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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 hand-picked 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)

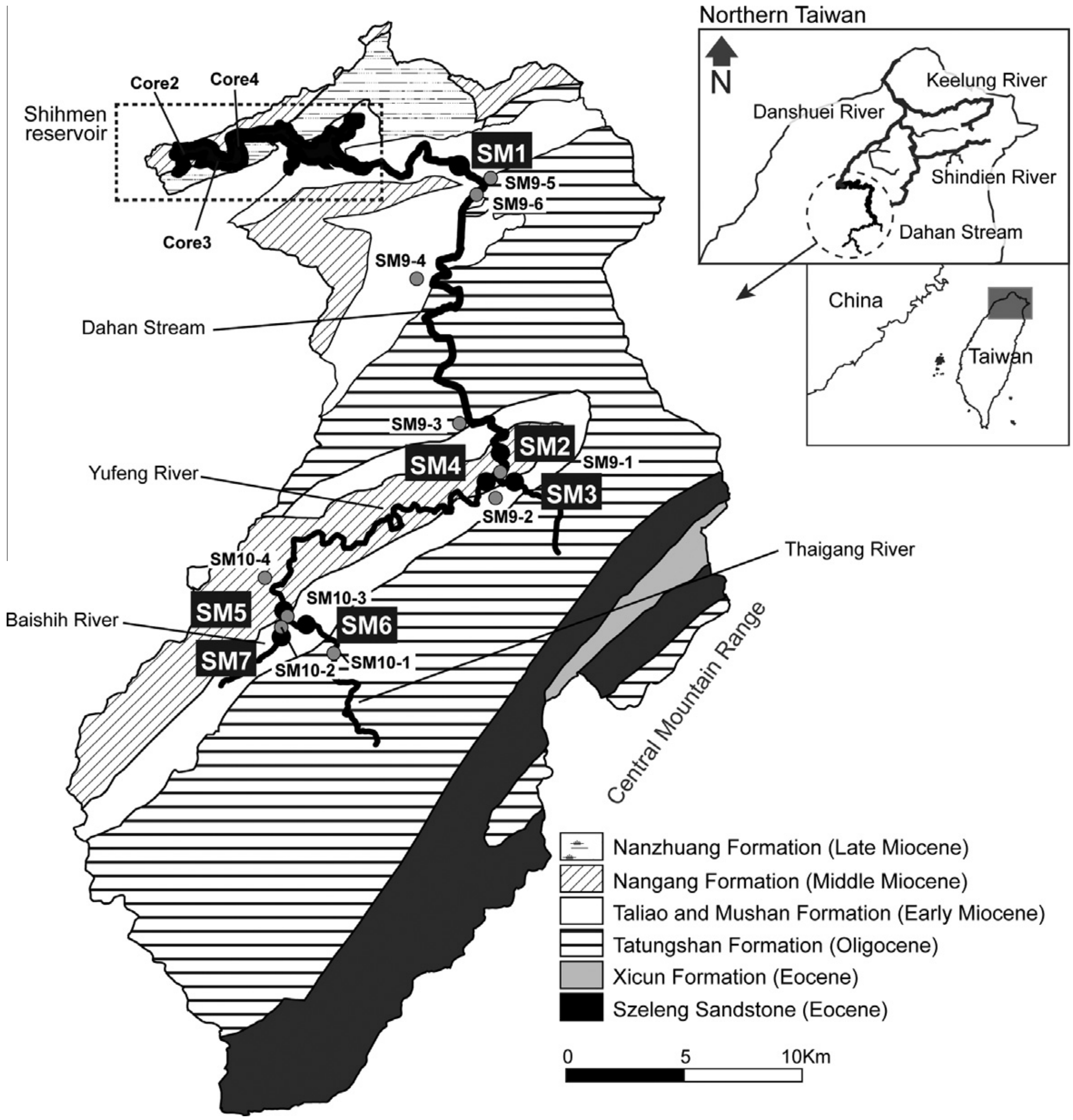

Fig. 1. Location map showing the geological setting and various sampling sites in Shihmen Reservoir and Dahan catchment. Three sediment cores were collected inside the reservoir, water samples and streambed sediments (bold face) were sampled in the main stream and tributaries.

technique was applied to evaluate the domain components to affect the chemical compositions in stream water. Factor analysis (FA) was performed using statistical analysis software SPSS 17.0 (SPSS Inc. USA). The major ions and Sr in stream waters were selected as variance elements, the varimax rotation was carried out and the maximum number of factors was set to extract under the accumulation of 95% of total variance.

For Sr isotopic determination, purification procedure was carried out using Eichrom Sr^{spec} resin (bed volume about 0.6 ml) on class-10 working benches to isolate the matrix components and Rb. The $^{87}Sr/^{86}Sr$ ratios were measured by thermo ionization mass spectrometer (Thermo Scientific, Triton TI) or multi-collector inductively coupled plasma mass spectrometer (Thermo Scientific, Neptune). Both mass spectrometers equipped with 8 movable Faraday cups ($10^{11}\ \Omega$ resistors) and one fixed central cup. ^{84}Sr, ^{86}Sr, ^{87}Sr, and ^{88}Sr ion-beams were collected simultaneously in a static mode, and ^{85}Rb ion-beam was monitored for correction of ^{87}Rb contribution. ^{83}Kr ion-beam was monitored for ^{86}Kr correction (assumed 0.664740 for $^{83}Kr/^{86}Kr$) in Neptune measurements. The gains of amplifiers were calibrated daily and the baselines of detectors were measured in each analytical session. A virtual amplifier technique was used to eliminate any amplifier bias. The $^{86}Sr/^{88}Sr$ isotopic ratios were normalized to 0.1194 where potential mass fractionation artifact was corrected by exponential law. Long-term replicated measurement of NIST SRM987 standard showed an average $^{87}Sr/^{86}Sr$ of 0.710247 ± 10 (2σ) for Triton TI and 0.710275 ± 20 (2σ) for Neptune, respectively.

