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RESEARCH ARTICLE

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Quantifying the glacial meltwater contribution to mountainous streams using stable water isotopes: What are the opportunities and limitations?

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Abstract

This study aims to determine the opportunities and limitations of using stable water isotopes to quantify the glacial meltwater contribution to mountainous streams. For this purpose, three partially glaciated catchments in the Swiss Alps were selected as the study area. In the three catchments, stable isotope analysis ($\delta^{18}O$ and $\delta^{2}H$) was conducted of the streams and the end-members that contribute to the stream discharge (glacial meltwater, rain, snow). The investigations revealed that the contribution of glacial meltwater to mountainous streams can be quantified using stable water isotopes if three criteria are met: (A) The snow meltwater contribution to mountainous streams must be negligible due to its highly variable stable isotope signature; (B) the groundwater input needs to be either insignificant during this snow-free period or the groundwater residence time must be short such that groundwater contribution does not delay the end-member signal arriving in the streams; and (C) the isotope signal of the glacial melt end-member needs to be distinct from the other end-members. One of the three investigated catchments fulfilled these criteria in August and September, and the glacial meltwater contribution to the mountainous streams could be estimated based on stable water isotopes. During this time period, the glacial meltwater contribution to the stream discharge corresponded to up to 85% ± 2% and to 28.7% ± 10% of the total annual discharge, respectively. This high glacial meltwater contribution demonstrates that the mountainous stream discharges in August and September will probably strongly decrease in the future due to global warming-induced deglaciation. Overall, this study demonstrates that many hydrogeological conditions need to be met so that stable water isotopes can be used to quantify the glacial meltwater contribution to mountainous streams. This highlights the challenges when using stable water isotopes for hydrograph separation and serves as a guide for future stable water isotope studies in mountainous regions.

KEYWORDS
discharge separation, glacial meltwater discharge, mountainous catchments, stable water isotopes

1 | INTRODUCTION

Mountainous regions provide crucial water resources for downstream valleys and can act as a buffer during meteorological droughts (Beniston et al., 2011; Rohrer et al., 2013). Mountainous water reservoirs are also of major importance for hydropower production since they are the main contributors to artificially dammed lake reservoirs being used to produce hydroelectric energy. Mountainous water reservoirs are drained by streams, whereby several 'end-members' contribute to the stream discharges including rain, snow, and glacial meltwater as well as groundwater representing a mixture of the former three end-members. There is a broad agreement that the relative contribution of rain, snow melt-, glacial melt- and groundwater to mountainous streams will change in the future due to global warming (Arnoux et al., 2020; Bolch et al., 2012; Bombelli et al., 2019; Bradley et al., 2006; D'Agata et al., 2018; Finger et al., 2012; Orlove, 2009; Patro et al., 2018; Puspitarini et al., 2020). To quantitatively predict the future discharge of mountainous streams during climate change, it is of major importance to gain knowledge about the current mountainous stream components. In particular, it is crucial to quantify the relative contribution of glacial meltwater to mountainous streams since this component will likely disappear in the future caused by global warming. Currently, it is estimated that about 119 million people worldwide live in catchments, in which glacial meltwater is responsible for at least 50% of the total discharge for at least 1 month per year (Schaner et al., 2012). Moreover, since glacial meltwater is a non-renewable resource, the quantification of the contribution of glacial meltwater to the discharge in mountainous watersheds is an essential part of climate change risk assessment and sustainable water management in glacierized catchments (Miller et al., 2012; Schaner et al., 2012; Viviroli et al., 2011).

Up to present, several methodological approaches have been used to estimate the relative contribution of glacial melt, snow melt- and rainwater to the total discharge of mountainous streams including glaciological approaches, hydrological balance methods, hydrological modelling, and hydro-chemical tracer methods (Frenierre & Mark, 2014). Most of these glacial meltwater studies were conducted in the Himalaya, the Alpine region, the Rocky Mountains, and the Andes, whereby the glaciological approaches, hydrological balance, and hydrological tracer methods were mainly applied in the Rocky Mountains, the Andes, and the Himalaya Region and the hydrological modelling approaches were primarily used in the Alpine and Himalaya region. Among the hydro-chemical tracer method studies, a high number of studies applied stable water isotopes to estimate the relative contribution of glacial meltwater to mountainous streams (Cable et al., 2011; Kong & Pang, 2012; Mark & Mckenzie, 2007; Nolin et al., 2010; Ohlanders et al., 2013; Pu et al., 2017; Wagner et al., 2021; Zongxing et al., 2015; Zuecco et al., 2019). However, all of these carried out studies encountered challenges in applying stable water isotopes such as a high isotopic variability of the end-members, especially of snow, or the input of groundwater representing a mixture of the different end-members, which complicates the quantification of the glacial meltwater contribution to mountainous streams. To overcome these challenges, some of the studies, which were conducted in dry regions, neglected the input of ground- or rain-water (Mark & Mckenzie, 2007; Ohlanders et al., 2013). Other studies combined the stable water isotope analysis with numerical simulations to increase the robustness of the results (Nolin et al., 2010; Wagner et al., 2021), considered the snow- and glacial meltwater as one end-member (Pu et al., 2017; Zongxing et al., 2015) or neglected the contribution of snow meltwater to mountainous streams completely (Kong & Pang, 2012; Yang et al., 2016). Despite these efforts, none of these studies evaluated systematically under which hydrological conditions stable water isotopes can be used to quantify the glacial meltwater contribution to mountainous streams. However, this information would be of particular importance to gain knowledge about the opportunities and limitations, of the stable water isotope method to quantify the input of glacial meltwater to mountainous streams, which is a prerequisite for future studies. In addition, the knowledge about when and where the stable isotope method can be applied is also crucial since the method provides several advantages compared to other methods including a low dependency on existing data, the possibility to capture temporal and spatial variations of the different contributions to total discharge, and the applicability from micro to mesoscale (Frenierre & Mark, 2014).

To address this research gap, this study aims to systematically determine the hydrogeological conditions under which the stable water isotopes can be used in mountainous regions to quantify the glacial meltwater contribution to streams. For that purpose, three partially glaciated Alpine watersheds were selected in the central Alpine region in Switzerland as study areas. In these three catchments, the opportunities and limitations of the stable water isotope method were evaluated by stable water isotope ($\delta^{18}O$ and $\delta^{2}H$) measurement in the three catchments between July 2019 and March 2020 combined with electrical conductivity (E.C.) and stream discharge analysis. The measurements provide detailed insight into the parameters and processes controlling the stable water isotope composition in Alpine streams and under what conditions stable isotopes can be used for discharge separation. Moreover, this study provides information on the continuous monitoring of mountainous catchments for the quantification of the glacial meltwater contribution to mountainous streams using stable water isotopes leading to a better validation of available modelling studies.

