

## CHEMICAL COMPOSITIONS OF HOT SPRING WATERS IN THE TATUN VOLCANIC AREA, NORTHERN TAIWAN

YAW-LIN CHEN<sup>1</sup>, HUANN-JIH LO<sup>1</sup>, YUE-GAU CHEN<sup>1</sup>, SHAO-WEI CHENG<sup>2</sup>, TSUNG-KWEI LIU<sup>1</sup>, CHEN-FENG YU<sup>3</sup>,  
SHENG-RONG SONG<sup>1</sup>, JIANN-NENG FANG<sup>4</sup> AND CHI-CHI HUANG<sup>2</sup>

1. Department of Geosciences, National Taiwan University
2. Institute of Statistical Science, Academia Sinica
3. Department of Earth Sciences, National Cheng Kung University
4. Department of Earth Science, Taiwan Museum

### ABSTRACT

Spring waters from thirteen sites in the volcanic terrane of the Tatun volcanic area of northern Taiwan were collected monthly from January to December of 2000 for analyses of Na<sup>+</sup>, K<sup>+</sup>, Mg<sup>+2</sup>, Ca<sup>+2</sup>, Al<sup>+3</sup>, Si<sup>+4</sup>, Cl<sup>-</sup> and SO<sub>4</sub><sup>-2</sup>. In the meantime, some water samples, collected from both the volcanic terrane and sedimentary one, were analyzed for sulfur isotopes. It is found that chemical concentrations of the spring waters show variations with both time and space. High precipitations in the 1st, 2nd, 4th and 7th weeks are usually accompanied with low chemical concentrations of the spring waters. Average chemical concentrations are higher in the dry-periods than in the wet-periods. Yet, ionic ratios of the wet-/dry-periods fall in a narrow range. It is also clear that values of  $\delta^{34}\text{S}$  of the spring waters in the sedimentary terrane (+25 to +29 permils) are much higher than those of the spring waters in the volcanic terrane (+1 to +8 permils).

It is considered that mixing of the spring waters in the dry-periods with different amounts of meteoric water can produce those with the observed chemical concentrations. Chemical variations among the spring waters may be attributed to the contribution of different amounts of chemical components of volcanic gases and volcanic rocks to spring reservoirs. The values of  $\delta^{34}\text{S}$  of the spring waters suggest that spring reservoirs in the sedimentary and volcanic terranes may be formed, respectively, of sedimentary and volcanic rocks.

Reservoirs in the volcanic rocks may mainly be supplied by meteoric water which runs through different paths taking different time to contribute to the springs. It is presumable that anionic

components such as  $\text{SO}_4^{2-}$  and  $\text{Cl}^-$ , and cationic components such as  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$ ,  $\text{Al}^{3+}$  and  $\text{Si}^{4+}$  of the spring waters in the volcanic terrane may essentially be contributed from volcanic gases and altered volcanic rocks, respectively.

**Key words:** Tatun volcanic area, chemical components, spring waters

## INTRODUCTION

Springs in the Tatun volcanic area of northern Taiwan occur essentially in the volcanic terrane, and partly in the sedimentary one (Fig. 1). Those of the former are studied in this investigation, while those of the latter have recently been studied (Chen *et al.*, 2001a). Though intermittent studies on the springs have been reported (MRSO, 1969, 1970, 1971, 1973; Cherng, 1978; Chen and Yang, 1984; Liu *et al.*, 1984; Wang *et al.*, 1987; Wang, 1991; Chen, 1994; Fang *et al.*, 1998), a long-term investigation on them has rarely been carried out. In this study, the spring waters in the volcanic terrane were collected monthly from January to December of 2000 for analyses of essential chemical components. In the meantime, some water samples, collected from both the volcanic terrane and sedimentary one, were analyzed for sulfur isotopes.

