

# Impact of the East Asian summer monsoon on long-term variations in the acidity of summer precipitation in Central China

B. Z. Ge<sup>1,4</sup>, Z. F. Wang<sup>1</sup>, X. B. Xu<sup>2</sup>, J. Tang<sup>2</sup>, Y. J. He<sup>3</sup>, I. Uno<sup>5</sup>, and T. Ohara<sup>6</sup>

<sup>1</sup>State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry (LAPC), Institute of Atmospheric Physics, Chinese Academy of Science, Beijing 100029, China

<sup>2</sup>Key Laboratory for Atmospheric Chemistry, Centre for Atmosphere Watch and Services, Chinese Academy of Meteorological Sciences, China Meteorological Administration, Beijing, China

<sup>3</sup>Chinese Research Academy of Environmental Sciences, Beijing, China

<sup>4</sup>Graduate University of the Chinese Academy of Sciences, Beijing, 100049, China

<sup>5</sup>Research Institute for Applied Mechanics, Kyushu University, Fukuoka, Japan

<sup>6</sup>National Institute of Environmental Science, Tsukuba, Japan

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**Abstract.** The acidity of precipitation has been observed at stations of the Acid Rain Monitoring Network run by the China Meteorological Administration (CMA-ARMN) since 1992. Previous studies have shown that different long-term trends exist in different regions but detailed analysis of the causes of these is lacking. In this paper, we analyze summertime precipitation acidity data from the CMA-ARMN during 1992–2006 using EOFs and show that the summertime pH in China had different trends before and after 2000. The most significant decrease of pH is found in Central China. To investigate the causes of this decrease of pH in summer, we explore the relationship between changes in the pH value, the East Asian summer monsoon index, rainfall data, and pollutants emissions. We find that the East Asian summer monsoon can significantly affect the acidity of summer precipitation in Central China. In strong monsoon years, the pH in Central China is about 0.33 lower than that in weak monsoon years. Chemical transport model simulations using fixed emissions indicate that about 65% of the pH value difference (i.e., 0.22) is related to the summer monsoon, and constitutes 18–36% of the observed pH change (0.6 ~ 1.2) in Central China during 1992–2006. Further studies reveal a relationship between the pH in Central China and the rainfall in the middle and lower reaches of the Yangtze River (MLYR), which can explain about 24% of the variance of pH in Cen-

tral China. Simulations using an annually varying emission inventory show that at least 60% of the variation in precipitation acidity in Central China can be attributed to changes in pollutant emissions. Therefore, the increase in emissions of acidic species is the most important cause for the observed decrease of pH in Central China, and changes in meteorological factors, such as rainfall and other parameters related to the East Asian summer monsoon, play a less important but still significant role.

## 1 Introduction

Acid rain has been one of the most significant environmental problems in China since the late 1970s. Rapid economic development and population growth has led to increases in the emission and subsequent deposition of sulfur, and widespread acid rain was observed in southern and southwestern China between the 1980s and the mid 1990s (Ding et al., 1997, 2004; Wang and Wang, 1995, 1996; Zhao et al., 1988; Huang et al., 1995). The rapid expansion of the acid rain zone exposes citizens and ecosystems to great risk in southern and southwestern China, and after the mid 1990s the region of acid rain extended into Central China (Zhang et al., 2007; Xie et al., 2009). In order to reduce the emission of SO<sub>2</sub> and mitigate acid rain in China, the Chinese government issued the “Atmospheric Pollution Prevention and Control Act of the Peoples Republic of China” in 1995. Following



Correspondence to: Z. F. Wang  
(zifawang@mail.iap.ac.cn)

this act, a national control measure known as “Two Control Zones” (TCZ) was enacted in 1998 with the aim of strengthening control of SO<sub>2</sub> emissions (Liu et al., 1998). Meanwhile, municipal governments attempted to improve air quality in different ways to strengthen the control of the emissions of gaseous pollutants and particulate matter. Although these efforts had some positive effects in the first few years, rapid economic development and an increased demand for energy have caused further increases in the emissions of SO<sub>2</sub> and NO<sub>2</sub> over the last decade. A large number of reports (Zhang et al., 2008; Xie et al., 2009; Wang and Xu, 2009; SEPA; Gao et al., 2009; Larssen et al., 2006) reveal that nationwide emissions of SO<sub>2</sub> and NO<sub>2</sub> have increased significantly since 2000, following the temporary decrease in emissions in the 1990s.

Most studies on acid rain link its variation to the emissions and transport of SO<sub>2</sub> and NO<sub>2</sub> (Khemani et al., 1994; Browning et al., 1991; Wang et al., 2007; Vogt et al., 2006; Quan et al., 2007; Liu et al., 2008; Larssen et al., 2006), but few consider the impacts of climatic conditions (Xie et al., 2009; Dayana and Lamb, 2008; Rai et al., 2009; Yeung et al., 2007; Tiwari et al., 2007; Ravichandran and Padmanabhamurty, 1994; Davies et al., 1991). Previous studies have shown that before the 1990s acid rain occurred mainly in southern and southwestern regions of China. More recently, however, acid rain has seriously affected the central parts of China, especially in eastern areas (Xie et al., 2009). Tang et al. (2010) used the Man-Kendall method to obtain a trend in precipitation acidity in China, and show that during 2000–2006 acid rain moved from southwestern to eastern and central China. We analyzed the change in precipitation acidity in China between 1992 and 2006, and found that the most distinct increase occurred in Central China. What are the causes of this increase and are they related to changes in climatic conditions? In this paper, we report the potential contributions of the summer monsoon and rainfall in the middle and lower Yangtze River regions to precipitation acidity in Central China. We also evaluate the influence of variations in emissions on changes in the acidity of precipitation in Central China.

## 2 Data and methodologies

### 2.1 Data

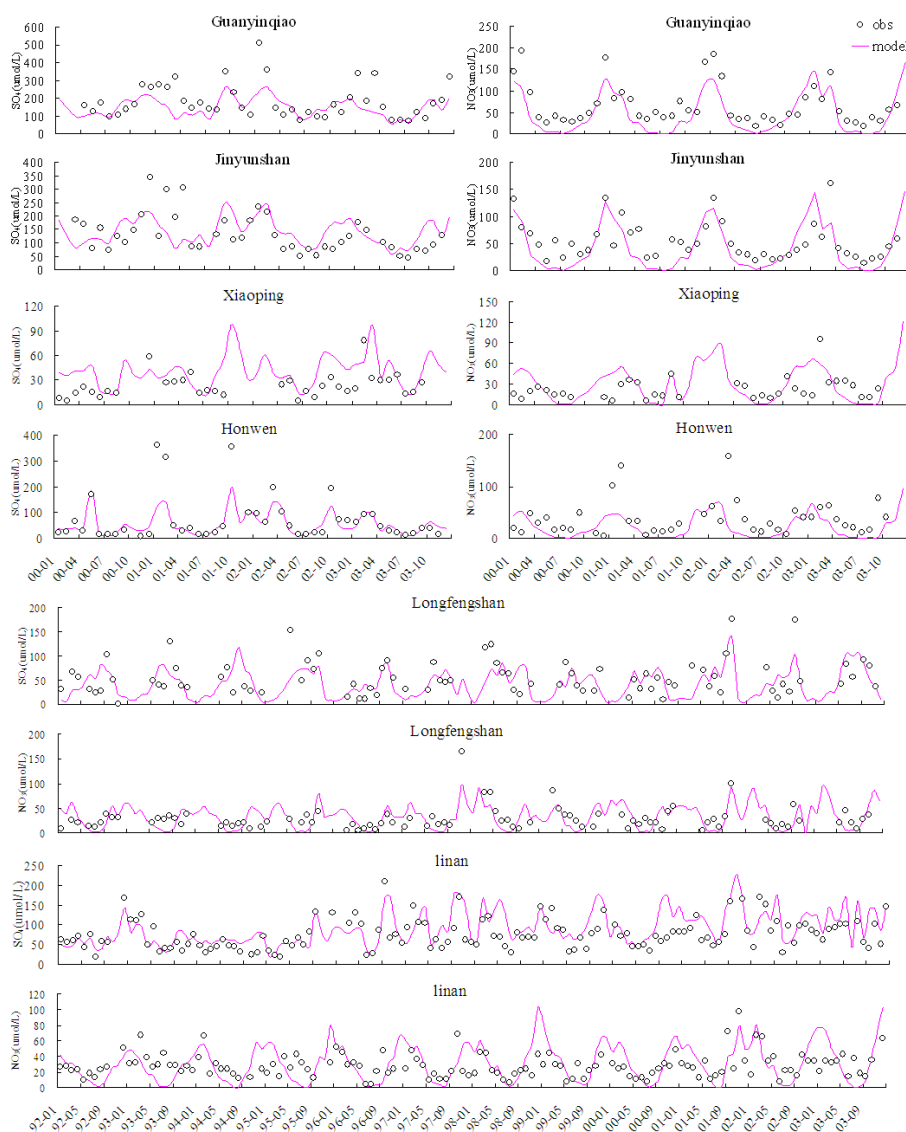
It is well known that soil aerosol/dust originating from northern China can affect precipitation acidity over almost all of the country in winter and springtime under conditions of continental outflow during the East Asian winter monsoon (Wang et al., 2002; Rodhe et al., 2002; Wai and Tanner, 2005; Yeung et al., 2007). In order to exclude this factor we choose to focus on summertime (from June to August) and study the contributions from the East Asian summer monsoon and pol-

