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Influence of hydrological and hydrogeochemical parameters on arsenic variation in shallow groundwater of southwestern Taiwan

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SUMMARY

Shallow groundwater in the Choushui river alluvial fan of southwestern Taiwan contains high As contents. Arsenic concentrations in most of wells varied spatial–temporally. The purpose of this study is to quantitatively evaluate the hydrological and hydrogeochemical factors that influence As concentration in the shallow groundwater of southwestern Taiwan. Dynamic factor analysis (DFA), a dimension reduction technique that considers the time component for studying multivariate time series, is adopted to obtain underlying common trends of response time series and influences of explanatory variables. The major As-associated hydrological and hydrogeochemical parameters were identified and applied to predict the variation of As concentrations in groundwater. The results showed that As fluctuation in groundwater were governed by the rainfall and river water recharges in both dry and wet seasons. Rapid infiltration of surface water caused a rise in groundwater elevation in the rainy season and created a reductive environment, inducing reductive dissolution of As-bearing Fe (hydr)oxides. Dissolved As concentration was subsequently transported by fast groundwater flow to downstream areas. Moreover, extensive pumping for agricultural and aquacultural demands masked the influences of groundwater elevation on variations of As concentrations and led to critical seawater intrusion. In non-saline wells, Fe reduction and subsequent secondary mineralization between dry and wet seasons was responsible for As mobility. Sulfate reduction in saline wells promoted sulfide precipitation and restrained As mobility in wet season. Collectively, hydrological and hydrogeochemical variables controlled the temporal variation and spatial distribution of As concentration, respectively, in the shallow groundwater of southwestern Taiwan.

1. Introduction

Arsenic is one of the main inorganic contaminants in the continental water bodies. Arsenic can present in an inorganic or organic forms, depending on redox conditions in natural water. The inorganic form of As encountered in groundwater with high Eh condition is associated with arsenate, whereas arsenite predominates in reducing condition (Bose and Sharma, 2002). In addition, the redox potential controls the solubility and mobility of inorganic As (Signes-Pastor et al., 2007). Widespread and non-point source of As contamination in shallow groundwater poses health risk to the millions of people in many regions of the world, particularly in the southeast Asia (Berg et al., 2006; Buschmann et al., 2008; Guo et al., 2008; Polya et al., 2008). In Taiwan, shallow groundwater (<60 m) in southern part of the Choushui river alluvial fan contains high As concentration (Liu et al., 2006). From 1999 to date, a comprehensive investigation of groundwater quality was undertaken by the Water Resource Agency of Taiwan. Around 46–61% of shallow wells (<60 m) in southern Choushui river alluvial fan, the groundwater As concentrations exceeded WHO recommended guideline of 10 μg/L. Approximately 86–93% of high As well water had the reduction potential with the average Eh from −75.5 to −139.7 mV (Agricultural Engineering Research Center, 2008). Moreover, the reduction condition resulted in that the dominate As species was arsenite (∼67% of total As) (Agricultural Engineering Research Center, 2007). Wang et al. (2007) and Lu et al. (2011) reported that As reduction conditions are of critical in shallow groundwater of southern Choushui river alluvial fan, especially in its downstream flood plain and the reductive dissolution of

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As-rich Fe oxy-hydroxides from the subsurface is responsible for the mobility of As in groundwater.

Since 2000, the Environmental Protection Agency of Taiwan undertook the seasonal investigations of shallow groundwater quality in the southern Choushui river alluvial fan. The average groundwater As concentrations of all 10 wells from 2002 to 2008 were 22.5 ± 30.0 μg/L (<http://wqshow.epa.gov.tw/>). Groundwater As concentration in these wells change temporally. The spatial–temporal variation of As concentration may be influenced by the complicate regional biogeochemical conditions (Harvey et al., 2002; Islam et al., 2004; Rowland et al., 2007), and hydrological and human-related factors (García et al., 2007), resulting a change of redox potential. To infer spatial–temporal variation of As concentration in shallow groundwater, time-varied hydrological and hydrogeochemical parameters need to be comprehensively assessed.

Dynamic factor analysis (DFA) is a dimension reduction technique that considers time component for assessing multivariate time series responses of hydrological and water quality data. Multivariate time series may be analyzed as a response of functions assuming that there are common driving forces behind them, i.e. underlying factors that determine the variation of individual observations with time. These factors can be described by trends and/or explanatory variables. Recently, DFA has been applied in large oceanographical time series (Mendelssohn and Schwing, 2002), fisheries applications (Zuur et al., 2003a; Zuur and Pierce, 2004), groundwater and surface water level changes (Ritter and Muñoz-Carpena, 2006; Kaplan et al., 2010), groundwater quality trends (Muñoz-Carpena et al., 2005; Ritter et al., 2007; Kuo and Chang, 2010), topsoil water dynamics (Ritter et al., 2009), and oceanic ecology (Kuo and Lin, 2010). The analysis provides information including underlying common patterns in the response time series, interactions between response time series, and influences of considered explanatory variables. Because the analysis of large water quality data sets is complex and many characteristic factors affect the variation of chemical concentration in the system, DFA can effectively handle such data sets and identify the dominant effects controlling the observed variation. The purpose of this study is to quantitatively evaluate the influence of hydrological and hydrogeochemical parameters on As contents in shallow groundwater of southern Choushui river alluvial fan of Taiwan. The analysis was conducted in three steps: (1) identification of the common trends of groundwater As concentrations; (2) development of the dynamic factor model (DFM); and (3) examination of the results from the optimal DFM with local hydrological and hydrogeochemical settings. The results can be applied to evaluate As contamination potential and develop effective management scheme for As-affected ground water aquifers.

2. Material and methods

2.1. Study area

Southern part of the Choushui river alluvial fan is located in southwestern Taiwan and surrounded by the Taiwan Strait to the west and the Central Mountain to the east (Fig. 1). The southern Choushui river alluvial fan has an area of around 1290 km^2, and extends 48 km from east to west and 24 km from north to south. Choushui river and Peikang river are the two major rivers flowing through this area. The former is the longest river (186.6 km) in Taiwan with a drainage area of 3156.9 km^2. The average river flow is up to 24,000 m^3/s and provides respectable recharge to the shallow aquifer. Rainfall infiltration is also a source of groundwater recharge in this area. Rainy (April–September) and dry (October–March) have average cumulative rainfall of 1450 and 260 mm/yr, respectively (Agricultural Engineering Research Center, 2008).

