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Classification of Stable Isotopes and Identification of Water Replenishment in the Naqu River Basin, Qinghai-Tibet Plateau

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Received: 29 August 2018; Accepted: 24 December 2018; Published: 28 December 2018



Abstract: The stable isotopic study of the mechanism of runoff replenishment in the Qinghai-Tibet Plateau is a time-consuming and complicated process requiring complex monitoring data and scientific evaluation methods. Based on the data of water stable isotopes (^{18}O and ^2H) in the Naqu River basin, the present paper developed a framework of the variable fuzzy evaluation model (VFEM) to provide a method to classify stable isotopes and generalize the source identification of water replenishment by rainfall or snowmelt in the Naqu River basin. The grade eigenvalues of tributaries were ranked from low to high as follows: 1, 1.005, 1.089, 1.151, 1.264, 1.455 and 2.624. Three sets of tributaries were distinguished. The grade eigenvalues of the Najinqu, Bazongqu, Mumuqu, Chengqu and Gongqu Rivers were small, indicating that these tributaries were strongly supplemented by precipitation and snowmelt; the grade eigenvalue of the Zongqingqu River was in the medium range (1.455); the third group included the Mugequ River with a high status value (2.624). This study mainly highlighted the combination of the classification of stable isotopes and plots of $\delta^2\text{H}$ vs. $\delta^{18}\text{O}$ in the source identification of water replenishment, which will be helpful for studying runoff replenishment and the evolution mechanism in the Qinghai-Tibet Plateau.

Keywords: stable isotopes; variable fuzzy evaluation method; water replenishment; Naqu River basin; Qinghai-Tibet Plateau

1. Introduction

The study of water cycle evolution to solve water resource and related environmental issues has become a significant task for hydrological observation processes. However, traditional hydrological observation is inconvenient in ungauged catchments, especially in the arid region with extreme climate conditions [1].

Variations in the stable isotopes of water, such as ^{18}O and ^2H , are widely used as environmental tracers in studying hydrological processes [2–6]. Short and long-term isotope signals in precipitation are thus transmitted through the whole catchment [7]. Isotopes of particular interest for hydrological studies include the stable isotopes of water (^{18}O , ^2H), which are incorporated within the water molecule (H_2^{18}O , $^1\text{H}^2\text{H}^{16}\text{O}$), and exhibit systematic spatial and temporal variations as a result of isotope fractionations that accompany water-cycle phase changes and diffusion [8]. Stable- and radio-isotope tracers have been widely applied in Earth systems studies, including hydrological and climatological research, for their ability to provide a sharper focus on some of the underlying processes

that control the chemical and physical behaviour of elements and compounds in nature. Spatial and temporal variations in hydrogen and oxygen isotopic compositions ($\delta^2\text{H}$ and $\delta^{18}\text{O}$) could be attributed to equilibrium exchanges occurring at all phase transitions and kinetic isotopic fractionation during transformation from the ocean surface to the atmosphere, small rainfall amounts when rain drops fall below the cloud [9]. For example, ratios of stable isotopes of hydrogen and oxygen ($^2\text{H}/^1\text{H}$ and $^{18}\text{O}/^{16}\text{O}$) in river waters were measured to investigate the hydrological pathway of the Xijiang River, in southwest China [10]; groundwater was found to be caused principally by the mixing of rainfall and water stored before the event in the Meilin watershed [11]; $\delta^2\text{H}$ and $\delta^{18}\text{O}$ were used for evaluating the source of water [12–15] and exploring the interaction between surface water and groundwater [16–19] by using the change in isotopic composition due to evaporation or mixing by different waters.

The Qinghai-Tibet Plateau has long been the focus of international academic concern because it possesses a unique high plateau climate and is very sensitive to global climate change and human activities. Isotope studies on the processes of rivers and lakes in the plateau area have achieved significant progress since the 1960s [20]. Zhang et al. [21] investigated the variation of $\delta^2\text{H}$ and $\delta^{18}\text{O}$ in glacial meltwater at Mount Everest and found that $\delta^2\text{H}$ in ice and snow is larger than that in river and lake water compared to the change of $\delta^{18}\text{O}$. Regarding precipitation, the variation of $\delta^{18}\text{O}$ is subject to the effects of both temperature and rainfall quantity [22]. By analyzing the variation of $\delta^{18}\text{O}$ in the Naqu River basin in the plateau, Tian et al. [23] proved that the effects of lake water and river runoff on stable isotopes differ considerably. Pang et al. [24] undertook a detailed examination of the isotope fractionation process occurring at the ice–snow phase transition zones at the Baishui Glacier. While isotopic characteristics of river water have been described in general, few studies have investigated the classification of stable isotopes for the source identification of water replenishment.

Due to the particularity of its geographical location and the lack of the data from hydrological stations, it is very difficult to explore the hydrological cycle in the Naqu River basin. The variable fuzzy evaluation model (VFEM) can reasonably identify the relationship degree and relationship function between the sample index and standard interval of each level index. The assessment level of each sample by varying the model and its parameters are properly determined, so that the reliability of assessment can be improved. The proposed method has been applied to assess the water resources carrying capacity of the Huaihe River basin [25].

This paper presents a set of criteria using VFEM to determine the classification of stable isotopes for the identification of water replenishment in the Naqu River basin. It can be concluded that VFEM is useful for the classification of water replenishment. The research result may provide support to runoff replenishment by rainfall or snowmelt in the Naqu River basin.

2. Description of the Study Area

The Naqu River basin is located in the North Tibet Plateau in the Tibet Autonomous Region of China with its domain between $30^{\circ}54'–32^{\circ}43'$ N and $91^{\circ}12'–92^{\circ}54'$ E (Figure 1), and with a catchment area of 16,350 km². The average elevation of the study area is higher than 4600 m. The length of the river is 460 km with a 920-m drop. The mean annual rainfall is 477.8 mm, and the mean annual temperature is -0.6 °C. The annual rainfall is concentrated in the summer (from June to October) and accounts for about 81.9% of the total annual rainfall. Thus, the period from June to October has been defined as the wet season, and the remaining months as the dry season. Although snowfall mostly occurs in the non-flood season and total snowfall accounts for less than 20% of the precipitation amount, snowmelt flow contributes significantly to the streamflow in May and June. The Mugequ, Chengqu and Gongqu Rivers are the main sub-basins of the Naqu River basin and have catchment areas of 2103, 1090 and 1232 km², respectively. The average annual runoff of the Naqu River basin is 3.1 billion cubic meters.