lution to the variation in the acidity of precipitation in Central China.

Precipitation acidity, conductivity and rainfall are observed using polyethylene polymer bucket of 40 cm diameter at stations of the Acid Rain Monitoring Network run by the China Meteorological Administration (CMA-ARMN), which share the facilities of the meteorological observation stations, and are normally located in suburb of a city or a town, thus much less directly influenced by urban emissions. During the period 1992–2006, event samples were collected before 2005 and daily precipitation samples were collected during 2006 at 88 network stations. Strict quality assurance and quality control was applied to the dataset from the CMA-ARMN. Data for 14 of the 88 stations were found to be unsuitable for long-term trend analysis (Tang et al., 2007, 2010) and precipitation acidity data from the 74 remaining stations are used in this study. In details, 37 among the 74 sites are located in relatively sparsely populated areas, including 3 GAW (Global Atmosphere Watch) regional stations and 3 weather stations on top of mountains, the rest 37 are city or near-city sites. Although the sites are of different characteristics, they are mixed and distributed in different regions. In this paper, we focus on variations of precipitation acidity and meteorological parameters in larger regions and the relationship between these quantities. The quality of data from all the 74 sites was checked by series procedures (Tang et al., 2010) and is believed meeting our purpose. For meteorological data we use reanalysis data for 850 hPa wind fields from the National Centers for Environmental Prediction (NCEP) and rainfall at 753 stations selected by CMA. To define the monsoon intensity and to investigate the influence of the monsoon on precipitation acidity in China, we used a dynamical normalized seasonality Monsoon Index (MI), which was developed by Li and Zeng (2002) on the basis of grid-by-grid and season-by-season calculations of the climatological wind field.

### 2.2 Modeling

The modeling system used in this study is CMAQ version 4.4 (Byun and Ching, 1999) driven by meteorological fields calculated by the Regional Atmospheric Modeling System (RAMS) Version 4.3 (Cotton et al., 2003; Pielke et al., 1992), with initial and boundary conditions defined by NCEP reanalysis data. The horizontal resolution of the model is 80 km, and there are 19 layers in the vertical following a sigma-z coordination system up to 23 km, the modeling domain is shown as sector area in Fig. 2 (for more details see Yamaji et al., 2006). The model uses the Regional Emission Inventory in Asia (Ohara et al., 2007), which is based on energy statistics, emission factors, and other socioeconomic information and covers the years 1980–2003 (He et al., 2007, 2008; Yamaji et al., 2006). For the purpose of comparison, we conducted three simulations to derive SO<sub>2</sub>, NO<sub>2</sub>, SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> concentrations as monthly averages

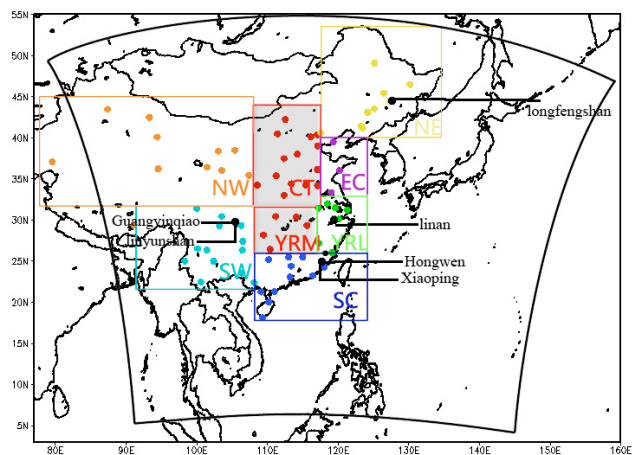


**Fig. 1.** Time series of modeled and observed monthly volume-weighted mean concentrations of sulfate and nitrate for EANET sites and GAW background stations.

for 1980–2003, but do not continue beyond 2003 in the absence of a suitable emission inventory. In the first simulation annually varying meteorological conditions were used along with the 2000 emission inventory, that is, only meteorological conditions change (hereafter MC); in the second, meteorological conditions for 2000 were used along with annually-varying emissions (hereafter EC); and in the third both meteorological conditions and emissions varied from year to year (hereafter Normal). It is worthy to mention that, we aimed to exclude the influence of pollutants emission for MC, and the meteorological factors for EC. According to that, the lowest emission and the modest value of summer monsoon in 2000 is ideal for our purpose.

The CMAQ model is widely used and is well validated with measurements from Chinese sites, e.g., Tieshanping in

the Sichuan basin and Caijiatang in Hunan province (Quan et al., 2008), and it was found that simulated  $\text{SO}_2$  and  $\text{SO}_4^{2-}$  agree well with observations. Ohara (2010) found that the model can reproduce the observed temporal variations in monthly wet deposition of  $\text{SO}_4^{2-}$  at long-term acid deposition monitoring sites in East Asia since 1988. Lin et al. (2008) using this model to investigate the Long-range transport of acidifying substances in East Asia and found the model is able to capture major spatial and seasonal variations of  $\text{SO}_2$ ,  $\text{NO}_2$  and wet sulfate and nitrate deposition over East Asia, although there exist an underestimation on nitrate caused by the uncertainties in emission of precursors for nitrates. In addition, many studies have validated  $\text{O}_3$  and related species, which affect atmospheric oxidation, using measurements at EANET observation stations (Tanimoto et al., 2009;



**Fig. 2.** Distribution of stations of CMA-ARMN and four EANET and two GAW stations marked by black dot, the shadow area in CT and YRM means the key area of our study, modeling domain is sector area.

Ohara et al., 2008), at the mountain sites Taishan, Huashan and Huangshan in China (He et al., 2008) and observations during the TRACE-P measurement campaign (Zhang et al., 2006b). These studies all show that the model can reproduce the tempo-spatial distributions of these species reasonably well. Thus, the model reproduces many of the important features of  $\text{SO}_2$  and  $\text{SO}_4^{2-}$  chemistry seen in the observations (Zhang et al., 2004; Zhang et al., 2006a). We have also compared the rainwater concentrations of  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  from the CMAQ model with four EANET observation stations in China, e.g., Hongwen and Xiaoping in Xiamen and Guanyinqiao and Jinyunshan in Chongqin, and two GAW sites, e.g., Linan in Yangtze River Delta and Longfengshan in northeast of China, and the trends of our model results are similar to that observed (see Fig. 1). The correlations between model and observations were greater than 0.5 (significant at the 95% level) and the bias of the mean values were less than 30% (see Table 1).