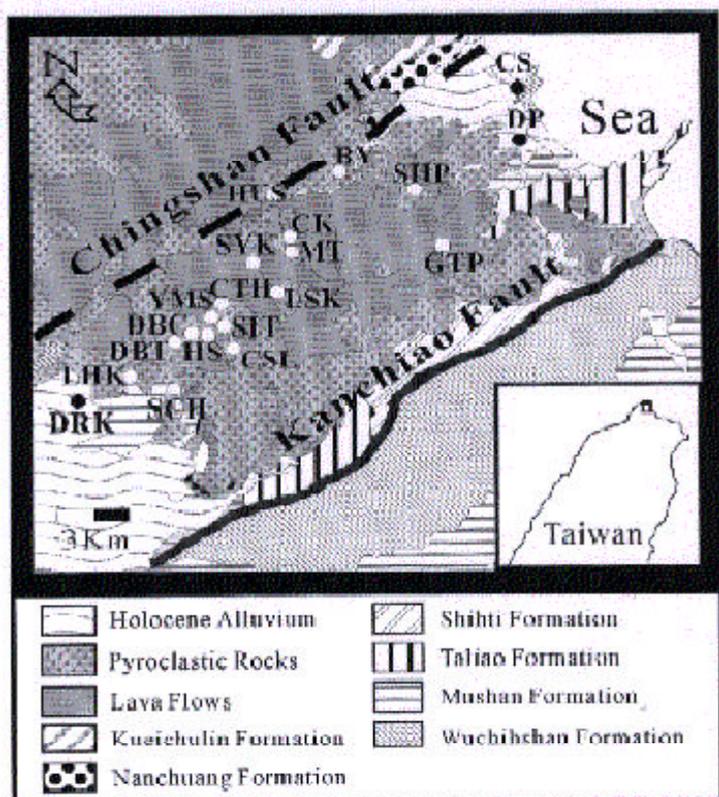


Figure 1. Spring sites and the geological map (modified from C.G.S., 1998) of the Tatun volcanic area. Abbreviations used for the springs. BY: Bayang, CK: Chiku, CSL: Chungshanlao, CTH: Chutzehu, CS: Chingshan, DBC: Dingbichao, DBT: Dingbeitou, DP: Dapu, DRK: Direku, GTP: Gentzeping, LHK: Lenguangku, LSK: Lengshuikeng, HS: Hoshan, HUS: Hushan, MT: Matsao, SCH: Swuangchunghsi, SHP: Shihuangping, SIT: Siao intan, SYK: Siao yukeng, YMS: Yangmingshan

Using the results of this study and other data available, this paper deals with: 1) time and space variations of chemical concentrations of the spring waters, 2) the relationships between chemical concentrations of the spring waters and local precipitations, 3) reservoirs of the springs, and 4) sources of materials that contribute chemical components to the spring waters.

## SAMPLING AND METHODS

Spring waters from thirteen sites in the volcanic terrane of the Tatun Volcanic area were collected for chemical analyses of major components. The spring sites and the geological map are shown in Figure 1. The Environmental Protection Administration's procedures (EPA, 1994) were followed for sampling as reported elsewhere (Chen *et al.*, 2001a)

Concentrations of  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Mg}^{+2}$ ,  $\text{Ca}^{+2}$ ,  $\text{Al}^{+3}$  and  $\text{Si}^{+4}$  were determined by using Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES). Analytical uncertainties are estimated to be  $\pm 1\%$  of the reported values, but they increase as concentrations decrease. On the other hand, concentrations of  $\text{Cl}^-$  and  $\text{SO}_4^{-2}$  were determined by using Ion Chromatography (IC) with analytical uncertainties of  $\pm 2\%$  and  $\pm 5\%$ , respectively. pH values were measured in situ with a portable pH-meter. Data of the precipitation (CWB, 2000) were taken from the Chutzehu Weather Station of the Central Weather Bureau of R.O.C..

Water samples from the sites of CS, DP, DRK (in the sedimentary terrane), LHK, LSK, MT, SHP and SYK (in the volcanic terrane) (see Figure 1) were selected for analyses of sulfur isotopes. The analytical procedures were the same as those described by Chen *et al.* (1997). Sulfur isotopic compositions were analyzed in the laboratory of Illinois State Geological Survey and analytical uncertainties are estimated to be  $\pm 0.3\%$  of the reported values.