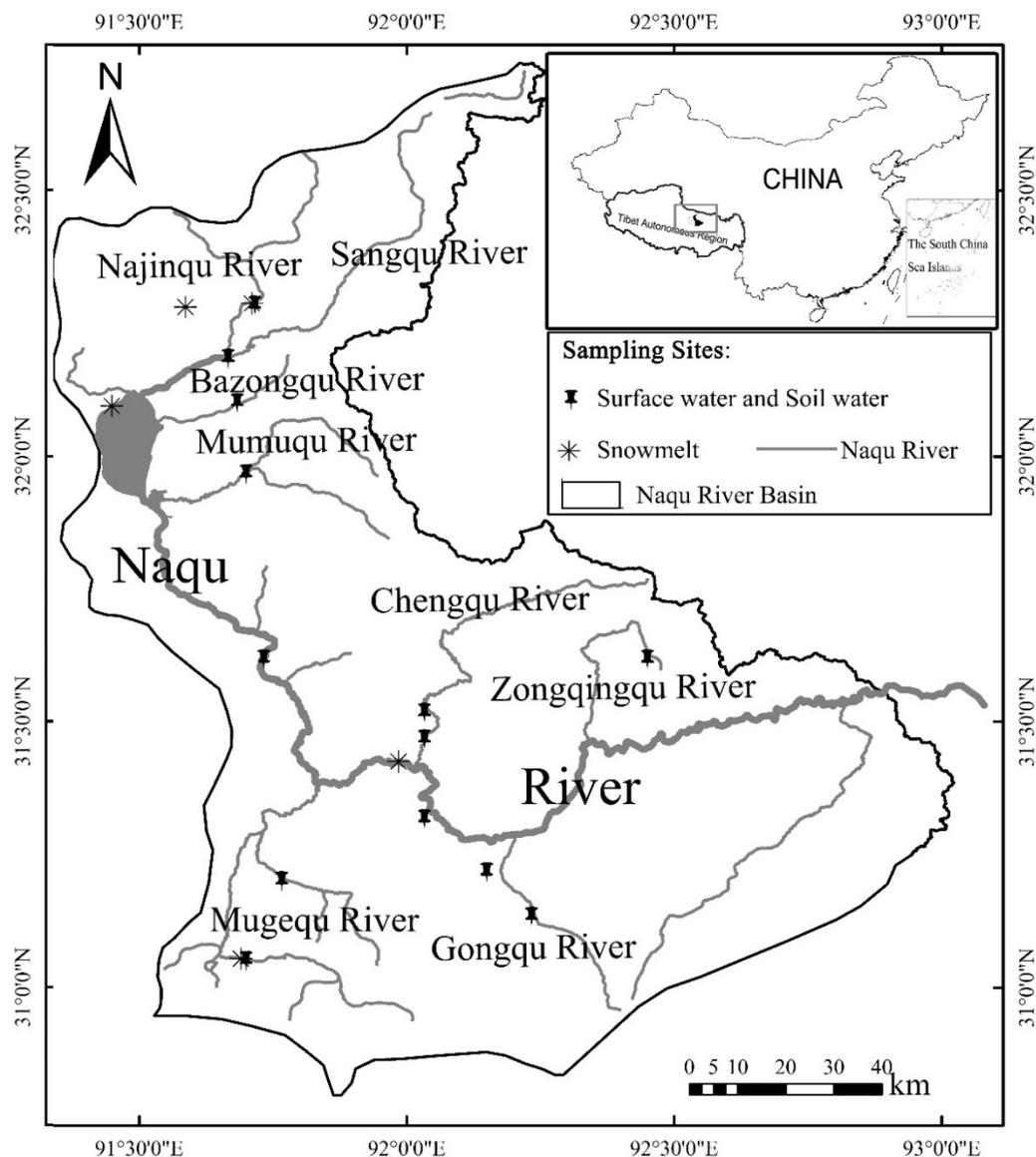


Figure 1. Research area and the sampling positions distribution.

3. Materials and Methods

3.1. Sampling and Analytical Methods

River and soil waters were sampled on 20–23 August 2016 in the Naqu River basin, and further sample collection from upstream to downstream in the Naqu River basin was conducted on 4–10 January 2017. In total, we collected 27 samples of surface water (14 samples in January, 13 samples in August), 13 samples of soil water in August, and 5 samples of snowmelt in January. Thirteen sample sites were chosen around the mainstream and tributary of the Naqu River basin. The sampling sites are shown in Figure 1. Twelve water samples were collected from the main stream and three tributaries, and then stored in glass sampling bottles for ^2H and ^{18}O analysis.

Twelve soil samples were also collected near the water sampling sites for water extraction. The soil was drilled at the top layer of soil (0–10 cm) and then immediately stored in polytetrafluoroethylene bottles to prevent evaporation. Soil samples were frozen and then thawed overnight before water was extracted using the cryogenic vacuum distillation method.

Water samples were analyzed for ^2H and ^{18}O through wavelength-scanned cavity ring down spectroscopy (WS-CRDS) (Picarro L1115-I, Picarro, Santa Clara, CA, USA). The $\delta^2\text{H}$ and $\delta^{18}\text{O}$ were

corrected using VSMOW (Vienna Standard Mean Ocean Water) ($\delta^2\text{H} = 0\text{‰}$, $\delta^{18}\text{O} = 0\text{‰}$) and Standard Light Antarctic Precipitation ($\delta^2\text{H} = -428\text{‰}$, $\delta^{18}\text{O} = -55.5\text{‰}$). The analytical precision was generally 0.5‰ for $\delta^2\text{H}$ and 0.1‰ for $\delta^{18}\text{O}$. $(^2\text{H}/^1\text{H})_{\text{sample}}$ or $(^{18}\text{O}/^{16}\text{O})_{\text{sample}}$ is the stable isotope ratio in the sampled water, and $(^2\text{H}/^1\text{H})_{\text{standard}}$ or $(^{18}\text{O}/^{16}\text{O})_{\text{standard}}$ is the stable isotope ratio in standard mean ocean water. The final results are presented as

$$\delta^2H_{V-SMOW} = \left(\frac{^2\text{H}/^1\text{H}_{\text{sample}}}{^2\text{H}/^1\text{H}_{\text{standard}}} - 1 \right) \times 1000(\text{‰})$$

$$\delta^{18}\text{O}_{V-SMOW} = \left(\frac{^{18}\text{O}/^{16}\text{O}_{\text{sample}}}{^{18}\text{O}/^{16}\text{O}_{\text{standard}}} - 1 \right) \times 1000(\text{‰})$$

3.2. Methods

The concept of fuzzy sets depicting imprecision or vagueness was introduced by Zadeh, which gives fuzziness a scientific description and has generated great interest in academic circles. An extended Zadeh’s fuzzy set theory and the engineering fuzzy set theory were established according to relative membership function. Afterwards, the theory of variable fuzzy sets (VFS) was presented by Chen [26], which can scientifically and reasonably determine membership degrees and relative membership functions of disquisitive objectives (or indices) at level interval relating to the engineering system. This can fully use one’s experience and knowledge as well as qualitative and quantitative information of the index system to obtain weights of objectives (or indices) [27]. In this paper, a VFEM based on variable fuzzy sets was proposed to identify the classification of stable isotopes in the Naqu River basin, and the theory and methodology of the proposed model is presented here.

3.2.1. The Definition of Variable Fuzzy Sets

In order to define the concept of VFS, we suppose that U is a fuzzy concept, and A and A^C represent attractability and repellency, respectively. To any elements $u(u \in U)$, $\mu_A(u)$ and $\mu_{A^C}(u)$ are the relative membership function of elements u to A and A^C that express degrees of attractability and repellency, respectively, where, $\mu_A(u) + \mu_{A^C}(u) = 1$, and $0 \leq \mu_A(u) \leq 1, 0 \leq \mu_{A^C}(u) \leq 1$.