According to the hydrogeological settings, the southern Choushui river alluvial fan is classified mainly into proximal-fan, the mid-fan and the distal-fan areas (Central Geological Survey, 1999). The hydrogeological formation of the proximal-fan, which consists entirely of gravel and sand, is an unconfined aquifer. In the distal-fan and the mid-fan areas, the formation is divided into six inter-layered sequences, including three marine sequences and three non-marine sequences. The non-marine sequences compose of coarse sediment, ranging from medium sand to highly permeable gravel, are regarded as aquifers, whereas the marine sequences with fine sediments are regarded as aquitards. The depths interval of the three aquifers were <60, 120–200, and 280–350 m, respectively (Agricultural Engineering Research Center, 2008). The hydraulic conductivity in the shallowest aquifer ranged from 0.25×10^{-3} to 1.04×10^{-3} m/s, indicating that surface water can easily infiltrate into this aquifer (Central Geological Survey, 1999).

Due to deficient surface water and rapid direct runoff, the substantial amount of groundwater was extracted for the agricultural and domestic needs. More than 90% of shallowest groundwater with high As concentration (>10 μg/L) was used for agriculture and aquaculture in the coastal areas. Intensive pumping has caused abrupt groundwater level drop, serious seawater intrusion and land subsidence (Liu et al., 2001; Lin et al., 2006). Moreover, the use of As-contaminated groundwater for agriculture and aquaculture has allowed As to indirectly enter the food chain via various paths and pose health risks to local residents.

2.2. Dynamic factor analysis

Time series are time dependent data showing a systematic and a non-systematic variations. Time series analysis is generally based on decomposing the characterization of the deterministic and random variations. However, the analysis of multivariate time series by classical methods is difficult because of data stationarity (Zuur et al., 2003a). These methods required a long time series data and were unable to recover the missing values (Solow, 1994; Zuur et al., 2003a). DFA is designed to identify the underlying common trends or latent effects in several time series and interactions among them. It allows for the evaluation of the effect of explanatory variables. DFA is similar to other dimension reduction techniques like factor analysis or redundancy analysis, but it considers the time component and thus can be adopted for analysis of non-stationary time series. The DFA is based on the so-called structural time series models (Harvey, 1989) that provides descriptions of the measured time series data of N response variables and the scheme of DFA is described as follows (Zuur et al., 2003b):

$$N \text{ time series} = \text{linear combination of} M \text{ common trends} + \text{level parameters} + \text{explanatory variables} + \text{noise} \quad (1)$$

The DFA choose minimum M to yield a reasonable model fitting result. The mathematical formulation of DFM is given by

$$S_n(t) = \sum_{m=1}^{M} \gamma_{m,n}\alpha_m(t) + \mu_n + \sum_{k=1}^{K} \beta_{k,n}x_k(t) + \varepsilon_n(t) \quad (2)$$

$$\alpha_m(t) = \alpha_m(t-1) + \eta_m(t) \quad (3)$$

where $S_n(t)$ is the value of the nth response variable at time t, which represents herein the groundwater As concentration at the nth well and time t. $\sum_{m=1}^{M}\gamma_{m,n}\alpha_m(t)$ is a linear combination of common trends, in which $\gamma_{m,n}(t)$ is the factor loading and $\alpha_m(t)$ is the mth unknown common trend at time t. μ_n is the nth constant level parameter (intercept term) to move up or down each linear combination of common trends. $\sum_{k=1}^{K}\beta_{k,n}x_k(t)$ represents a linear

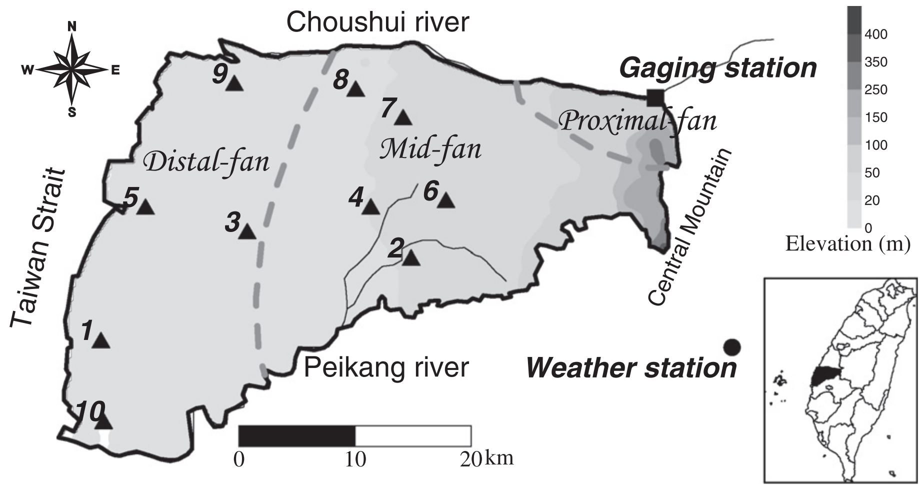

Fig. 1. Locations of 10 shallow monitoring wells (▲), a weather station (●) and a river flow gauging station (■) in the Yun-Lin county of Taiwan. The contour map and dashed line denote geographical elevations and hydrogeological boundaries, respectively.

combination of explanatory variables, in which $x_k(t)$ is the value of the kth explanatory variable at time t and $\beta_{k,n}$ is the regression coefficients for the K explanatory variables. When the time series explanatory variables are normalized, the magnitude of $\beta_{k,n}$ represents the influence of each explanatory variable $x_k(t)$ on the corresponding $S_n(t)$. $\varepsilon_n(t)$ and $\eta_m(t)$ are noise components which are assumed to be independent on each response time series, homogeneity, and normally distributed with mean zero and a covariance matrix. Notably, if seasonal or cyclic components are present in the time series, they will be masked and included in the trend component of Eq. (3). The mth trend at time t is equal to the mth trend at time $t-1$ plus a contribution of the noise component, $\eta_m(t)$. If the corresponding diagonal element of Q is relatively small, then the contribution of the error component will be small for all t, and the mth trend will be a smooth curve. Hence, the trends are smoothing functions over time and are independent on each other. This study used the Brodgar statistical software (Highland Statistics Ltd., Newburgh, UK, www.brodgar.com) to conduct DFA.