2 | MATERIALS AND METHODS

2.1 | Site description

The three selected catchment areas are located in the Gadmen Valley in the Central Swiss Alps and are named Steinwasser, Giglibach, and Wendenwasser (Figure 1). Moreover, they are all situated in the Aar massif, consisting of metamorphic Gneiss and Granites. In the Giglibach catchment, the Aar massif is dominated by the Erstfeld Gneiss-complex, while in the Wendenwasser and Steinwasser catchment, the Aar massif mainly consists of the Innertkirchen Migmatite (Swisstopo, 2023). In all three catchments, the Aar massif is partly overlain by moraine and talus material with various thicknesses. The

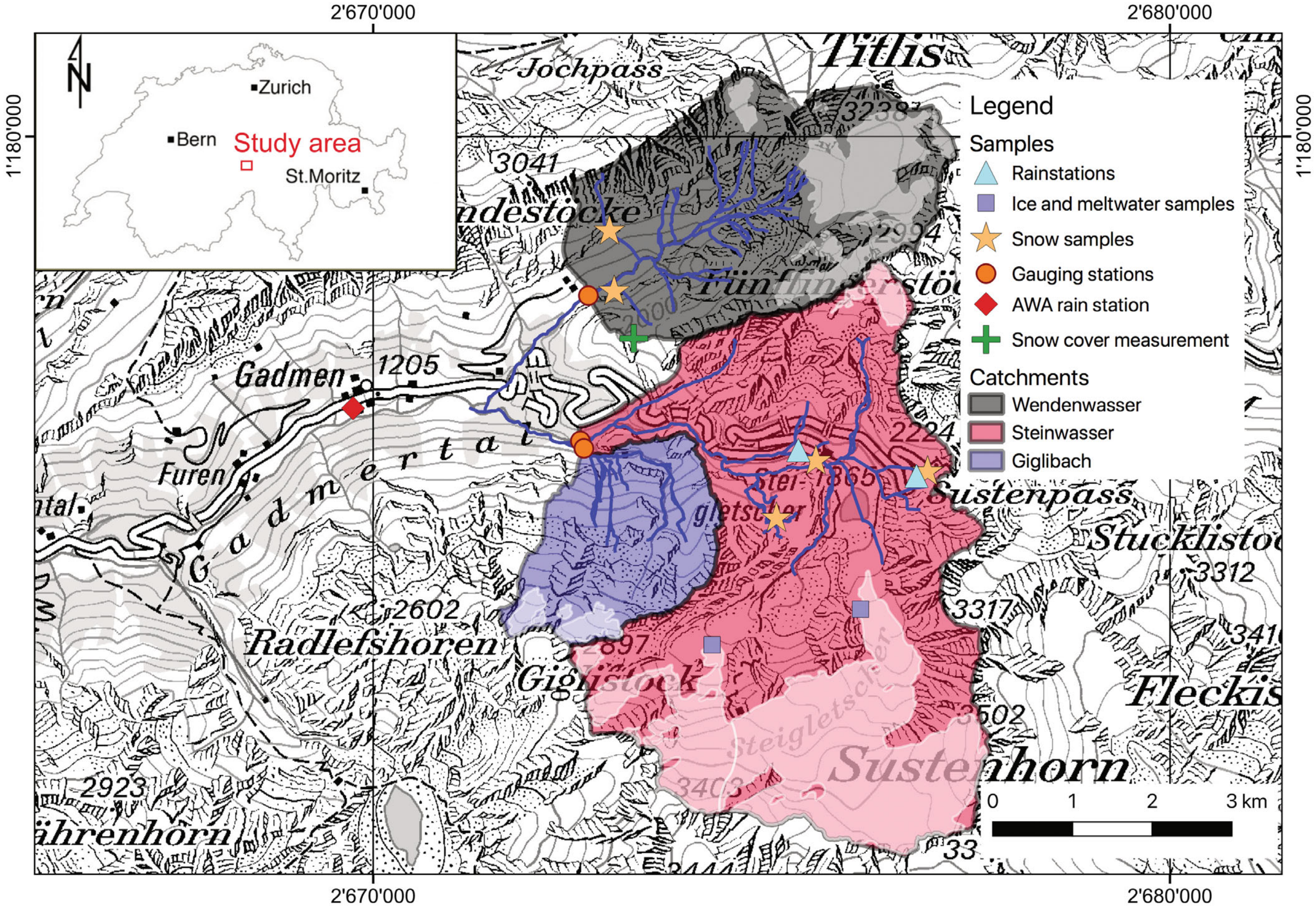

FIGURE 1 Areas of the Wendenwasser (pink), Steinwasser (grey), and Giglibach (blue) catchment, which are located in the Gadmen valley in the central part of the Swiss Alps and the sampling locations of the ice (pink squares), snow (orange stars) and rain (turquoise triangles) end-members as well as the catchment effluent measuring stations (orange circles). The red diamond represents the AWA rain station, whereas the green cross indicates the location of the Gschletteregg snow measuring station, and the white areas show the glaciated parts of the three catchments.

three catchments are located adjacent to each other (Figure 1) and show elevation ranges of 1425–3502 masl (Steinwasser), 1440–2897 masl (Giglibach), and 1539–3238 masl (Wendenwasser), respectively. The catchment areas differ in their size and degree of glaciation, whereby the Steinwasser catchment shows the largest size and degree of glaciation (24.2 km^2; 28.0%), followed by the Wendenwasser (11.2 km^2; 14.9%) and Giglibach catchment (4.9 km^2; 6.0%). The difference in sizes and degrees of glaciation areas provide the advantage that the opportunities and limitations of the stable isotope method to quantify the glacial meltwater contribution to the stream discharges can be evaluated under various conditions.

2.2 | Field measurements and sampling

2.2.1 | End-member sampling

To characterize the stable water isotope composition of the end-members (rain, glacial meltwater, snow) that contribute to the streams in the three catchment areas and to analyze their potential spatio-temporal evolution, each end-member was sampled and analysed several times at various locations in the three catchment areas (Figure 1). Groundwater was not considered as an end-member since it represents a mixture of the three end-members (rain, ice, snow) and only represents an additional flow path from the end-member sources to the streams supplementary to the surface run-off. Furthermore, during groundwater recharge and the migration in the groundwater system, the isotopic signal of the three end-members remains unchanged, justifying the consideration of snow, ice, and rain as the ultimate end-members. The rain end-member was sampled by using a 1B Palmex rain collector at the merging effluents of the Steinwasser and Giglibach catchment (1430 masl) and a Young rain collector (Nr. 55203) at the effluent of the Wendenwasser catchment (1542 masl; Figure 1). In addition, two Young rain collectors (Nr. 55203) were installed in the Steinwasser catchment at two different altitudes (1842 and 2210 masl) to capture potential changes of the δ^{18}O and δ^{2}H rain signal as a function of the altitude. The rain was sampled from the rain collectors in 16 days intervals between

July and October 2019 and stored in 300 mL plastic bottles prior to analysis. Besides, precipitation data was acquired from June 2019 to March 2020 from the precipitation measuring station in Gadmen (Figure 1) operated by the Office of Water and Waste (AWA) of the Canton of Bern, Switzerland (Cantonal Office of Waste and Water (AWA) of Bern, 2020). To determine the stable isotope signature of the snow end-member, 20 snow samples were taken at various locations in the three catchments covering an elevation range between 1541 and 2169 masl between February 2019 and March 2020 (Figure 1). During winter, the snow samples were collected by helicopters as the walking trails were not accessible. At each location, the snow was sampled vertically from the snow surface to the bottom of the snow cover using a Standard Federal Snow Tuber (SFST) resulting in a sample volume of 500 mL. After sampling, the snow was transferred into a wide-mouth PET bottle, which was closed immediately after filling to ensure that the snow melts in a closed container to avoid an evaporation-induced alteration of the sample prior to laboratory analysis. In addition to the taken snow samples, the thickness of the snow cover was measured every 10 min at the Gschletteregg measuring station at 2063 masl (Figure 3) being operated by the Swiss Institute for Snow and Avalanche Research (SLF). For measuring the isotopic signature of the ice-end-member, ice samples were taken from the glaciated areas in the three catchments using an ice pick between August and September 2019 (Figure 1). To ensure that the taken ice samples are representative for the meltwater component in the streams, solid ice as well as glacial meltwater samples were taken from the ablation zone of the glaciated area. To ensure that the solid ice samples are unaltered, the uppermost centimeters were scraped off and not used during sampling. Afterwards, the solid ice was sampled with an ice pick, and around 100 g of the solid ice was transferred into a plastic container and afterwards immediately closed to avoid that any melting-induced alteration takes place of the sample prior to the laboratory analysis. To sample the glacial meltwater, 300 mL plastic bottles were filled with the glacial meltwater and immediately closed to avoid an evaporation-induced change of the samples before they were analysed in the laboratory.