Let

$$D_A(u) = \mu_A(u) - \mu_{A^C}(u) \tag{1}$$

where $D_A(u)$ is defined as the relative difference degree of u to A .

Then, mapping,

$$\left\{ \begin{array}{l} D_A(u) : D \rightarrow [-1, 1] \\ u \mapsto D_A(u) \in [-1, 1] \end{array} \right. \tag{2}$$

is defined as the relative difference function of u to A . Since $\mu_A(u) + \mu_{A^C}(u) = 1$, then:

$$D_A(u) = 2\mu_A(u) - 1 \tag{3}$$

Let,

$$\tilde{V} = \{(u, D) | u \in U, D(u) = \mu_A(u) - \mu_{A^C}(u), D \in [-1, 1]\} \tag{4}$$

$$A_+ = \{u | u \in U, 0 < D(u) < 1\} \tag{5}$$

$$A_- = \{u | u \in U, -1 < D(u) < 0\} \tag{6}$$

$$A_0 = \{u | u \in U, D(u) = 0\} \tag{7}$$

where \tilde{V} denotes VFS, and A_+ , A_- , and A_0 indicate attracting sets, repelling sets and the balance and qualitative change boundary of VFS, respectively [28]. The details of the definition of VFS are also available in these references.

3.2.2. The Relative Membership Degree of Indices

Suppose the object evaluated has m indices eigenvalue as follows:

$$X = \{x_1, x_2, \dots, x_m\} \tag{8}$$

The indices can be evaluated by c levels, and the criteria interval matrices of m indices and c levels can be expressed as follows:

$$I_{ab} = ([a_{ih}, b_{ih}]) \tag{9}$$

where $i = 1, 2, \dots, m; h = 1, 2, \dots, c$. Level 1 means the superior level and level c means the inferior level. If the index is the larger the better, then $a > b$. If the index is the smaller the better, then $a < b$. For every $[a_{ih}, b_{ih}]$, the range of interval $[c_{ih}, d_{ih}]$ can be determined according to the upper and lower bound of its adjacent intervals.

$$I_{cd} = ([c_{ih}, d_{ih}]) \tag{10}$$

(1) When $h = 1$, then its criteria interval of the index $I_{ab} = [a_{i1}, b_{i1}]$. Obviously, for A , the relative membership of the upper bound a_{i1} , $\mu_A(a_{i1})$ equals 1, and the relative membership degree of the lower bound b_{i1} , $\mu_A(b_{i1})$ equals 0.5.

Suppose that M_{i1} is a point value belonging to the interval $[a_{i1}, b_{i1}]$, and its relative membership degree to A is 1. Since level 1 is the superior level, and the relative membership degree of the left bound point a_{i1} to A is 1, then $M_{i1} = a_{i1}$; that is, M_{i1} is located at the left bound point of $[a_{i1}, b_{i1}]$.

When the index eigenvalue x_i of the object evaluated belongs to the interval $[a_{i1}, b_{i1}]$, x_i must be located at the right of M_{i1} . The relative membership degree of x_i to level 1 can be calculated as follows:

$$\mu_A(x_i)_1 = 0.5 \left[1 + \left(\frac{x_i - b_{i1}}{M_{i1} - b_{i1}} \right)^\beta \right]; x_i \in [M_{i1}, b_{i1}] \tag{11}$$

The above equation satisfies the conditions that when $x_i = a_{i1}$, $\mu_A(a_{i1}) = 1$, and when $x_i = b_{i1}$, $\mu_A(b_{i1}) = 0.5$.

When x_i belongs to adjacent level interval $[a_{i2}, b_{i2}]$, x_i must be located at the right of M_{i1} . The relative membership degree of x_i to level 1 can be calculated as follows:

$$\mu_A(x_i)_1 = 0.5 \left[1 + \left(\frac{x_i - b_{i1}}{d_{i1} - b_{i1}} \right)^\beta \right]; x_i \in [b_{i1}, b_{i1}] \tag{12}$$

The above equation satisfies the conditions that when $x_i = b_{i1}$, $\mu_A(b_{i1}) = 0.5$, and when $x_i = d_{i1}$, $\mu_A(d_{i1}) = 0$ where β is a non-negative number. When $\beta = 1$, Equations (11) and (12) become linear functions.

(2) When $h = c$, then its criteria interval of the index $I_{ab} = [a_{ic}, b_{ic}]$. Obviously, for A , the relative membership of the upper bound b_{ic} , $\mu_A(b_{ic})$ equals 1, and the relative membership degree of the upper bound a_{ic} , $\mu_A(a_{ic})$ equals 0.5.

Suppose that M_{ic} is a point value belonging to the interval $[a_{ic}, b_{ic}]$, and its relative membership degree to A is 1. Since level c is the superior level, and the relative membership degree of the left bound point b_{ic} to A is 1, then $M_{ic} = b_{ic}$; that is, M_{ic} is located at the right bound point of $[a_{ic}, b_{ic}]$.

When x_i belongs to the interval $[a_{ic}, b_{ic}]$, x_i must be located at the left of M_{ic} . The relative membership degree of x_i to level c can be calculated as follows:

$$\mu_{AC}(x_i)_c = 0.5 \left[1 + \left(\frac{x_i - b_{ic}}{M_{ic} - b_{ic}} \right)^\beta \right]; x_i \in [M_{ic}, b_{ic}] \tag{13}$$

The above equation satisfies the conditions that when $x_i = b_{ic}$, $\mu_{AC}(b_{ic}) = 1$, and when $x_i = a_{ic}$, $\mu_{AC}(a_{ic}) = 0.5$.

When x_i belongs to adjacent level interval $[a_{i(c-1)}, b_{i(c-1)}]$, x_i must be located at the left of M_{ic} . The relative membership degree of x_i to level c can be calculated as follows:

$$\mu_{AC}(x_i)_c = 0.5 \left[1 + \left(\frac{x_i - a_{i1}}{c_{ic} - a_{ic}} \right)^\beta \right]; x_i \in [c_{ic}, a_{ic}] \tag{14}$$

(3) When c is the middle level, suppose that c is an odd number, and the middle level $l = (c + 1)/2$ means not good and not bad. For the criteria interval of the index $[a_{il}, b_{il}]$, its relative membership degree of the upper bound a_{ic} and lower bound b_{ic} , $\mu_A(a_{ic})$ equals 0.5 and $\mu_{AC}(b_{ic})$ equals 0.5, respectively. However, they are not the same as the relationship with the adjacent level, viz., a_{il} and $b_{i(l-1)}$ are coincident, and b_{il} and $a_{i(l+1)}$ are coincident. The relative membership degree of the former is less than 0.5 to level $(l - 1)$, and the relative membership degree of the latter is less than 0.5 to level $(l + 1)$.

Suppose that M_{il} is a point value of $\mu_A(u) = 1$ in the interval $[a_{il}, b_{il}]$, and M_{il} can be selected as the midpoint value of the interval $[a_{il}, b_{il}]$ because the change from the superior to inferior levels is gradual; that is, $M_{il} = (a_{il}, b_{il})/2$.