## EXPERIMENTAL RESULTS

Total dissolved chemical components (TDCC), including  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Mg}^{+2}$ ,  $\text{Ca}^{+2}$ ,  $\text{Al}^{+3}$ ,  $\text{Si}^{+4}$ ,  $\text{Cl}^-$  and  $\text{SO}_4^{-2}$ , of the spring waters from January to December of 2000 versus time are plotted in Figure 2. Clearly, the amounts of TDCC fluctuate with time and vary from spring to spring. In other words, chemical concentrations of the spring waters show variations with both time and space. It is noted that better correlations generally occur between chemical concentrations of the spring waters and total precipitations of one week in the studied area as revealed from our previous studies (Chen *et al.*, 2001a, b). So, total precipitations of 1 to 7 days (1st week), 8 to 14 days (2nd week), 15 to 21 days (3rd week), 22 to 28 days (4th week), 29 to 35 days (5th week), 36 to 42 days (6th week), 43 to 49 days (7th week) and 50 to 56 days (8th week) before sampling are chosen for correlating with TDCC of the spring waters and the results are shown in Figure 3. As indicated in the figure, better correlations occur at the 1st, 2nd, 4th and 7th weeks and the best one occurs at the 1st week (Figure 3 and Table 1). In other words, high precipitations at the 1st, 2nd, 4th and 7th weeks are accompanied with low TDCC of the spring waters. Since the two-best correlations occur at the 1st and 2nd weeks (Figure 3 and Table 1), the total precipitations of 14 days before sampling are chosen to define the wet- and dry-periods in this study. So, the wet- and dry-periods are arbitrarily taken to be more than 3000mm (Tab. 2) and less than 300mm (Tab. 2) of the total precipitations of 14 days before sampling, respectively.

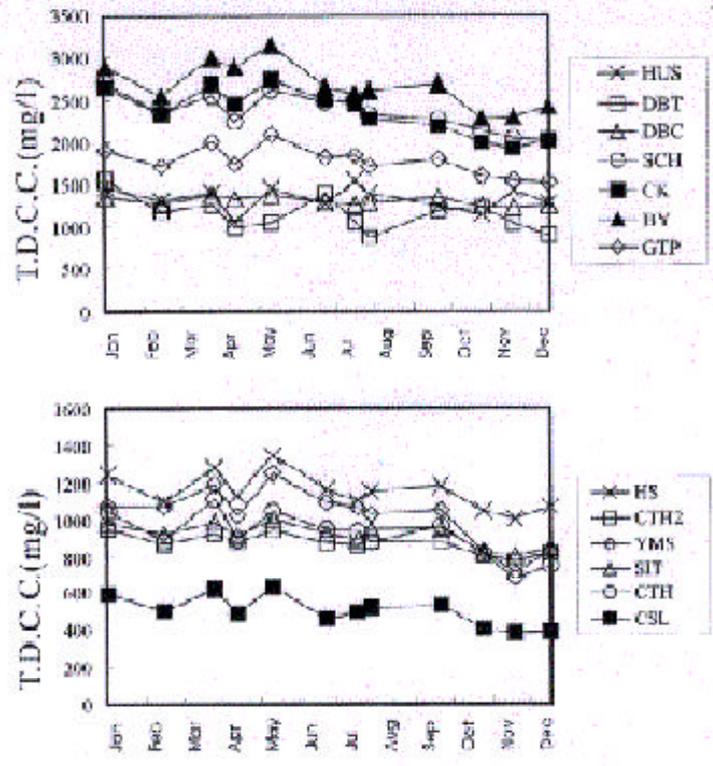


Figure 2. Monthly variations of total dissolved chemical components (TDCC) of the spring waters in 2000 (dry-periods: Jan., Mar., May, and wet-periods: Oct., Nov., Dec. from Table 2).

