



# Application of water chemistry as a hydrological tracer in a volcano catchment area: A case study of the Tatun Volcano Group, North Taiwan



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## ARTICLE INFO

### Article history:

Received 5 May 2012

Received in revised form 13 October 2013

Accepted 16 February 2014

Available online 22 February 2014

This manuscript was handled by Laurent Charlet, Editor-in-Chief, with the assistance of Tamotsu Kozaki, Associate Editor

### Keywords:

Hydrochemistry

Rare earth elements

Natural tracer

Tatun Volcano Group

## SUMMARY

In this paper, water chemistry is successfully applied to elucidate hydrological processes through the use of natural tracers in a hydrological system. The concept of a natural tracer is principally based on water–rock interaction. In this case, a volcanic watershed in the Tatun Volcano Group is examined with the hydrochemistry of the Peihuang Creek system analyzed in terms of acidic hydrothermal water. The application of principal component analysis demonstrates that the hydrochemistry of Peihuang Creek is dominated by mixing among three end members, Lujiaoken seep water, Matsao seep water and shallow circulated water. Conservative ions, such as halogens, reveal that recharge of shallow circulated water with low ionic concentration is dominant in the mountain area and gradually becomes insignificant in the plains area. Rare earth elements also confirm this derivation. In addition, rare earth elements demonstrate parallel pattern along the tributaries, which implies that the fractionation of rare earth elements is not considerable and dilution is the major factor attenuating the concentrations of rare earth elements. Therefore, the constant slope of REE pattern allows for semi-quantitative estimation of mixing proportion of the two major tributaries. The results show that Lujiaoken Creek supplies about 50% of waters to the downstream Peihuang Creek. Comparing commonly used nature tracers, such as  $\text{Cl}^-$  and environmental isotopes, this study demonstrates that rare earth elements have the advantage of very low background concentrations and easily defined sources if fractionation is not considerable. Under this circumstance, the calculation of water mixing is applicable.

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## 1. Introduction

Hydrochemical compositions are widely used as natural tracers in hydrological systems. The most commonly used natural tracers include conservative ions (e.g.  $\text{Cl}^-$ ,  $\text{Br}^-$  and  $\text{F}^-$ ), stable isotopes (e.g.  $^{18}\text{O}/^{16}\text{O}$ ,  $^2\text{H}/^1\text{H}$ , and  $^{87}\text{Sr}/^{86}\text{Sr}$ ) and radio isotopes (e.g.  $^3\text{H}$  and  $^{14}\text{C}$ ). Although some large-ion cations (e.g.  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Rb}^+$  and  $\text{Cs}^+$ ) are incompatible with mineral phases, cations are generally not suitable as natural tracers due to the chemical processes of sorption, ion exchange, and/or surface complexation (Essington, 2003; Weiner, 2008). The most used conservative ion as a natural tracer is  $\text{Cl}^-$ , which has relatively high abundance in natural water masses. It is widely applied in the estimation of groundwater recharge rates and evaluation of mixing ratios involving seawater or brine (e.g. Murphy et al., 1996; Sami and Hughes, 1996; Klaus et al., 2008; Waber and Smellie, 2008). Stable isotopes of hydrogen and oxygen are ideal conservative tracers of water sources because they are part of the water molecule itself. The stable isotope ratios

are mainly controlled by the processes of evaporation and condensation. As a result, in different parts of the hydrologic cycle, water is naturally characterized by isotopic fingerprints and its route through the hydrologic cycle can be traced (Gat, 1970; Sklash et al., 1976; Fritz, 1981; Clark and Fritz, 1997; Coplen et al., 1999). Radioisotopes can be applied to hydrological studies in two principal ways: as a tracer to identify different water masses, or more directly as an age measuring device. Of the commonly used radioisotopes,  $^3\text{H}$  is most widely used because it forms part of the water molecule and travels with ground water (Fontes and Garnier, 1979; Michel, 2005).

In general, the natural tracers mentioned above are characterized by being conservative, non-reactive and abundant in spite of radioisotopes. In some cases, trace elements and rare earth elements (REEs) are also effective natural tracers. However, these cations may be removed by the chemical reactions of adsorption and precipitation depending on hydrochemical conditions such as pH. For example, in the drainage from acid mines or geothermal water in volcanic areas, where oxidation of sulfide to sulfate leads to acidic water (pH values of 2–3) (Delmelle and Bernard, 2000; Taran and Peiffer, 2009), trace metals are relatively conservative

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in terms of chemical attenuation reactions. The reason for this is that high concentrations of protons ( $H^+$ ) in low-pH water compete with positive-charged trace metals for adsorption sites and ligands (Essington, 2003; Weiner, 2008). In addition, trace metals show very low background concentrations in water and are solely supplied by easily defined sources with higher concentrations. In the calculation of mixing between precipitation (in sub-ppt level) and geothermal water/leachate in a volcano watershed or at a mining site (in ppb level), trace metals and REEs have an advantage in that one can ignore background concentrations (Lewis et al., 1998; Gammons et al., 2005).