2.2.2 | Field station sampling and measurements

To measure the discharges and the electrical conductivity in the three catchments' effluents and for taking stream samples for stable water isotope analysis, three field measuring stations were deployed at the effluents of the three catchments (Figure 1). The measurements were conducted between June 2019 and March 2020, whereby the exact sampling period for each parameter and catchment effluent is provided in Table 1.

The stream discharges and the electrical conductivity were measured every 10 s and averaged over a 10 min interval during the monitoring periods. The stream discharges were determined via stream level measurements using the discharge/water level (P/Q) relationship, whereby the P/Q relation in the three streams was determined using the salt dilution method at various stream water levels (Wyss, 2020).

TABLE 1 Sampling periods for different parameters in catchment effluents.

Catchment	Parameter	Sampling period
Wendenwasser	Discharge	31 July 2019 to 21 March 2020
Wendenwasser	Electrical conductivity	13 August 2019 to 7 November 2019
Wendenwasser	Stable water isotopes ($\delta^{18}O$, $\delta^{2}H$)	31 July 2019 to 9 March 2020
Steinwasser	Discharge	19 June 2019 to 2 March 2020
Steinwasser	Electrical conductivity	19 June 2019 to 29 February 2020
Steinwasser	Stable water isotopes ($\delta^{18}O$, $\delta\ ^{2}H$)	18 June 2019 to 9 March 2020
Giglibach	Discharge	17 July 2019 to 21 March 2020
Giglibach	Electrical conductivity	17 July 2019 to 9 March 2020
Giglibach	Stable water isotopes ($\delta^{18}O$, $\delta^{2}H$)	17 July 2019 to 9 March 2020

The electrical conductivity of the streams was measured using a Campbell Scientific probe. Samples for stable water isotope analysis (oxygen and hydrogen) were taken from the catchments' effluents between June 2019 and February 2020. The samples were taken every second day between June and October 2019 using an autosampler design from the University of Freiburg, Germany, and afterwards manually every second week since the increasing snow cover prevented the continuous automatic monitoring by using the autosampler. To avoid evaporation between stream water sampling and analysis, 180 drops of Paraffin were added to the empty sample bottles prior to stream sampling as conducted by previous studies (Michelsen et al., 2018; Ohlanders et al., 2013). The water-insoluble Paraffin remains on top of the water during sampling due to its lower density compared to water preventing the evaporation and alteration of the sample.

2.3 | Laboratory analysis

2.3.1 | Stable oxygen and hydrogen water isotope measurements of end-members and streams

The stable oxygen and hydrogen isotope ratios of rain, snow, ice, and stream discharge samples were analysed using a Picarro L2120-I cavity ring-down spectrometer (CRDS) with vaporization module V1102-I at the Institute of Geological Sciences, University of Bern, Switzerland. The measured stable oxygen and hydrogen isotope ratios were expressed in the delta notation ($\delta = (R/R_{Std} - 1) \times 1000$ (‰)), where R and R_{Std} are the isotope ratios of the sample and the standard, respectively. Raw $\delta^{18}O$ and $\delta^{2}H$ values are obtained by a tenfold measurement of each sample followed by a post-run correction (memory and drift) according to van Geldern and Barth (2012). To obtain $\delta^{18}O$ and $\delta^{2}H$ values on the international Vienna Standard Mean Ocean Water (VSMOW) scale, raw delta values were calibrated against two internal standards, which were referenced to the VSMOW scale using international IAEA standards. The two standards used for calibration differed in their isotope composition and span a calibration interval between −27.41‰ and −2.65‰ for $\delta^{18}O$ values and between −209.8‰ and −13.9‰ for the $\delta^{2}H$ values, respectively. The analytical uncertainty of the $\delta^{18}O$ and $\delta^{2}H$ measurements was determined based on multiple internal and IAEA standard analysis and corresponds to 0.10‰ and 1.5‰, respectively.

2.4 | Discharge separation based on stable isotope measurements

The contribution of the glacial melt end-member to the discharges of the three catchments was quantified based on the $\delta^{18}O$ and $\delta^{2}H$ signature of the end-members (snow, rain, glacial meltwater) and the $\delta^{18}O$ and $\delta^{2}H$ measurements in the catchments' effluents. Afterwards, the results were evaluated for their plausibility to determine the hydrological conditions under which the stable isotope method can be used to estimate the contribution of glacial meltwater to mountainous streams. To quantify the glacial melt end-member discharge contribution, we focused on the snow-free time period between September and August (see detailed justification in section 3.3) and hence, we used a two-component hydrograph separation by considering the two end-members (rain and glacial meltwater) that contributed predominately to the stream discharges during this time period.

$$I_{\text{Effluent}} = X \cdot I_{\text{End-member1}} + (1 - X) \cdot I_{\text{End-member2}} \quad (1)$$

where I_{Effluent} is the isotopic composition of the catchment's effluent, $I_{\text{End-member1}}$ and $I_{\text{End-member2}}$ are the isotopic compositions of the considered end-members (rain and ice) and X is the contribution of the glacial melt end-member to the effluent.

To quantify the contribution of the glacial melt end-member to the catchment's effluent, Equation (1) was resolved to X:

$$X = (I_{\text{Effluent}} - I_{\text{End-member2}}) / (I_{\text{End-member1}} - I_{\text{End-member2}}) \quad (2)$$

To determine the total glacial meltwater discharge for a catchment over a specific time period, the relative glacial meltwater water contribution (X Equation 2) was multiplied by the total discharge and by summing them up over time for the considered time period according to:

$$Q_{\text{Meltwater}} = \sum_{i=1}^{n} X_i \cdot q_i \cdot (t_{i+0.5} - t_{i-0.5}) \quad (3)$$

where $Q_{\text{Meltwater}}$ (m^3) is the total glacial meltwater volume, X (Equation 2) is the relative glacial meltwater contribution, i is the number of samples, q (m^3/s) is the total discharge rate, and t (s) is the time of sampling.

3 | RESULTS AND DISCUSSION

3.1 | Analysis of the isotopic composition of end-members

The temporal stable water isotope evolutions ($\delta^{18}O$ and $\delta^{2}H$) of the three end-members (rain, snow, glacial meltwater) are illustrated in Figure 2. The rain end-member showed average $\delta^{18}O$ and $\delta^{2}H$ values of −8.64‰ and −58.1‰, respectively, during the sampling period (8 August–18 October; Table 2). At the effluent location of the Wendenwasser catchments and at the merging effluent location of the Giglibach and Steinwasser catchment the rain end-member showed similar average $\delta^{18}O$ and $\delta^{2}H$ values of −8.14‰, −54.1‰, −8.24‰ and −52.4‰, respectively (Figure 1). Compared to the effluent locations, the rain was more depleted in ^{18}O and ^{2}H at higher altitudes at the low and high Steinwasser rain sampling location during the early stage of the sampling period (8–29 August 2019; Figure 1) showing a $\delta^{18}O$ shift of 0.27‰/100 m and a $\delta^{2}H$ shift of 1.8‰/100 m, respectively. The depletion of ^{18}O and ^{2}H in the rain with increasing altitude can be attributed to the altitude isotope effect, which includes the preferential precipitation of heavy isotopes during the continuous orogenic uplift of humid air masses (Clark & Fritz, 1997). However, in contrast to the early stage of the sampling period, no altitude isotope effect was observed during the later stage of the sampling period (30 August–3 October), which likely results from different meteorological conditions such that no continuous orogenic uplift of the humid air masses and precipitation occurred. Consequently, a continuous depletion of heavy isotopes with increasing altitude could not be observed for the entire sampling period and no overall $\delta^{18}O$ and $\delta^{2}H$ altitude correction factor could be established for the three catchments. As opposed to the ambiguous $\delta^{18}O$ and $\delta^{2}H$ variations as a function of the altitude, a more distinct temporal $\delta^{18}O$ and $\delta^{2}H$ evolution was observed in the three catchments during the sampling period. While the rain was enriched in ^{18}O and ^{2}H in June 2019, ($\delta^{18}O = -4.19$‰; $\delta^{2}H = -26.9$‰), it became progressively lighter with increasing time reaching delta values of $\delta^{18}O = -12.26$‰ and $\delta^{2}H = -84.3$‰, respectively, in October 2019 (Figure 2).

