Warming and monsoonal climate lead to large export of millennial-aged carbon from permafrost catchments of the Qinghai-Tibet Plateau

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Abstract
Permafrost carbon pool destabilization causes substantial fluvial export of soil carbon, yet the export patterns and magnitudes are not well understood. Here we investigated the radiocarbon ($^{14}$C) in dissolved organic and inorganic carbon (DOC and DIC, respectively) exported from a mid-sized river in the central Qinghai-Tibet Plateau (QTP) permafrost region. We utilized the radiocarbon dating technique to reveal the ages of riverine dissolved carbon and a statistical model to partition the riverine carbon from different age categories. DOC and DIC showed bomb-depleted $^{14}$C signatures corresponding to millennial ages. Seasonally, $^{14}$C-depleted DOC and DIC ages were associated with active layer thaw and flow path deepening. Spatially, older DOC and DIC were found in the valley sites correlated with warmer permafrost and higher groundwater flow. Further, isotopic mixing models suggested that 83 ± 27% of riverine DOC was derived from active layer and permafrost layer aged carbon. DIC export was comprised of a smaller portion of aged carbon (47.3 ± 2.6%) but a much larger flux of aged carbon due to higher annual DIC export. Interestingly, approximately 56% of annual aged DOC and DIC were exported in the short summer season (July to September). The monsoon climate-induced overlap of high discharge and maximum active layer thaw depth in summer enhanced the remarkably rapid fluvial export of millennial-aged carbon. Annual aged carbon yields in YRSR (275 ± 90 and 1661 ± 91 kg km$^{-2}$ yr$^{-1}$ for DOC and DIC, respectively) are much larger than those of Kolyma River (160 ± 89 and 234 ± 105 kg km$^{-2}$ yr$^{-1}$ for DOC and DIC, respectively). These results suggest a unique old carbon loss pattern in the QTP permafrost region compared to higher latitude permafrost regions with a non-monsoonal climate. As climate warms, more old carbon export is expected, which may affect the permafrost carbon pool and the river biogeochemical processes.

1. Introduction
The vast permafrost carbon pool in cryosphere regions is being mobilized due to climatic warming-induced permafrost thaw and active layer deepening (Schuur et al 2008, 2015, Yang et al 2010a, Turetsky et al 2019). Fluvial export is one of the most important pathways for permafrost carbon loss (Plaza et al 2019, Wild et al 2019) due to a concurrent change of the hydrology and biogeochemistry in permafrost regions (Frey and McClelland 2009, Vonk et al 2015, Walvoord and Kurylyk 2016). Although old carbon loss from thawed permafrost soil has been demonstrated in many regions (Schuur et al 2009, Feng et al 2013, Wang et al 2018a), both old and young carbon were found in rivers draining permafrost landscapes (Benner et al 2004, Guo et al 2007, Qu et al 2017, Drake et al 2018, Dean et al 2018, Wang et al 2018b, Wild et al 2019), and currently the overall response of the lateral export of carbon due to permafrost
warming remains unclear. Dissolved organic and inorganic carbon (DOC and DIC, respectively) are two main forms of carbon exported by permafrost rivers draining permafrost (Striegel et al. 2005, Dornblaser and Striegel 2015, Song et al. 2020), but their sources and export patterns are still inadequately constrained, especially for the alpine permafrost region.

The Qinghai-Tibet Plateau (QTP) has the largest alpine permafrost distribution in the world with a permafrost area of 1 060 000 km$^2$ (Zou et al. 2017). The QTP permafrost stores approximately 12.7 Pg C soil organic carbon (Zhao et al. 2018) and 15.2 Pg C soil inorganic carbon storage (Yang et al. 2018b) in the top 1-meter of soil. Compared to the Arctic permafrost, the QTP permafrost is generally warmer and has thicker and drier active layers (Hinkel and Nelson 2003, Zhang et al. 2005, Wu and Zhang 2010). The monsoonal climate over the QTP can bring both high temperature and rainfall in the summer (Tang and Reiter 1984, Yao et al. 2013), which allows the maximum active layer thaw depth period to overlap with the period of high discharge (Wang et al. 2009, Wu and Zhang 2010). This may enhance the carbon export from deeper thawed soil layers during summer. In contrast, Arctic rivers export the majority of water and carbon during the spring freshet period (Raymond et al. 2007, Vonk et al. 2015). In the Arctic permafrost region, the active layer thaw depth reaches its maximum during late summer accompanied with aged carbon release (Vonk et al. 2015), but the lower discharge in this period compared to spring freshet may limit the older carbon export. These differences may cause different aged carbon export patterns between QTP alpine permafrost and Arctic permafrost. Warming-induced permafrost thaw and active layer seasonal fluctuations have been mobilizing the QTP carbon pools and causing CO$_2$ or CH$_4$ emission (Mu et al. 2016, 2017b) and fluvial carbon export (Qu et al. 2017, Mu et al. 2017a, Song et al. 2019). However, the current understanding of aged carbon loss through rivers across the QTP is incomplete. The permafrost-derived versus active layer-derived carbon age proportions in QTP riverine carbon export are still unknown.

