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The contributions of socioeconomic and natural factors to the acid deposition over China

Lulu Cui\textsuperscript{a,1}, Jianhong Liang\textsuperscript{d,1}, Hongbo Fu\textsuperscript{a,b,c,*}, Liwu Zhang\textsuperscript{a,**}

\textsuperscript{a} Shanghai Key Laboratory of Atmospheric Particle Pollution and Prevention, Department of Environmental Science & Engineering, Institute of Atmospheric Sciences, Fudan University, Shanghai, 200433, P.R. China

\textsuperscript{b} Shanghai Institute of Pollution Control and Ecological Security, Shanghai 200092, P.R. China

\textsuperscript{c} Collaborative Innovation Center of Atmospheric Environment and Equipment Technology (CICAET), Nanjing University of Information Science and Technology, Nanjing 210044, P.R. China

\textsuperscript{d} Institute of Karst Geology, Chinese Academy of Geological Sciences, Guilin 541004, China

Abstract

China has experienced severe acid rain pollution during the past decades due to excessive sulfur oxides (SO\textsubscript{2}) and nitrous oxides (NO\textsubscript{x}) emissions, which further caused lake acidification, biodiversity losses and climate change. Although the major sources of acid deposition have been clarified previously, the contributions of socioeconomic (natural) factors to the regional acid deposition remained unknown. Therefore, a series of valuable data including socioeconomic (natural) variables and measured pH value in the rainwater at the city level were collected to identify the key factors influencing the rainwater pH value at the national and the regional scale using the spatial econometric model/geographical detector technique and geographical weight regression (GWR) model, respectively. The results showed that the annual mean pH value in the
rainwater in China was 6.54 ± 0.72. The rainwater pH in winter (6.01 ± 0.41) was significantly
lower than those observed during summer (6.74 ± 0.64), spring (6.71 ± 0.71) and autumn (6.71 ±
0.69). The spatial econometric model indicated that socioeconomic indicators including per capita
gross industrial production (GIP), ratio of built-up area to the urban land (RBU), foreign direct
investment (FDI), SO₂ emission, and meteorological factors of annual mean precipitation (AMP),
and annual mean relative humidity (AMRH) were the main factors for the acid deposition. The
geographical detector technique implied that the power of determinants were in the order of
AMRH (10.00%) = AMP (10.00%) > SO₂ emission (8.51%) > FDI (8.32%) > RBU (7.64%) > per
capita GIP (7.00%). The GWR implied that GIP, FDI, and SO₂ emission made relatively higher
contribution to acid deposition in East China relative to other regions owning to the huge
population and the higher energy consumption. The higher rainfall amount and RH in Southeast
China significantly increased the pollutant deposition fluxes and promoted the heterogeneous
transformations of precursors of acid rain, respectively. The findings herein shed light upon the
socioeconomic forces for the acid deposition in China for the first time and provided the new
information for government sectors to control the acid rain pollution in the future.

Keywords: Acid deposition; socioeconomic factors; natural factors; GWR model; China
* Corresponding author: fuhb@fudan.edu.cn (Hongbo Fu); zhanglw@fudan.edu.cn (Liwu Zhang)

1 These authors contributed equally to this work
1. Introduction

During the past decades, China has suffered from severe acid rain pollution due to the large emissions of its precursors including sulfur oxides (SO\textsubscript{2}) and nitrous oxides (NO\textsubscript{x}) along with the rapid development of industry and agriculture (He et al., 2006; Jia et al., 2014; Kuribayashi et al., 2012; Yu et al., 2016, 2017). Acid precipitation deriving from heterogeneous chemical processes of SO\textsubscript{2} and NO\textsubscript{x} have exerted adverse effects on the terrestrial and aquatic ecosystem, including the severe soil (lake) acidification (Richter et al., 2005), the inhibition of plant growth (Stevens et al., 2004), and the biodiversity loss (Sala et al., 2000). Although a series of policies prohibiting SO\textsubscript{2} emission have been implemented since 2000, the acid rain pollution is still severe in many parts of China (e.g., Sichuan Basin and Yangtze River Delta) (Kuang et al., 2016; Liu et al., 2016b; Qiao et al., 2018). In this context, investigating regional characteristics and potential drivers of the acid deposition is highly needed so as to provide scientific basis for formulating targeted mitigation policies in future.