4. Results

4.1. Elemental and isotopic compositions

Results for surface water Na, Mg, Ca, SO_4, K, Cl, Si, and Sr concentrations are listed in Table 1. The part of major ions, Na showed a clear spatial variance with the highest concentration in downstream site and the lowest concentration in the most up-stream. For the trace element Sr varied between 1.5 μM and 2.4 μM. All

Table 1
Major elements, Sr and $^{87}Sr/^{86}Sr$ in stream waters in Dahan catchment.

Site	Sampling date	Na (μM)	Mg (μM)	Ca (μM)	SO_4 (μM)	K (μM)	Sr (μM)	Si (μM)	Cl (μM)	$^{87}Sr/^{86}Sr$
SM 1	2006/4/6	213	219	314	91	22.3	1.5	160	102	0.712956
SM 2	2006/4/6	215	330	551	179	23.8	2.4	121	34	0.713293
SM 3	2006/4/6	188	159	281	86	12.2	1.5	123	67	0.713170
SM 4	2006/4/6	173	289	485	187	23.3	2.2	98	26	0.713491
SM 5	2006/4/6	173	247	416	152	19.9	2.1	95	66	0.712966
SM 6	2006/4/6	164	254	396	142	18.0	2.3	112	66	0.713203
SM 7	2006/4/6	193	302	543	246	23.6	2.1	105	73	0.713103

Site	Na/Cl	Mg/Cl	Ca/Cl	SO_4/Cl	K/Cl	Sr/Cl	References
SM 1	2.10	2.15	3.09	0.89	0.22	0.01	
SM 2	6.37	9.75	16.31	5.30	0.71	0.07	
SM 3	2.82	2.38	4.21	1.29	0.18	0.02	
SM 4	6.57	10.98	18.44	7.10	0.89	0.08	
SM 5	2.63	3.74	6.30	2.31	0.30	0.03	
SM 6	2.49	3.87	6.02	2.16	0.27	0.03	
SM 7	2.63	4.12	7.40	3.35	0.32	0.03	
World mean rivers	1.34	0.64	1.58	0.52	0.15	3.43	Meybeck (1987)
Seawater	0.86	0.10	0.02	0.05	0.02	0.0002	IAPSO (OSIL, UK)
Taipei acid rain				0.40			Li et al. (1997)

Table 2
Results of principal component analysis of stream chemistry.

	Factor 1	Factor 2	Factor 3
Na	−.119	**.916**	−.267
Mg	**.945**	.299	−.009
Ca	**.971**	.188	.010
SO4	**.908**	.050	.305
K	**.707**	.597	.164
Sr	**.926**	−.148	−.081
Si	−.650	**.707**	−.122
Cl	−.689	.330	.625
%Var[a]	61.60	24.42	7.55

Bold face type indicates values of factor loadings higher than 0.7.
[a] Indicates percentage of total variance.

Table 3
Clay minerals, illite crystallinity, and illite chemistry index in Dahan catchment.

Site	Compositions of clay minerals				Illite crystallinity (°Δ2θ)	Illite chemistry index
	Smectite%	Illite%	Kaolinite%	Chlorite%		
SM1	0.0	57.7	0.0	42.3	0.21	0.33
SM2	0.0	57.9	0.0	42.1	0.24	0.32
SM3	0.0	45.2	0.0	54.8	0.18	0.41
SM4	0.0	60.0	0.0	40.0	0.23	0.32
SM5	0.0	54.1	0.0	45.9	0.30	0.40
SM6	0.0	59.0	0.0	41.0	0.25	0.33
SM7	0.0	61.7	0.0	38.3	0.36	0.34
SM9–2	0.0	60.3	0.0	39.7	0.26	0.30
SM9–3	0.0	79.0	0.0	21.0	0.31	0.33
SM9–5	32.4	44.1	0.0	23.5	0.29	0.38
SM10–1	0.0	56.2	0.0	43.8	0.27	0.41
SM10–3	28.6	52.9	0.0	18.5	0.42	0.38
SM10–4	0.0	87.5	0.0	12.5	0.48	0.46

the element/Cl ratios significantly shifted away from the values of seawater and world mean rivers (Meybeck, 1987). We used PCA to examine the main factors affecting the chemical composition of stream water and the results are summarized in Table 2. The first 3 factors can explained 94% of the total variance, in particular the former 2 factors (86% of the total) showed lots of variance elements' factor loading value higher than 0.7.

The Sr isotopic data are listed in Tables 1 and 4, where the $^{87}Sr/^{86}Sr$ ratios in stream waters, bed loads, and sediment cores are fall in a range of 0.71296–0.71349, 0.71714–0.71843, and 0.71187–0.72647, respectively. According to the ANOVA test of sampling site (the strata) and Sr isotopic ratios, the sediments sampled in Oligocene strata show the most radiogenic Sr isotopic compositions and the less radiogenic values were appeared in the sediments collected from early to middle Miocene strata. Stream waters $^{87}Sr/^{86}Sr$ ratios are plotted against Cl/Sr ratios and show a mixing trend in the catchment (Fig. 6). Dissolved Sr isotopes display high correlation with the corresponding bed loads compositions (Fig. 7). The $^{87}Sr/^{86}Sr$ ratios in the size separated bed loads are spread in the range of (1) 0.71709–0.72021 for <0.25 mm fraction, (2) 0.71724–0.72007 in 0.25–0.35 mm, (3) 0.71606–0.71986 in 0.35–0.5 mm, (4) 0.71606–0.71915 in 0.5–0.59 mm, and (5) 0.70967–0.72096 in >0.59 mm (Fig. 5). There are significant Sr isotopic differences in the bulk sediments at the same sampling sites (see Table 4 for details).

