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Source variability of sediments in the Shihmen Reservoir, Northern Taiwan: Sr isotopic evidence

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ABSTRACT

High turbidity in water reservoirs during the monsoon season has caused serious problems for drinking water quality in Taiwan. In this study, we collected stream waters and streambed sediments, including bed loads and fresh landslide deposits, in the Dahan Stream and analyzed 101 $^{87}Sr/^{86}Sr$ ratios in different grain-size fractions in the streambed sediments from the upper catchment and 3 sediment cores from the Shihmen Reservoir, as well as dissolved major and trace elements in stream waters, to identify possible changes of sediment sources in the reservoir. The $^{87}Sr/^{86}Sr$ ratios in stream water and streambed sediments ranged from 0.71296–0.71349 and 0.71714–0.71843, respectively. The large Sr isotopic difference between the paired stream water and streambed sediments are evidence for disequilibrium water/rock interaction in the weathering limited regions. The Sr isotopic compositions in bulk sediments and various grain-size fractions showed no clear spatial variability. However, there is an interesting relationship between grain sizes and Sr isotope ratios, implying effects of mineral sorting during sediment transportation. The illite crystallinity and the illite chemistry index of streambed sediment suggest moderate to strong chemical weathering in this regions. The dissolved constituents in the Dahan stream support that silicate weathering is the predominated controlled mechanism and only minor carbonate dissolution occurred near the Central Mountain Range. The $^{87}Sr/^{86}Sr$ ratios in the sediment cores suggest that modern reservoir sediment is mainly derived from the upper watershed, composed of primarily Oligocene sedimentary rocks. However, the sediment sources have changed significantly since the reservoir built 37 years ago, the $^{87}Sr/^{86}Sr$ ratios were spread widely outside the present-day observations in the Dahan catchment. The homogeneous distribution of $^{87}Sr/^{86}Sr$ in the upper reservoir cores reflected disturbance due to recent turbidity events within the reservoir. These new chemical and isotopic data provide useful spatial and temporal information of weathering sources in a high denudation sub-tropical mountainous watershed.

1. Introduction

Quantifying the mechanisms that controlling the chemical composition of surface waters has received strong attention in recent years, these dissolved products of weathering will be transported into the oceans eventually. Previous geochemical investigations in large river systems have focused on the chemical weathering and physical erosion rates from various types of catchments (Gaillardet et al., 1999; Martin and Meybeck, 1979; Negrel et al., 1993). Climatic variability, lithology, regional tectonic, hill-slope morphology and biological activity all impact the degree of weathering. However, other factors affect also the dissolved constituents in waters including atmospheric inputs, rock weathering, water/sediment interactions, redox condition, biological cycles, and anthropogenic pollution in watersheds (Gibbs, 1970; Li, 1992; Meybeck, 1987; Ozturk, 1995; Stallard and Edmond, 1983).

Strontium (Sr) isotopes are useful geochemical tracers for gaining a better understanding of water/rock interactions, water mass sources and their migration pathways in nature (Bain and Bacon, 1994; Bullen et al., 1996; Palmer and Edmond, 1992). The Sr isotopic variations in nature are produced mainly by the radioactive decay of ^{87}Rb to ^{87}Sr (Faure, 1986). This ratio remains in equilibrium and unchanged in the weathering products due to the long half-life of ^{87}Rb (half-life = 48.8 Ga, Banner, 2004) when compared with the time scales of sediment genesis and transport. More importantly, Sr isotopic ratios are not fractionated by low-temperature abiotic chemical reactions, phase separation, evaporation, or biological assimilation (Graustein, 1989). Hence, $^{87}Sr/^{86}Sr$ in surface waters reflects regional chemical weathering process and the mixing of water masses. The $^{87}Sr/^{86}Sr$ in minerals are dominantly controlled

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by the ages and the Rb/Sr ratios in source rocks, and influence the Sr isotopic compositions in surface waters during chemical exchange processes. Land et al. (2000) showed that differential weathering susceptibility of minerals in substrates imparted distinct $^{87}Sr/^{86}Sr$ ratios to different mixing processes with temporal variation. The weathering of Rb enriched minerals, such as micas and feldspars, will increase $^{87}Sr/^{86}Sr$ ratios in surface water (Aberg et al., 1989; Aubert et al., 2002; Bain and Bacon, 1994; Blum et al., 1993; Land et al., 2000). The $^{87}Sr/^{86}Sr$ ratios in riverine suspended particles and marine sediments have been used to identify provenance of sediments in freshwater and estuarine ecosystems, and further to study particle-associated material dynamics (Douglas et al., 2003; Ingram and Lin, 2002; Singh and France-Lanord, 2002). The relationships between grain size and particle Sr isotopic composition have been emphasized in the marine sediments, and were correlated with the particle Rb/Sr ratio and mineralogical compositions (Eisenhauer et al., 1999; Revel et al., 1996). However, the particle size relationships in streambed sediments Sr isotopes are not well known in watersheds with high denudation rates.