Analytical results of DFA are interpreted in terms of regression parameters $\beta_{k,n}$, the canonical correlation coefficients $\rho_{m,n}$, and the match between model estimations and observed values. The goodness-of-fit of model is assessed by visual inspection between the observed and predicted N time series, the coefficient of efficiency (Ce) (Nash and Sutcliffe, 1970) and Akaike's Information Criterion (AIC) (Akaike, 1974). The Ce is a relative assessment of the model performances and has been applied to assess the predictive power of hydrological and water quality models (Chiang et al., 2007; Harmel and Smith, 2007; McCuen et al., 2006). It compares the variance of 1:1 line (perfect agreement) to the variance of observed data. Statistical significance (p-value) for Ce was estimated with the bootstrap percentile-t method (Zoubir and Boashash, 1998). The AIC is a statistical criterion for model selection that combines the measure of fit with a penalty term based on the number of parameters used in the DFM. The more trends or explanatory variables were used, the better of the fit. However, the penalty also increases for each parameter added into the model. To compare two or more models, the DFM with the lowest AIC value represents an appropriate model. The common trends, $\alpha_m(t)$, are functions that represent the patterns of data that cannot be described with the explanatory variables included in the model. Canonical correlations coefficients ($\rho_{m,n}$) are used to assess the cross-correlation between the response variables (S_n) and the common trends (α_m). If $\rho_{m,n}$ is close to 1, it indicates stronger correlation between the corresponding response variable time series and the common trend. The terms 'high', 'moderate', and 'weak' correlation are applied to $|\rho_{m,n}| > 0.75$, 0.5–0.75, and 0.3–0.5, respectively. The influences of each explanatory variable x_k on each S_n is given by the regression parameters, $\beta_{k,n}$.

2.3. Analysis procedure and compilation of DFM

Representative response and explanatory variables were collected from available geochemical data and hydrological records. Multivariate time series analysis was conducted by three steps. First, relevant explanatory and response variables were identified according to our previous studies (Liu et al., 2006; Wang et al., 2007). Time series of As concentrations were considered as response variables. The explanatory variables were reduced by using variance inflation factor (VIF) analysis (Zuur et al., 2007), which eliminated the collinear variables. Second, goodness-of-fit indicators (AIC and Ce) were used to determine the optimal DFM. These models were obtained by the various combinations of common trends and explanatory variables. Then, Ce and root mean square error (RMSE) were used to evaluate the goodness-of-fit of each response variable in each well. In addition, the selected DFM performance was evaluated by visual inspection of the match between fitted and observed As contents. Third, the obtained results from the optimal DFM were comparatively examined with local hydrological and geochemical settings.

In this study, the seasonal groundwater quality data of mid- and distal-Choushui river alluvial fan areas were collected from Environmental Protection Agency during the period of 2001–2008. The field sampling and the chemical analyses of groundwater followed the NIEA code W103.50B and APHA (2005), respectively. For all chemicals, a total of 15 samples including blank, spike, duplicate and check samples (standard solutions from Merck) were measured sequentially (APHA 2005). Table 1 shows the mean, minimum, maximum, and samples number of obtained groundwater quality data within this period. All data used in this study are higher than the detection limit of chemicals. For missing values of variables, the algorithm for Kalman filtering and smoothing approaches of DFA were adopted for dealing with missing data (Zuur et al., 2003b). High average As concentrations were found in shallow well Nos. 5 (89.7 ± 52.2 μg/L) and 10 (28.6 ± 10.2 μg/L). The correlations between As concentration with total organic carbon (TOC) ($R = 0.55$) and inorganic carbon (alkalinity) ($R = 0.35$) are significant (both p-values < 0.01) (Wang et al., 2007). Stüben et al. (2003) suggested that the presence of NH_4^+ re-

Table 1
Analytical results of groundwater quality and hydrological parameters during 2001–2008.

Wells	As (μg/L)	Fe (mg/L)	TOC (mg/L)	NH_4^+ (mg/L)	GE (m) [d]
1	5.80 ± 2.25 (1.6–10.8) [a] *n* = 28 [b]	3.76 ± 3.26 (0.12–13.60) *n* = 25	0.99 ± 0.35 (0.33–2.13) *n* = 32	2.89 ± 1.47 (0.13–6.88) *n* = 32	−3.2 ± 0.5 (−4.37 to −2.17) *n* = 29
2	16.59 ± 8.13 (6.7–38.7) *n* = 28	3.32 ± 1.66 (0.31–7.67) *n* = 25	1.98 ± 0.58 (0.97–3.73) *n* = 32	0.53 ± 0.30 (0.12–1.48) *n* = 32	7.8 ± 2.5 (2.68–12.85) *n* = 30
3	15.01 ± 23.08 (ND-112.0) [c] *n* = 28	2.07 ± 2.13 (0.01–7.37) *n* = 25	1.73 ± 0.65 (0.54–3.82) n = 32	0.78 ± 0.65 (0.03–2.75) *n* = 32	−8.5 ± 2.8 (−13.66 to −3.93) *n* = 30
4	6.50 ± 2.91 (3.0–15.1) *n* = 28	0.80 ± 0.83 (0.01–4.46) *n* = 25	1.95 ± 0.58 (1.07–3.53) *n* = 32	2.00 ± 1.71 (0.56–7.80) *n* = 32	9.5 ± 0.9 (7.53–11.05) *n* = 30
5	89.67 ± 52.15 (ND-261.0) *n* = 28	1.15 ± 1.01 (0.14–4.72) *n* = 25	1.71 ± 0.57 (0.85–2.81) *n* = 32	0.40 ± 0.21 (0.17–1.03) *n* = 32	−6.0 ± 2.6 (−9.51 to −0.72) *n* = 30
6	4.24 ± 13.59 (ND-70.6) *n* = 27	2.34 ± 5.58 (ND-19.10) *n* = 24	2.33 ± 1.25 (0.56–5.96) *n* = 31	0.15 ± 0.27 (0.02–1.48) *n* = 31	28.8 ± 0.8 (27.14–30.36) *n* = 30
7	13.23 ± 6.32 (5.1–27.5) *n* = 28	1.94 ± 1.12 (0.22–4.54) *n* = 25	1.70 ± 0.72 (0.57–3.64) *n* = 32	0.15 ± 0.09 (0.05–0.52) *n* = 32	19.0 ± 0.8 (17.33–20.26) *n* = 29
8	22.40 ± 12.58 (3.1–73.7) *n* = 28	3.57 ± 1.24 (1.28–5.40) *n* = 25	2.06 ± 0.79 (0.80–3.74) *n* = 32	0.43 ± 0.15 (0.13–0.84) *n* = 32	9.1 ± 1.2 (6.87–10.93) *n* = 30
9	22.03 ± 17.07 (3.9–79.8) *n* = 28	2.50 ± 2.38 (ND-10.10) *n* = 25	2.04 ± 0.76 (0.78–4.04) *n* = 32	3.05 ± 1.77 (0.38–8.75) *n* = 32	4.3 ± 1.8 (0.97–7.50) *n* = 30
10	28.56 ± 10.20 (ND-50.4) *n* = 28	8.92 ± 7.03 (0.34–19.60) *n* = 25	1.65 ± 0.84 (0.66–4.67) *n* = 32	7.99 ± 3.73 (4.25–27.0) *n* = 32	−25.8 ± 4.6 (−37.25 to −17.8) *n* = 29