This progressive depletion of ^{18}O and ^{2}H over time can be associated with seasonal changes and the accompanying temperature decrease between June and October (Clark & Fritz, 1997). To characterize the snow end-member in the three catchments areas, the stable oxygen and hydrogen isotope ratios of the snow were measured during the snow accumulation and ablation period, respectively as the isotopic signal of snow can differ significantly during these two periods (Beria et al., 2018; Cooper, 1998; Dietermann & Weiler, 2013; Lee et al., 2010; Zhou et al., 2008). Based on the monitoring of the snow thickness at the Gschletteregg measuring station (Figure 1), the snow accumulation period during which the snow becomes progressively thicker was identified from early November until April, whereas the snow ablation period during which the snow cover becomes continuously thinner due to sublimation, melting and redistribution processes was observed between May and October (Figure 3).

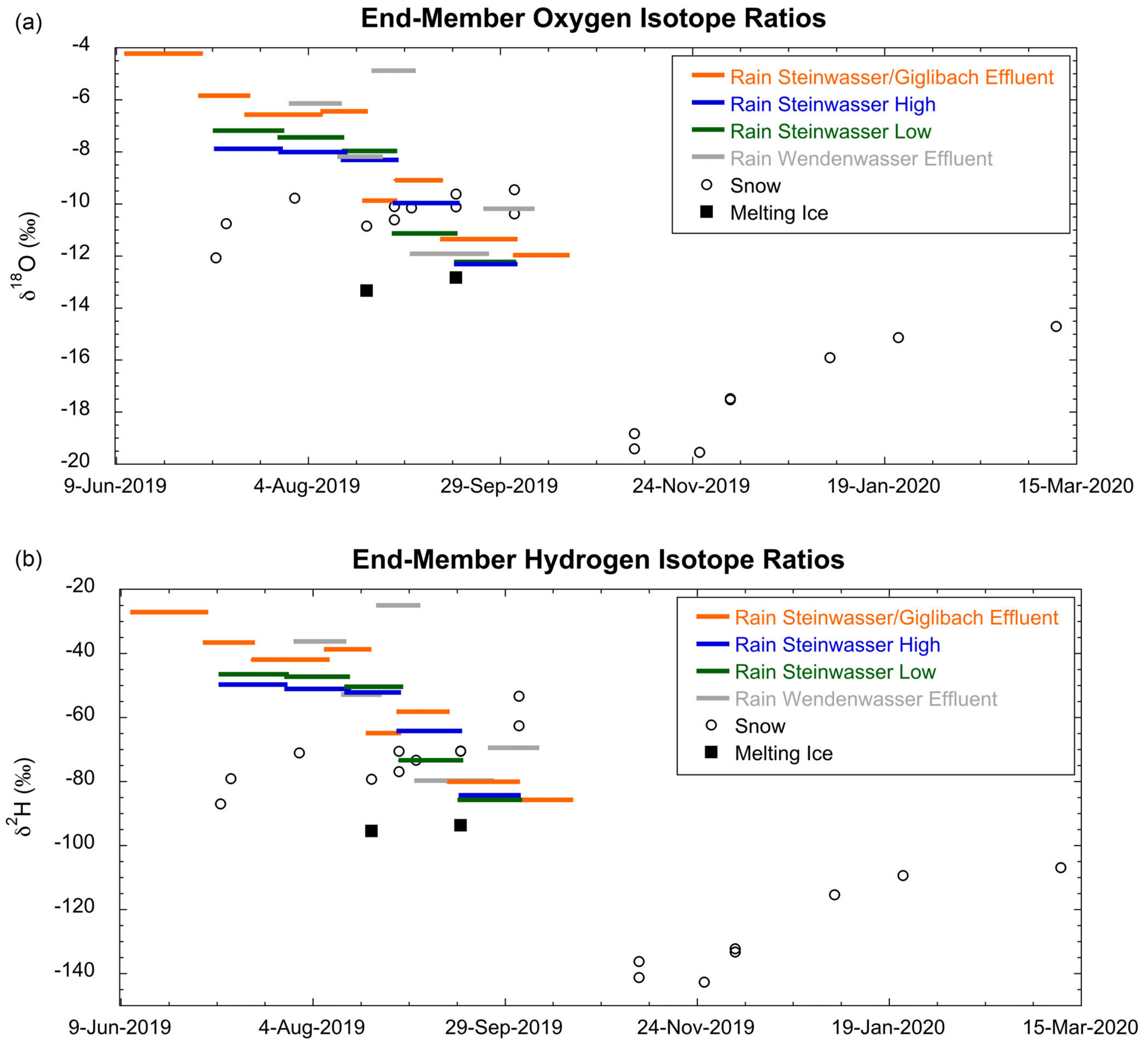

FIGURE 2 Temporal $\delta^{18}O$ (a) and $\delta^{2}H$ (b) evolution of the end-members including rain (solid lines), snow (open circles), and melting ice (filled squares) during the sampling period (June 2019 until March 2020) in the Steinwasser, Wendenwasser, and Giglibach catchment.

TABLE 2 Number of samples, average $\delta^{18}O$ and $\delta^{2}H$ values, standard deviation, as well as min and max $\delta^{18}O$ and $\delta^{2}H$ values of end-members and catchment discharges samples during the entire sampling period.

Water source	Number of samples	Average $\delta^{18}O$	Standard deviation $\delta^{18}O$	Min $\delta^{18}O$	Max $\delta^{18}O$	Average $\delta^{2}H$	Standard deviation $\delta^{2}H$	Min $\delta^{2}H$	Max $\delta^{2}H$
Rain	23	−8.64	2.38	−12.26	−4.19	−58.1	18.7	−85.7	−24.6
Snow	20	−13.48	3.77	−19.54	−9.44	−96.9	30.1	−142.7	−53.3
Ice	3	−13.43	0.65	−14.12	−12.83	−96.9	4.2	−93.6	−101.7
Wendenwasser	26	−12.02	0.62	−12.96	−12.96	−83.2	4.7	−90.4	−69.3
Steinwasser	45	−12.45	0.60	−13.27	−11.20	−87.1	4.4	−93.3	−77.4
Giglibach	50	−12.62	0.37	−13.09	−11.53	−88.3	2.8	−92.7	−79.8

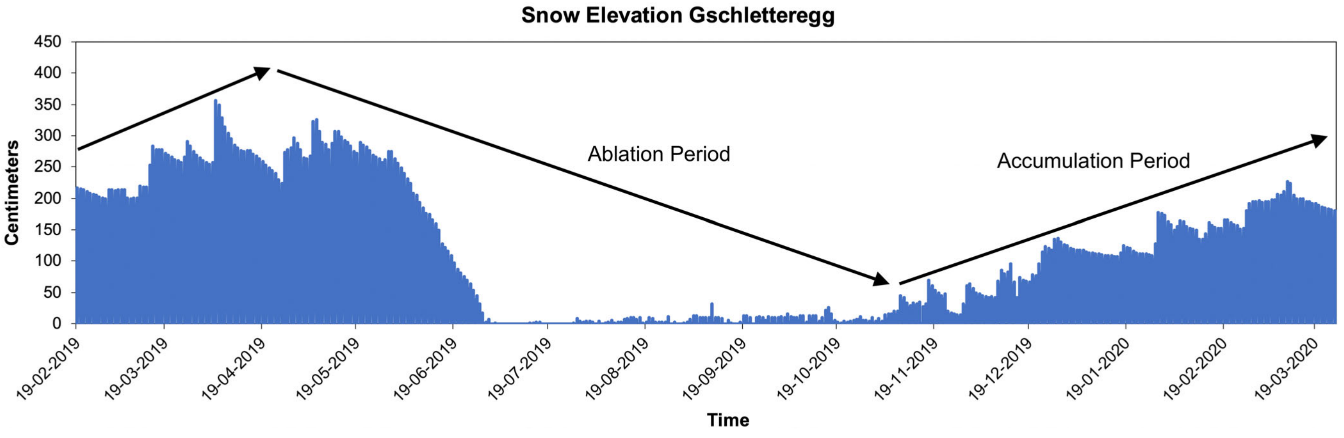

FIGURE 3 The measured snow thickness over time at the Gschletteregg measuring station being representative for the three catchment areas. Between November and April snow accumulation occurs, while from May to October snow ablation takes place.