Here we investigated the DOC and DIC ages and source compositions from a typical river from the QTP permafrost region by utilizing radiocarbon ($^{14}$C) measurements in DOC and DIC at different seasons. The $^{14}$C dating technique is powerful and has been applied to various rivers to distinguish carbon sources and processes (Raymond and Bauer 2001b, Raymond et al. 2007, Wild et al. 2019). Aged DOC in Arctic rivers was hard to detect due to a large contribution from sources with significant amounts of bomb carbon (Neff et al. 2006, Raymond et al. 2007, Aiken et al. 2014) and the preferential mineralization of old organic carbon (Drake et al. 2015, Mann et al. 2015, Spencer et al. 2015). Conversely, aged DOC in the QTP rivers was detected in recent studies (Qu et al. 2017, Wang et al. 2018a), which makes it possible to link the active layer deepening to old carbon export in QTP rivers and partition the carbon sources from different ages. This study aimed to reveal the age variabilities of DIC and DOC, apportion their sources from different carbon pools, and assess the potential causes and implications of the aged carbon export by sampling rivers with variation in permafrost thermal characteristics.

2. Methods

2.1. Site description, field observation, and river sampling

The Yangtze River source region (YRSR) lies within the central QTP permafrost region (figure 1). YRSR has a drainage area of 1.38 × 10$^5$ km$^2$ and a mean elevation above 4000 m. The region has a continental monsoon climate characterized by warm and humid summers and cold and dry winters (Tang and Reiter 1984). The annual mean temperature and precipitation in YRSR are −1$^\circ$C and 387.7 mm, respectively. Due to the summer monsoon, 89% of the annual precipitation is received from May to September (Liang et al. 2008). The winter season of YRSR receives little precipitation and hence neither accumulated snowpack nor snowmelt freshet occurs during the spring. The dominant vegetation in YRSR is grassland, including alpine meadow and alpine steppe. The major soil type in YRSR is Gelisols according to the USDA classification.

The 6 catchments selected in this study are underlain by permafrost coverage ranges from 77% to 100% (figure S1, table 1). The thermal state of the permafrost was characterized by the mean annual ground temperature (MAGT), which was generated with the temperature at the top of permafrost model using satellite data (Obu et al. 2019). MAGT has been widely used as an indicator of permafrost thermal state (Romanovsky et al. 2010, Ran et al. 2018). Basin average MAGTs were calculated from the gridded MAGT data by Obu et al. (2019). The higher basin average MAGTs indicate warmer permafrost and a higher potential or extent of permafrost thaw. For QTP permafrost region, MAGT decreases with elevation, while higher MAGT areas also have thicker active layer (Qin et al. 2017). According to the average MAGT levels and site locations, we divided the sites as valley sites (i.e. ZMD, TTH, and YSP) and mountain sites (i.e. FM1, FM3, and FM5). The mountain and valley sites have basin average MAGT of −4.17 and −3.03 $^\circ$C, respectively. The active layer thickness in this area varies from 1.3 to 4.6 m (Wu and Zhang 2010), which is thickening due to climate warming. The active layer usually starts to thaw during April or May and starts to refreeze in late October, then remains frozen during the wintertime.
Figure 1. Maps of the study area, the Yangtze River source region (YRSR). (a) Map showing FM1, FM3, and FM5 watersheds. (b) Map showing TTH, YSP, and FM catchments are nested in YRSR (above ZMD hydrological station). The data of mean annual ground temperature (MAGT, °C) was obtained from a recently study (Obu et al., 2019). Maps were generated using ArcGIS 10.4.

Daily discharge of the investigated sites was obtained from the continuous water stage and velocity measurements from automatic hydrological stations (FM sites) or the hydrographic bureau of the Qinghai province (ZMD, TTH, and YSP). The typical hydrological year has two flood periods and three low-flow periods for YRSR above ZMD station (figure S2) due to the collective impacts of seasonal active layer thaw and precipitation change (2009, Wang et al., 2011). The annual study period is divided into four seasons: spring period (April to June), characterized by initial thaw of active layer and rising streamflow; summer period (July to September), when active layer is fully thawed and rivers have high-flow; autumn period (October to November), during which active layer refreezes and streamflow falls; and winter period (December to the next March), when the entire active layer is frozen and the rivers have a minimum or no streamflow.

To identify the runoff composition differences among the upstream and downstream basins, the flow duration curve (FDC) analyses were performed for each basin using daily discharge. FDC is the relationship between the frequency and magnitude of discharge (Smakhtin, 2001, Castellarin et al., 2004). A FDC is constructed by reordering the discharge time series values from high to low flow and assigning discharge values to class intervals and counting the occurrence numbers within each class interval (Smakhtin, 2001). A specific frequency discharge, Qp, indicates the streamflow at a given frequency p. For example, Q90 refers to the discharge at the 90% exceedance probability, which is often used as a proxy for groundwater flow (Smakhtin, 2001). The FDCs were calculated with the software FDC v2.1 (https://hydrooffice.org/Tool/FDC). After obtaining the FDCs (figure S3), we calculated the Q90/Q50 ratios to represent the portion of...
streamflow that originated from groundwater storage (Smakhtin 2001). The valley sites have a much higher average Q90/Q50 ratio (0.23) than the mountain sites (0.02), suggesting more deep groundwater flow from the sub-permafrost aquifer in valley sites.