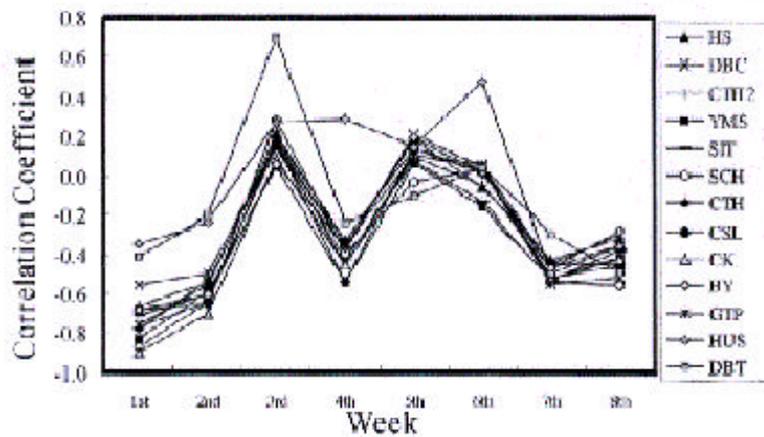


Figure 3. Correlation coefficients between TDCC of the spring waters and total precipitations of different durations before sampling.

Table 1. Correlation coefficients of TDCC and total precipitations of different durations before sampling.

Spring \ Duration	Week							
	1st	2nd	3rd	4th	5th	6th	7th	8th
BY	-0.74	-0.69	0.06	-0.35	0.20	0.01	-0.47	-0.27
CK	-0.83	-0.73	0.07	-0.47	0.04	0.07	-0.57	-0.40
CSL	-0.80	-0.77	0.20	-0.31	0.19	0.01	-0.49	-0.46
CTH	-0.91	-0.64	0.17	-0.52	0.19	-0.01	-0.41	-0.44
CTH2	-0.85	-0.74	0.17	-0.44	0.12	-0.06	-0.46	-0.35
DBC	-0.77	-0.64	0.18	-0.34	0.31	-0.01	-0.39	-0.30
DBI	-0.49	-0.25	0.68	-0.23	-0.07	0.02	-0.27	-0.49
GTP	-0.82	-0.64	0.21	-0.34	0.12	0.05	-0.55	-0.51
HS	-0.77	-0.65	0.18	-0.35	0.23	-0.08	-0.41	-0.31
HIS	-0.22	-0.20	0.24	0.27	0.07	0.51	-0.55	-0.33
SCH	-0.84	-0.68	0.26	-0.40	-0.08	0.06	-0.58	-0.55
SII	-0.87	-0.74	0.16	-0.40	0.14	-0.16	-0.49	-0.38
VMS	-0.92	-0.69	0.05	-0.53	0.09	-0.17	-0.51	-0.41

Table 2. Total precipitations of 14 days before sampling and wet-and dry-periods.

Sampling date	1/6	2/19	3/30	4/17	5/16	6/27	7/21	8/1	9/25	10/29	11/23	12/21
Total precipitation (mm)	205	614	115	1297	261	1309	1900	993	1905	3288	4177	4389
Wet- or dry-period	dry			dry			dry			wet		

Average chemical concentrations of the spring waters in dry- and wet-periods are listed in Table 3. In the meantime, ionic ratios of average chemical concentrations of wet- and dry-periods are shown in Figure 4. Obviously, average chemical concentrations are higher in the dry-periods than in the wet-periods. Yet, ionic ratios of the wet-/dry-periods fall in a narrow range.

Isotopic data of sulfur ( $\delta^{34}\text{S}$ ) of the spring waters in the volcanic and sedimentary terranes are shown in Table 4. It is clear that values of  $\delta^{34}\text{S}$  of the spring waters in the sedimentary terrane (+25 to +29 permils) such as CS, DP and DRK are much higher than those of the spring waters in the volcanic terrane (+1 to +8 permils) such as LHK, LSK, MT, SHP and SYK (see Fig. 1 for the spring sites).