4.2. Clay minerals

The clay assemblages in the Dahan streambed sediments are illite and chlorite, with average abundances of 59.7% and 35.7%, respectively (Table 3). Smectite is very rare and only found in SM9-5 and SM10-3, average value of 30.5%. Kaolinite was not found in any sample. The relative proportions of clay contents showed no significant changes by location in the watershed. The illite crystallinity and illite chemistry index were calculated from the X-ray diffractograms. The illite crystallinity was obtained from half height width of the 10 Å peak and illite chemistry index were defined as the ratio of the 5 Å and 10 Å peak. The illite crystallinity in the clay collected from the Dahan catchment ranged from 0.18–0.48°Δ2θ with mean value of 0.29°Δ2θ, and the illite chemistry index ranged from 0.30–0.46, average 0.36.

4.3. Particle size of streambed sediments

The particle sizes of streambed sediments in the Shihmen Reservoir sediment cores are all smaller than 0.25 mm, fine sand (Table 5). However, sediments collected from the upstream area of the Dahan watershed are poorly sorted. The high percentage of coarse sands in samples from the upper area causes difficulty for grain size analysis. All particle size distributions of the streambed and landslide sediments were calculated via the dry weight data and detailed are given in Table 4 together with the information of Sr isotopic compositions.

Table 4
$^{87}Sr/^{86}Sr$ in streambed sediments in Dahan watershed.

Site	Sampling date	>0.59 mm (>30 mesh)		0.50–0.59 mm (30–35 mesh)		0.35–0.50 mm (35–45 mesh)		0.25–0.35 mm (45–60mesh)		<0.25 mm (<60mesh)		Bulk Sediment
		$^{87}Sr/^{86}Sr$	Wt (%)	$^{87}Sr/^{86}Sr$	Wt (%)	$^{87}Sr/^{86}Sr$	Wt (%)	$^{87}Sr/^{86}Sr$	Wt (%)	$^{87}Sr/^{86}Sr$	Wt (%)	$^{87}Sr/^{86}Sr$
Tatungshan Formation (Oligocene)												
SM 9–3	2006/9/20	0.71895	44	0.71953	4	0.71985	26	0.72007	11	0.72021	15	0.71951[a]
SM 9–4	2006/9/20	0.72096	100									0.72096[a]
SM 10–1	2006/10/24	0.71859	86	0.71794	1	0.71814	3	0.71818	4	0.71782	6	0.71851[a]
											Average	0.71966
Taliao and Mushan Formation (Early Miocene)												
SM 1	2006/4/6	0.71602	7	0.71772	3	0.71774	8	0.71787	18	0.71776	64	0.71785
SM 3	2006/4/6	0.71810	81	0.71809	8	0.71798	6	0.71762	3	0.71780	1	0.71771
SM 6	2006/4/6	0.71915	44	0.71844	17	0.71896	21	0.71795	12	0.71798	6	0.71843
SM 7	2006/4/6	0.71710	14	0.71844	7	0.71687	57	0.71724	12	0.71751	9	0.71731
SM 9–2	2006/9/20	0.71831	30	0.71819	20	0.71813	35	0.71820	11	0.71811	5	0.71820[a]
SM 9–5	2006/9/20	0.71671	39	0.71606	6	0.71737	23	0.71766	14	0.71858	18	0.71730[a]
SM 9–6	2006/9/20									0.71798	100	0.71798[a]
SM 10–2	2006/10/24	0.71843	37	0.71825	4	0.71778	12	0.71790	23	0.71782	24	0.71807[a]
SM 10–3	2006/10/24	0.71989	100									0.71989[a]
											Average	0.71808
Nangang Formation (Middle Miocene)												
SM 2	2006/4/6	0.71883	28	0.71915	22	0.71841	32	0.71885	7	0.71826	11	0.71794
SM 4	2006/4/6	0.71965	6	0.71855	37	0.71890	38	0.71806	11	0.71827	8	0.71802
SM 5	2006/4/6									0.71808	100	0.71714
SM 9–1	2006/9/20	0.71821	77	0.71794	4	0.71318	8	0.71723	7	0.71732	5	0.71771[a]
SM 10–4	2006/10/24									0.71709	100	0.71709[a]
											Average	0.71758

[a] Represents the $^{87}Sr/^{86}Sr$ ratios calculated data based on the size separation samples.

Table 5
$^{87}Sr/^{86}Sr$ in Shihmen Reservoir sediment cores.

Depth (cm)	Grain size	$^{87}Sr/^{86}Sr$[a]
SMC Core 2		
20	<0.25 mm	0.72002
220	<0.25 mm	0.72306
420	<0.25 mm	0.71524
620	<0.25 mm	0.71667
820	<0.25 mm	0.71187
1020	<0.25 mm	0.72063
1220	<0.25 mm	0.71318
1420	<0.25 mm	0.71537
1620	<0.25 mm	0.72647
1820	<0.25 mm	0.71801
SMC Core 3		
20	<0.25 mm	0.71832
30	<0.25 mm	0.71907
40	<0.25 mm	0.71793
50	<0.25 mm	0.71808
60	<0.25 mm	0.71906
SMC Core 4		
10	<0.25 mm	0.71663
20	<0.25 mm	0.71762
30	<0.25 mm	0.71798
40	<0.25 mm	0.71755