The river water sampling was conducted during the open water seasons of 2017 in 6 sites across the YRSR (table 1, figure 1). The sampling dates for carbon concentration and radiocarbon analyses can be found in table S1 and table 2, respectively. Water samples were filtered immediately after sampling through pre-dried glass fiber filters (0.45 µm pore size) in the field. After filtration, ~120 ml of each water sample was split and gently filled into several pre-rinsed high-density polyethylene bottles with tight-fitting caps for different analysis purposes. The samples were stored under frozen and dark conditions until analysis to avoid microbial activities.

### 2.2. Isotopic analyses of δ¹³C and Δ¹⁴C

We used an Elementar Vario TOC Select Analyzer (Elementar, Germany) to analyse the DOC and DIC concentrations (figure S2, table S1). The δ¹³C and Δ¹⁴C values were determined by the following method as described before (Raymond and Bauer 2001a, Raymond et al. 2007). Briefly, samples were converted to CO₂ before isotopic analyses. For DOC, water samples were placed into reactors and acidified to pH ≈ 2 with 85% phosphoric acid and sparged with ultra-high purity helium gas for 10 min to remove air gases. Then the sample was acidified to pH ~2 with needle-injected 85% phosphoric acid and sparged with ultra-high purity helium gas for another 10 min to collect DIC-originated CO₂. The CO₂ gas was purified and collected with the aforementioned method. The CO₂ from DOC and DIC were divided into two portions at a 10:1 volume ratio. The smaller portions were used to determine δ¹³C-DIC and δ¹³C-DIC by the mini carbon dating accelerator mass spectrometry system (MICADAS) at the Laboratory of Ion Beam Physics, ETH Zürich. The radiocarbon data are reported as fraction modern (Fm) as described before (Stuiver and Polach 1977, Reimer et al. 2004). The ¹⁴C ages were expressed as years before present (BP), while the ‘present’ here means the year 1950. Radiocarbon data is also converted to Δ¹⁴C values as per mil (%o).

Flux-weighted mean Δ¹⁴C data were calculated with the corresponding DOC or DIC fluxes of the same sampling time (Hossler and Bauer 2013):

$$I_s = \frac{\sum_i (F_i(t) C_i(t) F_i(t))}{\sum_i (C_i(t) F_i(t))}$$

where $I_s$ is the flux-weighted isotopic signature for site $s$; $I_i(t)$ is the isotopic signature for site $s$ measured on sampling date $t$; $C_i(t)$ is the carbon concentration for site $s$ measured on sampling date $t$; $F_i(t)$ is the mean daily discharge for site $s$ measured on sampling date $t$.

### 2.3. Carbon flux estimate from LOADEST

Total DIC and DOC export fluxes for YRSR were calculated with discharge and concentrations using

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**Table 1.** Sample sites and basin average properties of the study basins. Permafrost coverage data is from recent published Qinghai-Tibetan plateau permafrost map (Zou et al. 2017). The percentage coverage of permafrost and mean elevation were calculated with ArcGIS 10.4. Mean basin slope was calculated as the elevation difference divided by mainstream length. Basin average MAGTs were calculated from the grided MAGT data by Obu et al. (2019). Q90 and Q50 are the discharge at the 90% and 50% exceedance probability, respectively. The Q90/Q50 ratio denotes the portion of streamflow that originated from groundwater storage.

| Sites Name (Abbreviation) | Area (km²) | Permafrost % | Basin mean Elevation (m) | Basin slope | Basin average MAGT (°C) | Flux-weighted Δ¹³C-DIC (%o) | Flux-weighted Δ¹⁴C-DIC (%o) |
|--------------------------|------------|--------------|--------------------------|-------------|-------------------------|----------------------------|----------------------------|
| Zhimenda (ZMD)           | 138 000    | 77           | 4761                     | 0.0024      | -2.3                    | 0.255                      | -430.7                     | -448.4                     |
| Tuotuohu (TTH)           | 16 949     | 92           | 4947                     | 0.0045      | -3.1                    | 0.200                      | -456.0                     | -454.6                     |
| Yanshiping (YSP)         | 4538       | 98           | 5152                     | 0.0057      | -3.7                    | 0.230                      | -441.3                     | -666.5                     |
| Fenghuo Mountain #1 (FM1)| 117.0      | 100          | 4925                     | 0.0191      | -4.2                    | 0.029                      | -135.3                     | -291.8                     |
| Fenghuo Mountain #3 (FM3)| 56.4       | 100          | 4955                     | 0.0216      | -4.4                    | 0.036                      | -229.2                     | -395.4                     |
| Fenghuo Mountain #5 (FM5)| 6.8        | 100          | 4863                     | 0.0408      | -3.9                    | 0.007                      | -208.8                     | -326.0                     |
Table 2. Concentrations, \(\delta^{13}C\), \(\Delta^{14}C\), and radiocarbon ages (years BP) of DIC and DOC across YRSR sites.