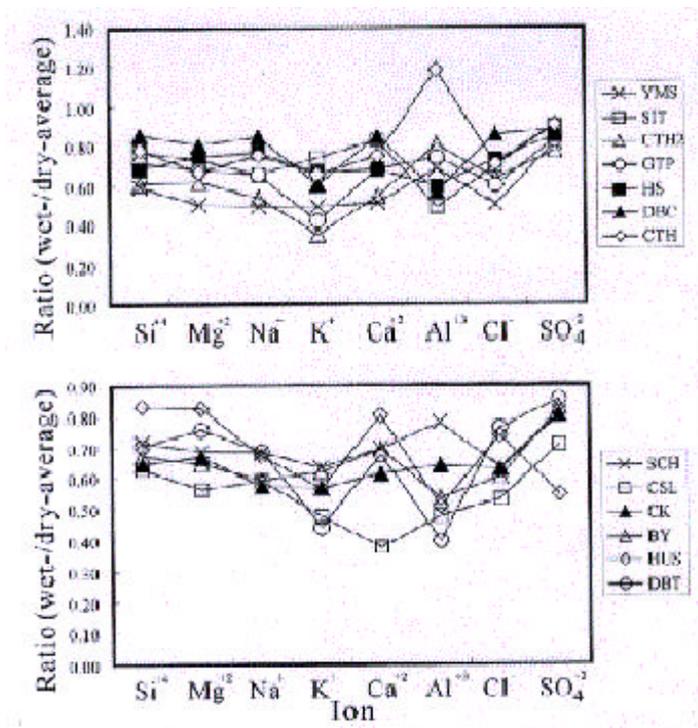


Figure 4. Ionic ratios of average chemical concentrations of wet- and dry-periods.

## DISCUSSION AND CONCLUSION

As mentioned previously, chemical concentrations of the spring waters fluctuate with time (Fig. 2) and the same results have already been reported (MRSO, 1969, 1970, 1971, 1973; Cherng, 1978; Wang *et al.*, 1987; Chen, 1994). It is found that chemical concentrations of the spring waters are lower in wet-periods than in dry ones (Tab. 3). Moreover, ionic ratios of wet- and dry-averages fall in a narrow range (Table 3 and Figure 4). Thus, a dilution process is suggested. In other words, mixing of the spring waters in dry-periods with different amounts of meteoric water can produce those with the observed chemical concentrations (Fig. 2). It is worthy to mention that the values of ionic ratios of  $\text{SO}_4^{2-}$  are much higher than others (Tab. 3). Similar results were also obtained in our previous study (Chen *et al.*, 2001a). Sulfate concentrations might mainly be controlled by a continuous supply of sulfur-bearing volcanic gases such as  $\text{H}_2\text{S}$  and  $\text{SO}_2$ . These gases are oxidized near the surface and keep the spring waters saturated with  $\text{SO}_4^{2-}$  so as to show higher ionic ratios of  $\text{SO}_4^{2-}$  as suggested in our previous paper (Chen *et al.*, 2001a). Variations of chemical concentrations among the springs are apparently shown in Figure 2. The same results have also been reported by previous workers (Chen and Yang, 1984; Liu *et al.*, 1984; Wang *et al.*, 1987; Wang, 1991; Chen, 1994; Fang *et al.*, 1998). These workers suggested that chemical variations might be attributed to water-rock interaction, incorporation of seawater, oxidation of volcanic gases and evaporation. Though no conclusive arguments can be drawn from this study, it is believed that the contribution of different amounts of chemical components of volcanic gases and volcanic rocks could play a very important role to the chemical variations. The volcanic gases and volcanic rocks are considered as main