[a] Minimum and maximum.
[b] Number of samples.
[c] ND denotes not detected.
[d] Groundwater elevation related to sea level.

flects the reducing condition for the release of As and Fe into groundwater. Moreover, hydrological properties including groundwater elevation, precipitation and river flow may also affect the As distribution and transformation (Ayotte et al., 2006). Since fluctuation of groundwater As concentration may be a function of regional hydrogeochemical processes governed by various hydrogeochemical factors, the As-related geochemical variables ([As], [Fe], [TOC], and [NH_4^+], i.e. the time series of As, Fe, TOC, and $NH_4{}^+$ concentrations, respectively) and redox-sensitive hydrological parameters ([GE], [P], and [R], i.e. the time series of groundwater elevation, precipitation and river flow, respectively) were used as the explanatory variables. Collectively, the selected explanatory variables are: (1) *As05* and *As10* ([As] of wells Nos. 5 and 10, μg/L); (2) *Fe05* and *Fe10* ([Fe] of wells Nos. 5 and 10, mg/L); (3) *TOC05* and *TOC10* ([TOC] of wells Nos. 5 and 10, mg/L); (4) *NH05* and *NH10* ([NH_4^+] of wells Nos. 5 and 10, mg/L); (5) *GE10* ([GE] of No.10 well, m); (6) *P* ([P] from the weather station, mm) and (7) *R* ([R] from the gauging station, m^3/s) are adopted for DFM to predict the response variables of the remaining eight As concentrations in well Nos. 1–4 and 6–9.

3. Results and discussion

3.1. Experimental time series data

Table 1 represents the mean, minimum, and maximum of considered response and explanatory variables and shows the high variability of most of variables. High groundwater As concentrations (>10 μg/L) were observed in seven monitoring wells. Complicated geochemical reactions and hydrological processes lead to weak correlations ($|r| < 0.6$) between As, Fe, TOC, NH_4^+ concentrations and groundwater elevation, though the reductive dissolution of Fe (hydr)oxides is regarded as the most plausible mechanism of As mobilization in this area (Wang et al., 2007). In general, average concentrations of As, Fe, TOC and $NH_4{}^+$ tend to decrease from the coastal area (well Nos. 1, 5, 9 and 10) to mid-fan area (well Nos. 2, 3, 4, 6, 7 and 8). Fig. 2 illustrates the time series data of quarterly precipitation, river flow, and groundwater elevations, average As, Fe, TOC, NH_4^+ concentrations of wells during the eight-year period. Groundwater elevation, precipitation and river flow can be clearly discriminated between dry and wet seasons (Fig. 2a–c). Although the peaks of As, Fe, TOC and NH_4^+ concentrations are not concentrated in dry or wet seasons, the seasonal variation of As, Fe, and NH_4^+ concentrations generally decreases from 2001 to 2008 whereas that of TOC exhibit a slight increase (Fig. 2d–g). Due to high variations of all response and explanatory variables, the original time series data were deseasonalized by using the LOESS smoothing method (Cleveland, 1993) to reduce outliers originated from the effect of seasonal variations. The deseasonalized response and explanatory variables were then standardized (mean value is 0 and standard deviation is 1), to perform DFA. Moreover, variance inflation factor (VIF) analysis was adopted to identify the collinearity of explanatory variables before conducting DFA (Zuur et al., 2007). The VIF values for the selected variables are given by $1/(1 - R^2)$. Hence, linear regression is used for predicting each explanatory variable as a function of others (Montgomery and Peck, 1992). All explanatory variables were considered in VIF analysis, including *TOC05*, *TOC10*, *Fe05*, *Fe10*, *NH05*, *NH10*, *As05*, *As10*, *GE10*, *P* and *R*. Only variables with VIF value <5 were used for the DFA (Table 2).

3.2. Performance of the optimal DFM

The optimal DFM was determined based on the different numbers of common trends and the combinations of explanatory

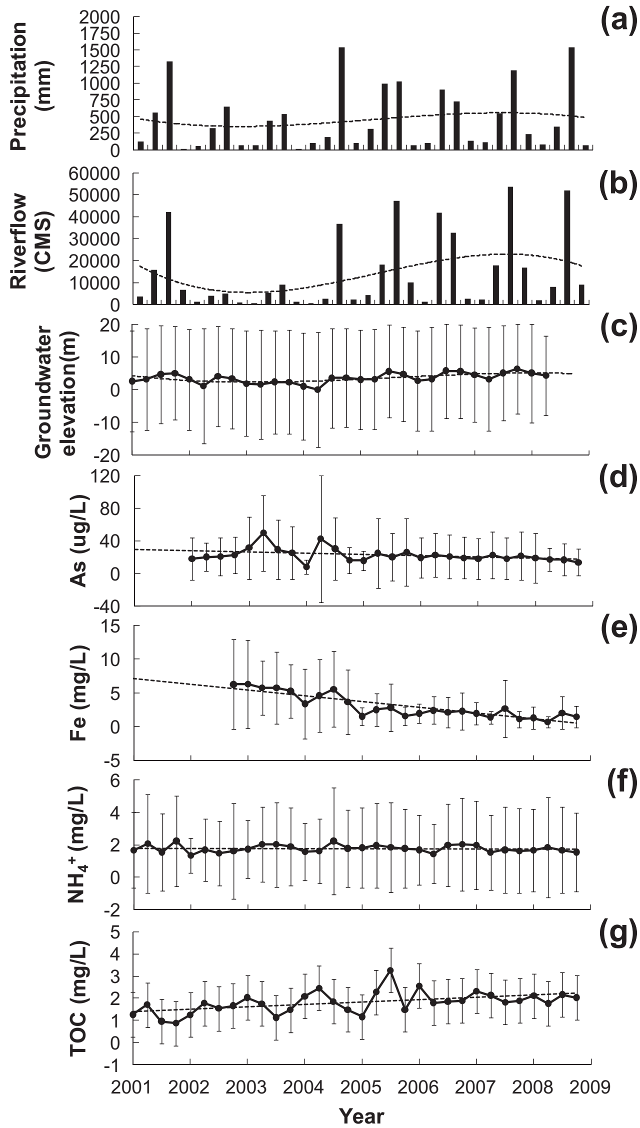

Fig. 2. Seasonal variations of the hydrological and geochemical explanatory variables during 2001–2008. (a) precipitation from a weather station, (b) river flow records from a river flow gauging station, and average (c) groundwater elevation, (d) As, (e) Fe, (f) TOC, (g) NH_4^+ of 10 shallow wells. The dashed lines denote the linear or polynomial regressions of explanatory variables. Error bars represent standard deviation from mean of variables.