Previous studies focusing on source identification of acid rain have found close relationships between acid precipitation and socioeconomic factors. For instance, Zhan et al. (2015) demonstrated that N fertilizer use and energy consumption played significant roles on the acid deposition. Yu et al. (2016) revealed that pH, SO\textsubscript{4}\textsuperscript{2-} and NO\textsubscript{3}- in precipitation in Chinese agricultural and natural ecosystems was closely associated with the level of economic development. Qiao et al. (2018) pointed that the acidic substances in the precipitation of Jiuzhaigou were mainly derived from the emission of local tour buses. Apart from anthropogenic activities, meteorological factors were also found to play an important role on the acid deposition by influencing the transport and dispersion of the precursors (Song et al., 2017; Xing et al., 2017).
Despite these findings, the joint effects of socioeconomic factors and meteorological conditions on regional acid deposition were not assessed yet. Besides, most of studies revealed the relationship between socioeconomic or meteorological factors and acid deposition using only simple regression models, which have the limitation of ignoring the spatial autocorrelation between acid deposition and explanatory variables and thus cause the assessment uncertainty. Spatiotemporal models are needed to clarify the relationship between acid precipitation and socioeconomic/natural indicators more accurately in future.

Nowadays, a wide range of models including spatial econometric model (Wang et al., 2018), land use regression model (Liu et al., 2016a), and geographically weighted regression model (Chen et al., 2018) etc. have been used to identify the socioeconomic driving factors of air pollution. Although these models showed good performance in analyzing the correlation between dependent and independent variables, they are based on the linear hypothesis which might not invariably in agreement with the practical situation. In fact, multicollinearity unavoidably exists between predicting factors, which causes bias in linear regression models. The geographical detector method (GDM) has been proven to be a robust technique for diagnosing collinearity of independent variables and thus enhancing the reliability of independent variable identification (Zhou et al., 2018). Given this, the GDM technique was used in this study so as to avoid duplication of information conveyed in the seemingly independent variables, and to explore the spatial correlations between acid precipitation and the finalized socioeconomic/natural indicators.

As a comprehensive indicator, the rainwater pH value can mirror the acid deposition status because it is extremely sensitive to the acidic substances in the precipitation (Charlson and Rodhe,
In this study, we measured the pH in the rainwater of 407 monitoring sites in 2010, and then predicted the rainwater pH values in 336 prefecture-level cities using spatiotemporal Kriging interpolation (STK) method. Based on these estimated data, we analyzed the spatiotemporal variations of the rainwater pH values to reflect the acid deposition status over China. Further, the underlying effects of the socioeconomic and natural factors on the rainwater pH values at a national scale were investigated using the spatial regression model in combination with the GDM. For validation, the local relationship between predictor variables and acid deposition was also assessed using the GWR model. The main objective of our study is to investigate the spatiotemporal variations of the rainwater pH across the whole China, and assess the comprehensive effects of socioeconomic and natural factors on acid precipitation at the city level in China. Results in this study could provide scientific basis for the government sector to formulate regional and national policy measures to reduce the acid deposition.

2. Material and methods

2.1 Data sources and preparation

2.1.1 The pH value of rainwater

In the present study, we collected the rainwater samples at 407 monitoring sites over China in 2010 using plastic buckets (diameter 30 cm) installed at 1.5 m above ground level only during rainfall events. After the sampling, the sample pH values were measured immediately using a pH meter (MP-6p, HACH, USA) at 20-25°C. Given the uneven distribution of the monitoring sites, we estimated the rainwater pH values with a higher spatial resolution of 0.25° × 0.25° over the whole China using the STK models. The STK models could be classified into two types including...
the separation models and the non-separation models. The separation STK models comprised of
Bilonick model (BM), Dagum model (DM), and Ma model (MM), and the non-separation models
was composed of Gneiting model (GM), Cressie-Huang model 1 (CH1), and Cressie-Huang
model 2 (CH2). The optimal STK model was selected based on the least fitting error of six models.
It has been demonstrated that this method showed a good predictive performance (Lin et al., 2018;
Xu et al., 2018a). The pH value of rainwater in each city was calculated based on the average of
pH values in the corresponding grids.