If x_i belongs to the interval $[a_{il}, b_{il}]$, and x_i is located at the left of M_{il} , the relative membership degree of x_i to level l can be calculated as follows:

$$\mu_A(x_i)_l = 0.5 \left[1 + \left(\frac{x_i - a_{il}}{M_{il} - a_{il}} \right)^\beta \right]; x_i \in [a_{il}, M_{il}] \tag{15}$$

If x_i belongs to the left adjacent level interval $[a_{i(l-1)}, b_{i(l-1)}]$, x_i must also be located at the left of M_{ic} . The relative membership degree of x_i to level l can be calculated as follows:

$$\mu_A(x_i)_l = 0.5 \left[1 - \left(\frac{x_i - a_{il}}{c_{il} - a_{il}} \right)^\beta \right]; x_i \in [c_{il}, a_{il}] \tag{16}$$

If x_i belongs to the interval $[a_{il}, b_{il}]$, and x_i is located at the right of M_{il} , the relative membership degree of x_i to level l can be calculated as follows:

$$\mu_A(x_i)_l = 0.5 \left[1 + \left(\frac{x_i - b_{il}}{M_{il} - b_{il}} \right)^\beta \right]; x_i \in [m_{il}, b_{il}] \tag{17}$$

If x_i belongs to the right adjacent level interval $[a_{i(l+1)}, b_{i(l+1)}]$, x_i must also be located at the right of M_{ic} . The relative membership degree of x_i to level l can be calculated as follows:

$$\mu_A(x_i)_l = 0.5 \left[1 - \left(\frac{x_i - b_{il}}{d_{il} - b_{il}} \right)^\beta \right]; x_i \in [b_{il}, d_{il}] \tag{18}$$

The M_{ih} is an important parameter and can be obtained according to the standard eigenvalue a_{ih} and b_{ih} as follows:

$$M_{ih} = \frac{c - h}{c - 1} a_{ih} + \frac{h - 1}{c - 1} b_{ih} \tag{19}$$

Equation (19) satisfies the above three suppositions: (1) when $h = 1$, then $M_{i1} = a_{i1}$; (2) when $h = c$, then $M_{ic} = b_{ic}$; (3) when $h = l = \frac{c+1}{2}$, then $M_{il} = \frac{a_{il} + b_{il}}{2}$.

3.2.3. Variable Fuzzy Evaluation Model (VFEM) for Classification of Stable Isotopes

Assume that there are n samples of stable isotopes which need to be assessed as follows:

$$X = \{x_1, x_2, \dots, x_n\} \tag{20}$$

The eigenvector of sample j is denoted by means of m indices eigenvalue as follows:

$$x_j = (x_{1j}, x_{2j}, \dots, x_{mj})^T \tag{21}$$

Then, the matrix of eigenvalues to all samples is obtained as follows:

$$X = \{x_{ij}\} \tag{22}$$

where x_{ij} is the eigenvalue of sample j of the j th index, $i = 1, 2, \dots, m$ and $j = 1, 2, \dots, n$. The evaluated samples are recognized according to c grades, so the standard eigenvalue matrix can be written as

$$Y = \{y_{ih}\} \tag{23}$$

where y_{ih} is the standard eigenvalue of index i about the level h , and $h = 1, 2, \dots, c$.

According to the standard eigenvalue matrix of indices and practical situation, the matrix of attraction range I_{ab} , the matrix of range I_{cd} and the point value matrix M can be computed as follows:

$$I_{ab} = ([a_{ih}, b_{ih}]), I_{cd} = ([c_{ih}, d_{ih}]) \tag{24}$$

$$M = (M_{ih}) \tag{25}$$

where $[a_{ih}, b_{ih}]$ and $[c_{ih}, d_{ih}]$ are the attraction scope and the range of M_{ih} is a point belonging to $[a_{ih}, b_{ih}]$, for which its relative membership degree equals 1.

The matrix of the relative membership degrees can be computed by Equations (11)–(18).

$$[U_h] = (\mu_A(x_{ij})_h) \tag{26}$$

where $\mu_A(x_{ij})_h$ is the relative membership degree of the sample j to the index i about the grade h .

Assume that the weight vector of the indices is:

$$\omega = (\omega_1, \omega_2, \dots, \omega_m) \tag{27}$$

and satisfies $\sum_{i=1}^m \omega_i = 1$, which can be obtained according to the order consistency theorem of the importance of deterministic index.

A synthetic relative membership degree vector can be obtained by the variable fuzzy evaluation model.

$${}_i u' = 1 / \left[\frac{1 + \sum_{j=1}^m [w_j (1 - \mu_A(x_{ij})_h)]^p}{\sum_{j=1}^m [w_j \mu_A(x_{ij})_h]^p} \right]^{\alpha/p} \tag{28}$$

where α is the rule parameter of model optimization ($\alpha = 1$ for the least-single method and $\alpha = 2$ for least squares method), and p is distance parameter ($p = 1$ denotes hamming distance and $p = 2$ denotes Euclidean distance).

After normalizing ${}_i u'$, the normalized synthetic relative membership degree of each index about the grade h is obtained:

$$U = ({}_i u) \tag{29}$$

$${}_i \mu_h = {}_i u'_h / \sum_{h=1}^c {}_i u'_h \tag{30}$$

The grade eigenvalues can be computed using Equation (27), and the evaluation level can be determined according to the value H .

$$H = (1, 2, \dots, c)U \tag{31}$$

3.2.4. Determination of the Weight of Index

In order to determine the weight vector of the two indices (δ^2H and $\delta^{18}O$), the consistency scale matrix of the importance of two indices is obtained according to the order consistency theorem of the importance of deterministic index:

$$F = \begin{vmatrix} 0.5 & 1 \\ 1 & 0.5 \end{vmatrix} \begin{matrix} \textcircled{1} \\ \textcircled{2} \end{matrix} \tag{32}$$

According to the ranking of the importance of matrix F and empirical knowledge, the index $\delta^{18}O$ is in a “slightly” important position compared with δ^2H . The non-normalized weight vector of the two evaluation indices can be obtained based on the relationship table between the mood operator and the relative membership degree [25]:

$$w' = \begin{vmatrix} 1 & 0.55 \end{vmatrix} \tag{33}$$

Further, the weights of δ^2H and $\delta^{18}O$ can be obtained by normalization:

$$w = \begin{vmatrix} 0.65 & 0.35 \end{vmatrix} \tag{34}$$

4. Results

4.1. Classification of Stable Isotopes in the Naqu River Basin Based on the Variable Fuzzy Evaluation Method

Based on the comprehensive analysis of the influence factors of stable isotopic concentrations in water in the Naqu River basin, referring to the range of the water isotopes in Qinghai-Tibet plateau, the selected index system included $\delta^{18}O$ and δ^2H . Figure 2a,b show the variation range of δ^2H and $\delta^{18}O$ in water. The variation range of $\delta^{18}O$ was -15.60‰ to 10.52‰ , and the variation range of δ^2H was -117.7‰ to -88.6‰ . Figure 2a,b refer to the sampled waters. The stable isotopes values are shown in Table 1, which were from the sampled waters.