Table 2
The calculated values of variance inflation factor (VIF) analysis for the explanatory variables.

Variable [a]	VIF
As05	2.51
As10	2.69
TOC05	3.25
TOC10	3.24
Fe05	1.47
Fe10	3.71
NH05	1.25
NH10	1.67
GE10	3.66
P	2.53
R	3.77

[a] Variables of *As*, *TOC*, *Fe*, *NH*, *GE*, *P*, and *R* denote the time series of As, TOC, Fe, NH_4^+ concentrations, and groundwater elevation, precipitation, river flow records, respectively. Numbers of *05* and *10* denote the monitoring wells of Nos. 5 and 10, respectively.

Table 3
Selection of dynamic factor models based on performance coefficients.

Trend	Explanatory variables	AIC	*Ce* [a]
1	–	561.5	**0.265**
2	–	538.1	**0.524**
3	–	537.1	**0.635**
4	–	547.1	**0.634**
1	*As(05, 10)* [b]	555.8	**0.412**
1	*As(05, 10), GE10*	547.0	**0.476**
1	*As(05, 10), GE10, TOC(05, 10)*	538.0	**0.545**
1	*As(05, 10), GE10, TOC(05, 10), Fe(05, 10)*	462.3	**0.600**
1	*As(05, 10), GE10, TOC(05, 10), Fe(05, 10), NH(05, 10)*	378.1	**0.732**
1	*As(05, 10), GE10, TOC(05, 10), Fe(05, 10), NH(05, 10), P*	374.5	**0.759**
1	*As(05, 10), GE10, TOC(05, 10), Fe(05, 10), NH(05, 10), R*	373.9	**0.764**
1 [c]	***As(05, 10), GE10, TOC(05, 10), Fe(05, 10), NH(05, 10), P, R***	347.3	**0.806**

[a] *Ce* was calculated with the combined set of predictive versus observed values for all the wells. Values of *Ce* in bold denote the significant correlation (p-value < 0.01).
[b] Number in the parentheses denotes well No.
[c] The best DFM for the consideration of explanatory variables.

Table 4
Optimal DFM results for groundwater As in eight wells.

S_n	As01	As02	As03	As04	As06	As07	As08	As09
$\gamma_{1,n}$	0.11	0.09	−0.15	−0.16	−0.01	0.00	0.07	0.03
μ_n	−0.01	0.10	−0.03	−0.04	0.02	−0.03	−0.03	0.04
$\beta_{GE10,n}$	−0.24	**−0.31** [a]	−0.08	**0.28**	−0.21	**0.50**	0.21	**−0.45**
$\beta_{P,n}$	**−0.33**	**0.42**	**−0.17**	**−0.23**	−0.01	**−1.52**	**0.58**	**0.52**
$\beta_{R,n}$	0.31	−0.26	0.11	0.23	**−0.45**	**0.78**	**−1.14**	**−0.42**
$\beta_{As05,n}$	0.08	0.03	0.17	**0.38**	0.10	**0.22**	−0.03	−0.02
$\beta_{As10,n}$	**0.57**	−0.01	0.13	0.16	0.20	0.37	−0.01	0.25
$\beta_{Fe05,n}$	**0.32**	−0.01	**0.89**	**0.57**	0.20	**0.47**	**0.34**	−0.26
$\beta_{Fe10,n}$	−0.07	**−0.57**	0.05	0.14	−0.20	0.11	**−0.63**	−0.20
$\beta_{TOC05,n}$	0.03	−0.07	**−0.50**	**−0.64**	−0.10	**−0.27**	**0.26**	**−0.37**
$\beta_{TOC10,n}$	−0.03	0.06	0.00	0.06	−0.01	0.04	0.12	**−0.41**
$\beta_{NH05,n}$	0.02	**−0.13**	**−0.12**	**−0.18**	−0.01	**−0.12**	0.07	0.01
$\beta_{NH10,n}$	**0.56**	**0.65**	0.06	0.12	0.14	0.40	0.05	**0.75**
Ce	**0.94**	**0.87**	**0.97**	**0.88**	0.39	**0.82**	**0.74**	**0.74**
RMSE [b]	0.20	0.24	0.13	0.21	0.50	0.32	0.40	0.38

[a] Regression coefficient of explanatory variables in bold denote significant value (t-value > 2).
[b] Root mean square error.

variables (Table 3). The *Ce* values of each DFM which were calculated by the predictive and observed data are all positive significant. For DFM without explanatory variables, the DFM using three common trends yielded the low AIC and high *Ce* values, indicating that several latent effects influence the variability of [As]. In this study, the hydrogeological conditions of 10 monitoring wells were similar, including the depths of screens, geological deposits, topographic elevations, land uses, precipitation, and evaporation. Hence, the DFM using one common trend is adopted for further consideration of explanatory variables. Table 3 shows that increasing explanatory variables in the DFM decreased the AIC values and increased *Ce* values. The DFM with one common trend and included all explanatory variables yielded the best description of time series of As concentrations in eight wells (AIC = 373.1 and *Ce* = 0.806). Table 4 summarizes the results obtained from the optimal DFM, including estimated regression parameters, factor loadings, canonical correlation coefficients, and level parameters. The optimal DFM was used to quantify the major factors affecting As concentrations and to predict As contents variations in the remaining eight wells. The As concentrations of seven wells were fitted by the optimal DFM with high *Ce* values (0.74–0.97) while for well No. 6 the fit was not satisfactory (*Ce* = 0.39). The explanatory variables

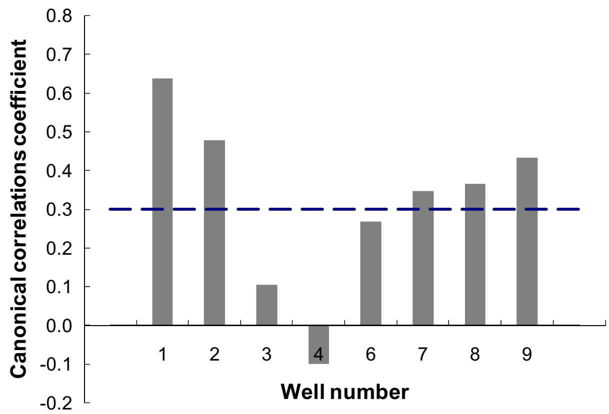

Fig. 3. Corresponding canonical coefficients for the best dynamic factor models. The dashed line indicates the threshold (0.3) for weak correlations with the common trend.

included in the DFM describe well [As] variations. The regression parameters ($\beta_{k,n}$) represent the intensity of the corresponding explanatory variable in the model. Relatively high values of $\beta_{P,n}$, $\beta_{R,n}$, $\beta_{GE10,n}$, $\beta_{Fe05,n}$, $\beta_{TOC05,n}$, and $\beta_{NH05,n}$ in most of S_n indicate that the hydrological parameters (precipitation, river flow and groundwater elevation) and geochemical variables (Fe, TOC and NH_4^+) are the predominate factors, governing the variation of As concentration in groundwater. Notably, *As06* time series is less influenced by most of the variables because well No. 6 located in upstream area of the mid-fan where the redox condition of shallow aquifer is in oxidative condition and anthropogenic activities is mild. Moreover, the corresponding canonical correlation coefficients ($\rho_{1,n}$) (Fig. 3) denote relationship between As concentrations of each wells and the estimated common trend. The [As] variations of well Nos. 1, 2, 7, 8 and 9 have weak to moderate correlations ($0.3 < |\rho_{1,n}| < 0.65$) with the common trend.

