ocean ridges (MOR) if considering (i) recently reported MOR CO₂ fluxes of $\sim 58 \text{ Mt yr}^{-1}$ (ref. 57), and (ii) an updated mantle ³He flux from MOR ($527 \pm 102 \text{ mol yr}^{-1}$; ref. 58) that corresponds to a mantle CO₂ flux of $\sim 46 \text{ Mt yr}^{-1}$.

We emphasize that more work is required to reduce the scaling-up uncertainties in CO₂ flux estimation for large-scale active collisional orogens such as the Himalayas and Tibet. For example, water-gas interaction (e.g., partial exsolution and dissolution of gas in groundwater) and subsurface calcite precipitation could lead to underestimation of the deep CO₂ output at the surface^{24,59,60} and need to be considered in future work. The effect of such secondary processes on CO₂ flux estimation is still challenging to be quantitatively constrained so far. Nevertheless, our up-to-date estimate of deep CO₂ emissions from extensional tectonics indicates that the Himalayan-Tibetan orogen is a globally important carbon source. Furthermore, the estimated deep CO₂ output is conservative because the rifts and normal fault systems in more northerly and easterly tectonic units (e.g., Qiangtang Block and Songpan-Ganzi Block) of the Tibetan Plateau were not included in our calculation (Methods). It is thus plausible that deep CO₂ output of the Himalayan-Tibetan orogen would be comparable in magnitude with the mantle CO₂ output from global MOR systems, even in case of comparison with the CO₂ flux estimate for the MOR ($\sim 97 \text{ Mt yr}^{-1}$) based on ³He flux of $1000 \pm 250 \text{ mol yr}^{-1}$ and CO₂/³He ratio of 2.2×10^9 (ref. 61).

Role of orogenic CO₂ emissions in global carbon cycle

By linking geochemically constrained CO₂ origins and field-based observations of CO₂ fluxes along a rift-related profile following the direction of Indian underthrusting, we demonstrate that geophysical and geo-tectonic processes in response to the underthrusting of India beneath Asia could stimulate the release of huge amounts of crustal carbon from Earth's largest active collisional orogen. Importantly, the outgassed CO₂ in the Himalayan-Tibetan orogen is primarily of crustal origins, with its total output ($\gg 37 \pm 21 \text{ Mt yr}^{-1}$) in comparable magnitude with the mantle CO₂ fluxes ($46\text{--}97 \text{ Mt yr}^{-1}$; Supplementary Fig. 6) from global MOR systems. We suggest that the massive crustal CO₂ emissions in tectonically active

collisional orogens must be considered equally important as the mantle CO₂ from MOR^{57,58,61} and intra-continental settings such as the East African rift^{53,62,63}. To thoroughly understand the framework of solid Earth degassing and carbon cycling feedbacks, an important theme of future work, tectonic CO₂ emissions from active collisional orogens should be combined with volcanic CO₂ outputs⁶⁴ (e.g., MOR⁵⁷ and subduction zones^{60,65}) to give an unbiased average $\delta^{13}\text{C}$ value of deep carbon for the global carbon isotope mass balance model^{66,67}. Our findings would provide new insights into how continental collision dynamics could influence the mobilization and emission of crustal carbon over an active orogen-wide scale, which is crucial to our understanding of solid Earth degassing and deep carbon cycling processes from a whole-Earth plate tectonic point of view.

Methods

A series of near N-S striking extensional rifts have developed in northern Himalaya and southern Tibet (Fig. 1a) as a result of orogen-parallel extension since Miocene time²². These rifts are roughly spaced by a distance of $\sim 150\text{--}200 \text{ km}$ and are large-scale focal zones of modern hydrothermal activities (hot and boiling springs, geysers, steam fissures, and fumaroles; refs. 11–13,34), as well as diffuse soil CO₂ emissions. Centering in the major seven rift systems, the TTYR consists of several kinematically linked rift segments, which could be divided into the southern, central, and northern TTYR (Fig. 1b). Our field campaign in the TTYR was conducted from May to July 2021, covering the entire rift from the northern Himalaya to southern Tibet, to quantify the CO₂ outgassing fluxes and decoding the origins of the CO₂-bearing fluids.