| Sites | Date (2017/m/d) | DIC (mg/L) | \(\delta^{13}C\)-DIC (‰) | \(\Delta^{14}C\)-DIC (‰) | Age ± sd (years BP) | DOC (mg/L) | \(\delta^{13}C\)-DOC (‰) | \(\Delta^{14}C\)-DOC (‰) | Age ± sd (years BP) |
|-------|-----------------|------------|-------------------|------------------|------------------|-------------|------------------|------------------|------------------|
| ZMD   | 2/15            | 20.76      | 1.8               | -553.4 ± 10      | 6411 ± 81        | 1.00        | -27.6           | -320 ± 9.8       | 3033 ± 79        |
| ZMD   | 6/15            | 27.71      | -2.7              | n.d.             | n.d.             | 1.95        | -27.4           | -382.2 ± 9.5     | 3803 ± 77        |
| ZMD   | 7/30            | 26.91      | -1.9              | -460.3 ± 9.3     | 4889 ± 76        | 1.97        | -29.8           | n.d.             | n.d.             |
| ZMD   | 9/15            | 27.73      | -1.9              | -394.9 ± 8.9     | 3971 ± 72        | 4.39        | -28.1           | -491.3 ± 10.3    | 5364 ± 83        |
| ZMD   | 10/30           | 28.29      | -0.5              | -452 ± 9.1       | 4766 ± 74        | 1.38        | -21.1           | -211.4 ± 9.4     | 1843 ± 76        |
| ZMD   | 12/15           | 28.72      | 0.4               | -510.5 ± 10.1    | 5673 ± 81        | 1.92        | -24.9           | -369 ± 9.8       | 3633 ± 79        |
| TTH   | 5/28            | 12.81      | 5.5               | -576.2 ± 9.9     | 6831 ± 80        | 2.25        | -29.0           | -671.2 ± 12.1    | 8869 ± 98        |
| TTH   | 7/16            | 22.82      | -4.8              | -447.9 ± 8.5     | 4706 ± 69        | 1.27        | -24.5           | -387.8 ± 9.7     | 3876 ± 79        |
| TTH   | 8/25            | 21.93      | -2.4              | -432.7 ± 9.2     | 4488 ± 75        | 6.72        | -37.6           | n.d.             | n.d.             |
| TTH   | 10/9            | 34.68      | -0.6              | -510.8 ± 10.1    | 5678 ± 82        | 3.43        | -27.2           | -426.2 ± 10      | 4397 ± 81        |
| YSP   | 5/28            | 20.49      | -0.4              | -435.1 ± 9.9     | 4523 ± 80        | 2.53        | -25.9           | -53 ± 8.8        | 373 ± 72         |
| YSP   | 7/16            | 17.15      | -1.1              | -425.5 ± 9.5     | 4387 ± 77        | 2.71        | -31.6           | -805.6 ± 18.2    | 13 089 ± 147     |
| YSP   | 8/25            | 20.17      | -1.5              | -437.7 ± 7.8     | 4560 ± 63        | 2.16        | -32.3           | -766.1 ± 17.2    | 11 607 ± 140     |
| YSP   | 9/25            | 27.89      | -5.8              | -450.3 ± 10.1    | 4741 ± 82        | 2.16        | -30.9           | -684.8 ± 13.1    | 9209 ± 106       |
| FM1   | 5/26            | 8.53       | -2.5              | -337.8 ± 9.9     | 3246 ± 80        | 2.31        | -25.2           | -397 ± 10        | 4011 ± 81        |
| FM1   | 7/17            | 29.59      | -3.0              | -430.6 ± 9.6     | 4459 ± 78        | 2.04        | -25.9           | -283.3 ± 11.1    | 2610 ± 90        |
| FM1   | 8/28            | 34.85      | -6.7              | -121.5 ± 8.1     | 976 ± 66         | 5.39        | -29.8           | -290.6 ± 8.6     | 2693 ± 70        |
| FM1   | 10/9            | 31.46      | -3.9              | -392.6 ± 9.4     | 3939 ± 76        | 1.96        | -26.2           | -360.6 ± 9.5     | 3528 ± 77        |
| FM3   | 5/26            | 12.68      | -2.9              | -153.2 ± 9.3     | 1271 ± 75        | 3.10        | -28.0           | -448.8 ± 10.5    | 4719 ± 85        |
| FM3   | 7/17            | 21.68      | -2.0              | -324 ± 8.9       | 3080 ± 72        | 2.51        | -29.2           | -328.4 ± 9.8     | 3133 ± 79        |
| FM3   | 8/28            | 37.44      | -6.6              | -229 ± 8.5       | 2024 ± 69        | 5.28        | -29.0           | -395.9 ± 11.5    | 3983 ± 93        |
| FM3   | 10/9            | 31.15      | -2.9              | n.d.             | n.d.             | 1.38        | -27.9           | -311.1 ± 9.4     | 2929 ± 76        |
| FM5   | 5/26            | 8.26       | -4.3              | -180.9 ± 8.4     | 1538 ± 68        | 4.71        | -26.6           | -185.6 ± 8.8     | 1584 ± 71        |
| FM5   | 7/17            | 24.80      | -4.0              | -386 ± 9.4       | 3853 ± 76        | 2.92        | -22.4           | -222.3 ± 9.6     | 1965 ± 78        |
| FM5   | 8/28            | 35.91      | -6.8              | -201 ± 9         | 1737 ± 73        | 4.46        | -29.1           | -326.6 ± 9.5     | 3111 ± 77        |
| FM5   | 10/9            | 36.50      | -0.6              | -382.1 ± 9.8     | 3802 ± 80        | 2.51        | -28.4           | n.d.             | n.d.             |