Taiwan is situated at the collision boundary between the Philippine Sea Plate and the Eurasian Plate. As a typical active orogenic belt, it has the highest uplifting rate, average 2–10 mm $year^{-1}$ in the world (Liu and Yu, 1990). Similarly, the rates of physical denudation and chemical weathering in this region are both the highest in Asia (Dadson et al., 2003; Li, 1976; You et al., 1988). The Central Mountain Range (CMR) located in central Taiwan has a well developed radial drainage system which transports riverine materials to the coast through the Danshuei River, the largest watershed in northern Taiwan. Some aspects of water chemistry in the upper parts of the watershed have been previously studied and results showed that solute concentrations were predominately controlled by the degree of ambient rock weathering and that the seasalt contribution increases downstream to the estuary (Chu and You, 2007). The Dahan catchment located at the upward watershed of the Danshuei River drainage system has an average altitude of 2000 m with steep slope. The Shihmen Reservoir effectively accumulates transported riverine sediments and freshwater. The purpose of this study is to analyze dissolved constituents and Sr isotopic composition in the Dahan Stream and Danshuei River system to identify the factors controlling water chemistry. We were focused to delineate the relationship between grain size and Sr isotopic compositions, and to trace the provenance of sediment in the Shihmen Reservoir since it was built 37 years ago.

2. Study area

The Shihmen Reservoir (135 m in altitude, operating since 1974) is the largest artificial dam of drainage area and water storage in the northern Taiwan which supplies drinking and industrial water to Taipei basin with a few millions of residents (Fig. 1). The Dahan Stream headwater is the main drainage system to supply water for the reservoir and originates from the eastern part of CMR at 3514 m. The drainage area is approximately 1163 km^2, 135 km in mainstream length, and 62.1 $m^3 s^{-1}$ of average discharge rate. The Dahan Stream consists of 2 major headwater tributaries, Baishih River and Thaigang River (note that the Yufeng River is another name for the upper Dahan Stream). This catchment is located in a subtropical climate region, the lowest temperature is 10.8 °C in January and the highest temperature is 23.2 °C in July. The mean annual precipitation in the watershed is up to 2382 mm, focused in May–October (Yu and Wang, 2009).

In early Pleistocene, strong orogenic activity uplifted the original sedimentary basin as part of CMR and resulted in complicated geological formations to form 2 main faults and 3 folds in the drainage area which caused >60.6% of the catchment is steeper than 55° in slope. After the event of 7.3 magnitude Chi–Chi earthquake (1999), there were frequent landslides occur widely inside the Dahan catchment. Thus, in rain seasons and typhoon events, sometimes even with small local rainfall in mountain area, abundance fine-grained suspended materials and bed load sediments would be supplied to the downstream and the reservoir.

The main stratigraphic units in the Dahan catchment are composed of low-grade metamorphic sediments containing slate, shale, sandstone and alluvial sand in early Miocene, Oligocene and Eocene formation. The stratum ages and the grade of metamorphism are older and stronger from the west part catchment to the nearby CMR. The Szeleng Sandstone (15.3%) is the oldest Eocene metamorphic sandstone with slate and shale; the Tatungshan Formation (50.7%) is the major strata unit in entire catchment and consists of Oligocene argillite, sandy shale sandstone with little metamorphic sediment; the Taliao and Mushan Formation (14.8%) and Nangang Formation (13.4%) are composed of Miocene shale; and others are small modern terrace deposits.

3. Methodology

Seven pairs of stream waters and streambed sediments were collected in the mainstream and the tributaries in the Dahan catchment in April and October 2006. Additional streambed sediments were collected from 8 landslide sites and smaller tributaries along the mainstream channel in October 2007. Three sediment cores (about 20 m in length) were collected in the Shihmen Reservoir in 2008. Sediment cores were sub-sampled at 200 cm intervals (core 2) or 10 cm (cores 3 and 4). All sample pretreatment, chemical analysis, and Sr isotopic analysis were carried out in the Isotope Geochemistry Laboratory (IGL) at National Cheng Kung University, Taiwan. The clay mineralogical compositions were analyzed by the State Key Laboratory of Marine Geology at Tongji University, China. Detailed procedures for clay determination are given elsewhere (Liu et al., 2008).

3.1. Sample pretreatment

All experimental procedures conducted in this study were carried out in a clean room (class 1000–10000 or class 10 working benches). Water samples were filtered through 0.45 μm nylon membranes immediately after collection and stored in acid-cleaned PE bottles, then acidified with purified nitric acid (purified from GR grade acid by laboratory) before 4 °C preservation. Streambed sediments and core segments were sorted into 5 different size fractions (<0.25 mm, 0.25–0.35 mm, 0.35–0.5 mm, 0.5–0.59 mm and >0.59 mm). Large stones or pebbles were handpicked before wet sieving. Streambed particles were pulverized. Approximately 100 mg of sediment (dry weight) was dissolved in mixed acids as following procedures. 2 ml 35% supra pure H_2O_2 was added to remove organic matter and dried overnight. Then, samples were transferred to clean Teflon digestion vessels using 2 ml HF (~20 N) and 5 ml purified HNO_3 acid (~10 N). Microwave digestion was done using a CEM MARS 5 digestion oven. The digested solutions were evaporated to dryness, and then heated with 3 ml purified concentrated HCl acid for several hours. This procedure was repeated if a sample showed signs of incomplete digestion.

3.2. Major and trace elements and Sr isotopic measurement

Major elements, Rb and Sr concentrations were measured using a high resolution sector field inductively coupled plasma mass spectrometer (Thermo Scientific, Element 2) with analytical precision better than 2%. The Principal component analysis (PCA)