In the recent years, the use of REEs as natural tracers in ground and surface waters have been frequently reported (Johannesson et al., 1997; Åström, 2001; Gruau et al., 2004; Davranche et al., 2011). REEs' two major characteristics are: (1) different types of rocks have distinctive REE patterns; therefore, the source of REE can be definitively determined; and (2) the cation radii of REEs steadily decrease with increase in atomic number (from La to Lu – the so-called lanthanide contraction) and all REEs form trivalent cations (+3) with the exceptions of  $Ce^{4+}$  and  $Eu^{2+}$ . This leads to gradual fractionation from light REEs (LREE) to heavy REEs (HREE) in hydrochemical reactions (Cook and Herczeg, 1999; Johannesson et al., 2005). Based on the fractionation of REEs (REE pattern), dominant hydrochemical reactions can be identified. As mentioned, REEs are not conservative and may be adsorbed onto suspended materials. Predominant suspended materials include hydrous ferric oxides (HFO), hydrous aluminum oxides (HAO) and clay minerals. Both HFO and HAO mainly appear as colloids of grain size  $<0.45 \mu m$  (Bozau et al., 2008; Kay et al., 2011) and gradually transfer into oxides (Weiner, 2008). According to Steinmann and Stille (2008), LREE are preferably removed by HFO and HAO, resulting in LREE-depleted water. On the other hand, in acid mine drainage or volcanic watersheds, adsorption of REE may not be significant if there is no fractionation of REE in water. Under the circumstance, fractionation of REE is a good indicator for evaluating the effectiveness of HFO and HAO adsorption. However, this process is highly dependent on pH and low pH values generally prevent REE from

attenuation and fractionation in water (Lewis et al., 1998), which is the case in this study.

The Tatun Volcano Group (TVG) is located at the northern tip of Taiwan, which lies on the western edge of the Pacific “rim of fire”. The volcano group is composed of a cluster of dormant andesitic volcanoes including more than 20 volcanic craters. Although the most recent volcanic activity in TVG occurred about 200,000 years ago, extensive post-volcanic activity can be observed with the emission of fumarolic gases and the appearance of thermal springs. The volcano group covers an area of about 150 km<sup>2</sup> and is mainly drained by the Peihuang Creek that flows northward into the East China Sea. The Peihuang Creek is named for its location and color. It translates to North Yellow Creek in English. Its yellow color comes from the sulfur content. The hydrochemistry of the Peihuang Creek presents the typical chemical characteristics of water in a volcanic catchment region and the creek system should provide suitable conditions for studying REEs as natural hydrological tracers. Accordingly, the recharge area of the Peihuang Creek watershed can be identified and the mixing proportion of two major tributaries (Lujiakou Creek and Matsao Creek) can also be estimated if they have distinct REE signatures.

## 2. Regional hydrology

Most of the volcanic peaks in the TVG have elevations of 1000–1200 m above sea level. During the spring time, there is generally a stationary cold front over the TVG which produces a good deal of precipitation. Additionally, typhoons in summer and fall bring heavy rains to the area. In winter, monsoonal winds propagate southward bringing moisture to the TVG. However, comparing the intensity of precipitation in summer and fall, the period of winter and early spring has the lowest monthly precipitation of 190 mm, which is  $<1/3$  of that in summer and fall. Accordingly, the seasonal variation of precipitation is significant. Because of its high topography precipitation in the region is promoted and the above scenarios result in a large number of rainy days annually.

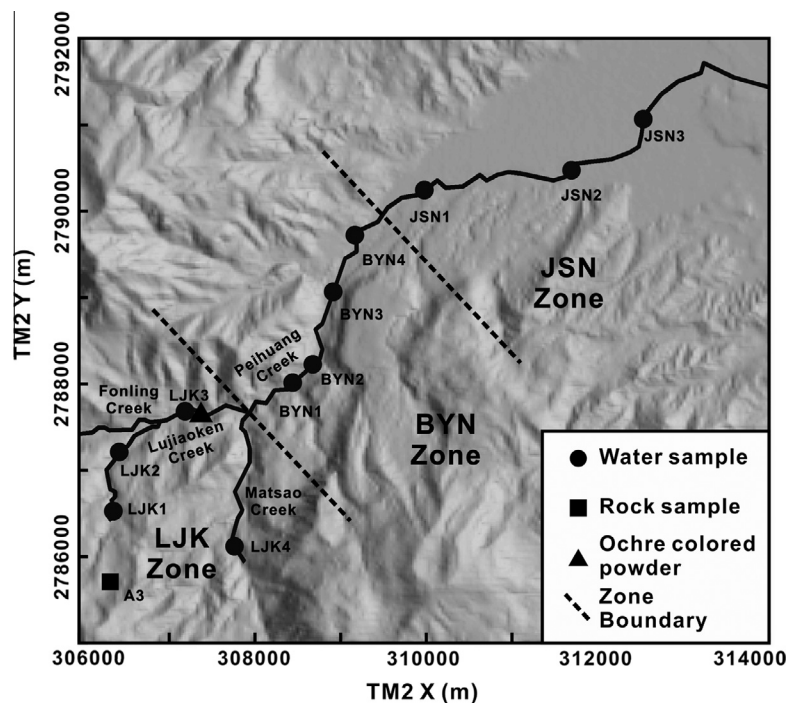


Fig. 1. Topographical map of TVG watershed showing the sampled localities. TM2 stands for two degree zone Transverse Mercator projection with Geodetic Reference System 1967. The locality of rock sample A3 was published by Wang et al. (2004).

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