Values of  $\delta^{34}\text{S}$  of the spring waters in the sedimentary terrane (CS, DP and DRK in Table 4) and in the volcanic one (LHK, LSK, MT, SHP and SYK in Table 4) are similar to those of sedimentary and volcanic materials, respectively (Hoefs, 1987). This may imply that reservoirs of the springs in the sedimentary terrane may be in sedimentary rocks, while those of the springs in the volcanic terrane may be in volcanic ones. In addition, the same conclusion is also suggested from altitudes of the springs and the sedimentary rocks in the studied area as illustrated in Figure 5. It is noted that altitudes of the springs in the sedimentary terrane are lower than 100 meters, while those of the springs in the volcanic terrane are higher than 280 meters above the sea level. In the meantime, the highest surface outcrops, which appear in the northern- and southern-most parts of the studied area (Fig. 1), of the sedimentary rocks of different strata range mostly from 200 to 280 meters (except S.F.) as shown in Figure 5. So, the sedimentary rocks distributed in the northern- and southern-most parts of the studied area may be the potential sources for recharging the spring reservoirs in the sedimentary rocks with meteoric water because of eastward-dipping of the sedimentary strata (Chen and Wu, 1971; C.G.S. 1998). Consequently, it is inferred that the springs with altitudes higher than 280 meters in the volcanic terrane can hardly be supplied from the sedimentary reservoirs, though those with altitudes lower than 100 meters in the sedimentary terrane are possible. In other words, the springs in the volcanic terrane are most probably supplied from the volcanic rocks. Besides, ionic ratios also suggest a different source of the springs in the volcanic and sedimentary terranes as indicated in Figure 6.

As shown in Figure 3, better correlations between amounts of TDCC of the spring waters and total precipitations of one week occur at the 1st, 2nd, 4th and 7th weeks. This may imply that reservoirs in the volcanic rocks are mainly supplied from meteoric water which runs through different paths taking different time to contribute to the springs. It should be noted that most of the springs appear in pyroclastic rocks (Fig. 1). So, it is reasonable to assume that the pyroclastic rocks could be the most potential reservoirs for the springs in the volcanic terrane because the pyroclastic rocks are loosely consolidated (Chen and Wu, 1971; MRSO, 1973) and are generally considered to be more permeable than massive lava flows (Fisher and Schmincke, 1984). As a consequence, the springs are concentrated in the southwestern part where the pyroclastic rocks are dominated in the studied area (Fig. 1). The subsurface distribution of the pyroclastic rocks is not clearly known though their surface distribution has been mapped (Chen and Wu, 1971; C.G.S. 1998). So, the reasons for how the pyroclastic rocks to influence the springs showing a common character of better correlations between amounts of TDCC and total precipitations of one week occurring at the 1st, 2nd, 4th and 7th weeks are not clear and, certainly, further studies are encouraged to solve the problem.

Table 4. A comparison of  $\delta^{34}\text{S}$  (in permil) of the spring waters in the sedimentary and the volcanic terranes.

Terrane	Sedimentary			Volcanic				
	CS	DP	DRK	LHK	LSK	MT	SHP	SYK
Spring								
$\delta^{34}\text{S}$ (‰)	+24.5	+28.5	+26.1	+2.4	+7.8	+0.8	+3.3	+3.4

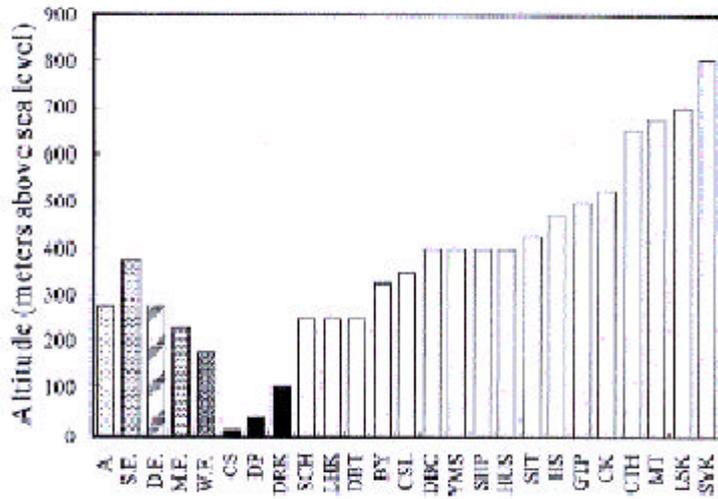


Figure 5. Altitudes of the highest outcrops of sedimentary strata and of the springs in the studied area. A : alluvium. S.F. : Shidi Formation. D.F. : Daliao Formation. M.F. : Mushan Formation. W.F. : Wuchihshan Formation. Other abbreviations being the same as in Figure 1.