5. Discussion

5.1. Water chemistry in the Dahan catchment

The factors that might to exert control on dissolved solutes in Dahan Stream water chemicals include (1) the type and different degree of local bedrock weathering; (2) seasalt input; (3) anthropogenic pollution. Concentrations of Na, Mg, Ca, K, SO_4 and Sr in streams in the Dahan catchment show large variation in concentrations and the element/Cl values shift away from the world-wide average value for river water (Meybeck, 1987) and seawater (IAPSO, OSIL, UK). Because there are no Cl-bearing minerals in this area, dissolved Cl is assumed to be entirely originated from seasalt and used for marine contribution calculation. The non-seasalt fraction is denoted as ns-X = X − Cl*(X/Cl)$_{seawater}$, where X is the given elemental concentrations in stream waters, and (X/Cl)$_{seawater}$ is the element/Cl ratio in seawater (Li et al., 1997). For most elements, the marine contributions are <10%. However, significant Na (15% in average, about 40% in most downstream site) is derived from seasalt. The dissolved Na is functions of distance from western to eastern Central Mountain, indicating a diminishing seasalt input away from the coast (Li et al., 1997). Additionally, this area suffers less human perturbation as the mean altitude is higher than 2000 m. These dissolved SO_4/Cl ratios differ significantly from the rain waters collected from Taipei basin (Li et al., 1997), indicates minor contribution from the urban pollutions. Consequently, the chemical compositions in the Dahan Stream waters are mainly driven by the local bedrock weathering (Fig. 2).

The non-seasalt fraction of dissolved Mg, Ca and HCO_3^- in rivers is mainly due to carbonate and silicate weathering in Taiwan (Chu and You, 2007; Chung et al., 2009; Li et al., 1997). The ns-Mg and ns-Ca normalized by ns-Na in the Dahan Stream water suggests about 90% total cation is silicate weathering origin (Fig. 4). A highly positive linear relationship is observed in the plot of ns-Ca/ns-Na and ns-Mg/ns-Na, range of 2.14–4.16 and 1.17–2.31, respectively. The result shows relative strong carbonate weathering compared with others downstream rivers of the Shindien River and the Keelung River (Fig. 4). Strong carbonate dissolution occurs normally in high denudation mountain areas (Jacobson et al., 2003). In the Dahan Stream, the dissolved chemical constituents are mainly derived from silicate weathering, although the carbonate contribution increases gradually towards the CMR area. We used multivariate statistics to evaluate the covariance of elemental compositions in surface water to understand the potential controlling factors to influence the water chemistry. Two major sources, the value of factor loading is larger than 1, were identified in the upstream of the Dahan catchment. These two factors explain 86.0% of total variances. The first factor (61.6% of the total) shows a high correlation among Mg, Ca, SO_4, Sr, and K. A linear relationship

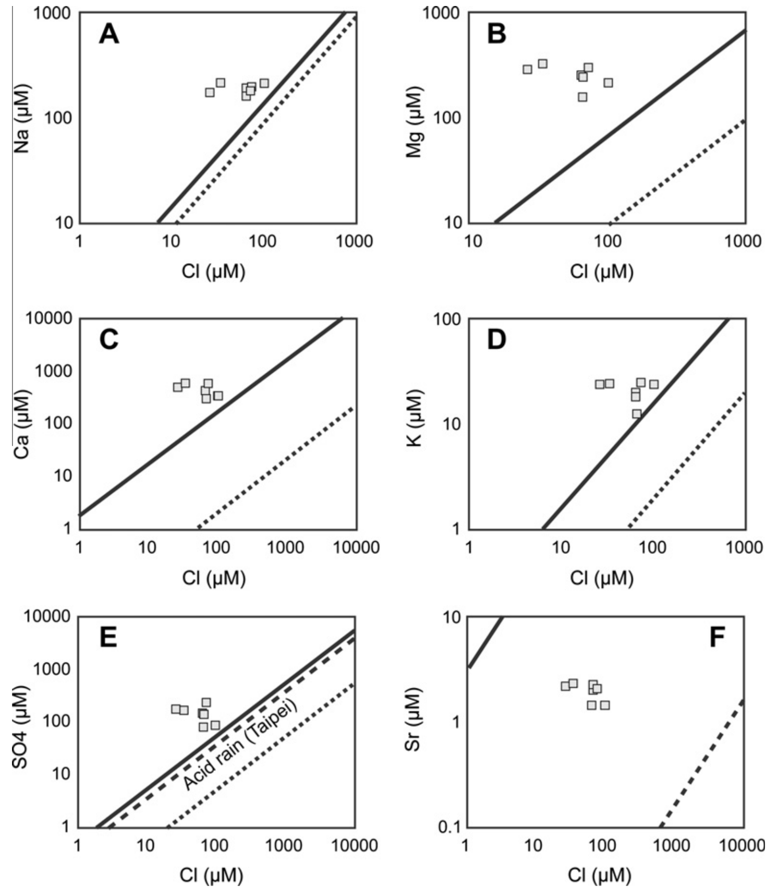

Fig. 2. Scatter plots for dissolved constituents (X) versus Cl. The solid line (river water) and the dotted line (seawater) represent the world mean X/Cl ratio. In (E), the dashed line shows the SO_4/Cl ratio rain in the Taipei basin (Li et al., 1997).

between ns-Mg, ns-Ca, or ns-Mg + ns-Ca and ns-SO_4 indicates possible weathering of sulfide minerals in alumino-silicate rocks (Fig. 3), i.e. schists and gneiss with outspread pyrite clastic in the CMR (Ho, 1975). Carbonate weathering of marbles and limestone will release Ca and Mg into river water. However, marbles are seldom found in northern Taiwan, the carbonate weathering contribution might be derived from dissolution of secondary calcite, marine carbonate deposits in the past, or carbonate veins in the source rock (White et al., 1999). Although a major proportion of Na is seasalt origin, significant excess Na must be released from the ambient rocks. In factor 2 (24.4% of the total), the moderate to high correlation among Na, K, Mg and Si probably implies weathering of silicate minerals, i.e. feldspars or micas.