Sample collection and analysis

Temperature, pH, and electrical conductivity (EC) were measured for the main spring outlets of thermal springs in the field (Supplementary Data 1). Free gas samples ($n = 16$) from bubbling springs were collected into low-He diffusivity glass containers by water displacement method. Standard procedures of sample collection in the field and timely laboratory analysis after sampling were adopted to minimize air contamination. Soil gas samples ($n = 24$) were collected from depths of 20–90 cm in soils of the measurement sites for analysis of CO₂ concentrations and carbon isotopic compositions (Supplementary Fig. 7). Gas chemistry, He, and C isotope analysis were conducted at Oil and Gas Research Center, Northwest Institute of Eco-Environment and Resources, Chinese Academy of Sciences. Major gas components (such as CO₂, N₂, CH₄, O₂, and Ar) were determined using a MAT 271 mass spectrometer. About 1 mL of gas was extracted from the

sample bottle using a syringe and then injected into the sample entrance line connected to the mass spectrometer. Repeated analysis of air standard yields analytical error <2% for major gas species (N₂, O₂, Ar, and CO₂). Carbon isotopic compositions in CO₂ of thermal spring gases and soil gases were analyzed by Agilent 6890 gas chromatograph (GC) coupled to ThermoFinnigan Delta Plus-XP Isotope Ratio Mass Spectrometer (IRMS). An analytical uncertainty of GC-IRMS is < ±0.3% based on repeat measurements of gas standards. Helium isotopes were determined by a Noblesse noble gas mass spectrometer equipped with a two-stage gas separation line and purification system. Before measurement by the mass spectrometer, the free gases (~500 μL) were purified by exposure to a spongy titanium furnace heated to 800 °C, removing most active gases (e.g., H₂O, O₂, N₂, and CO₂). H₂ in the gases was eliminated by a Zr-Al getter running at room temperature. Next, the purified noble gases were trapped by a cryogenic trap filled with activated charcoal. He and Ne were released into the mass spectrometer at a cryogenic trap temperature of 70 K. Then, ⁴He, ²⁰Ne, and ²²Ne were examined with a Faraday collector, and ³He and ²¹Ne were analyzed with an electron multiplier. The stability of the Noblesse Mass Spectrometer was monitored by repeated analysis of standard air before and after analyzing the sample every day. The air collected from the top of Gaolan Mountain, South of Lanzhou City, was analyzed to meet laboratory standards during routine measurement. Repeated analysis of the standard air yields an analytical error of less than 1.5% for the ³He/⁴He value. The analytical data of bubbling gases from thermal springs are summarized in Supplementary Data 1. For comparison, gas geochemical data reported in the literature are compiled in Supplementary Data 2. In addition, volume fraction and carbon isotopic composition of CO₂ in soil gases are summarized in Supplementary Data 3.

Quantifying diffuse soil CO₂ emissions and uncertainty assessment

A total of 939 points of in-situ diffuse soil CO₂ fluxes (Supplementary Data 4) were obtained from the TTYR between May and July 2021, following the accumulation chamber method⁶⁸. Specifically, soil CO₂ fluxes were measured by a West Systems LI820 infrared CO₂ detector with a CO₂ detection range of 0–2% and an accuracy of 4%. An open-bottomed accumulation chamber with a volume of $6.19 \times 10^{-3} \text{ m}^3$ was placed directly on the soil surface to form a tight seal (Supplementary Fig. 7), and the CO₂ flux was calculated by the linear segment that represents the natural accumulation of CO₂ concentration with time in the chamber. To minimize the biological influence on accumulated CO₂ in the chamber, we selected measurement points in non-vegetation areas or removed the vegetation cover and the uppermost few centimeters of the soils. During our fieldwork, there was little precipitation in the TTYR, showing typical meteorological features in the Tibetan Plateau that are suitable for soil CO₂ flux measurements considering the influence of soil moisture on CO₂ outgassing via micro-seepage. To constrain the distribution and magnitude of CO₂ fluxes in the rift, we employed a sampling grid of 20–50 m in hydrothermal fields. For active faults, the measurement spacing was 50–500 m for observational profiles of soil CO₂ fluxes that are generally perpendicular to the fault strike (Fig. 1 and Supplementary Fig. 8). Moreover, in areas with less assessable topography and transportation limitations, the spacing between adjacent soil CO₂ profiles may extend up to 5 km. The measurement points were selected depending on accessibility, geomorphic expression of the fault trace, and/or natural environmental conditions.