Table 1. The characteristic values of $\delta^{18}O$ and δ^2H in the rivers.

| The Evaluation Index | Evaluation of Regional (‰) | | | | | | | | | |
|----------------------|----------------------------|--------------|----------------|--------------|---------------|--------------|---------------|----------------|--------------|------------------|
| | Najinqu River | Sangqu River | Bazongqu River | Mumuqu River | Middle Stream | Mugequ River | Chengqu River | The Downstream | Gongqu River | Zongqingqu River |
| $\delta^{18}O$ | -15.3 | -14.9 | -15.4 | -15.0 | -10.5 | -13.2 | -14.8 | -12.3 | -15.6 | -15.0 |
| δ^2H | -116 | -113 | -117 | -115 | -89 | -107 | -117 | -100 | -118 | -110 |

Two indices were divided into four levels according to the principle of proportion (Table 2). The higher the level was, the greater the concentration of heavy isotopes in the river after evaporation. This means that the river was under a non-equilibrium process. If the level was lower, it meant that the river was characterized by isotope dilution and replenished by precipitation or snowmelt.

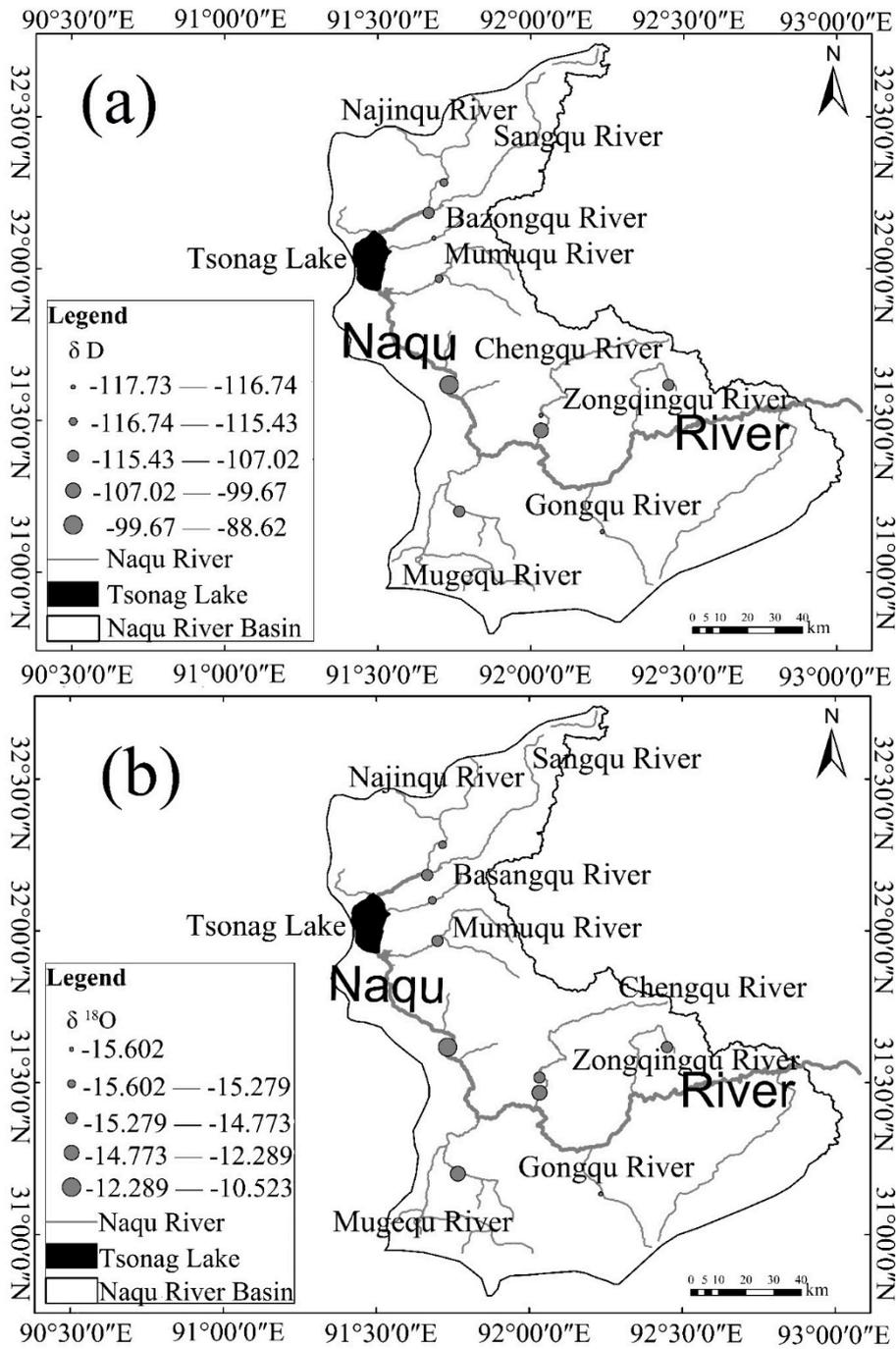


Figure 2. Spatial variation of stable isotopic enrichment in the Naqu River basin.

Table 2. The standard value of stable isotopes in the river.

| The Evaluation Index | Index Standard Value (‰) | | | |
|-----------------------|--------------------------|--------------------|--------------------|----------------|
| | Level 1 | Level 2 | Level 3 | Level 4 |
| $\delta^{18}\text{O}$ | ≤ -14.6 | (-14.6) to (-13.6) | (-13.6) to (-12.6) | $0 \geq -12.6$ |
| $\delta^2\text{H}$ | ≤ -111 | (-111) to (-104) | (-104) to (-97) | ≥ -97 |

According to the above analysis, matrices I_{ab} , M , and I_{cd} can be determined as follows

$$I_{ab} = \begin{vmatrix} [-15.6, -14.6] & [-14.6, -13.6] & [-13.6, -12.6] & [-12.6, -10.52] \\ [-117.73, -110.73] & [-110.73, -103.73] & [-103.73, -96.73] & [-96.73, -88.62] \end{vmatrix}$$

$$I_{cd} = \begin{vmatrix} [-15.6, -13.6] & [-15.6, -12.6] & [-14.6, -10.52] & [-13.6, -10.52] \\ [-117.73, -103.73] & [-117.73, -96.73] & [-110.73, -88.62] & [-103.73, -88.62] \end{vmatrix}$$

$$M = \begin{vmatrix} -15.6 & -14.6 & -13.6 & -10.52 \\ -117.73 & -110.73 & -103.73 & -88.62 \end{vmatrix}$$

In the first place, it should be judged whether the evaluating index x_{ij} is located at the left or right of point M_{ih} . Then, the relative membership degree can be computed by using Equations (11)–(18).