In the Dahan drainage area, steep slopes and flashy hydrologic responses will enhance physical erosion, and further strengthen the chemical weathering. The illite crystallinity and illite chemistry

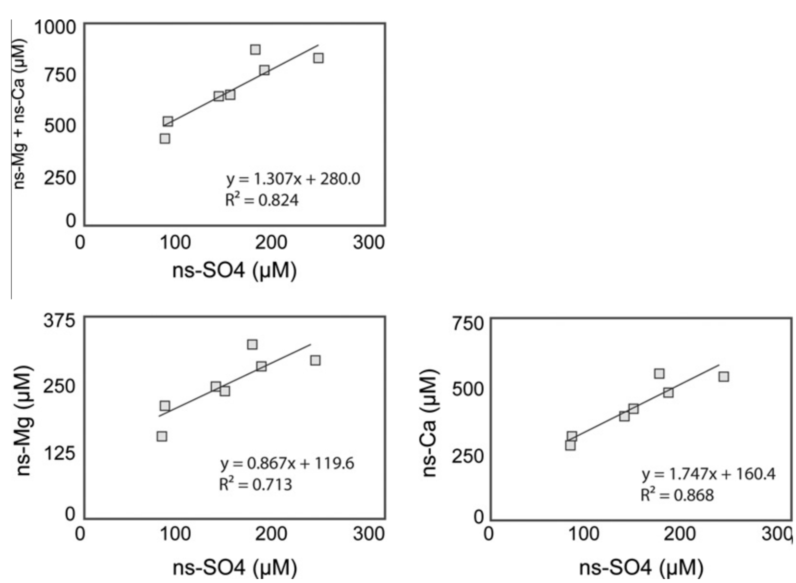

Fig. 3. Scatter plots for ns-Mg + ns-Ca, ns-Mg and ns-Ca with ns-SO_4.

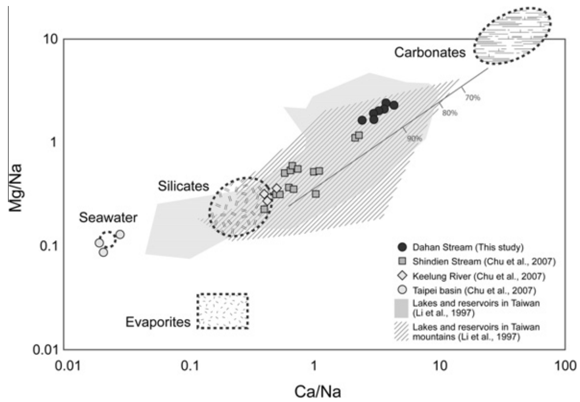

Fig. 4. Diagram of Mg/Na versus Ca/Na for rivers in northern Taiwan. Solid points represent the samples in Dahan catchment in this study, squares, diamond, are the samples in the downstream rivers of Dahan Stream (Chu and You, 2007). The areas in gray and dotted lines are the ranges of the given ratios in plain and mountain lakes/reservoirs in Taiwan, respectively (Li et al., 1997). The end-member compositions of carbonates, silicates and evaporites weathering are from (Gaillardet et al., 1999). Numerical calculations based on Mg/Na – 0.35, Ca/Na – 0.6 for silicates (Taylor and McLennan, 1985), and Mg/Na – 10, Ca/Na – 50 for carbonates (Stallard and Edmond, 1987).

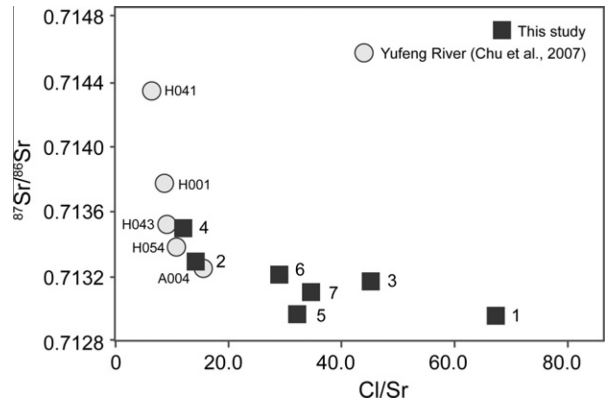

Fig. 6. Plot of $^{87}Sr/^{86}Sr$ versus Cl/Sr in Dahan watershed. Circles from published data (Chu and You, 2007). Squares show sites SM-2 and SM-4 data in Yufeng River in this study.

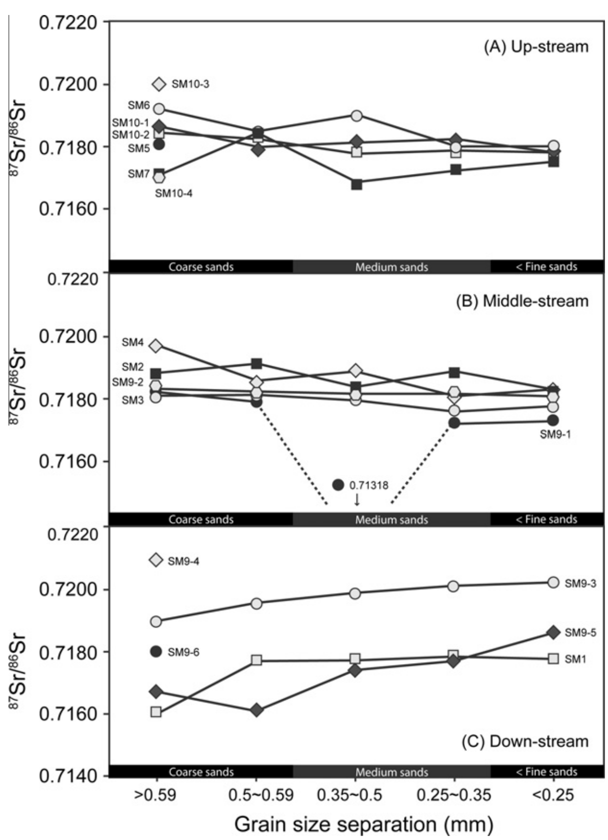

Fig. 5. Sr isotope measurements according to particle size in Dahan streambed sediments. (A) and (B) are samples collected in the upper and middle-reaches of Dahan stream. (C) All data for streambed sediments Sr isotopic ratios in the downstream area.