Note: sd is the standard deviations of the analytical uncertainty, n.d. means no data.

![Figure 2.](image)

Figure 2. \(\Delta^{14}C\) versus \(\delta^{13}C\) plots for DOC and DIC in YRSR and other basins across the globe. Carbon-14 and carbon-13 isotope data other than this study were obtained from a large global radiocarbon dataset (Marwick et al 2015) and recent studies (Smits et al 2017, Drake et al 2018, 2019, O’Donnell et al 2020, Blattmann et al 2019).

LOADEST model (Runkel et al 2004) that was integrated within the LoadRunner tool (version 1.2b, https://environment.yale.edu/loadrunner/). Briefly, both carbon concentration and daily discharge data were input into LOADEST. LOADEST can select the best regression model from 9 predefined regression models and use the best-fitted model to calculate the carbon flux. The model selections were based on the Akaike Information Criterion and Schwarz Posterior Probability Criteria (Runkel et al 2004). In this study, the fitted DOC and DIC models for ZMD have R² values of 0.981 and 0.958, respectively (table S2), demonstrating high goodness-of-fit of the models. LOADEST can output daily, monthly and annually flux values using maximum likelihood estimation (MLE), adjusted maximum likelihood estimation.
Figure 3. Flux-weighted mean $\Delta^{14}C$ values and corresponding radiocarbon ages for DOC (a) and DIC (b) in YRSR at different seasons. Error bars represent standard errors.

Figure 4. Comparisons of flux-weighted mean $\Delta^{14}C$-DOC (a) and $\Delta^{14}C$-DIC (b) and corresponding ages in mountain sites and valley sites. The diamond points are mean values.

2.4. Sources apportionment by isotopic mixing models

DOC source apportionment was determined with a Bayesian tracer mixing model (MixSIAR) using dual carbon isotopes ($\Delta^{14}C$ and $\delta^{13}C$). The $\Delta^{14}C$-DOC and $\delta^{13}C$-DOC end-members were obtained from the various published soil isotopic measurements from the QTP (Mu et al. 2014, Yu et al. 2017, 2018a, Wang et al. 2018b) (table S3). DOC was assumed from three potential source pools including (1) recent fixed modern organic carbon from topsoil (0–10 cm), with a mean $\Delta^{14}C$ value of 32 ± 23‰, corresponding to a few years to decades of age; (2) active layer that contains old carbon, with a mean $\Delta^{14}C$ value of −196 ± 130‰, corresponding to 1690 years; and (3) ancient carbon from permafrost soil, which were formed during Holocene for QTP permafrost (Jin et al. 2007), which has a mean $\Delta^{14}C$ value of −551 ± 128‰ and corresponding to 6370 years old. The active layer and permafrost soil $\Delta^{14}C$ end-member values are close to those recently used for the Siberia Arctic region (Wild et al. 2019). DIC sources were estimated using the $\Delta^{14}C$-DIC based on a simple two-source mixing model with MixSIAR. We postulated that DIC originated from two age categories: geogenic source from fossil carbonate dissolution with $^{14}C$-dead $\Delta^{14}C$ of −1000‰ (Clark and Fritz 1997, Marwick et al. 2015); and modern source ($\Delta^{14}C = 0‰$) from modern organic carbon mineralization, atmospheric CO$_2$, and/or silicate weathering. The isotopic data from different sources and the $\delta^{13}C$ and $\delta^{14}C$ data of DIC or DOC were used as input in the model. The Bayesian mixing models were solved with a Markov Chain Monte Carlo (MCMC) simulation approach using MixSIAR as integrated within the R package 'MixSIAR' (Stock et al. 2018). The uncertainties in source values and model errors were incorporated into the MixSIAR models. We used the $\delta^{13}C$ and $\delta^{14}C$ data of DIC and DOC in ZMD station to apportion the source compositions of the YRSR since ZMD is the outlet of the YRSR. The source compositions...
of the sub-basins were also solved with the respective sub-basin isotopic data.