Fig. 4a shows the model fit results derived from the optimal DFM with the selected explanatory variables. Chang et al. (2010) reported that artificial neural networks (ANNs) were useful for predicting the spatial distribution of groundwater As concentration in the south-western coastal area of Taiwan. Several strategies were proposed to recover the missing data and to alleviate over-fitting problem, including leave-one-out cross-validation, modified performance function, and principal component analysis. However, estimations by using ANNs for some groundwater [As] variations were unreliable, especially when variables contained high variations and short non-stationary time-series characteristics. In this study, the DFM improved the aforementioned defects and satisfactorily matched the groundwater [As] fluctuations with high *Ce* values except for a few peaks. Moreover, the prediction by DFM with deseasonalized time series (Fig. 4a) is better than with original time series (Fig. 4b), indicating the LOESS smoothing method (Cleveland, 1993) can effectively reduce outliers originated from the effect of seasonal variations. The determined common trend from optimal DFM (Fig. 5) showed that groundwater As concentration in this area gradually reduce from 2001 to 2008. With the lowest common trend on 2008, however, groundwater As concentrations of eight wells were still higher than 10 μg/L. The common trends in wet season were higher than those in dry season of the year (t-test, $p < 0.05$) indicating that high groundwater As concentration mostly occurred in wet season.

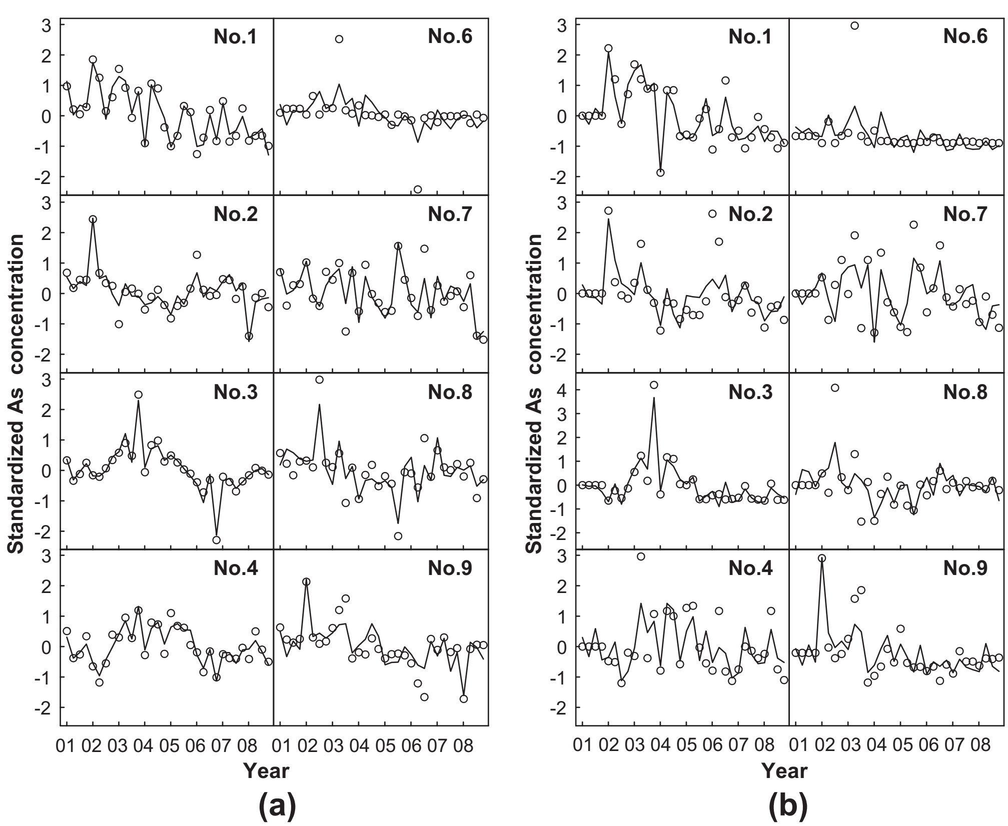

Fig. 4. Dynamic factor model fits for groundwater As of eight shallow wells in standardized concentration by using (a) deseasonalized time series and (b) original time series. The open circles and solid lines denote the measured and predicted As concentration, respectively.

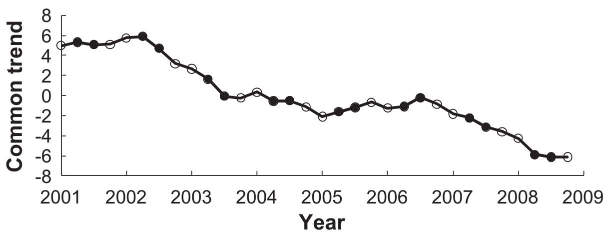

Fig. 5. Common trends of the best dynamic factor models. The open and solid circles denote the dry and wet seasons, respectively.