index are used as indicators for the extent of hydrolysis condition (Krumm and Buggisch, 1991). The Illite chemistry index refers to a ratio of the 5 Å and 10 Å areas. The ratio below 0.5 represents Fe-Mg-rich illite (biotite and mica) which implies the characteristics of physical erosion, and above 0.5 represents the Al-rich illite that indicates strong chemical weathering of hydrolysis (Gingele et al., 1998). In addition, the illite crystallinity obtained from half height width of the 10 Å peak also has been utilized as an indicator of hydrolysis condition (Krumm and Buggisch, 1991; Ehrmann, 1998). The higher illite chemistry index corresponds to the lower illite crystallinity, which imply stronger hydrolysis process and vice versa. The mean illite crystallinity and illite chemistry index of streambed sediment in Dahan Stream is 0.29°$\Delta 2\theta$ and 0.36, respectively and suggest moderate to strong chemical weathering in this regions, compared to the other main rivers in the south-eastern Asia (Liu et al., 2008). The lower illite crystallinity value detected here is an additional evidence to infer that huge amount of fine-grained particles have supplied by the shale hydrolysis in the Dahan drainage system.

5.2. Grain size effects and Sr isotopes in bed load sediments

The degree of sediment sorting is rather poor for bed loads in the Dahan drainage basin. Large accumulation of mud associated with landslides near the river banks may exacerbate this phenomenon. Detailed information on relative proportions of the grain-size distribution in bed load particles are summarized in Table 4 and show significant differences with upstream specimens. In general, fine-grained particles are rare in upstream stations where clays increase toward the downstream. This is our reason to selected large particles (sub-mm levels) in this study compared with previous (sub-mm to μm). The coarser sands or pebbles are deposited immediately during transport in streams or near the reservoir inlet. Only fine-grained particles (<0.25 mm) can be transported into the reservoir, either accumulated at the bottom or suspended in the water during/after rainfall events. This explains the mean particle size of less than 0.25 mm in the sediment cores. The distribution of sediment particle size can record different sedimentation processes and climatic information (Revel et al., 1996). Sr isotopic composition in bulk sediments may represent in situ characteristics of water/rock interactions and fine-grained clays reflect sources of sediments.

Sequential extraction of Sr in sediments has shown that changes in the composition of different phases would lead to modifications in the $^{87}Sr/^{86}Sr$ ratios of bulk sediments (Xu and Marcantonio, 2004). The Sr isotopic compositions in the Fe–Mn oxyhydroxides and carbonates fractions reflect the labile Sr in mobile phase, emphasizing the importance of non-residual fraction in Sr isotopic compositions. In fact, the labile Sr is relatively small in bulk sediment. Another important mechanism to affect the Sr isotopic compositions is differential weathering of Sr-bearing minerals and grain-size effects (Eisenhauer et al., 1999; Innocent et al., 2000;

Revel et al., 1996; Schettler et al., 2009; Techer et al., 2009). The Rb/Sr ratio difference is the domain factor to influence $^{87}Sr/^{86}Sr$ ratios in sediments. Eisenhauer et al. (1999) reported $^{87}Rb/^{87}Sr$ and $^{87}Sr/^{86}Sr$ variation is a function of particles grain size in the Arctic Ocean sediments. In the Murray-Darling River, Australia, weathering of plagioclase is the main source of less radiogenic Sr in the colloidal (<1 μm) and the dissolved phases (<0.003 μm); the coarse particles are mainly derived from weathering of silicate materials, i.e. relatively more radiogenic Sr from K-feldspar and mica (Douglas et al., 1995). Furthermore, Brass (1975) suggested released of less radiogenic Sr than the residual fraction, resulted in appreciably more radiogenic clay minerals in the weathering profiles. In this study, the $^{87}Sr/^{86}Sr$ ratios in bulk sediments fall in a narrow range, 0.71714–0.71843, which is lower than any other sub-size fraction (Fig. 5). No clear $^{87}Sr/^{86}Sr$ trend was observed in the entire sets of different grain-size fraction sediments collected from the Dahan Stream catchment. This is probably resulted from combined effects of sediment sources, mineral compositions and chemical weathering. Besides, repeated tectonic formation of folds in the drainage area and widespread landslide deposits nearby the mainstream channel are long term factors that may complicated the Sr isotope distribution in streambed sediments. Only the down-stream sites, which contain relative large proportion of fine-grained sediments, show significant correlation between grain sizes and Sr isotopes. It is noticed that short runoff distance would result in weak mineral sorting to reduce the observed relationships. Also the Sr isotopes in bed loads may have experienced differential weathering or mineral sorting during the transportation/deposition processes. For convenient discussion, $\Delta^{87}Sr$ is used in the following sessions to express the extent of Sr isotopic differences between two samples at the ppm level. $\Delta^{87}Sr = [(^{87}Sr/^{86}Sr)_{sub\text{-}fraction}/(^{87}Sr/^{86}Sr)_{bulk} - 1] \times 10^6$ (ppm), where $(^{87}Sr/^{86}Sr)_{sub\text{-}fraction}$ and $(^{87}Sr/^{86}Sr)_{bulk}$ are the Sr isotopic ratios in sub-fraction and its cohesive bulk sample, respectively. The large extent of Sr isotopic variability was found in the coarse fractions (>0.59 mm) and relative small in others. Based on the results presented in Lan et al. (2002), the heterogeneous distribution of $^{87}Sr/^{86}Sr$ ratios in same stratum is within ±0.001 (or 1000 ppm) in this region. However, the largest isotopic difference in the separated particles can exceed 2500 ppm in our study (i.e. the $\Delta^{87}Sr$ in SM1 is 2500 ppm). In the case of Shihmen Reservoir, these new results emphasize the importance of size effects for the application of Sr isotopes to trace potential sediment sources.