3. Results

3.1. Millennial-aged DOC and DIC in YRSR
The YRSR consistently displays depleted $\Delta^{14}$C-DOC signatures with millennial ages across different sites and seasons (table 2 and figure 1). Across our study systems, the flux-weighted mean $\Delta^{14}$C-DOC ranges from $-666\%$ to $-292\%$, corresponding to $^{14}$C age from 9847 to 2707 years BP. Holocene DOC with an age of more than 10 000 years BP is found at the YSP site during summer. Averaging these flux-weighted means across sites gives a $\Delta^{14}$C-DOC of $-430\%$ and $^{14}$C age of 4899 years BP for DOC. Compared to $\Delta^{14}$C-DOC signatures across global rivers, $\Delta^{14}$C-DOC ages in YRSR are among the oldest (figure 2(a)).

Similar to DOC, $\Delta^{14}$C-DIC signatures are also bomb-depleted across the YRSR (table 2). The flux-weighted mean $\Delta^{14}$C-DIC ranges from $-456\%$ to $-135\%$ for the investigated sites, with corresponding $^{14}$C ages from 4846 to 1127 years BP (figure 1). The averaged flux-weighted $\Delta^{14}$C-DIC and $^{14}$C age for all sites are $-317\%$ and 3153 years BP, respectively. Compared to riverine DIC radiocarbon age from other regions of the world, $\Delta^{14}$C-DIC ages in YRSR are among the oldest (figure 2(a)).

Across the four seasons, flux-weighted mean $\Delta^{14}$C-DOC shows a sharp decrease from spring to summer (figure 3(a)). The decreasing $\Delta^{14}$C-DOC through seasons suggests that older carbon is being released when the active layer gradually thaws to deeper soil horizons with deeper flow paths. Flux-weighted mean $\Delta^{14}$C-DIC increases from spring to summer then declines rapidly from autumn to winter when active layer thaw and flow path become deeper (figure 3(b)). Overall, the consistent old riverine carbon across seasons and sites suggest relatively continuous sources of old carbon throughout the year.

Spatially, $\Delta^{14}$C-DOC and $\Delta^{14}$C-DIC ages are more depleted (older) in the valley sites (i.e. ZMD, TTH, and YSP) than the mountain sites (i.e. FM1, FM3, and FM5, see figure 1 and figure 4). The differences of carbon ages between valley and mountain sites are statistically significant. The older carbon in valley sites suggest that the release of old carbon is associated with warmer permafrost since the valley sites have higher MAGT. Besides, the DOC and DIC ages are negatively correlated with Q90/Q50 ratio (figure 5). The higher Q90/Q50 ratios in valley sites can increase the aged carbon export via deep groundwater flow (Barnes et al 2018). Overall the dissolved carbon ages are older in basins have overall warmer permafrost and higher groundwater flow.

3.2. Contributions of aged carbon components to DOC and DIC export
Quantifications of the relative contributions for DOC and DIC export from different carbon pools can further elucidate the patterns of aged carbon mobilization and export. For the entire YRSR, isotopic mixing model suggests that the old carbon from the active layer and ancient carbon from the permafrost layer contribute 45.2 ± 23.6% and 37.8 ± 13.3% to DOC export, respectively (figure 6(a)). The sub-catchments showed slightly different source compositions. The overall source proportions for DOC have a pattern of old > ancient > modern except TTH.
where ancient carbon is the largest composition (figure S4a). The DIC sources proportions consistently have higher modern carbon contributions than fossil carbon contributions. The valley sites have higher portions of fossil DIC than the mountain sites (figure S4b). Higher proportions of aged carbon in the valley sites (figure S4) are consistent with the bulk $\Delta^{14}$C-DOC and $\Delta^{14}$C-DIC signatures, which are correlated with warmer permafrost and more groundwater flow in the valley sites that bring higher proportions of aged carbon. To the best of our knowledge, this is one of the first efforts to apportion the aged carbon component in QTP riverine carbon export.

The DOC and DIC fluxes show pronounced seasonality with the highest fluxes occurring in the later summer for all the sites (figure S5). Annual DIC flux ($485 \pm 29$ Gg C yr$^{-1}$) from YRSR is over 10 times the DOC flux ($46 \pm 5$ Gg C yr$^{-1}$), both of which contain significant portions of old and/or ancient carbon (figures 6(d)–(e)). Across seasons, the majority of the annual DIC and DOC export occurs in the summer, regardless of the age categories (figures 6(b)–(c)). The modern DOC and DIC export fluxes are $7.8 \pm 5.9$ and $255.4 \pm 12.6$ Gg C yr$^{-1}$, respectively. The aged (i.e. old, ancient, or fossil) carbon export are $38 \pm 12$ and $229 \pm 13$ Gg C yr$^{-1}$, for DOC and DIC respectively. The DOC and DIC export for each age category were calculated based on the source portions and total carbon export. Summer had the highest aged carbon export flux among the four seasons. Together, DOC and DIC export 56% (151 Gg C) of annual aged carbon in summer.