$$U_1 = \begin{vmatrix} 0.84 & 0.63 & 0.92 & 0.72 & 0 & 0 & 0.59 & 0 & 1 & 0.69 \\ 0.87 & 0.63 & 0.98 & 0.84 & 0 & 0.23 & 0.93 & 0 & 1 & 0.48 \end{vmatrix}$$

$$U_2 = \begin{vmatrix} 0.16 & 0.38 & 0.09 & 0.28 & 0 & 0.30 & 0.42 & 0 & 0 & 0.32 \\ 0.13 & 0.37 & 0.02 & 0.16 & 0 & 0.73 & 0.07 & 0.21 & 0 & 0.98 \end{vmatrix}$$

$$U_3 = \begin{vmatrix} 0 & 0 & 0 & 0 & 0 & 0.71 & 0 & 0.43 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0.27 & 0 & 0.71 & 0 & 0 \end{vmatrix}$$

$$U_4 = \begin{vmatrix} 0 & 0 & 0 & 0 & 1 & 0.21 & 0 & 0.35 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0.29 & 0 & 0 \end{vmatrix}$$

When $\alpha = 1$ and $p = 2$, according to Equations (28) and (29), the synthetically relative membership degree matrix U can be determined as follows:

$$U = \begin{vmatrix} 0.970 & 0.736 & 0.995 & 0.911 & 0 & 0.007 & 0.849 & 0 & 1 & 0.545 \\ 0.030 & 0.264 & 0.005 & 0.089 & 0 & 0.385 & 0.151 & 0.008 & 0 & 0.455 \\ 0 & 0 & 0 & 0 & 0 & 0.585 & 0 & 0.735 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 0.023 & 0 & 0.256 & 0 & 0 \end{vmatrix}$$

The eigenvector of level H was computed according to Equation (30)

$$H = \begin{pmatrix} 1 & 2 & 3 & 4 \end{pmatrix} \times \begin{vmatrix} 0.970 & 0.736 & 0.995 & 0.911 & 0 & 0.007 & 0.849 & 0 & 1 & 0.545 \\ 0.030 & 0.264 & 0.005 & 0.089 & 0 & 0.385 & 0.151 & 0.008 & 0 & 0.455 \\ 0 & 0 & 0 & 0 & 0 & 0.585 & 0 & 0.735 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 0.023 & 0 & 0.256 & 0 & 0 \end{vmatrix}$$

$$= \begin{pmatrix} 1.089 & 1.030 & 1.264 & 1.005 & 4 & 2.624 & 1.151 & 3.248 & 1 & 1.455 \end{pmatrix}$$

The grade eigenvalues of the stable isotopes in the river were obtained according to VFEM, as shown in Table 3. The grade eigenvalues in Table 3 based on Equation (30) were from sampled river waters in the summer. According to VFEM, this paper realized the quantitative classification of stable isotopes in different river sections. The status value in different river sections was ranked from low to high as follows: 1, 1.005, 1.089, 1.151, 1.264, 1.455 and 2.624.

Table 3. The grade eigenvalues of the stable isotopes in the river.

| Evaluation of Regional | Status Value | Rating |
|------------------------|--------------|--------|
| Najinqu River | 1.089 | 1 |
| Sangqu River | 1.03 | 1 |
| Bazongqu River | 1.264 | 1 |
| Mumuqu River | 1.005 | 1 |
| Middle stream | 4 | 4 |
| Mugequ River | 2.624 | 2 |
| Chengqu River | 1.151 | 1 |
| The downstream | 3.248 | 3 |
| Gongqu River | 1 | 1 |
| Zongqingqu River | 1.455 | 1 |

4.2. Local Meteoric Water Line (LMWL) in the Naqu River Basin

The $\delta^2\text{H}$ and $\delta^{18}\text{O}$ in precipitation are currently measured at over 300 stations across the globe as part of the Global Net-work for Isotopes in Precipitation (GNIP), which cooperates with the International Atomic Energy Agency (IAEA) and the World Meteorological Organization (WMO) [29]. Despite the complexity of global hydrological processes, Craig [30] found that stable isotope ratios of hydrogen and oxygen in monthly precipitation correlate on a global scale in a linear relationship known as the global meteoric water line (GMWL):

$$\delta^2\text{H} = 8\delta^{18}\text{O} + 10$$

The best-fit line of this regression is the local meteoric water line (LMWL), which can be compared to the GMWL [31]. The isotopic characteristics of precipitation can be shown by the regression of the LMWL [32]. However, there is no suitable station having at least one full-year record in the Naqu River basin. So we analyzed the data for Lhasa to replace the LMWL because Lhasa and the Naqu River basin are located near each other in the middle of the Qinghai–Tibet Plateau, with a distance of approximately 300 km. Furthermore, the water vapor source of these two places is similar. The Lhasa LMWL can be described by the following equation [33]:

$$\delta^2\text{H} = 7.9\delta^{18}\text{O} + 6.29\text{‰}$$

The stable isotope values of precipitation are mainly determined by air temperature and moisture. An LMWL can reveal the evaporation capacity in the localities. The slope and intercept of LMWL (7.9,6.29) are both smaller than the GMWL (8,10) which shows the stable isotopic characteristics of precipitation in an arid region. The heavy isotope is enriched because of evaporation during rainfall in an arid climate. The slope of the regression line corresponds to the GMWL, indicating that condensation and precipitation occur at full equilibrium between the vapor and precipitation phases [34]. In contrast to the GMWL, the slope is slightly lower, showing drier and stronger evaporation conditions. The intercept is higher, likely due to a higher deuterium d-excess [35,36].

Deuterium excess is defined as:

$$d = \delta^2\text{H} - 8\delta^{18}\text{O}$$

It is used to measure the deviation of a set of data points from a line with a slope of 8 in $\delta^2\text{H}$ vs. $8\delta^{18}\text{O}$ plots [37]. The d-excess value of GMWL is 10. The deuterium excess varies with humidity, wind speed and sea surface temperature during primary evaporation and averages to 10‰ on a global scale [38].

4.3. Stable Isotopic Composition of Surface Water, Soil Water and Snowfall

The stable isotopic composition of the the Naqu River is highly variable and is determined by the flow of water generated by heavy rainfall, soil water, and snowmelt events. These numbers came from

the average of the sampled waters, which were from sampled river waters, soil water and snowfall. As shown in Table 4, in August, the $\delta^{18}\text{O}$ values of the river waters varied from -15.9‰ to -10.5‰ , and the $\delta^2\text{H}$ values of the river waters varied from -119‰ to -89‰ . The composition of heavier isotopes did not follow the LMWL.

Table 4. The statistical table of stable isotopic composition in the Naqu River basin.

| Time | Samples | Range of Variation (‰) | |
|---------|---------------|------------------------|--------------------|
| | | $\delta^{18}\text{O}$ | $\delta^2\text{H}$ |
| August | Surface water | -15.6 to 10.5 | -118 to -89 |
| January | Surface water | -16.6 to -9.1 | -122 to -81 |
| August | Soil water | -17.3 to -11.2 | -151 to -96 |
| January | Snowfall | -15.0 to -7.6 | -101 to -63 |

The river water line is defined as:

$$\delta^2\text{H} = 5.5964\delta^{18}\text{O} - 30.942\text{‰} \quad (R^2 = 0.94)$$

The regression line for the soil water line is:

$$\delta^2\text{H} = 7.6452 \delta^{18}\text{O} - 8.8688\text{‰} \quad (R^2 = 0.91)$$

The regression line for the snow line is:

$$\delta^2\text{H} = 4.9117 \delta^{18}\text{O} - 24.915\text{‰} \quad (R^2 = 0.91)$$

Figure 3 is related to the sampled waters. As shown in Figure 3, the slope and intercept of both the river water and soil water lines were lower than those of GMWL (8,10) and the LMWL (7.9,6.29) in August. The average deuterium excess of surface water was 3.17‰ , which was smaller than the GMWL (10‰), implying a slower evaporation process in August. The slope of soil water regression was 7.65, which was larger than the Naqu River water line (5.60). The evaporation of soil water was not obvious. The slope (4.9) and intercept (-24.9) of the snow line were lower than the slope of GMWL (8,10) and the LMWL (7.9,6.29) in the winter.