### 2.3 Volume weighted average method

The volume weighted average (VWA) value (Huang et al., 2008; Yeung et al., 2007; Seqheira and Lai, 1998) of pH is used, as this eliminates the influence of different rainfall intensities on the pH. VWA pH at a station in a certain period is calculated by multiplying the volume of precipitation collected using the equation

$$\text{pH}_{\text{VWA}} = -\log\left(\frac{\sum (10^{-\text{pH}_i} \times V_i)}{\sum V_i}\right) \quad (1)$$

where  $\text{pH}_{\text{VWA}}$  represents the volume weighted average pH, and  $\text{pH}_i$  and  $V_i$  are the pH value and sample volume of the sample  $i$ , respectively.

### 2.4 EOFs and the SVD method

Empirical Orthogonal Functions (EOFs) are a set of independent and orthonormal eigenvectors that most efficiently represent a given set of data. The objective of the method is to split the temporal variance of spatially distributed data into orthogonal spatial patterns called EOFs or empirical orthogonal models. The relative importance of any individual eigenvector to the total variance in the field is measured by its associated eigenvalue or variance contribution. Each eigenvector, which may represent the spatial patterns of its variance, is associated with a series of time coefficients that describe the time evolution of that particular eigenvector (Gianelli et al., 2007; Calbet and Schluskel, 2006). In Sect. 3.1.1, the changes of summer precipitation pH in time and space are performed by this method.

Singular Value Decomposition (SVD) aims to find the linear combination or coupling mode of two sets of data. The objective is to separate the highly correlated areas of these two set of data (Jordi et al., 2009; Alter et al., 2000). The SVD was firstly used in diagnosing the relationship between monthly surface temperature in America and sea level pressure in north Pacific (Prokasha, 1976). After that, a lot of studies (Wang and Chen, 2004; Ding and Jiang, 1996) have used SVD analysis to investigate the correlations among meteorological parameters, i.e., rainfall,  $u$  and  $v$  wind, SST, and even aerosol (Shi et al., 2007). It is one of the best tools to study the correlations of two factors with large number of spatial and temporal data. Using this method, we can obtain a series of left and right singular vectors, standing for two factors respectively. The SVD defines the relations of left (right) variable or left (right) factor and time coefficients of right (left) singular vector (of the other factor) as the left (right) heterology correlation coefficients field. This one pair of heterology correlation coefficients field represent one kind of distribution of correlations between two factors, the area where correlated significantly is the key area of interaction between two factors. Thereafter, a pair of heterology correlation coefficients field stands for one type of relationship of two factors to some extent. We used the SVD method to investigate the relationship between rainfall in the middle and lower reaches of the Yangtze River (MLYR) and  $\text{pH}_{\text{VWA}}$  in Central China.

## 3 Result and discussion

### 3.1 Description of the dataset

#### 3.1.1 Spatial patterns of VWA pH

We divide mainland China into eight regions, shown in Fig. 2: the middle reaches of the Yangtze River (YRM, 109–117° E, 26–32° N); lower reaches of Yangtze River (YRL, 117–123° E, 26–33° N); Southern China (SC, 109–123° E, 20–26° N); Southwest China (SW, 95–109° E, 20–33° N);

**Table 1.** Statistics of model results and observations. (*N*, std, and *R* represent the number of months, standard deviation, and correlation coefficient, respectively).

Guanyinqiao		<i>N</i>	Mean ( $\mu\text{mol L}^{-1}$ )	std	Error	<i>R</i>
SO <sub>4</sub> <sup>2-</sup>	Model	48	161.5	52.55	-16.07%	0.60 (sig.95%)
	Obs	37	192.4	69.03		
NO <sub>3</sub> <sup>-</sup>	Model	48	48.6	46.71	29.26%	0.84 (sig.95%)
	Obs	48	68.7	49.82		
Jinyunshan						
SO <sub>4</sub> <sup>2-</sup>	Model	48	141.5	47.92	3.98%	0.43 (sig.95%)
	Obs	44	136.1	73.54		
NO <sub>3</sub> <sup>-</sup>	Model	48	43.3	44.39	-23.17%	0.78 (sig.95%)
	Obs	47	56.4	37.95		
Hongwen						
SO <sub>4</sub> <sup>2-</sup>	Model	48	78.5	34.99	10.91%	0.65 (sig.95%)
	Obs	43	69.9	63.16		
NO <sub>3</sub> <sup>-</sup>	Model	48	33.0	29.81	-11.80%	0.50 (sig.95%)
	Obs	44	37.4	32.81		
Xiaoping						
SO <sub>4</sub> <sup>2-</sup>	Model	48	38.2	17.63	69.13%	0.47 (sig.95%)
	Obs	35	22.6	14.66		
NO <sub>3</sub> <sup>-</sup>	Model	48	22.7	23.12	2.76%	0.60 (sig.95%)
	Obs	36	22.1	16.39		
Linan						
SO <sub>4</sub> <sup>2-</sup>	Model	144	91.2	42.34	20.28%	0.62 (sig.99%)
	Obs	134	75.8	38.01		
NO <sub>3</sub> <sup>-</sup>	Model	144	31.5	23.88	11.92%	0.50 (sig.99%)
	Obs	135	28.1	15.44		
Longfenshan						
SO <sub>4</sub> <sup>2-</sup>	Model	144	41.2	42.34	-24.91%	0.47 (sig.99%)
	Obs	86	54.8	29.34		
NO <sub>3</sub> <sup>-</sup>	Model	144	33.2	23.79	22.25%	0.53 (sig.99%)
	Obs	91	27.2	22.85		

Center of China (CT, 109–117° E, 32–42° N); East China (EC, 117–123° E, 33–40° N); Northeast China (NE, 117–135° E, 40–55° N); and Northwest China (NW, 72–109° E, 33–49° N). Statistics are collected for the whole country and for these different regions. Most of stations are distributed nearly equidistantly in Central and East China, where acid rain occurs at high frequencies. Although stations in Northwest China are sporadic and unevenly distributed, our results may not be significantly distorted since acid rain occurs only very scarcely in that region. The results for pH<sub>VWA</sub>, together with the arithmetic mean pH, rainfall, and the number of stations, are listed in Table 2. As can be seen in the table, the lowest pH value is in SW and YRM, while the highest is in NW and NE for both pH averaging methods. In this study, we focus on YRM and CT, and call them Central China.

In the following analysis, we use pH<sub>VWA</sub> as it eliminates the influence of different rainfall intensities. Fig. 3a shows the spatial patterns of pH<sub>VWA</sub> in China during the summers of 1992–2006 (using Cress-Man interpolation method for mapping the spatial patterns of pH<sub>VWA</sub>, and the same below). It is clear that acid rain occurred mainly in the south of China in summer during this period, with the most severe acid rain in Sichuan and Guizhou provinces, in the southwestern part of China. The other severe acid rain areas are the Yangtze River Delta, northwestern Guangxi and the Pearl River Delta, which are located in southeastern and southern China. This distribution is similar to that found by Xie et al. (2009).