4. Discussion

Permafrost carbon mobilization under climate warming and the subsequent ‘permafrost carbon-climate feedback’ (Schuur et al. 2015) are key issues in global change science. Many studies have revealed the permafrost carbon pool change in the terrestrial ecosystem (Ding et al. 2017, Plaza et al. 2019), while the lateral hydrological export of permafrost carbon is still elusive, especially for the alpine permafrost across QTP. We found that the DOC and DIC in YRSR have millennial ages, older than those of Arctic rivers. Sites with warmer permafrost and more groundwater flow tend to have older riverine DOC and DIC. We estimate that most of the aged DOC ($83 \pm 27\%$) originates from the active layer and permafrost layer leaching. A substantial portion ($47.3 \pm 2.6\%$) of DIC comes from fossil carbonates. Approximately 56% of the annual aged carbon export occurs during the short summer monsoon season. The aged carbon exports in QTP rivers offer an opportunity to improve our understanding of permafrost carbon pool vulnerability and evaluate aged carbon mobilization magnitude and patterns.

4.1. Riverine carbon age and warming

In permafrost regions, warming-induced seasonal active layer fluctuations, permafrost thaw, and sub-surface hydrology enhancement can release aged carbon to fluvial networks by lateral transport (Neff et al. 2006, Aiken et al. 2014, Wild et al. 2019). Aged
DOCSoc in QTP rivers from hundreds to thousands of years old has also been found in previous studies (Qu et al 2017, Wang et al 2018a). Although rare to see, very old DOC was also found in a northern Alaska river, where DOC age was 2170–4950 years BP (Guo et al 2007). The overwhelmingly old DOC in YRSR contrasts with most of the Arctic permafrost rivers where DOC is mainly young (Benner et al 2004, Raymond et al 2007, Wild et al 2019). The thicker and drier active layer in QTP can enhance deep aged carbon release with longer flow paths when the active layer thawed. The groundwater discharge in QTP rivers are likely to be more dominant than in Arctic rivers considering the thicker and drier active layer (Walvoord and Kurylyk 2016, Wang et al 2017), which can enhance aged carbon export from groundwater. The difference of the lability of aged DOC in the two regions may also cause the contrasting DOC age (Mann et al 2015, Wang et al 2018a). Old DIC was also detected in the Arctic (Drake et al 2018, O’Donnell et al 2020) and tropical (Mayorga et al 2005) rivers. As an integrator for multiple sourcing processes, riverine DIC is a mixture of various carbon ages including carbon from carbonate mineral, soil CO$_2$, and organic carbon. $\Delta^{14}$C-DIC was positively correlated with DOC concentration across seasons and sites ($R^2 = 0.34, p = 0.003$), which may indicate some degree of old DOC mineralization to DIC (Selvam et al 2017, Drake et al 2018).

At the seasonal timescale, the shift of carbon age in rivers is direct evidence of active layer thaw. Deeper active layer and permafrost layer generally hold older carbon sources. The gradual top-down thaw through seasons liberates more old carbon to the riverine carbon pool in late seasons. In summer and autumn, the thawed active layer allows deeper vertical infiltration and thereby aged carbon export from the entire thawed soil column by subsurface flow (Dean et al 2018, Barnes et al 2018). The frozen permafrost layer impedes deeper infiltration, which leads to the lateral hydrological export of thawed aged carbon. As a result, the DOC age increase through seasons. The decline of $\Delta^{14}$C-DOC (increase of $^{14}$C age) through the growing seasons was also found in Siberian rivers (Neff et al 2006, Wild et al 2019), Canadian Arctic rivers (Dean et al 2018), and a QTP stream (Wang et al 2018a). The leachates of old carbon from deep soil layer (Mu et al 2014) in groundwater may also supply old DOC during winter. The seasonal shift of $\Delta^{14}$C-DIC indicating the impacts of flow paths and DIC source change (Walvoord and Striegl 2007). The peak of DIC age in summer (figure 4) signaling the adding of young respired contemporary carbon CO$_2$ to river DIC pool via carbonate acid weathering. $\Delta^{14}$C-DIC declines rapidly from autumn to winter when active layer thaw and flow path become deeper.

Permafrost thaw is another process governing the older carbon release (Schuur et al 2009, Aiken et al 2014). The term permafrost thaw is used to describe the transition of permafrost layer to seasonally thawed active layer under warming, which should not be confused with the active layer thaw at seasonal timescale. Spatially, the higher MAGT and thicker mean active layer thickness in the valley sites allows aged carbon release in deeper soil layer during summer. A higher degree of permafrost thaw liberates the aged carbon at the top of permafrost layer with subsurface flow, which results in ancient DOC transport in rivers (Vonk et al 2015). As permafrost thaws, the release of older carbonates in deeper soil layers or deep aquifers can also increase the DIC age in rivers (Barnes et al 2018). In the valley sites where MAGTs are higher, the potential for permafrost thaw is likely to be higher. Thereby, the sub-permafrost aquifer can discharge groundwater flow as more flow paths open (Walvoord and Kurylyk 2016), resulting in higher Q90/Q50 ratios (figure 3). More deep groundwater flow can bring older aged carbon to river water (figure 5). In the mountain sites, sub-permafrost groundwater flow is limited as a result of lower MAGT, which is suggested by the younger carbon age than that in the valley sites (figure 4).