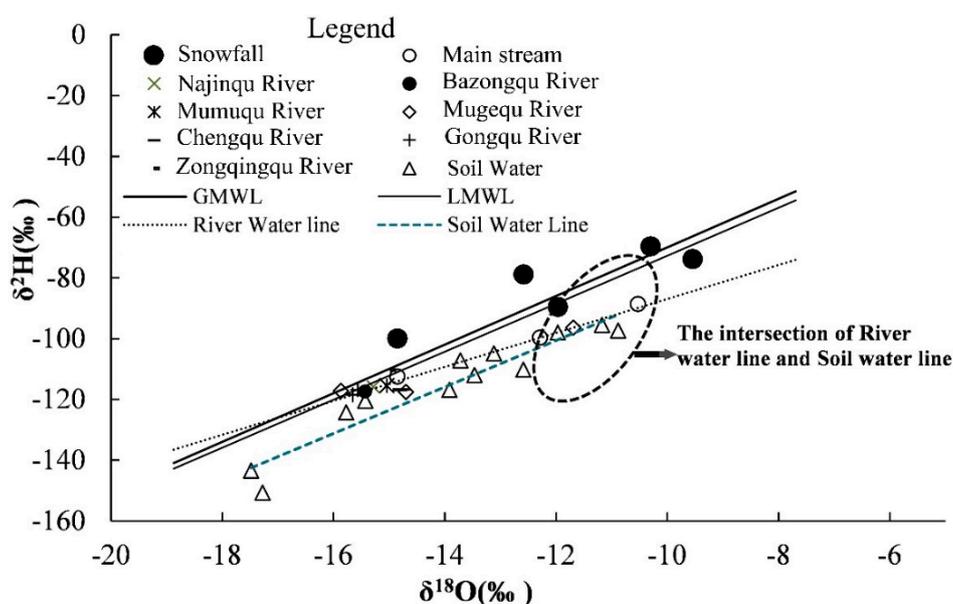


Figure 3. Plot of $\delta^2\text{H}$ vs. $\delta^{18}\text{O}$ for all water bodies in the Naqu River basin.

Compared to the LMWL, three sets of isotopic data were distinguished. The first group included data points with low $\delta^{18}\text{O}$ values and plots lay above the LMWL, which was from snowmelt. The second group, with medium $\delta^{18}\text{O}$ values, fit the LMWL, which was from river water, which means that the water originated from local rainfall. The third group with high $\delta^{18}\text{O}$ values, was below the LMWL, which was from soil water, signifying the effect of intensive evaporation processes.

5. Discussion

5.1. Analysis of Stable Isotopic Composition of Surface Water, Soil Water and Snowfall

The stable isotopic composition of the Naqu River is highly variable. There is no significant difference in $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values between river waters and soil waters. However, there is a significant difference in $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values between river waters and snowfall during the cold months, of which the mean value is -11.4‰ for $\delta^{18}\text{O}$ and -81‰ for $\delta^2\text{H}$. The characteristic analysis of the snow samples showed the enrichment of stable isotopes. We assumed that the snow samples collected were the condensation of water vapor generated by continental water vapor. In the winter, it has less precipitation and is relatively depleted because of the lower temperature and water vapor concentration. During periods of low flow in an arid environment, evaporation produces characteristic heavy-isotope enrichment in residual surface waters.

In arid or semi-arid climates, the stable isotopic composition of surface waters can be modified by non-equilibrium evaporation, leading the more highly-evaporated river waters to deviate substantially from the LMWL. In this non-equilibrium process, the boundary layer effects induce the kinetic fraction of hydrogen and oxygen isotopes, which acts unequally on both isotopes. After the evaporation of waters, the heavier isotopes (^{18}O and ^2H) in the remaining water are enriched. As shown in Figure 3, the slope and intercept of both the river water and soil water lines were lower than those of the GMWL (8,10) and LMWL (7.9,6.29) in August, indicating that precipitation after evaporation was the main source of river water in the study area. The average deuterium excess of surface water was 3.17‰ , which was smaller than the GMWL (10‰) implying a slower evaporation process in August. The intersection of the Naqu River water line and soil water line indicates that the surface water of the Mugequ River is replenished by the soil water. The slope of soil water regression was 7.65, which was larger than the Naqu River water line (5.60), indicating that surface water evaporated after soil water infiltration. The evaporation of soil water was not obvious. We assumed that soil water was replenished by rainfall. The slope (4.9) and intercept (-24.9) of the snow line were lower than the slope of the GMWL (8,10) and LMWL (7.9,6.29) in the winter. We assumed that the snow samples collected were the condensation of water vapor generated by continental water vapor, and that the river was only replenished by soil water in the winter. After the monsoon retreat, the Naqu River basin is mainly affected by continental water vapor, the atmospheric humidity is reduced, and the heavy isotope values of atmospheric water vapor are relatively high.

The study in the Naqu River basin showed the significant difference in mean values of both $\delta^2\text{H}$ and $\delta^{18}\text{O}$ along the river [38]. Therefore, the slope and intercept of the LMWL were lower than those of the refined GMWL. This deviation from the GMWL was mainly due to the different source of water replenishment with evaporation-induced kinetic fractionation effects [29]. Evaporation significantly affected both $\delta^2\text{H}$ and $\delta^{18}\text{O}$ values of flowing water and soil water, which were recharged by early snowmelt in the summer.

Compared to the LMWL, three sets of isotopic data are distinguished. Data points above the LMWL represent the surface water that mainly originated from snowmelt in mountains with low temperatures and absolute moisture content of air. Because the water exiting from the snowpack first during snowmelt was highly depleted isotopically, most of the river water plots above the LMWL were from the early melting water of snowpack and had depleted isotope values owing to the fractionation effect during the melting process [39]. The data fitting the LMWL means that the water

was supplemented by local rainfall. The last group, which was from Tsonag Lake water and the main stream, was below the LMWL, signifying the effect of intensive evaporation processes (Figure 3).

Figure 3 shows the relationship between $\delta^2\text{H}$ and $\delta^{18}\text{O}$ values of precipitation in the Naqu River basin. Referring to the LMWL, there was a higher evaporation process in the main stream and the Mugequ River. The intersection of the Naqu River water line and soil water line indicated that the surface water of Mugequ River is replenished by the soil water. $\delta^2\text{H}$ and $\delta^{18}\text{O}$ values between the Mugequ River and soil water were at the same level, which indicates the resupply between river and soil water. The Najinqu, Bazongqu, Mumuqu, Gongqu and Zongqingqu Rivers showed a lower level of heavy isotopes, which was the opposite of soil water, signifying the recharge of precipitation or snowmelt.