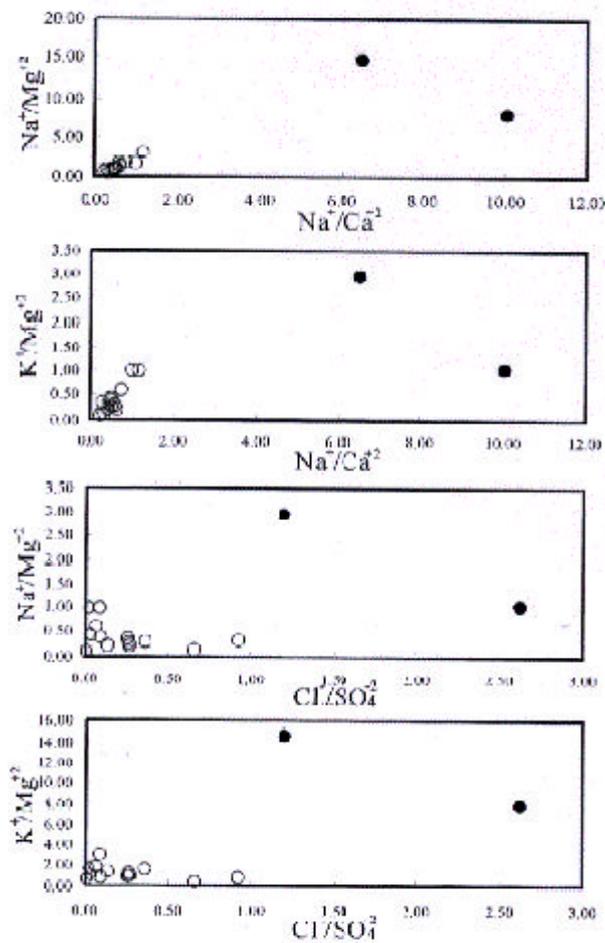


Figure 6. Plots of ionic ratios of the spring waters in the sedimentary and volcanic terranes.  $\circ$  : in the sedimentary terrane (data from Chen *et al.*, 2001a).  $\bullet$  : in the volcanic terrane

As started previously, chemical components of the spring waters may mainly be contributed from volcanic gases and altered volcanic rocks. Post-volcanic activities such as fumaroles, hot springs and hydrothermal alteration are common in the studied area (MRSO, 1969; 1971; 1973; Chen and Wu, 1971; Chen and Yang, 1984). Helium isotopes suggest a mantle origin of the fumarolic components such as CO<sub>2</sub>, H<sub>2</sub>S, SO<sub>2</sub> and HCl (Yang *et al.*, 1999; Ho, 2001). So, the volcanic gases should play an important role to the post-volcanic activities. The volcanic gases may dissolve in water to contribute chemical components such as SO<sub>4</sub><sup>-2</sup> and Cl<sup>-</sup> to the spring reservoirs and may heat up the spring waters as well. Such hot acidic waters may act as major liquid media for the hydrothermal alteration of the volcanic rocks. Chemical components such as Na<sup>+</sup>, K<sup>+</sup>, Mg<sup>+2</sup>, Ca<sup>+2</sup>, Al<sup>+3</sup> and Si<sup>+4</sup> etc. may be released to the spring reservoirs during the hydrothermal alteration of the volcanic rocks in acidic fluids as revealed by laboratory studies (Nogami and Yoshida, 1995; Fang *et al.*, 1998) and analyses of water samples from drilling wells in the studied area (MRSO, 1969; Chen and Yang, 1984). So, it is presumable that anionic components such as SO<sub>4</sub><sup>-2</sup> and Cl<sup>-</sup>, and cationic components such as Na<sup>+</sup>, K<sup>+</sup>, Mg<sup>+2</sup>, Ca<sup>+2</sup>, Al<sup>+3</sup> and Si<sup>+4</sup> of the spring waters in the volcanic terrane may essentially be contributed from the volcanic gases and altered volcanic rocks, respectively.

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