5.3. Spatial variation of Sr isotopic compositions

The isotopic characteristics of Nd and Sr of the early Cenozoic river bed sediments in East Asia have been preliminarily investigated at large scales. River sediments in Taiwan are primarily composed of continental detritus with $^{87}Sr/^{86}Sr$ ratios of 0.71099–0.72053 (Chen and Lee, 1990). Variable $^{87}Sr/^{86}Sr$ ratios in Oligocene meta-sediments have been reported for the Dahan drainage area and showed that (1) the $^{87}Sr/^{86}Sr$ ratio for early strata is 0.71995, (2) about 0.71678 in mid-strata, and (3) about 0.71878 in late strata (Lan et al., 2002). The $^{87}Sr/^{86}Sr$ ratios in bed loads obtained in this study, 0.71714–0.71843 for bulk samples, are similar with previous results. Our data further suggest that more radiogenic $^{87}Sr/^{86}Sr$ ratios are presented in Oligocene strata and less radiogenic values in Miocene strata. The sampling site of strata and Sr isotopic ratios were tested by analysis of variance (ANOVA, $\alpha = 0.01$) using SPSS 17.0 (SPSS Inc., USA) and suggested the p value <0.01, $F = 6.851$, and d. f. = 16. The $^{87}Sr/^{86}Sr$ ratios vary widely as functions of the degree of metamorphism and the rock ages (Faure, 1986). The sediments in Thaigang River have the most radiogenic $^{87}Sr/^{86}Sr$ ratios, whereas the lower $^{87}Sr/^{86}Sr$ ratios were found in tributaries in younger formations, such as the Taliao, Mushan, and Nangang Formations, with an exception of SM-3 which had an unexpected low $^{87}Sr/^{86}Sr$ ratio. This unexpected result may be the result of mixing with younger sediment or small particles from the Nangang Formation (Middle Miocene) or late strata due to the sampling site nearby the boundary of Nangang Formation and Mushan Formation. In the fine-grained size fraction (<0.25 mm), most samples have shown similar trend consistent with corresponding bulk samples. It indicates that most of the stream bed loads do reflect the source region in the Dahan catchment.

Compared to river sediments, the mechanisms influencing the Sr isotopic composition in surface water in this watershed appear to be complicated. All the weathering Sr releasing to water is less radiogenic (Brass, 1975). The $^{87}Sr/^{86}Sr$ ratios of stream water are affected by the mixing of water masses, the bed rock lithologies along the flow paths, and/or the differential dissolution of minerals (Bain and Bacon, 1994; Li et al., 2007). The activation energy in weathering reaction would control the participation chemical weathering of low resistant minerals. In this study, $^{87}Sr/^{86}Sr$ ratios in steam water and streambed sediments show a combined effect of disequilibrium of water/rock interaction and water masses mixing. From the up- to downstream, $^{87}Sr/^{86}Sr$ ratio versus 1/Sr diagram display a trend of three-steps mixing processes (Fig. 6). The first is the water mixing in the tributaries of Thaigang River and Baishih River (SM-5,6,7), where the $^{87}Sr/^{86}Sr$ ratio in Thaigang River sediment is more radiogenic than Baishih River. The second step occurs in Yufeng River (SM-4) with the most radiogenic $^{87}Sr/^{86}Sr$ ratio in dissolved loads, agree with Chu and You (2007). There is no major tributary flow to Yufeng River, however, many local flow path may carry landslide deposits to the main channel along the Yufeng River. This explains that even the bed load sediment may not be able to transport into main stream, the impact from the critical landslide deposits in this region still cannot be ignored in Yufeng River. The final step is the water mixing of Dahan Stream with Yufeng River and other smaller tributaries (SM-3). The $^{87}Sr/^{86}Sr$ ratios in the Dahan catchment exhibit a clear spatial variation of Sr isotopic compositions as follows: (1) high $^{87}Sr/^{86}Sr$ ratios in Thaigang River and Yufeng River, (2) moderate in Dahan Stream, and (3) low in Baishih River (Fig. 7).

5.4. Spatial and temporal variation of Sr isotopic composition in sediment cores

The particle sizes of specimens separated from sediment core Nos. 2–4 are all less than 0.25 mm, fine to very fine sand, in the Shihmen Reservoir. The $^{87}Sr/^{86}Sr$ ratios in these cores range from 0.71187–0.72647, having a wider range than the range observed in the present drainage system (0.71709–0.72021, <0.25 mm fraction). Several reservoir deposition clearance projects may have

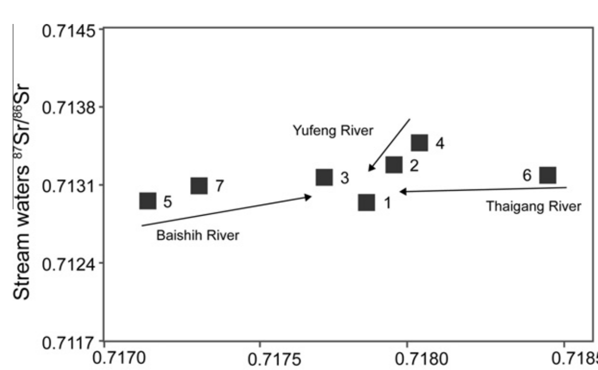

Fig. 7. Scatter plot of $^{87}Sr/^{86}Sr$ of streamwater versus streambed sediment in the Dahan watershed.