Diffuse CO₂ degassing from soils is commonly fed by different sources, such as biological carbon, hydrothermal carbon, and their mixture⁶⁹. To identify different populations in the soil CO₂ flux data, we followed the statistical method proposed by Sinclair⁷⁰. As shown by examples of soil CO₂ emissions from hydrothermal fields (Supplementary Fig. 4), the background populations of soil CO₂ fluxes (i.e., biogenic CO₂) could be well distinguished from the endogenic populations (i.e., hydrothermal CO₂). Average fluxes with 95% confidence intervals were calculated for the identified populations of hydrothermal fields following ref. 70. (Supplementary Table 3). High flux populations represent CO₂ derived from

endogenic sources, such as those observed in volcanic-hydrothermal systems⁶⁹. Low flux populations refer to CO₂ associated with biological activities in the soils, while intermediate populations are commonly interpreted as a mixture of biogenic and hydrothermal carbon. These average soil CO₂ fluxes ($F_{\text{hf,CO}_2}$ in $\text{g m}^{-2} \text{ d}^{-1}$) were then combined with estimated degassing areas ($S_{\text{hf,degassing}}$ in km^2) to obtain a CO₂ emission ($E_{\text{hf,CO}_2}$ in Mt yr^{-1}) for each hydrothermal field and the resulting CO₂ flux were summed together. The diffuse soil CO₂ emissions of $2.04 \pm 0.98 \text{ Mt yr}^{-1}$ for the hydrothermal fields in the entire TTYR (Supplementary Table 2).

For the profiles of soil CO₂ fluxes without visible hydrothermal manifestations, we classified the soil CO₂ flux data into two groups for southern, central, and northern segments of the TTYR (Supplementary Table 4): (i) background areas with comparable fluxes to biogenic values, and (ii) fault zone with higher average soil CO₂ fluxes (Supplementary Fig. 8). Considering the irregular spacing and sparse sampling used in regional surveys, we adopted the structural-area method proposed by Lee et al.⁵³ for estimating total CO₂ fluxes from fault zone and background area in the TTYR. To constrain spatial variations in CO₂ emission efficiency, we compared the average soil CO₂ flux of the fault zone profiles and background areas and found that diffuse soil CO₂ emissions are generally in the same magnitude among different segments of the TTYR. Specifically, the average soil CO₂ flux of the fault zone profiles is 36.3, 42.3, and $25 \text{ g m}^{-2} \text{ d}^{-1}$ for the southern, central, and northern segments of the TTYR, and that of the background area is 11, 8.6, and $10.4 \text{ g m}^{-2} \text{ d}^{-1}$ for southern, central and northern TTYR (see details in Supplementary Data 4). Therefore, the average soil CO₂ flux of the fault zones and background areas was multiplied by the area of soil degassing to estimate the corresponding total CO₂ output. The calculated soil CO₂ fluxes from the fault zone and background area were $1.40 \pm 0.53 \text{ Mt yr}^{-1}$ and $13.9 \pm 2.6 \text{ Mt yr}^{-1}$, respectively (Supplementary Table 2). The uncertainty in flux estimates is derived from the standard error of the average soil CO₂ fluxes. Detailed methods for estimation of soil degassing area and flux calculation (including equations and procedures) are provided in Supplementary Information. Finally, it should be emphasized that estimating CO₂ output for a rift such as the TTYR remains challenging, and assuming the measured flux data from a limited study area are representative of the entire rift is feasible at the current stage.

Carbon isotope mass balance models

For free gases from thermal springs, we combined He with C isotope systematics to calculate the proportions of different carbon source components using a ternary mixing model that involves the mantle (MORB), marine carbonate (CAR), and organic sediments (ORG) end-members³¹. Mantle, carbonate, and organic sediments are assumed to have CO₂/³He and $\delta^{13}\text{C}_{\text{CO}_2}$ values of 2×10^9 and $-6.5 \pm 2.5\text{‰}$, 1×10^{13} and $0 \pm 2\text{‰}$, as well as 1×10^{13} and $-30 \pm 10\text{‰}$, respectively (Fig. 2). This model indicates that the fraction of mantle carbon (M) + carbonate (C) is about 75–88%, and organic sediments (S) accounts for the rest fraction of carbon (12–25%; Supplementary Data 1). Notably, it is difficult to constrain the proportion of metamorphic carbon solely based on the model of Sano and Marty³¹, because the $\delta^{13}\text{C}$ value of metamorphic carbon could be close to that of either inorganic carbon or organic carbon depending on the protoliths of metamorphic decarbonation reactions^{71–73}.