The snow end-member showed average $\delta^{18}O$ and δ^2H values of −13.48‰ and −96.9‰, respectively, during the whole sampling period (Table 2). During the snow accumulation period (November to April), the snow samples revealed average $\delta^{18}O$ and δ^2H values of −17.31‰ and −127.1‰, respectively (Figure 2). The lowest $\delta^{18}O$ and δ^2H values (−19.40‰, −141.2‰) were detected in November at the beginning of the snow accumulation period. With increasing time, a continuous enrichment of ^{18}O and ^2H isotopes was observed in the snow reaching $\delta^{18}O$ and δ^2H snow values of −14.70‰ and −106.9‰, respectively, at the end of the accumulation period in April. The progressive enrichment of ^{18}O and ^2H in the snow occurred along the LMWL (Figure 4) and hence, it is likely not explainable by sublimation processes, which commonly leads to a righthand deviation from the LMWL (Beria et al., 2018). However, a recent study (Wahl et al., 2021) showed that the sublimation can also occur under equilibrium conditions and thus, not necessarily lead to deviation from the LMWL. Hence, the occurrence of snow sublimation cannot be completely ruled out.

In addition, the enrichment of ^{18}O and ^2H during the snow accumulation period cannot be attributed to sporadic rain events. Rain that falls during the accumulation period has a lighter isotopic signature compared to snow since the formation of snow from air moisture leads to a higher enrichment of heavy isotopes compared to the formation of rain (Clark & Fritz, 1997). Hence, the enrichment of ^{18}O and ^2H in the snow during the accumulation period is likely relatable to the moisture exchange with the atmosphere since these processes enrich the snow in ^{18}O and ^2H isotopes along the LMWL (Steen-Larsen et al., 2014). Compared to the snow accumulation period, more enriched $\delta^{18}O$ and δ^2H average values (−10.34‰, −72.1‰) were measured during the ablation period (May to October; Figure 2), which is in agreement with previous studies (Dietermann & Weiler, 2013; Lee et al., 2010; Zhou et al., 2008). Similar to the accumulation period, no significant deviation from the LMWL was observed during the snow ablation period, revealing that sublimation processes were likely not the predominant isotope fractionation process (Figure 4). The more enriched $\delta^{18}O$ and δ^2H snow values in the ablation compared to the accumulation period can be likely explained by the contribution of rain, which has a heavier isotopic signature compared to the snow during the ablation period. Besides, similar to the accumulation period the exchange with the atmosphere could also contribute to the enrichment of heavy oxygen (^{18}O) and hydrogen (^2H) isotopes during the snow ablation period.

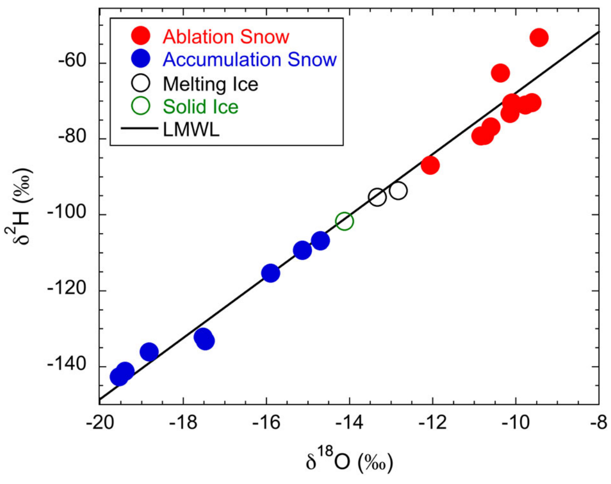

FIGURE 4 δ^2H and $\delta^{18}O$ measurements from the snow accumulation (blue filled circles) and snow ablation (red filled circles) period and δ^2H and $\delta^{18}O$ measurements of the solid (open green circle) and melting ice (open black circle). The solid black line represents the local meteoric water line (LMWL). The analytical uncertainty of the isotopic measurements corresponds to 0.10‰ for $\delta^{18}O$ and 1.5‰ for δ^2H, respectively, and is smaller than the size of the symbols.

For determining the $\delta^{18}O$ and δ^2H values of the glacial melt end-member both the solid and the melting ice were sampled. The ice samples showed average $\delta^{18}O$ and δ^2H values of −13.43‰ and −96.9‰, respectively (Table 2). Similar to the snow samples, no significant aberration of the $\delta^{18}O$ and δ^2H values from the LMWL was detected for both the solid and melting ice (Figure 4). This indicates that also for the glacial ice, sublimation processes played likely a minor