The changes in pH<sub>VWA</sub> during 1992–2006 are shown in Fig. 3b. In southwestern China, the acidity of precipitation decreases significantly during 1992–2006, with pH

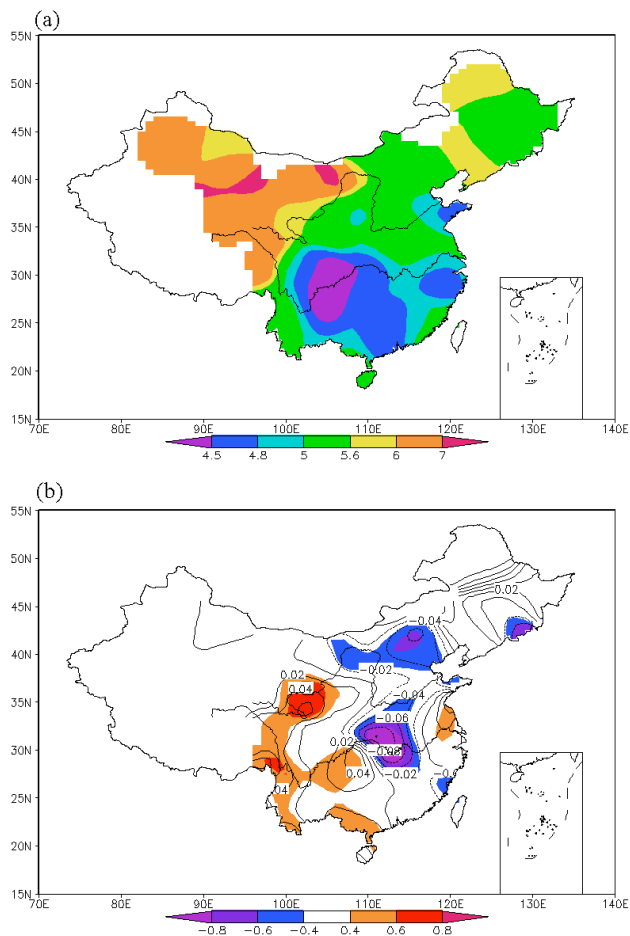
**Table 2.**  $\text{pH}_{\text{VWA}}$  and mean pH in summer during 1992–2006.

Defined region	YRM	YRL	SW	SC	NW	NE	CT	EC
$\text{pH}_{\text{VWA}}$ *	4.75	4.85	4.73	4.81	5.43	5.49	5.13	5.05
mean-pH**	4.79	4.89	4.75	4.86	5.74	5.55	5.34	5.41
Rainfall(mm)***	422.4	522.5	487.1	545.7	122.1	370.4	340.8	423.1
Number of stations	6	8	14	11	10	9	13	3

\*VWA average pH

\*\*direct average pH

\*\*\*total rainfall in summer per year per station



**Fig. 3.** Spatial patterns of the  $\text{pH}_{\text{VWA}}$  (a) and the changes in  $\text{pH}_{\text{VWA}}$  in China during the summer time of 1992–2006 (b). The shadowed color areas in (b) indicate the areas with changes significant at 90% level ( $\alpha = 0.10$ ) and the contour lines and colors represent the changes of pH per year and correlation coefficient.

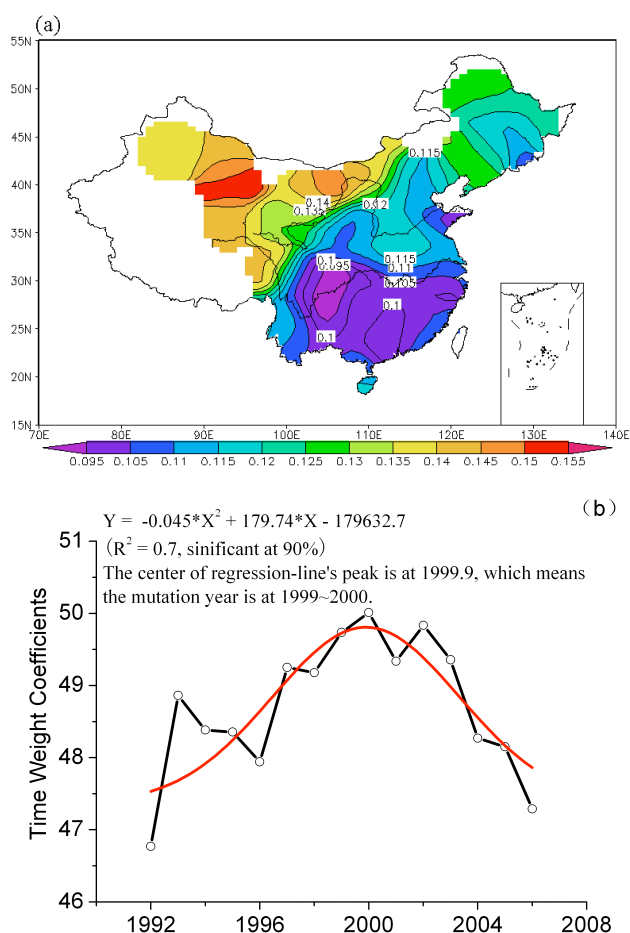
increasing by 0.3–0.6 (at  $\alpha = 0.10$ ). However, a substantial increase in the acidity of precipitation in summer was observed in Central China, as shown in Fig. 3b and in (Tang et al., 2010), with the pH value decreasing by 0.6–1.2 (at  $\alpha = 0.10$ ).

Using the EOF method, the summer pH dataset from the CMA-ARMN stations is separated into a set of spatial functions (or EOF eigenvectors) and a series of time weighting coefficients. Figure 4a and b shows the spatial distribution of the EOF eigenvectors and the variation of the time weighting coefficients, respectively. The variance contributions from the first EOFs eigenvectors of the original pH dataset is greater than 99%, and its spatial patterns are similar to those of  $\text{pH}_{\text{VWA}}$  (Shi et al., 2007; Gu et al., 2007) (see Fig. 3a). The variation in the time weighting coefficients in Fig. 4b symbolizes the overall variation in the acidity of summer precipitation in China. The variation in the time weighting coefficients follows a parabola, and can be divided into two different periods, i.e., 1992–1999 and 2000–2006., with pH increasing between 1992 and 1999 and decreasing between 2000 and 2006. This general trend based on summertime data is consistent with that obtained from the whole dataset by Tang et al. (2010).

To further investigate regional differences in the trends of acid rain in China, we calculated  $\text{pH}_{\text{VWA}}$  for the periods 1992–1999 and 2000–2006. The spatial distributions of the calculated  $\text{pH}_{\text{VWA}}$  values for the two periods are shown in Fig. 5a and b, and the differences between the distributions for both periods are shown in Fig. 5c. In southeastern China, the area with  $\text{pH}_{\text{VWA}}$  lower than 4.8 is significantly larger during 2000–2006 than during 1992–1999, indicating a worsening trend of acid rain in this area. As shown in Fig. 5c, the trend of precipitation acidity is very different from region to region. The contrasting trends become clearer on drawing a southeast–northwest dividing line from  $22^\circ \text{N}$ ,  $112^\circ \text{E}$  to  $42^\circ \text{N}$ ,  $105^\circ \text{E}$ . Northeast of the line, the acidity of precipitation increased (pH decreased) from 1992–1999 to 2000–2006, while southwest of the line, the opposite trend occurred. The most distinct acidification trends are found in Central China. In the SW area, the precipitation acidity decreased significantly (significant at a 90% level) from 1992–1999 to 2000–2006.