5.2. Estimation in Classification of Stable Isotopes in the Naqu River Basin Based on the Variable Fuzzy Evaluation Method

As shown in the Figure 4, it could be seen that the status values of the Najinqu, Bazongqu, Mumuqu, Chengqu and Gongqu Rivers were small, indicating that these tributaries were strongly supplemented by precipitation and snowmelt. The level of status value in the Mugequ River was 2.62 with a high level. In the lower reaches of the lake, the stable isotopes in the river were more abundant due to the evaporation of the lake and the role of stable isotopic fractionation.

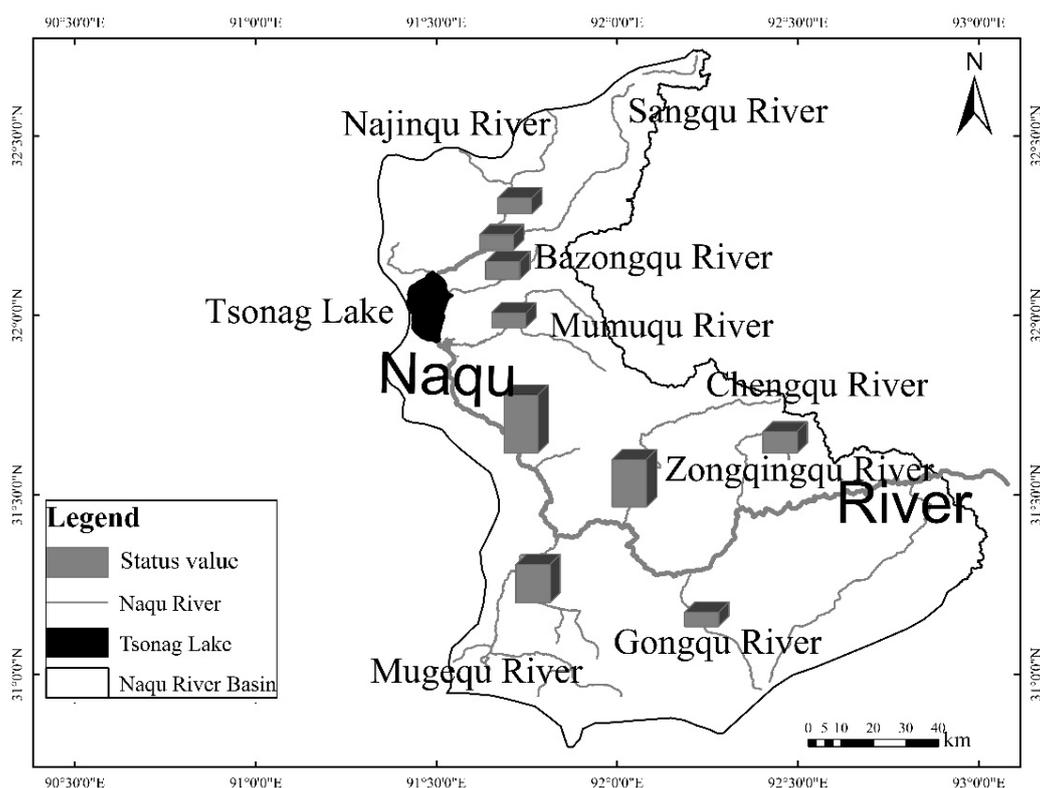


Figure 4. Spatial variation of the status values in the Naqu River basin.

The present paper developed a framework for analyzing stable isotopes according to VFEM, and generalized the source identification of water replenishment by rainfall or snowmelt in the Naqu River basin. This approach combined $\delta^2\text{H}$ and $\delta^{18}\text{O}$ together for the comprehensive evaluation of stable isotopes. Thus, the runoff replenishment by rainfall or snowmelt in the river can be visually determined by the status value of stable isotopes.

The contribution of this work is to transform double indices into the status value for the classification of stable isotopes in the source identification of water replenishment. For example, $\delta^2\text{H}$ and $\delta^{18}\text{O}$ were used for evaluating the source of water [12–14] by using the change in stable

isotopic composition due to evaporation or mixing by the different waters, which mainly focused on the exploration of the characteristics of water isotopes; Zhang et al. [21] investigated the variation of $\delta^2\text{H}$ and $\delta^{18}\text{O}$ in glacial meltwater at Mount Everest and found that the $\delta^2\text{H}$ in ice and snow was larger than in river and lake water relative to the change of $\delta^{18}\text{O}$, and $\delta^2\text{H}$ and $\delta^{18}\text{O}$ studied independently. This study mainly highlighted the discernible speed of runoff replenishment by rainfall or snowmelt based on the combination of the classification of stable isotopes and plot of $\delta^2\text{H}$ vs. $\delta^{18}\text{O}$ in the Naqu River basin.

However, according to VFEM and the results adopted in this paper, the rationality of the demarcation of the index standard value needs to be further improved. Furthermore, the data of stable isotope samples of river water, soil water and precipitation might need to be enlarged.

6. Conclusions

The classification of stable isotopes was obtained based on VFEM in the Naqu River basin. The grade eigenvalues in different river sections were ranked from low to high as follows: 1, 1.005, 1.089, 1.151, 1.264, 1.455 and 2.624. Compared to the LMWL, three sets of isotopic data were distinguished based on the plots of $\delta^2\text{H}$ vs. $\delta^{18}\text{O}$ because of different sources of water replenishment in the Naqu River basin. In the winter, snow is produced by the condensation of water vapor generated by continental water vapor, and the river is only replenished by soil water in the winter. In the summer, the status values of the Najinqu, Bazongqu, Mumuqu, Chengqu and Gongqu Rivers were small, indicating that these tributaries were strongly supplemented by precipitation and snowmelt. The level of status value in the Mugequ River was 2.62 with a high level. In the lower reaches of the lake, the stable isotopes in the river were more abundant due to the evaporation of the lake and the role of stable isotopic fractionation.

The present paper developed a framework of VFEM to provide a new method for the classification of stable isotopes. This research will be helpful for the study of runoff replenishment and the evolution mechanism in the Qinghai-Tibet Plateau. Furthermore, long-term and synchronous analysis of precipitations and runoff should be evaluated to enable a better understanding of runoff replenishment in the Naqu River basin.

Author Contributions: X.C. and F.W. conceived, designed, and drafted the manuscript; X.C. and G.W. planned and designed the methodology; X.C. revised the manuscript; F.W. guided and supervised the whole process; and all authors read and approved the final manuscript.

Funding: This work was supported by the Major Research Plan of the National Natural Science Foundation of China (91547209), National Key Research and Development Program of China (2016YFC0401401), the National Natural Science Foundation of People's Republic of China (51579101,51709111), and the Distinguished Young Scholar of Science and Technology Innovation (184100510014).

Acknowledgments: The authors would like to express their sincere gratitude to the anonymous reviewers for their constructive comments and useful suggestions that helped us improve our paper.

Conflicts of Interest: No conflict of interest exists in the submission of this manuscript, and the manuscript is approved by all authors for publication.

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