For diffuse soil CO₂ emissions, the deep CO₂ of hydrothermal origins could migrate toward the surface through the porosity of rocks and fractures/faults, which is expected to mix with CO₂ derived from biogenic sources and the atmosphere. Combined with the evaluation of the area of soil emanation (Supplementary Methods), the CO₂ outgassing fluxes in background areas, fault zones, and hydrothermal fields were estimated to be 13.9 ± 2.60 , 1.40 ± 0.53 , and $2.04 \pm 0.98 \text{ Mt yr}^{-1}$, respectively, including CO₂ from deep and shallow sources. To avoid overestimation of deeply-sourced CO₂ fluxes, we subtracted shallow carbon (e.g., biogenic CO₂) fractions from the total CO₂ output based on a carbon isotope mass balance model based on carbon isotopes ($\delta^{13}\text{C}$) and abundances of CO₂ in soil gases (Supplementary Fig. 9). The hydrothermal source is assumed to have 100 vol.% CO₂ and a $\delta^{13}\text{C}$ value of -2‰ (constrained by a scenario of batch

equilibrium CO₂ degassing from a DIC end-member with δ¹³C value of +2‰, i.e., equilibrium fractionation value Δ¹³C_{eq} = +4‰), while the air has 0.04 vol.% CO₂ and a δ¹³C value of −8‰. For the biogenic CO₂ end-member, we used a δ¹³C value (−23.6‰) reported for modern soils of the Tibetan Plateau⁷⁴ and assumed pure CO₂ production (i.e., 100 vol.% CO₂) for biogenic source components. This ternary mixing model yielded average deep CO₂ proportions of 62% and 33% for soil gas seeping in hydrothermal fields ($f_{\text{hf,deep}}$) and fault zones ($f_{\text{fz,deep}}$), respectively (Supplementary Data 3). In contrast, deeply-sourced CO₂ was remarkably low (9%) in soils of the background areas ($f_{\text{ba,deep}}$). Deep CO₂ fluxes from diffuse soil in the TTYR were calculated by summing the deep CO₂ fluxes of the hydrothermal fields ($E_{\text{hf,CO}_2} \times f_{\text{hf,deep}}$), fault zones ($E_{\text{fz,CO}_2} \times f_{\text{fz,deep}}$), and background areas ($E_{\text{ba,CO}_2} \times f_{\text{ba,deep}}$). The results show that deep CO₂ fluxes of soil seeping in the TTYR are 1.26 ± 0.60 Mt yr^{−1} in hydrothermal fields, 0.46 ± 0.18 Mt yr^{−1} in fault zones, and 1.25 ± 0.23 Mt yr^{−1} in background areas. Taken together, total deep CO₂ output through soils would be 2.97 ± 1.01 Mt yr^{−1} for the TTYR (Supplementary Table 2).

CO₂ flux extrapolation to the extensional tectonics

To extrapolate our measurements to extensional tectonics (mainly rifts and normal faults) of the Himalayan-Tibetan orogen, we compiled basic geological parameters such as the number of hydrothermal fields ($n = \sim 650$ in the China part of the Himalayan-Tibetan orogen¹³) and total area of major seven near N-S striking rifts (fault zone = ∼545 km²; background area = ∼13,563 km²) in northern Himalaya and southern Tibet and that of other smaller scale normal faults (∼337 km²) in the same region (Supplementary Table 5). A detailed procedure of the CO₂ flux extrapolation method was given in Supplementary Information. The estimated total deep CO₂ flux is 37 ± 21 Mt yr^{−1} for the rifts and normal faults in the northern Himalaya and southern Tibet. Most of the deep CO₂ output is accounted for by diffuse soil emanations from hydrothermal fields (Supplementary Table 6). Note that rift and normal fault systems in more northerly and easterly tectonic units (e.g., Qiangtang Block and Songpan-Ganzi Block) of the Tibetan Plateau and its surroundings are beyond the scope of this study and are not considered in flux extrapolation also due to the lack of observational data. Therefore, our estimate of deep CO₂ flux represents a conservative value of the entire rifts and normal fault systems of the Himalayan-Tibetan orogen. To reduce the error of deep carbon flux estimates, more work is required to investigate other rifts and normal faults in the Himalayan-Tibetan orogen.

Data availability

All data generated or analyzed in this study are provided in the online version of this article and Supplementary Information. Supplementary Data 1 to 4 (Excel spreadsheets) have been deposited at Zenodo (<https://doi.org/10.5281/zenodo.11125483>).

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Author contributions

S.X. and M.Z. conceived the idea and led the study. W.L., M.Z., L.C., and X.Z. collected samples and conducted in-situ CO₂ flux observations. W.L. and Y.L. (Yi Liu) prepared and analyzed all samples. W.L., M.Z., and Y.L. (Yi Liu) compiled data, performed geochemical modeling, and calculated CO₂ fluxes. M.Z. and W.L. wrote the manuscript with inputs from S.X., Y.S., X.Z., Y.L. (Ying Li), L.Z., Y.-C.L., and C.-Q.L. All authors contributed to data interpretations and commented on the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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