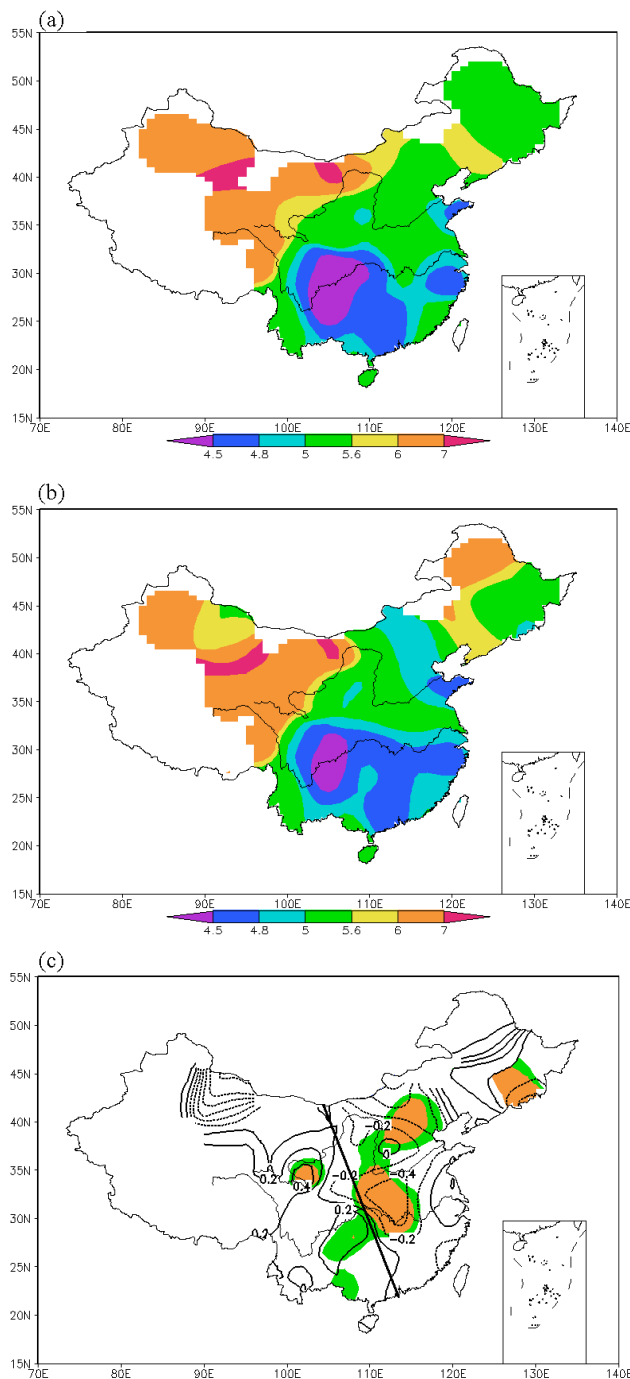
### 3.1.2 Acidity of precipitation in different areas

Figure 6 shows the time series of monthly  $\text{pH}_{\text{VWA}}$  in summer during 1992–2006 for different regions. In SW, the  $\text{pH}_{\text{VWA}}$



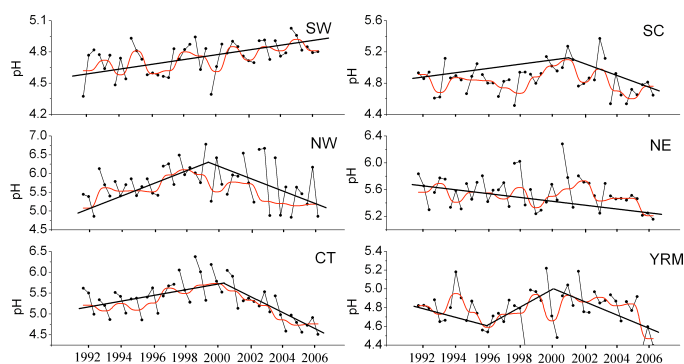
**Fig. 4.** Spatial distributions of the EOF eigenvectors of pH original dataset (a) and variation of time weighting coefficient (b). The black line with circles represents the actual variation of the time weight coefficients and the red line shows the regression line with  $R^2 = 0.70$  (significant at 90% level).

of precipitation increased gradually during 1992–2006, consistent with Tang et al. (2010) and Xie et al. (2009); in SC, NW and CT,  $\text{pH}_{\text{VWA}}$  first showed an increase and then a decrease, similar to the general trend over the whole country; in NE,  $\text{pH}_{\text{VWA}}$  decreased consistently. In CT,  $\text{pH}_{\text{VWA}}$  showed a regular pattern, with the lowest  $\text{pH}_{\text{VWA}}$  almost always occurring in August. In summer, the mean rain pH values decreased from June to August over CT. The substantial increase in the acidity of precipitation in this area might be explained by the following mechanism: developed cities inland emit heavy air pollution, in particular  $\text{SO}_2$  and  $\text{NO}_x$ , and this pollution is transported to Central China under summertime winds, leading to accumulation and to an increase in the acidity of precipitation, especially in years with a strong summer monsoon. Mean while, the strong summer monsoon can also decrease the volume of rainfall in MLYR which may further decrease the wet deposition of acidic pollutants in local and increase the transporting to Central China and then acidity of



**Fig. 5.** Spatial distributions of the  $\text{pH}_{\text{VWA}}$  values for 1992–1999 (a) and for 2000–2006 (b), and the differences between them (c). The green and yellow shadowed areas in (c) indicate areas in which the differences are significant at 90% and 95% level respectively, the real line is southeast-northwest line from 22° N, 112° E to 42° N, 105° E.

precipitation. The impacts of the East Asian Summer Monsoon and precipitation in MLYR on the variation in acidity of precipitation in Central China are addressed in the following sections.



**Fig. 6.** Three point dash-dotted time series of  $\text{pH}_{\text{VWA}}$  in the selected regions in summer during 1992–2006.

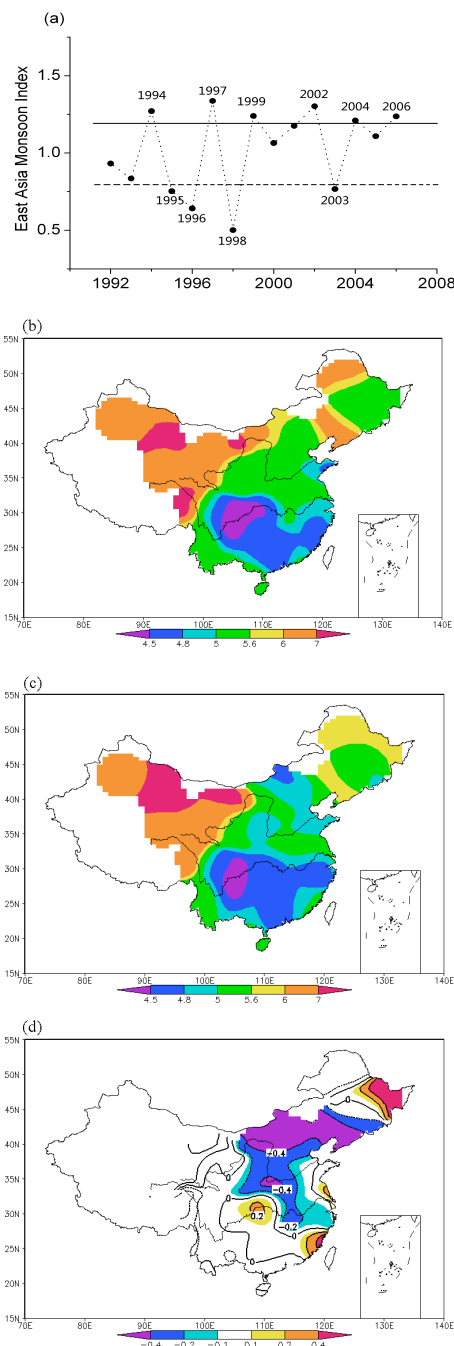
### 3.2 Effects of summer monsoon and rainfall in YRD

#### 3.2.1 Relationship between summer monsoon and pH

The monsoon is an important component of the global circulation system. In monsoon regions, the local weather and climate, and especially precipitation, are strongly influenced by the monsoon circulation. In order to study the potential effects of the summer monsoon on acid rain in China, the East Asian Monsoon Index (EAMI) (Li and Zeng, 2002), which characterizes the summertime climate in China (Li and Zeng, 2005), has been adopted by National Oceanic and Atmospheric Administration (NOAA), and is used to denote the intensity of summer monsoon.

Figure 7a shows the time series of EAMI (<http://www.lasg.ac.cn/staff/ljp/data-monsoon/EASMI.htm>). Based on this figure, we define years above the solid line (20 percent higher than the normal value “1”) as strong summer monsoon years (high MI), and those below the dashed line (20 percent lower than the normal value “1”) as weak summer monsoon years (low MI). Figure 7b and c shows average spatial patterns of summer  $\text{pH}_{\text{VWA}}$  in low and high MI years, respectively. The difference in summer  $\text{pH}_{\text{VWA}}$  between low and high MI years is shown in Fig. 7d. In MLYR, the  $\text{pH}_{\text{VWA}}$  values in high MI years are about 0.1~0.2 lower than in low MI years. In CT, the difference in  $\text{pH}_{\text{VWA}}$  values is 0.2~0.4, and in some areas it even exceeds 0.4. These results indicate that the precipitation in YRD and Central China is more acidic in strong summer monsoon years than in weak years. It is known generally that in the strong monsoon years the climate in the Yangtze River Delta (YRD) is characterized by strong winds and little precipitation (Li and Zeng, 2005). Model simulations with different scenarios in the following sections are used to estimate how much these climate features contribute to the worsening of acid rain in Central China.