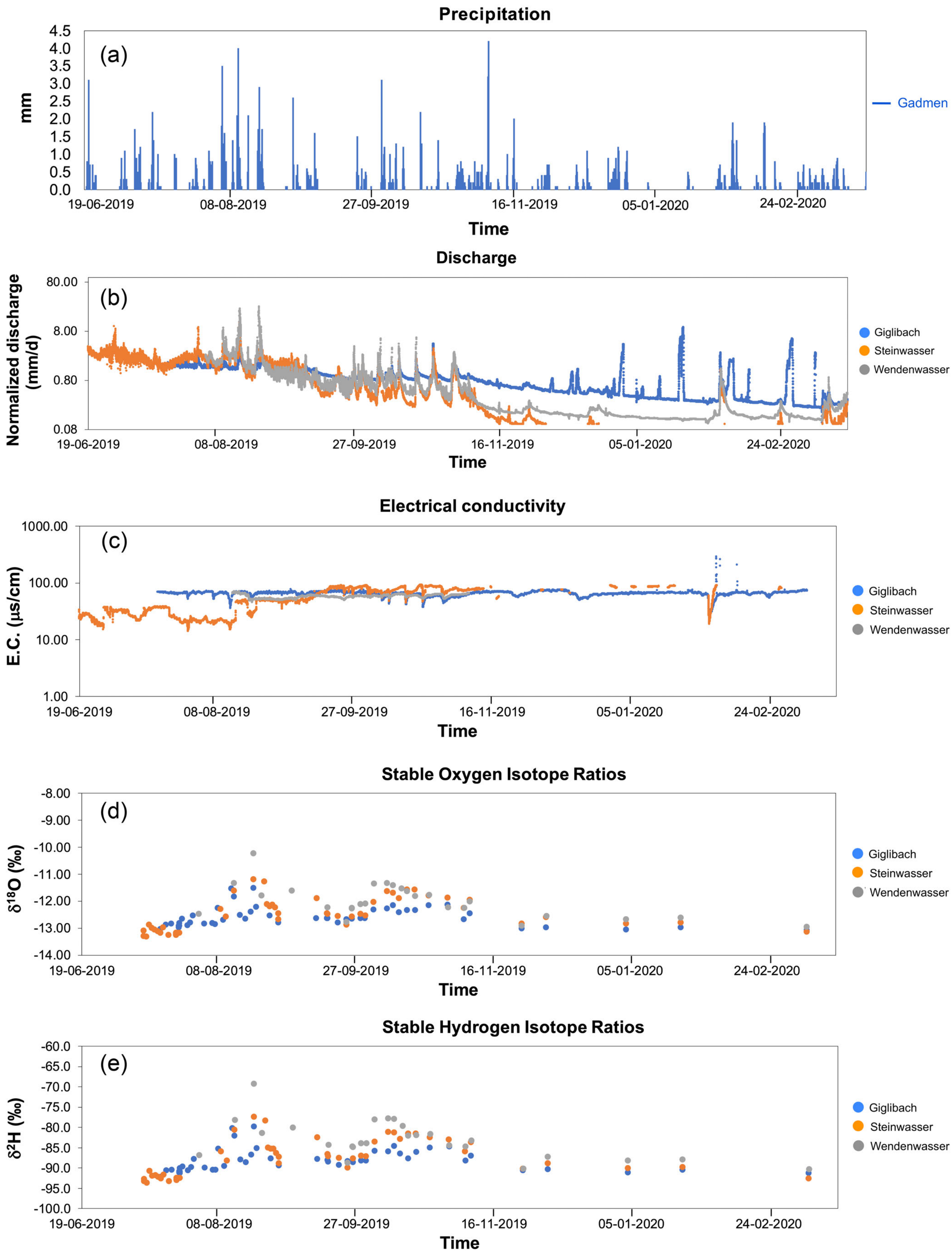

FIGURE 5 Legend on next page.

role and that the $\delta^{18}O$ and δ^2H glacial ice signatures were primarily controlled by melting/refreezing processes and the contribution of rain water and moisture. Compared to the solid ice ($\delta^{18}O = -14.12‰$ and $\delta^2H = -101.7‰$), the melted ice showed slightly more enriched $\delta^{18}O$ and δ^2H values showing a shift of $\Delta\delta^{18}O = 1.02‰$ and $\Delta\delta^2H = 7.2‰$, respectively in average (Figure 4). Additionally, the average $\delta^{18}O$ and δ^2H values (−13.42‰, −96.9‰) of the ice (solid and melting) were higher compared to the snow in the accumulation period and more depleted compared to the snow in the ablation period and the rain samples (Figure 2). The somewhat intermediate glacial $\delta^{18}O$ and δ^2H values compared to the snow in the ablation and accumulation period (Figure 4) is plausible since the glacial ice is formed from snow that originates from both the ablation and accumulation period (Beria et al., 2018).

3.2 | Qualitative discharge separation based on temporal stream analysis in the three catchment areas

To evaluate the different components of the mountain's streams in the three different catchments, highly temporally resolved precipitation, normalized discharge, E.C., and stable water isotope analysis ($\delta^{18}O$ and δ^2H) were conducted in the effluents of the three catchments between June/July 2019 and March 2020 in the Giglibach and Steinwasser catchment and between August 2019 and March 2020 in the Wendenwasser catchment (Figure 5).

In the Stein and Wendenwasser catchments, the stream discharges normalized to the catchment areas showed large temporal variations, whereby the discharge evolution can be divided into a high (June–August 2019), intermediate (September–October 2019) and low discharge time period (November–March 2020) based on sharp discharge changes over time (Figure 5b). During the high discharge period, the Stein and Wendenwasser normalized stream discharges ranged between 1 and 5 mm/day with a few peak discharges of up to 21 mm/day during heavy precipitation events (Figure 5a,b). During this time, the snow cover is rapidly decreasing (Figure 3) and the stream discharge in the Steinwasser catchment is likely dominated by snowmelt water flowing into the streams via surface run-off or groundwater, which is consistent with previous observations and simulations (Arnoux et al., 2020; Hydro-CH, 2018). The low $\delta^{18}O$ and δ^2H values (∼−12.50‰, −90.0‰) in the stream discharges of the Stein and Wendenwasser catchments during the high discharge period (June–August 2019) are also consistent with a significant snowmelt contribution to the stream discharge contribution via surface run-off and groundwater since the $\delta^{18}O$ and δ^2H values lie within the snow end-member values (Figure 4). Higher $\delta^{18}O$ and δ^2H values closer to the rain end-member (∼−7‰, −50‰) are only observed during heavy precipitation events in the high discharge period (Figure 5a,d,e). However, the input of the snowmelt via surface run-off versus via groundwater into the streams is challenging to unravel during the high discharge period based on the discharge and isotope data only. In contrast, the E.C. measurements in the streams can provide some evidence whether snow meltwater contributes to the stream discharges predominately via surface run-off or via groundwater inflow since groundwater has usually a higher E.C. value than snowmelt water (Krainer & Mostler, 2002; Zuecco et al., 2019) due the dissolution of solids during its migration through the subsurface. However, the E.C. of the groundwater is not constant and proportional to the sum of solutes dissolved in the groundwater, which is in turn directly proportional to the residence time of the groundwater in the subsurface because mineral dissolution reactions are kinetically- and not solubility-controlled. Due to this reactive and non-constant behaviour, the E.C. can only be used as a qualitative and not as a quantitative tracer for the groundwater for estimating its contribution to mountainous streams. The E.C. in the Steinwasser catchment shows a distinctively lower value (∼30 μs/cm) during the high discharge period compared to the Wendenwasser catchment (Figure 5c). This indicates that during this time period, the stream discharge in the Steinwasser catchment is dominated to a higher extent by snowmelt water that originates from surface run-off compared to snowmelt water that enters the streams via the groundwater. This conclusion is further supported by E.C. measurements of two springs close to the outlet of the Stein and Wendenwasser catchment representing groundwater and showing higher E.C. values of 148 and 149 μs/cm, respectively compared to the discharge in the Steinwasser catchment (∼30 μs/cm).

The intermediate discharge period (September–October 2019) in the Stein and Wendenwasser catchment was dominated by short discharges peaks (up to 5.4 mm/day), which were also related to precipitation events (Figure 5a) followed by baseflow recessions to normalized discharges of around 0.3 mm/day (Figure 5a,b). During this time period, the snow cover has disappeared (Figure 3) and the surface run-off input into streams is likely dominated by glacial melt or rain water, whereas snowmelt water could still contribute to the stream discharge via groundwater inflow if it is temporally stored as groundwater and subsequently released into the streams. In this intermediate discharge phase, the observed increased E.C. values to approximately 100 μs/cm, especially in the Steinwasser catchment (Figure 5c), reinforces the lower input of snowmelt water via surface run-off to the streams compared to the high discharge period between June and August 2019.

FIGURE 5 Temporal evolution of the precipitation in the study area (a) as well as discharge normalized to the catchment areas (b), electrical conductivity E.C. (c), stable oxygen (d), and stable hydrogen isotope ratios (e) measurements in the effluents of the Giglibach (blue circles), Steinwasser (orange) and Wendenwasser (grey circle) catchments between June 2019 and March 2020. The temporal precipitation evolution (a) was acquired from the precipitation measuring station in Gadmen operated by the Office of Waste and Water (AWA) of the Canton of Bern, Switzerland.

In the low discharge period, corresponding to the wintry baseflow period (November 2019–March 2020), the normalized discharge measurements were associated with some uncertainties due to the partial freeze of the measuring stations and the missing calibration measurements. This is also the reason why some data points are missing, especially for the Steinwasser catchment. Nevertheless, from the normalized discharge measurements in the wintry baseflow period, it can be seen that the discharge in the Stein and Wendenwasser catchment was lower compared to the preceding high and intermediate discharge phases (Figure 5b). Moreover, in the wintry baseflow period, the normalized discharge is systematically higher in the Wendenwasser compared to the Steinwasser catchment. This indicates that the groundwater contribution to the streams discharge is higher in the Wendenwasser compared to the Steinwasser catchment since it can be assumed that during the wintry baseflow period, groundwater is the main contributor to the streams in the two catchments (Cochand et al., 2019) and representative for the entire year (Fetter, 2018).