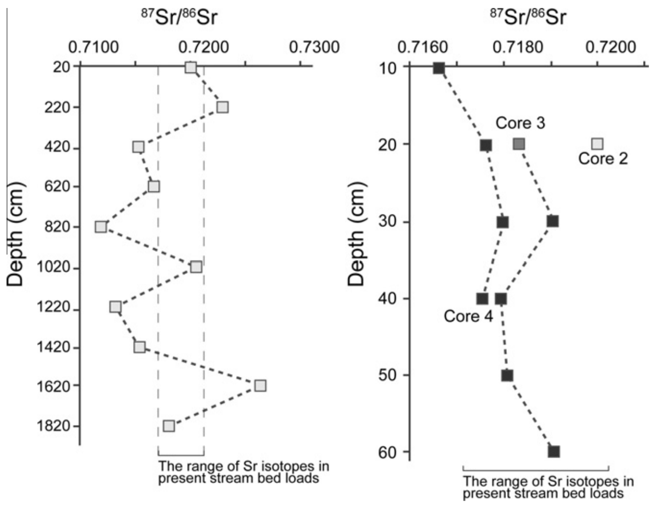

Fig. 8. Sr isotope sediment profiles in Shihmen Reservoir sediment cores. (Left) Shows the long core 2. (Right) Shows the high resolution cores 3 and 4.

resulted in discontinuous sedimentation records in the Shihmen Reservoir and exacerbated the complicated age dating of sediment cores. However, typhoons and heavy rainfall events have caused huge amount of sediment transported into reservoir and thick layer of deposition in very short time. Thus, we can compare directly the sediment provenance in most recent climatic event, upper core sediment, with those occurred previously. The high resolution sediment cores Nos. 3 and 4 show relative small Sr isotopic variability in the upper 60 cm (Fig. 8). The Sr isotope ratios in the core sediments are in agreement with the range reported for the Oligocene and Miocene formations in drainage system, mixed contribution from those strata in the upper catchment. The less variable Sr isotopic compositions in the upper cores may result from the homogeneous sedimentation and/or similar sediment sources. Based on the x-radiograph of sediment cores, little bioturbation phenomenon but abundant turbidity layers have documented in the upper sediment cores indicate frequent turbidity events have occurred in the recent past.

The core No. 2 sampled in the middle reservoir nearby the edge of the dam shows different sediments sources deposited during the past few decades (Fig. 8). Compared the Sr isotopic characteristics in the upper core sediments and the streambed sediments, it is implied that the more radiogenic Sr isotope is possibly derived from the Oligocene sedimentary strata of Tatungshan Formation. However, at a few intervals in the core (such as 420–820 cm), extremely low ^{87}Sr/^{86}Sr ratios were found suggesting another unidentified sediment sources that is out offside the range of Sr isotope ratios in present-day streambed sediments. Several possible factors controlling the spatial and temporal variation of Sr isotopes in sediment cores could include (1) impact of the recent tectonic activity, the Chi–Chi earthquake (magnitude 7.3) in 1999, caused serious and widely spread landslide events in the Dahan catchment and resulted in large amount of newly exposed sediments and further transported into stream channels, in which source of sediment was changed compared with environmental condition in the past. (2) Human developments or land use, (3) artificial sediment sources, such as direct input of fine sands and soils near the reservoir, and (4) in-sufficient sampling density which may have excluded sediment sources from debris flows and landslides nearby the stream channel. The former one might have relative large impact rather than the other factors, and the last case is unlikely due to that we have conducted close examination of samples in the landslide areas. The contributions from factors 2 and 3 are difficult to evaluated, but seems to play minor role compared with factor 1. These new results emphasize the importance of environmental impacts on spatial and temporal variations of Sr isotopes in the sediment cores. The data from this study suggests that turbidic water inputs during the heavy rainfall seasons were triggered by bottom density current within the water reservoir and the Oligocene formation is the mainly sediment sources in Shihmen Reservoir.

6. Conclusions

The illite crystallinity and illite chemistry index of these selected sediments imply moderate to strong chemical weathering in the Dahan Stream drainage catchment. Dissolved chemical compositions further suggest that most of the dissolved constituents were derived from silicate weathering and emphasize the importance of water/sediment interactions. Since the carbonates were solely found in the sedimentary strata of Northern Taiwan. A highly positive linear relationship of ns-Ca/ns-Na and ns-Mg/ns-Na in the most upper stream stations indicates that carbonate dissolution has occurred preferentially in the high denudation mountain area, i.e. Central Mountain Range.

The ^{87}Sr/^{86}Sr ratios in the Dahan drainage waters and bed loads fall in a range of 0.71296–0.71349 and 0.71714–0.71843, respectively. These data suggest a disequilibrium water/rock interaction and caused clear spatial Sr isotopic compositions variation in the Dahan catchment. The Sr isotopic analyses performed in different particle size fractions display a rather large Sr isotopic variability. The ^{87}Sr/^{86}Sr ratios in stream bed load show no relationship with particle sizes, and probably resulted from weak mineral sorting in the high denudation and tectonically active mountainous region.

Comparison of Sr isotopes in the fine-grained fraction particles in the Dahan drainage area and upper sediment cores within the Shihmen Reservoir suggests the present day sediment source is mainly from the Oligocene strata. In addition, Sr isotopes in reservoir sediment cores support that source sediments have varied spatially and temporally to the reservoir. The less variable ^{87}Sr/^{86}Sr ratios in all the upper cores reflect disturbance of fine-grained sediment that might be an important evidence for turbidity events occurred within the reservoir during the past few decades.

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