China relies heavily on coal combustion for energy and recently the volume of motor vehicles has increased sharply, leading to significant increases in emitted  $\text{SO}_2$  and  $\text{NO}_x$  (Hao

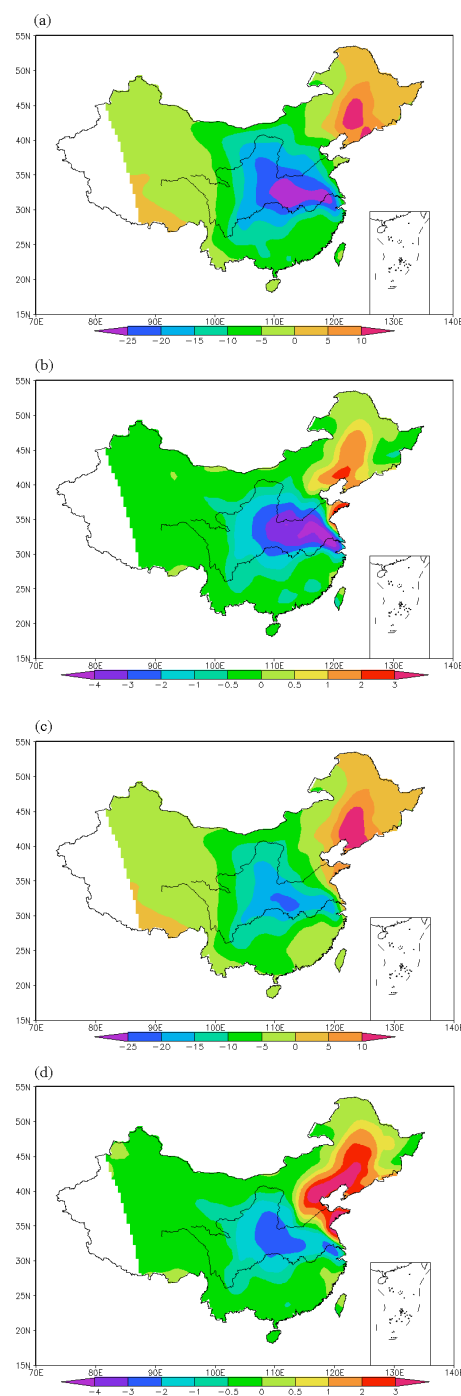


**Fig. 7.** Time series of EAMI (a), average spatial patterns of summer  $\text{pH}_{\text{VWA}}$  in weak (b) and strong (c) summer monsoon years, and the difference between strong and weak monsoon years (d).

et al., 2001; Zhao et al., 2008). These air pollutants are the dominant precursors of acidic species, such as  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  ions, which control almost 90% of the acidity of precipitation in Southern and Central China (Wang and Wang, 1995, 1996; Wang and Xu, 2009; Zhang et al., 1996; Lei et al., 1997). We simulate the distribution of these two key gaseous



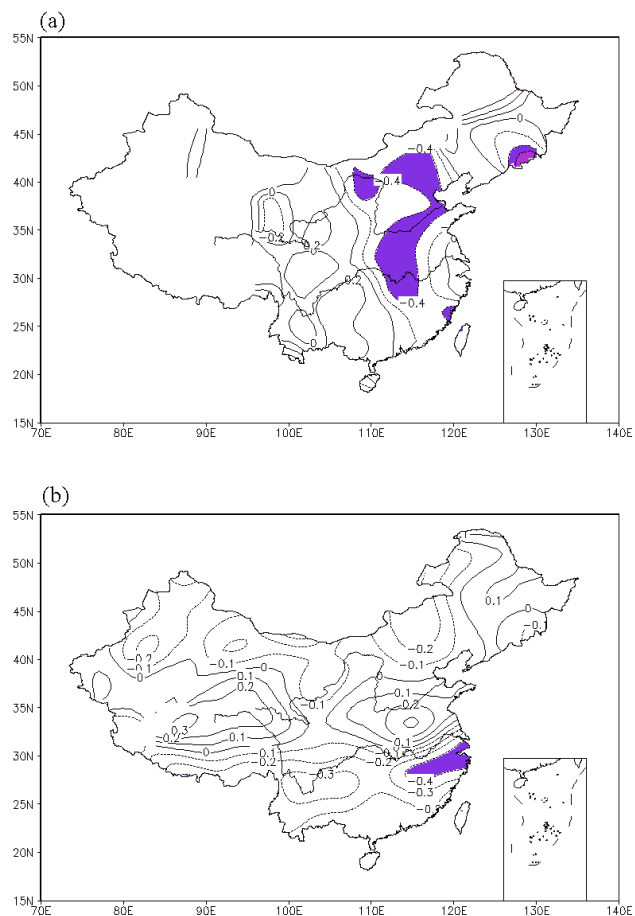
pollutants ( $\text{SO}_2$  and  $\text{NO}_2$ ) and ions ( $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$ ) in low and high MI years for different scenarios (see Sect. 2.2) using the CMAQ model. Note that the neutralization of rain by aerosols is not considered here as alkaline aerosol are not significant in this area (only about 11%–25%) compared to northern China (60%–70%) (Wang and Wang, 1995; Qin and Huang, 2001; Terada et al., 2002). Figure 8 shows the differences in the simulated  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  concentrations in rainwater between low and high MI years for the Normal and MC Scenarios. For the Normal Scenario, the simulated  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  concentrations in Central China are clearly lower in low MI years than in high MI years, as shown in Fig. 8a and b. The differences in the simulated  $\text{SO}_2$  and  $\text{NO}_2$  concentrations (not shown) have similar patterns. These differences may be attributed to differences in meteorological conditions, or pollution emissions, or both, between weak and strong monsoon years. The simulations for MC (varying meteorological conditions and fixed emission inventory) show similar patterns to the differences in the simulated  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  concentrations, but with reduced extent, as shown in Fig. 8c and d. This suggests that meteorological conditions related to the summer monsoon have caused higher concentrations of  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  in rainwater (and hence higher precipitation acidity) in Central China in high MI years. The differences in the  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  concentrations simulated for MC can be attributed solely to changes in the intensity of the summer monsoon, while those simulated for Normal can be attributed to intensity changes of the summer monsoon and to pollutant emissions. Supposing that the differences in the simulated  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  concentrations result in corresponding changes in the precipitation acidity in Central China, we can estimate the contribution of the summer monsoon to the change of precipitation acidity by comparing the values in Fig. 8c and d with those in Fig. 8a and b. We obtain a summer monsoon contribution of almost 65% (regional average of  $-12.0 \mu\text{mol L}^{-1}$  in Fig. 8c and  $-18.3 \mu\text{mol L}^{-1}$  in Fig. 8a;  $-1.2 \mu\text{mol L}^{-1}$  in Fig. 8d and  $-2.0 \mu\text{mol L}^{-1}$  in Fig. 8b) to the change of precipitation acidity in Central China, specifically for stations in Central China the contributions are 16.23%–95.35% and are increased by closer to summer monsoon zone. Since the overall difference in  $\text{pH}_{\text{VWA}}$  between low and high MI years is 0.33 (0.13–0.65 for stations in Central China), we deduce a regional average summer monsoon contribution of 0.22 (0.12–0.42 for stations in Central China) to the  $\text{pH}_{\text{VWA}}$  difference, about 18–36% of the observed  $\text{pH}_{\text{VWA}}$  change (0.6–1.2, see Fig. 1b) in Central China during 1992–2006. It is worthy to know that summer monsoon often affect the transportation of sulfate and nitrate in southeastern coast regions into Central China, while the alkaline substances in China originate mainly from northwest of China which are less influenced by summer monsoon. So, as we focused on the summer monsoon contribution on the precipitation acidity in Central China, the two dominantly anions (sulfate and nitrate) are facility to meet our purpose.