As opposed to the Stein and Wendenwasser catchment, a lower temporal normalized discharge variation was observed in the Giglibach catchment during the measurement period showing an average discharge of 0.8 m^3/s between July and October 2019. However, similar to the Stein and Wendenwasser catchment, $\delta^{18}O$ and δ^2H values within the snow and glacial melt end-member values were measured between July and October 2019 in the Giglibach catchment. This suggests that also in the Giglibach catchment, the snow, and glacial meltwater significantly contributes to the discharge between July and October 2019 either via surface run-off or via groundwater. Moreover, during the wintry baseflow period (November 2019–March 2020), a higher normalized discharge was observed in the Giglibach compared to the Stein and Wendenwasser catchment (Figure 5b). Interestingly, this provides evidence that the relative groundwater contribution to discharge in the Giglibach catchment is higher in comparison to the Stein and Wendenwasser catchment despite the overall lower absolute discharge. Since the groundwater acts as a buffer for the stream discharges, this is likely also the reason why the overall temporal variation of the stream discharge and E.C. measurements in the Giglibach catchment is lower compared to the Stein and Wendenwasser catchment.

Overall, it can be concluded that the three investigated catchments have varying contributions of groundwater to their discharge providing an excellent opportunity to investigate how groundwater—mountainous stream interactions potentially impact the quantitative evaluation of the glacial meltwater component in mountainous streams using stable water isotopes.

3.3 | Quantitative discharge separation based on stable water isotope ratios in the catchment's effluents

To determine the hydrological conditions under which the stable water isotope method is applicable and to evaluate its opportunities and limitations, an attempt was made to quantify the glacial meltwater contribution to the mountain streams based on the stable water isotope measurements in all three investigated catchments followed by a plausibility analysis. To this end, the period between August and September 2019 was selected. This time period is of special interest since the glacial meltwater contribution to the stream discharges is (a) likely highest throughout the year due to elevated temperatures; (b) subject to disappearance in the future due to climate change-induced deglaciation; and (c) the snow end-member is absent, which would otherwise hamper the quantification of the glacial meltwater contribution to the streams since the isotopic signal of the snow overlaps with the other two end-members (rain and glacial melt; Figure 2).

To quantify the glacial meltwater contribution to the stream discharges in August and September in the three catchments, first, the temporal evolution of the relative glacial meltwater contribution to the streams was determined. This was conducted based on the temporal stable isotope ratio measurements ($\delta^{18}O$ and δ^2H) in the catchment effluents (Figure 5d,e) and the isotopic signature of the rain and glacial meltwater end-members (Figures 2 and 4) combined with Equation (2). For the rain end-member, the temporal variation of the $\delta^{18}O$ and δ^2H values in each of the catchment outlets was taken into account, while the altitude effect was not considered as no overall altitude correction factor could be established (Figure 2). For the $\delta^{18}O$ and δ^2H signature of the glacial melt end-member, constant $\delta^{18}O$ and δ^2H values representing the average of the two taken samples ($\delta^{18}O = -13.08$‰ and $\delta^2H = -94.5$‰; Figure 4) were used for the discharge separation calculations. These two glacial melt end-member samples were taken in August and September covering the period during which the discharge separation was conducted and represent a temporal sampling frequency of one sample per month being comparable with other studies that carried discharge separation of mountainous streams (Zuecco et al., 2019).

The uncertainty of determined glacial melt water contribution was evaluated based on the uncertainty of the stable isotope measurement in the stream discharges and the contributing end-members (rain and glacial meltwater) combined with the Gaussian error propagation law, which was applied to Equation (2). This resulted in an average methodological uncertainty of the determined glacial meltwater contribution of ±2%. After determining the temporal evolution of the relative glacial meltwater contribution, the total glacial meltwater discharge was calculated for the three catchments in August and September by multiplying the relative glacial meltwater water contributions by the measured total discharges (Figure 6b) and by summing them up over time between August and September (Equation 3). This resulted in glacial meltwater discharges of 3.5 Mio m^3 for the Giglibach, 17.9 Mio m^3 for the Steinwasser, and 9.6 Mio m^3 for the Wendenwasser catchment in August and September whereby the uncertainty corresponded to 10%. The uncertainty was calculated by applying the Gaussian error propagation law to Equation (3), whereby the uncertainties of the relative meltwater contribution calculations (X_i; Equation 3) and the total discharge measurements (q_i; Equation 3) were considered. The determined glacial meltwater discharge values for the Giglibach and Wendenwasser catchment are not plausible

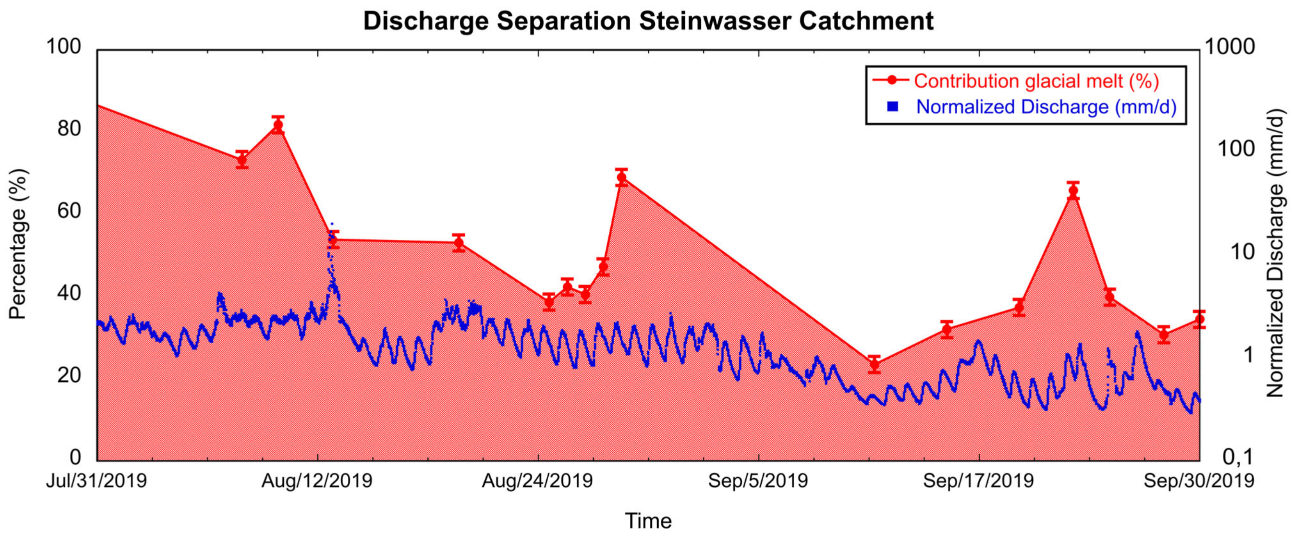

FIGURE 6 Quantification of glacial meltwater contribution to the Steinwasser catchment discharge in August and September 2019 using stable isotope ratios measurements δ^2H and $\delta^{18}O$) in the catchment effluents and the end-members using Equation (2) (red area). The average uncertainty of the determined glacial meltwater contribution corresponds to ±2%, which is indicated by the vertical error bars. The blue line represents the normalized stream discharge measurements at the effluent of the Steinwasser catchment.