3.3. Effects of hydrological variables on temporal variation of groundwater As

The regression parameters of precipitation and river flow in Table 4 ($\beta_{P,n}$ and $\beta_{R,n}$) were pronounced in most [As] time series. The contribution of river flow to the wells nearby the Choushui river (well Nos. 6–9) are more relevant than those to the remaining wells (well Nos. 1–4). High hydraulic conductivities of shallow aquifers in mid- and distal-fan areas (average of 4.2×10^{-4} and 2.5×10^{-4} m/s, respectively) provide a rapid infiltration path of rainfall and river water. Infiltrated rainfall water within a rainy season (~5–6 months) may result in rising the groundwater elevation and accelerating As release in the reductive environment. The rising groundwater elevation, concerning with increasing rainfall in recharge periods, leads to the increase of As concentration (Costa Goncalves et al., 2007) and Fe concentrations (Itai et al., 2008) via the reductive dissolution of As-rich Fe oxy-hydroxides. The distribution of groundwater ^{18}O isotope composition indicated that the infiltration of river water, especially the Choushui river, and the precipitation of mountainous area are the main sources of groundwater recharge (Wang et al., 2005). Chiang et al. (2005) indicated that 39% of recharged groundwater in Yun-Lin county is derived from the Choushui river water and 28% from the rainfall by the mass balance analysis of ^{18}O isotope composition. However, common trend of groundwater [As] variation (Fig. 5) is inversely correlated with the regressions of precipitation ($R = -0.14$, p-value > 0.01) and river flow variations ($R = -0.25$, p-value > 0.01) (Fig. 2a and b). The quarterly rainfall variation and river flow progressively increased from 2002 to 2008, whereas the common trend decreased correspondingly. Fast groundwater flow (approximately 142 and 56 m/yr, respectively) in mid- and distal-fan areas (Central Geological Survey, 1999) may transport the As concentration liberated during wet season to the downstream area (Rodriguez et al., 2004; Greif et al., 2008). Collectively, the fluctuation of As concentrations are likely affected by the infiltration from precipitation and river water, and the movement of groundwater flow.

According to the DFA results in this study, the contribution of groundwater elevation on As concentration changes are not relevant ($\beta_{GE10,n}$ values in Table 4). The oscillations of groundwater elevations are caused by groundwater recharge, discharge, and man-made pumping. Itai et al. (2008) indicated that spatial variation of redox condition is associated with the recharge/discharge cycle of groundwater. Groundwater abstraction by local inhabitants increases due to the rapid population growth and the lack of surface water resources during dry season. Around 60% and 76% water demands for agriculture and aquaculture, respectively, depends on groundwater in Yun-Lin county. Man-made excessive pumping may accelerate the rate of vertical infiltration of surface water (Lambrakis et al., 1996) and promote water–sediment interaction in aquifer (Petalas and Lambrakis, 2006). The intensive groundwater extraction hinders the effect of natural recharge and discharge on the variation of groundwater elevation and masks the correlation between As concentrations and groundwater elevation. Moreover, excessive abstraction of groundwater may cause downward movement of Fe-reducing conditions and mobilization of As to deep aquifers (Polizzotto et al., 2005; Berg et al., 2008). Anthropogenic influences of pumping for irrigation have been proposed to explain the elevated groundwater As concentration in many regions (Harvey et al., 2005; Klump et al., 2006; Neumann et al., 2010). Over-pumping in Yun-Lin county hence has acutely led to the groundwater drawdown, land subsidence, seawater intrusion, and change of redox condition and As geochemical cycle. To comply with the drinking water guideline of As concentration (10 μg/L), effectively conjunctive utilization of surface water and groundwater resources is one of the most important issues in future, especially during drought periods.

3.4. Implication of As distribution and geochemical conditions

The regression parameters of geochemical variables in Table 4 showed that variations of Fe, TOC, and NH_4^+ concentrations significantly affect the elevated As concentration of shallow groundwater in this area. The similar variations of the common trend and Fe concentration (Figs. 5 and 2e) ($R = 0.77$, p-value < 0.01) supported that the reductive dissolution of As-rich Fe (hydr)oxides in anaerobic groundwater is responsible for the mobility of As in groundwater of this area (Wang et al., 2007). Microbial degradation of organic matter is regarded as a biogeochemical factor of reductive reaction in anaerobic groundwater (Harvey et al., 2002; Akai et al., 2004). Long-term operation of farmed fish ponds caused the infiltration of labile organic carbon into shallow aquifer (Benner, 2010; Neumann et al., 2010). From 2001 to 2008, increased TOC concentration (Fig. 2g) recharged from paddy fields and fish ponds in this area provides sufficient electron donors for Fe reduction under a stable reducing environment (Fig. 2f) and driven As mobilization, especially in wet seasons.

However, the influence of the geochemical variables in well No. 5 is greater than those in well No. 10 for most [As] time series (Table 4). According to geochemical characteristics of groundwater in this area (Fig. 6), the monitoring wells were divided into saline wells (Nos. 1 and 10) and non-saline wells (Nos. 2–9). This is also evidenced by the significant regression parameter between $\beta_{As10,n}$

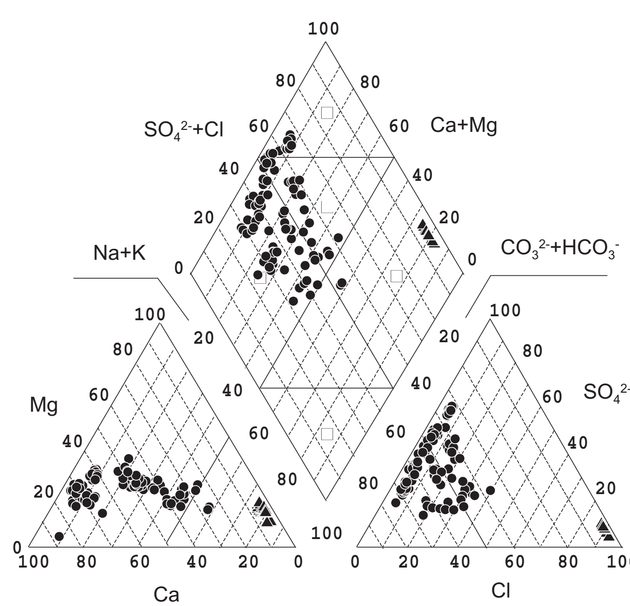

Fig. 6. Piper diagram of 8-years groundwater quality data of 10 monitoring wells. Solid triangles (▲) and circles (●) denote saline wells (Nos. 1 and 10) and non-saline wells (Nos. 2–9), respectively.

and *As01* time series. The similar tendencies of average Fe, TOC, NH_4^+, and SO_4^{2-} concentrations between saline and non-saline wells (Fig. 7) described the common effect of hydrological condition on geological setting and long-term geochemical variations in this area, even the extent of groundwater salinization in each well is different. Average groundwater As concentration of non-saline wells (23.8 ± 9.7 μg/L) was significantly higher than that of saline wells (17.2 ± 5.6 μg/L) (*t*-test, $p < 0.05$), while average Fe concentrations (2.2 ± 1.3 mg/L) of non-saline wells were lower than those of saline wells (6.3 ± 4.6 mg/L) (*t*-test, $p < 0.05$). In non-saline wells, the NH_4^+ concentrations in wet season was close to that in dry season (*t*-test, $p > 0.05$). The stable reducing environment suggested that As-bearing Fe oxyhydroxides acted as a major source of As