**Fig. 8.** Differences in the  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  concentration ( $\mu\text{mol L}^{-1}$ ) between weak and strong summer monsoon years, simulated for the Normal Scenario (a, b) and the MC (c, d).

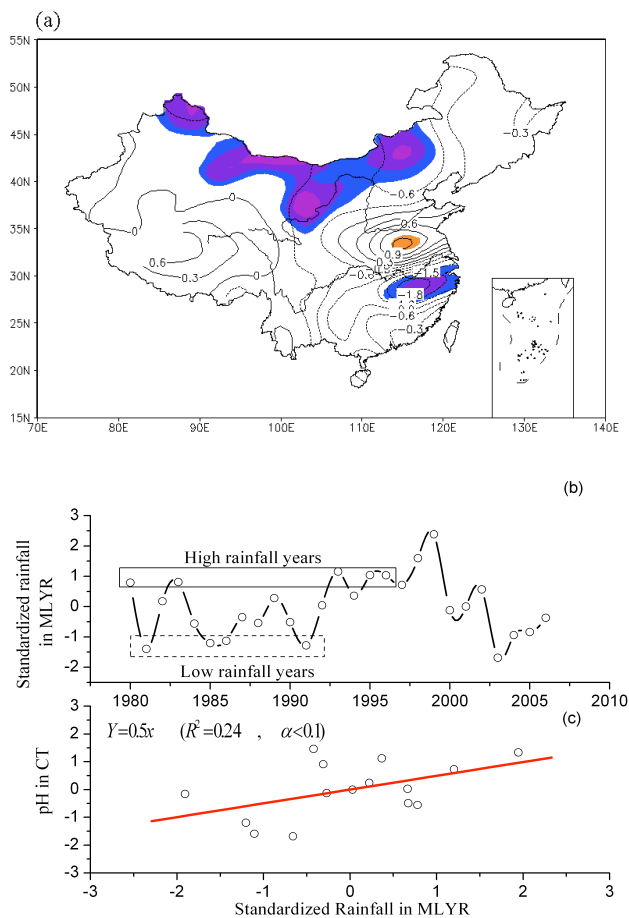
### 3.2.2 Influence of MLYR rainfall on $\text{pH}_{\text{VWA}}$ change in Central China

We used the SVD method (Sect. 2.4) to investigate the relationship between rainfall and  $\text{pH}_{\text{VWA}}$  in China. The first pair



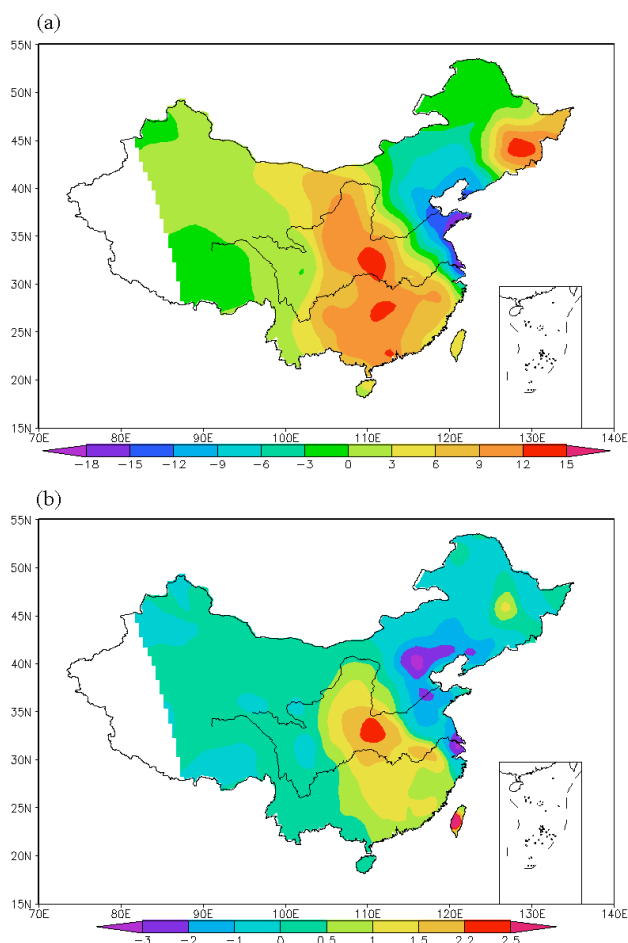
**Fig. 9.** The spatial patterns of left of first SVD eigenvectors (LFSE) (a) and right of first SVD eigenvectors (RFSE) (b) of summer rainfall and  $\text{pH}_{\text{VWA}}$  in China.

of heterology correlation coefficients field is shown in Fig. 9 and the variance contribution is greater than 30%. Fig. 9 shows that the spatial patterns of left of first SVD eigenvectors (LFSE), which stands for the correlations of  $\text{pH}_{\text{VWA}}$  and time coefficient of rainfall in China, and right of first SVD eigenvectors (RFSE), which stands for the correlations of rainfall and time coefficient of  $\text{pH}_{\text{VWA}}$  in China. Obviously, the  $\text{pH}_{\text{VWA}}$  value in Central China is positively correlated to the rainfall in the MLYR (the correlation coefficients of two fields are both 0.4). This suggests that there is a relationship between the observed decrease of the  $\text{pH}_{\text{VWA}}$  value in Central China during 1992–2006 and the decrease of rainfall in the MLYR. Actually, a decreasing trend of the rainfall was observed in the MLYR, as shown in Fig. 10a. The rainfall decreased significantly in this area with the rate of 1.2–1.8 mm per year during 1992–2006. As we discussed above, the pH value decreased significantly either in Central China. These two evidences prove the relationship mentioned above in detail.



**Fig. 10.** (a) Changes in rainfall ( $\text{mm year}^{-1}$ ) during 1992–2006 with the shadowed areas indicating values significant at 90% level; (b) time series of the standardized rainfall in the MLYR during 1980–2006; (c) the correlation between rainfall in MLYR and  $\text{pH}_{\text{VWA}}$  in Central China during 1992–2006.

To further support our hypothesis of the relationship, we calculated the total rainfall in the MLYR during 1980–2006 and standardized the time series by taking the difference between the total rainfall each year and the mean value over 1980–2006 and dividing by the standard deviation over this period. Figure 10b shows that the standardized rainfall varied within the range  $-1.5$  to  $1.5$  before 1998, underwent a sudden increase in 1998–1999, and then dropped to normal levels. For years up to 1997, we separately identify low and high rainfall years in Fig. 10b. We simulated the concentrations of  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  in rainwater for these low and high rainfall years using MC. Figure 11 shows the differences in the simulated rainwater  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  concentrations between the low and high rainfall years ( $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  concentrations in low rainfall years minus that in high rainfall years). It shows that both  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  concentrations have the greatest differences in Central China. Similar results are obtained by comparing the  $\text{SO}_2$  and  $\text{NO}_2$  concentrations simulated