In addition to warming-induced active layer and permafrost thaw, we do not rule out other factors that can affect aged carbon sources. For instance, the long-range transport and deposition of black carbon from fossil fuel combustion (Li et al 2016, Yan et al 2019) may also fuel ancient organic carbon in river water via precipitation and runoff generation processes. The decomposition of thaw-released old soil organic carbon may produce $^{14}$C-depleted CO$_2$, which can weather minerals and enter river networks. Although we are unable to assess this effect on the DIC age due to the lack of soil $^{14}$C-CO$_2$ data (Vaughn and Torn 2018).

The proportions of carbon from permafrost and active layer can further disentangle how fluvial export affects the permafrost carbon pool (Wild et al 2019). The high aged DOC source proportions are in contrast to the Arctic rivers where modern carbon constituted the majority portion of DOC (Wild et al 2019). The dominant active layer old carbon source for DOC is consistent with molecular evidence that showed active layer leachates can be a major source for stream dissolved organic matter (Wang et al 2018a). The substantial proportions of DOC from permafrost layer suggest the current mobilization of deep permafrost soil carbon in central QTP.

4.2. Monsoon drives large export of millennial-aged carbon

The seasonality of aged carbon export is an important feature that integrates the impacts of aged carbon sources and hydrological processes. The high proportions of aged DOC and DIC export in YRSR suggest rapid exports of millennial-aged carbon annually. But how rapid and why so rapid? We compare the annual aged carbon export yield per unit...
Table 3. Aged carbon yield of YRSR compares to the Kolyma River. Values are means ± standard deviations. For Kolyma River, total and aged yields of DOC were obtained from table S9 of Wild et al (2019); total DIC yield was obtained from the table 1 of Tank et al (2012); Old DIC proportion (15.2 ± 6.8%) was calculated from Drake et al (2018) data. Runoff data of the Kolyma river was obtained from Shiklomanov et al (2018). The active layer thickness of YRSR is from Wu and Zhang (2010). The active layer thickness for Kolyma is from the Circumpolar Active Layer Monitoring Network data (https://www2.gwu.edu/~calco/).

| River      | YRSR       | Kolyma      |
|------------|------------|-------------|
| Drainage area (km²) | 138 000 | 526 000     |
| Runoff in 2017 (mm) | 125 | 294         |
| MAGT (°C) | −2.3 | −6.6        |
| Active layer thickness (m) | 1.3−4.6 | 0.3−1.1    |
| DIC yield (kg km⁻² yr⁻¹) | 3512 ± 209 | 1540 ± 200 |
| Fossil DIC yield (kg km⁻² yr⁻¹) | 1661 ± 91 | 234 ± 105  |
| DOC yield (kg km⁻² yr⁻¹) | 331 ± 36 | 1555 ± 553 |
| Old and ancient DOC yield (kg km⁻² yr⁻¹) | 275 ± 90 | 160 ± 89   |

The processes controlling a large export of aged carbon reported here has important implications for permafrost carbon mobilization, riverine biogeochemical cycles, and aquatic ecosystem processes. If a large portion of thawed aged carbon is hydrologically exported before its decomposition, then the gaseous carbon exchange with the atmosphere from thawed old carbon may be less than expected. Furthermore, the thawed aged carbon has been found to be microbially labile and therefore will lead to a higher riverine CO₂ and CH₄ efflux downstream areas of the fluvial networks, leading to translocated greenhouse gas emissions from permafrost carbon (Venk and Gustafsson 2013). This additional fluvial carbon emissions from translocated permafrost carbon can potentially enhance the current atmospheric CO₂ and CH₄ levels with carbon assimilated thousands of years ago. Finally, more old carbon in fluvial networks can cause biotic assimilation of old carbon and further affect the stream nutrient status and fish growth (Fellman et al 2014, O’Donnell et al 2020). The consequences of riverine exported old carbon on aquatic ecosystems need further research.

5. Conclusions

This study investigates the ages and source compositions of DOC and DIC in YRSR, a mid-sized river in the QTP. Bulk Δ¹⁴C-DOC and Δ¹⁴C-DIC signatures suggest millennial-age carbon in QTP rivers and streams. Older riverine carbon ages are associated with active layer thaw, more groundwater flow, and higher MAGT. The YRSR exports more aged dissolved carbon fluxes than the Kolyma River mainly due to the unique summer monsoon across the QTP. Our results suggest that the QTP permafrost carbon pool is being rapidly exported via hydrological pathways upon thaw. The unique old carbon loss pattern in QTP is different from the higher latitude permafrost regions with a non-monsoonal climate. Future works are needed to understand the long-term and large-scale fluvial mobilization mechanisms of alpine permafrost carbon pool under a changing climate.
The consequences of old carbon in rivers should also be investigated in future studies.

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Data availability statement

The carbon isotopic and concentration data in this study is available from the main text and supplementary information. Other data are available from the corresponding author upon reasonable request.

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