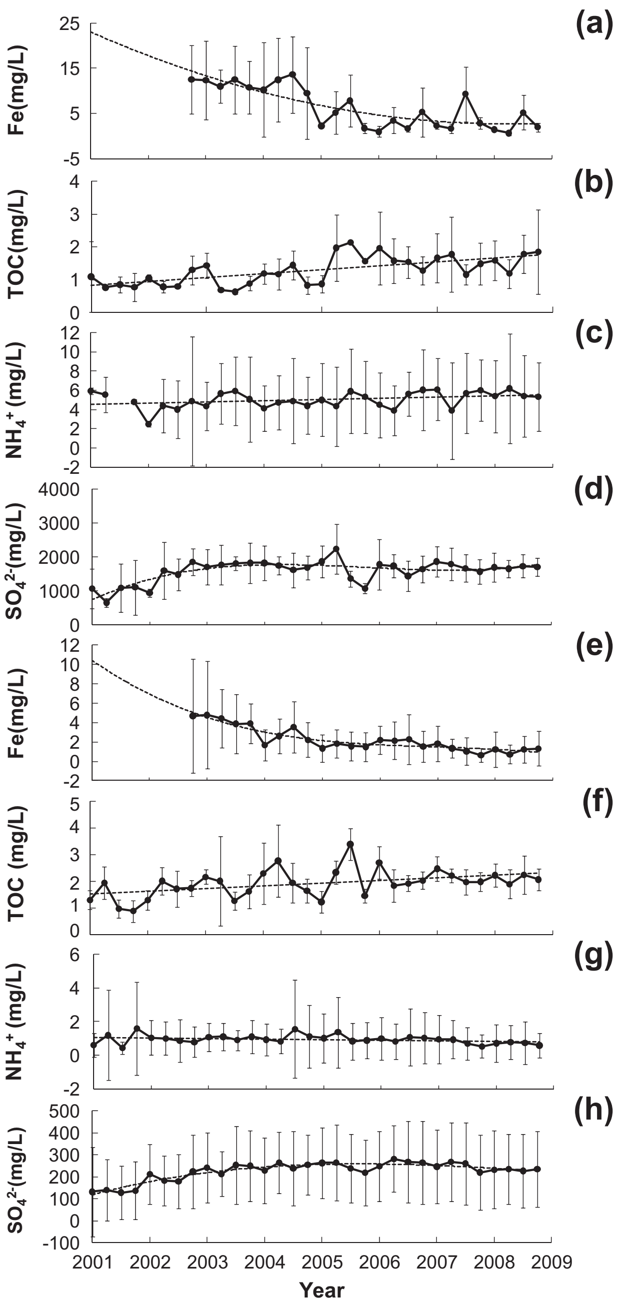

Fig. 7. Seasonal variations of average Fe, TOC, NH_4^+, and SO_4^{2-} concentrations in saline wells (a–d) and non-saline wells (e–h). The dashed lines denote the linear or polynomial regressions of explanatory variables. Error bars represent standard deviation from mean of variables.

(Lu et al., 2010) and the presence of high As concentrations (27.4 ± 11.5 μg/L) during wet seasons (*t*-test, $p < 0.05$). Besides, the occurrence of secondary mineralization of dissolved ferrous iron (Hansel et al., 2003) explained low Fe concentrations in both of dry and wet seasons in non-saline wells (~2.2 mg/L) (*t*-test, $p > 0.05$).

Furthermore, average TOC and NH_4^+ concentrations of saline wells in wet season were higher than those in dry season (*t*-test, both $p < 0.05$). The reducing environment in the wet seasons was formed by the risen groundwater elevation via rainfall infiltration and river water recharge. Presence of high SO_4^{2-} concentration in saline wells may act as an electron acceptor in the reduction process (Nath et al., 2008). High Fe (7.0 ± 4.7 mg/L) and sulfate concentrations in wet season could increase the ion strength in groundwater and trigger the mobility of As (Keon et al., 2001), but microbial-mediated sulfate reduction could also result in the formation of pyrite with co-precipitation or sorption of As (Mallick and Rajagopal, 1996) and reduce As concentration in groundwater. The following oxidation of As-bearing pyrite in dry season may act as a source of As (Chowdhury et al., 1999; Schreiber et al., 2000; Lowers et al., 2007) and elevated As concentration. However, the elevated As concentrations were not observed during the dry season in this study (*t*-test, $p > 0.05$). Lu et al. (2010) indicated that sulfate reduction occurred in distal-fan, and showed that the primary sulfide mineral, pyrite, was identified as authigenic framboidal type. Hence, As mobility in saline wells is primarily associated with the cyclic redox reactions of Fe oxyhydroxides and As bearing sulfides (Anawar et al., 2003).

4. Conclusion

In this study hydrogeochemical multivariate time series of high As groundwater, collected from the downstream area of Choushui river alluvial fan in Taiwan, were evaluated by DFA. The optimal DFM successfully identified the trend of groundwater [As] fluctuation. Arsenic concentrations observed in eight wells from 2001 to 2008 were mainly governed by precipitation, river flow, groundwater elevation, Fe, TOC and NH_4^+ contents. The results supported the hypothesis that the plausible As release mechanism is the reductive dissolution of As-bearing Fe (hydr)oxides in anaerobic groundwater. According to the evaluation of hydrological variables in DFM, [As] fluctuation were governed by the surface water recharges during dry and wet seasons and the hydrogeological conditions, including hydraulic conductivities and groundwater flow patterns. Rapid infiltration of surface water, including rainfall and river water caused a rise in groundwater elevation in the rainy season and created a reductive environment, inducing Fe reductive dissolution and As liberation from sedimentary deposits to groundwater. Fast groundwater flow may transport the dissolved As concentration to the downstream areas. However, the impact of groundwater elevation on [As] variation is less significant. Intensive pumping for agricultural and aquacultural needs masked the influences of groundwater fluctuation on [As] variation and led to critical seawater intrusion in some wells.

According to the geochemical characteristic of groundwater, monitoring wells in this area were classified into saline and non-saline wells. For non-saline wells, high-As and low-Fe groundwater under a stable reducing environment suggested that the Fe reduction in wet seasons and subsequent secondary mineralization in dry seasons was responsible for As mobility. Additionally, amount of sulfate in saline groundwater may act as an electron acceptor and promoted co-precipitation of sulfide minerals. Formation of sulfide minerals restrained the liberated As in the wet season and may oxidatively release As to groundwater in the following dry season. The redox cyclic interactions of iron and sulfur between dry and wet seasons resulted the low-As and high-Fe concentrations in saline wells. Thus, hydrological and geochemical variables, respectively, controlled the temporal variation and spatial distribution of groundwater As concentration. To comply with the As guideline concentration of drinking water (10 μg/L), conjunctive utilization of surface water and groundwater resources at saline/non-saline areas during drought/flooding periods will be one of the effective management scheme in southwestern coastal area of Taiwan.

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