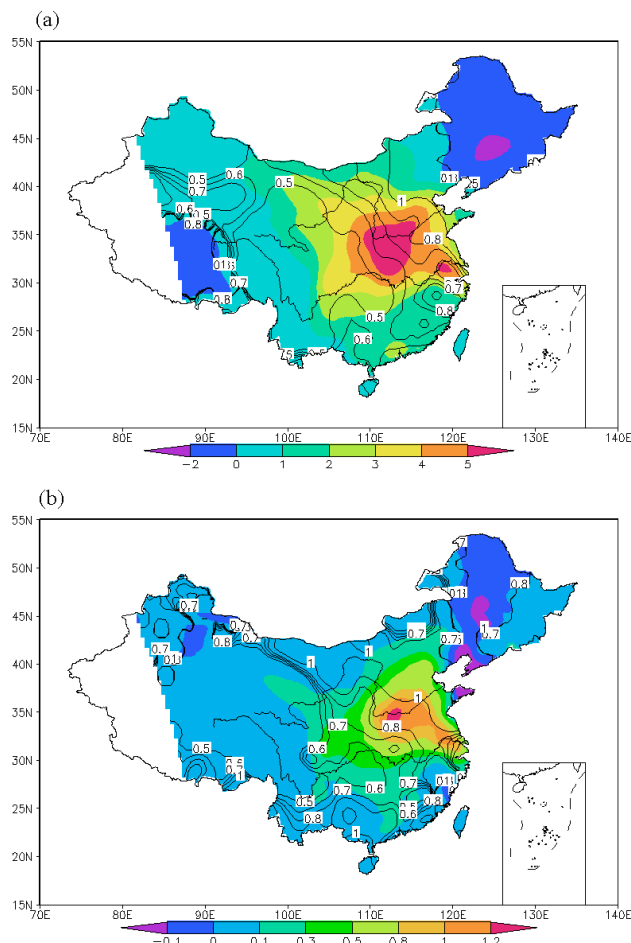


**Fig. 11.** Differences in the simulated rainwater  $\text{SO}_4^{2-}$  (a) and  $\text{NO}_3^-$  (b) concentrations ( $\mu\text{mol L}^{-1}$ ) between the low and high rainfall years.

for low and high rainfall years. Moreover, we find that the  $\text{pH}_{\text{VWA}}$  in Central China is positively correlated with rainfall in the MLYR with  $R^2 = 0.24$  (significant at a 90% level) and explains 24% of the variance in pH in Central China, as shown in Fig. 10c. Based on these results we conclude that less rainfall in the MLYR may enhance the concentrations of acidic gases and their products in Central China, and hence increase the acidity of precipitation in this region. Statistically, about 24% of the variance in pH in Central China can be explained by this fluctuation in rainfall in the MLYR.

### 3.3 Characteristics of modeled $\text{SO}_4^{2-}$ and $\text{NO}_3^-$ ion concentration during 1992–2003

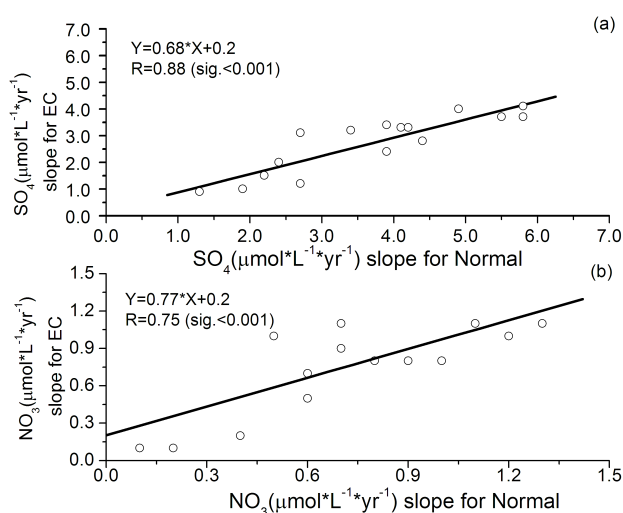
In Sect. 3.2, the contributions of changes in the summer monsoon and rainfall in the MLYR to the increase in precipitation acidity in Central China are studied using fixed pollutant emission intensities. However, the emissions of acidic gases ( $\text{SO}_2$  and  $\text{NO}_x$ ) are not stable, and hence the



**Fig. 12.** Annual changes in rainwater  $\text{SO}_4^{2-}$  (a) and  $\text{NO}_3^-$  (b) concentrations ( $\mu\text{mol L}^{-1} \text{y}^{-1}$ ) during 1992–2003.

impacts of changes in the emission intensities on precipitation acidity should also be addressed. For this purpose, we simulated rainwater  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  concentrations during 1992–2003 using the EC scenario (i.e., changing the emission inventory and using fixed meteorological conditions for 2000). Figure 12 shows the simulated annual changes in rainwater  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  concentrations during 1992–2003. The  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  concentrations in Central China show distinct increases at a rate of  $3.1\text{--}5.1 \mu\text{mol L}^{-1} \text{y}^{-1}$  and  $0.6\text{--}1.3 \mu\text{mol L}^{-1} \text{y}^{-1}$  (significant at a 95% level) for  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$ , respectively.

To estimate the relative contribution of pollutants emissions to the changes in  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  concentrations, we divided the rates of change of  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  concentrations simulated for EC by those simulated for the Normal scenario. The results are shown as contours in Fig. 10 and suggest that about 60%–80% of the changes in  $\text{SO}_4^{2-}$  concentration and 70%–80% of the changes in  $\text{NO}_3^-$  concentration in Central China are attributable to the changes in emissions.



**Fig. 13.** Scatter plot between EC and Normal scene in 19 CMA-ARMN stations located in Central China of the slope of annual changes in rainwater  $\text{SO}_4^{2-}$  (a) and  $\text{NO}_3^-$  (b) concentrations ( $\mu\text{mol L}^{-1} \text{y}^{-1}$ ) during 1992–2003.

In addition, we extract the 19 CMA-ARMN stations located in Central China to further support our results. Figure 13 shows the scatter plot between EC and Normal scene for sulfate and nitrate, 16 and 14 stations left for sulfate and nitrate respectively after removing any one that the correlation is not significant at the level of 99%. We can conclude that the sulfate and nitrate are changed by 68% and 77% respectively which are within our scope. This confirms that most of the changes in  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  concentrations in Central China are caused by changes in emissions, at least during 1992–2003. Assuming that these changes in  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  ion concentrations contributed to changes in the pH of precipitation, we can conclude that changes in pollutant emissions contributed at least 60% of the variance of precipitation acidity in Central China during 1992–2003.

#### 4 Conclusions

Summertime precipitation acidity data from CMA-ARMN stations between 1992 and 2006 have been analyzed using the EOFs method. We find that summertime pH in China experienced two different periods of variation, with pH increasing during 1992–1999 and decreasing during 2000–2006, which is similar to results obtained using the whole dataset (Tang et al., 2010). The most significant decrease of pH is found in Central China. To investigate the causes of the observed decrease of summertime pH in this region, the East Asian summer monsoon index and rainfall data are analyzed, together with the pH data, and simulations using CMAQ are performed for different scenarios.

We find that the East Asian summer monsoon can significantly affect the acidity of summer precipitation in Central China. For example, there is a difference of 0.33 in pH in Central China between years with weak and strong summer monsoons, and about 65% of this difference (i.e., 0.22) is related to the summer monsoon when using simulations with fixed emissions, accounting for 18–36% of the observed pH variation (0.6–1.2) in Central China during 1992–2006. Our SVD analysis and simulations reveal that there is a relationship between the pH in Central China and the rainfall in the MLYR. The pH value of precipitation in Central China is positively correlated with the rainfall in the MLYR, with  $R^2 = 0.24$  (significant at a 90% level), suggesting that 24% of the variance in pH in Central China can be explained by the fluctuation in rainfall in the MLYR. However, the East Asian summer monsoon influences the rainfall in the Yangtze River regions, so the contribution of rainfall to the precipitation acidity in Central China may partially overlap with that of the summer monsoon.

The impacts of pollutant emissions on precipitation acidity in Central China are investigated using the CMAQ model with an annually varying emission inventory, and show that at least 60% of the variance of precipitation acidity in Central China can be attributed to changes in pollutant emissions. Therefore, increasing emissions of acidic species are the dominant factor contributing to the observed significant decrease of pH in Central China, and changes in meteorological factors, such as rainfall and other parameters related to the East Asian summer monsoon, play a less important but still significant role. Although this study estimates the impacts of the East Asian summer monsoon, rainfall in MLYR and pollutants emissions on the variation in pH in Central China, further studies are needed to confirm these effects and to investigate the type of acid rain in this area.

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