since they indicate a glacial meltwater production of 3.5 Mio and 9.6 Mio m^3 from glaciated areas of 0.29 and 1.67 km^2, respectively, which corresponds to glacial meltwater production of 12.0 and 5.8 m over the glaciated areas. This glacial meltwater production is 4.4 and 2.1 times higher than the observed average glacial meltwater production of 2.75 m in Switzerland in 2019 being estimated based on mass balance calculations (GLAMOS, 2020). This indicates that in the Giglibach and Wendenwasser catchments, the glacial meltwater contribution to the streams is strongly overestimated probably due a snowmelt water input from previous periods, which was temporarily stored as groundwater and released to the streams in August and September, and which has a similar isotopic signature than the glacial meltwater. In contrast to the Giglibach and Wendenwasser catchment, the glacial meltwater production in the Steinwasser catchment in September and August corresponds to 2.6 m over the glaciated area, which is consistent with the observed average glacial meltwater production of 2.75 m for glaciers in Switzerland in 2019 (GLAMOS, 2020). This demonstrates that the groundwater input from previous snowmelt periods to the stream in the Steinwasser catchment is absent or smaller than in the Giglibach and Wendenwasser catchment and not leading to an overestimation of the glacial meltwater contribution to the mountainous streams. This is also consistent with the normalized discharge measurements of the three catchments, especially during the wintry baseflow showing that groundwater contribution in the Steinwasser catchment is lowest compared to the Giglibach and Wendenwasser catchment (Figure 5b). Hence, based on the agreement of these two independent parameters (normalized discharge and plausible glacial meltwater input), it can be assumed that the stable water isotopes can be used to quantify the glacial meltwater contribution to mountainous streams if the normalized discharge during the wintry baseflow period is not exceeding 0.1 mm/day (Figure 5b). It seems that at a normalized discharge rate below 0.1 mm/day, the groundwater input to mountainous streams is sufficiently low such that it is not impairing the quantification of the glacial meltwater input to streams using stable water isotopes, especially for granitic bedrock catchments as present in our study.

Due to the plausible results for the Steinwasser catchment, it is also worth discussing the temporal evolution of the glacial meltwater contribution to the stream in the Steinwasser catchment in August and September (Figure 6). The highest glacial melt contribution to the stream discharges in the Steinwasser catchment was observed at the beginning of August likely due to the elevated temperatures, reaching values between 85%±2% (Figure 6). In late August and early September, the relative glacial melt water discharge contribution to the stream discharges in the Steinwasser catchment was lower compared to early August but remained above 40% whereas towards the end of September, the relative glacial meltwater contribution to the catchments' discharges further decreased to 30%. The higher contributions of glacial meltwater to the mountainous streams at the beginning of August and September (Figure 6) compared to the end of September can be likely related to higher temperatures accelerating the glacial melting (Figure 5a).

If we assume that the glacial meltwater mostly contributes to the stream discharges in August and September (Hydro-CH, 2018; Pfaundler & Schönenberger, 2013), the determined glacial meltwater water contribution to the stream discharge in the Steinwasser catchment (Figure 6) can be considered as the minimum annual glacial meltwater discharge volume (mAGMD). The determined mAGMD can then be related to the total annual stream discharge volume of the Steinwasser catchment to estimate the share of glacier meltwater in the total annual stream discharge volume. The total annual stream discharge volume for the Steinwasser catchment is estimated based on discharge simulations by the Federal Office for the Environment of Switzerland (Pfaundler & Schönenberger, 2013). These simulations revealed an annual discharge volume of 62.4 Mio m^3 for the Steinwasser catchment, which was consistent with annual stream discharge volume measurements by the Kraftwerke Oberhasli AG (personal communication) reinforcing the robustness and the representativity of these annual stream discharge volumes for the year 2019. The relation of the total annual discharge volume (62.4 Mio m^3) to the mAGMD for the Steinwasser catchments (17.9 Mio m^3) results in an annual glacial melt water discharge contribution of 28.7% ± 10%. This relatively high annual glacial meltwater discharge contribution in the Steinwasser catchments also reinforces the hypothesis that discharge regimes in mountainous catchments will change in the future when these glacial meltwater contributions cease caused by climate change (Muelchi et al., 2021).

3.4 | Limitations

For the carried-out hydrograph separation in this study, we relied only on two glacial melt samples for characterizing the glacial melt end-member. The isotopic signature of the glacial meltwater end-member did not overlap with the other end-member, which is a prerequisite for the discharge separation based on stable water isotopes. We acknowledge that the encountered low isotopic variability of the glacial meltwater end-member and its difference from the other end-members is a 'best-case scenario'. However, in other catchments, there is the possibility that this 'best-case scenario' is not met, and for future studies, the different isotopic signature of the glacial melt-end member compared to the other end-members needs to be considered as an additional criterion for the successful application of the stable water isotopes to quantify the contribution of the glacial meltwater to mountainous streams.

4 | CONCLUSIONS

The evaluation of the opportunities and limitations of using the stable water isotope method to quantify the glacial meltwater contribution to streams in Alpine regions revealed that the method can be successfully applied if the following three criteria are met: (a) The snow meltwater contribution to mountainous streams must be negligible due to its highly variable stable isotope signature; (b) the groundwater input needs to be either insignificant during this snow-free period (normalized discharge during wintry baseflow <0.1 mm/day) or the groundwater residence time must be short such that groundwater contribution does not delay the end-member signal (glacial melt and rainwater) arriving in the streams; and (c) the isotope signal of the glacial melt end-member needs to be distinct from the other end-members in the considered period.

In the three investigated catchments, the three criteria for successfully applying the stable isotope method were not met in Wendenwasser and Giglibach catchment since significant amounts of snow meltwater were stored as groundwater and released during the glacial meltwater period hampering the quantification of the glacial meltwater contribution to the mountainous streams. In contrast to the Wendenwasser and Giglibach catchments, the groundwater input was low in the Steinwasser catchment, which opened-up the possibility to quantify the glacial meltwater contribution to the mountainous streams in August and September when the snowmelt contribution was minimal. The resulting high stream contribution of glacial meltwater (up to 85% ± 2% corresponding to a minimum glacial meltwater contribution to the total annual discharges of 28.7% ± 10%) shows that the flow regime of mountainous streams will change in the future since the high contribution of glacial meltwater will disappear due to climate change-induced deglaciation. It is expected that these high glacial meltwater contributions to mountainous stream discharges will decrease not only in our study area but also in other Alpine regions during the next decades due to global warming. Moreover, the peak discharges in the mountainous streams will likely occur earlier in the year (May/June) compared to today (June/July) due to the earlier occurrence of the snowmelt caused by global warming. However, predictive discharge simulations for Alpine catchments suggest that the annual mountainous stream discharge volumes will not significantly decrease despite the ceasing glacial meltwater contributions as they will be compensated by higher discharge volumes in winter and spring (Hydro-CH, 2018).

Overall, the outcome of this study shows that many hydrogeological criteria need to be met so that stable water isotopes can be used to quantify the glacial meltwater contribution to mountainous streams. This highlights the great challenges of using stable water isotopes alone for discharge separation and the need for additional hydrological tracers. Nevertheless, the obtained hydrological knowledge about when and where stable water isotopes can be used in mountainous areas to quantify the glacial meltwater contribution to streams will provide guidance for future stable isotope studies evaluating the different contributing components to mountainous streams. Such guidance for future studies will allow the identification of mountainous catchments where the stable isotope method can be applied and will lead to a better interpretation of the results and associated simulations. An improved application of stable isotope methods in mountainous catchments in future studies based on the findings of this study will be crucial to better determine the effect of climate change on the glacial meltwater contribution to mountainous streams and its impact on high-altitude ecosystems.

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DATA AVAILABILITY STATEMENT

The data that support the findings of this study are openly available in Zenodo at https://doi.org/10.5281/zenodo.5571465.

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