2.1.2 The socioeconomic and natural factors

The socioeconomic and natural factors were selected based on two criterions. Firstly, the
selected indicators should be related with the rainwater pH value. Through reviewing a large
amount of references (Zhou et al., 2018; Chen et al., 2018), some common factors including per
capita gross domestic production (GDP), per capita gross industrial production (GIP), urban level,
and energy consumption which have been demonstrated to be tightly associated with acid
deposition or air pollution were integrated into the model (Zhou et al., 2018; Chen et al., 2018). In
addition, some potential factors affecting the SO$_2$ and NO$_x$ emissions such as foreign direct
investment (FDI) and road density were also considered into the model. On the other hand, the
data availability should be considered and most of the data could be drawn from the China
Statistical Yearbook. All of the variables in the present study were selected based on previous
work (Fang et al., 2015). The independent variables in the present study consisted of eight
socioeconomic indictors (e.g., GDP, GIP) and five natural factors (annual mean precipitation
(AMP)). The detailed selection basis was summarized as follows:
The socioeconomic factors comprised of per capita GDP (Unit: $10^4$ yuan/person), per capita GIP ($10^4$ yuan/person), the ratio of built-up areas to urban areas (RBU) (%), per capita electricity consumption (kW/person), foreign direct investment (FDI) ($10^4$ dollars), population density (person/km$^2$), road density (km/km$^2$), and SO$_2$ emission (ton). The Environment Kuznets Curve (EKC) suggested that the pollutant levels were closely associated with the economic development (Song et al., 2013). To remove the effect of city size on rainwater pH values, the per capita GDP was used as an important explanatory variable instead of GDP. It was well known that more than 60% of total SO$_2$ emission were released from the industrial activities (Yang et al., 2017), which could play an important role on the acid deposition. Therefore, the SO$_2$ emission and per capita GIP remained the higher priority in examining their effects on the acid deposition. In addition, some studies suggested that the urban expansion increased the emissions of acidic substances such as SO$_2$ and NO$_x$, both of which could decrease the rainwater pH values (Wang et al., 2018). Hence, we employed RBU as a proxy for urban expansion. Zhou et al. (2018) demonstrated that population density and residential energy consumption were related to the air pollutants. Among all of the energy types, only electricity consumption data was available at a city level. Thus, the population density and electricity consumption were incorporated into the final model. Transportation serves as one of the most important emission source of NO$_x$, which could affect the rainwater pH value. We used the road density as an indicator to reflect traffic intensity in the cities.

In recent years, the international trade has been paid more attention because it promoted the economic growth along with the environmental pollution (Guan et al., 2014). Liu et al. (2018) revealed that the inflow of FDI posed distinct effects on the air pollution (Liu et al., 2018). In the present study, we selected the FDI as a proxy for international trade.
Five natural factors including AMP, annual mean temperature (AMT), annual mean wind speed (AMWS), annual mean relative humidity (AMRH), and net primary production (NPP) were selected to assess their effects on acid deposition. AMP exhibited the two-way effects on the acid deposition. First of all, AMP promoted the wash-out effect of acidic pollutants (Vet et al., 2014), which could increase the rainwater pH values. However, the acidic deposition fluxes displayed the significant increase with the precipitation increase, which could aggravate the acid deposition. AMT, AMWS, and AMRH usually influence the acid deposition through changing the atmospheric mixing state (Li et al., 2019). The NPP was often selected as an important index to reflect the dynamic variation of green vegetation. In general, the severe acid deposition caused the NPP decrease (Liu et al., 2011). Finally, eight socioeconomic indicators and five natural variables were incorporated into the models.

2.2 Methods

2.2.1 The ordinary least square (OLS) regression

The OLS regression was generally used to examine the linear relationship between the independent and dependent variables. The partial regression coefficients could be estimated based on the independent and dependent variables, which revealed the importance of each predictor variable. The detailed formulas are as follows:

\[ Y = AX + b + \epsilon \]  
\[ A = (X^T X)^{-1} X^T Y \]

where \( Y \) represents the dependent variable (rainwater pH value); \( X \) denotes the predictor variables (e.g., AMT and per capita GDP); \( A \) is the partial regression coefficient matrix; \( b \) denotes the
intercept; \( \varepsilon \) represents the random error subjected to the Gauss-Markov distribution.

Although the OLS regression assessed the effects of predictor variables on the rainwater pH values, the method neglected the spatial autocorrelation of the rainwater pH values at a national scale. First of all, we used the Moran’s \( I \) statistic to assess the spatial autocorrelation of the rainwater pH values. The Moran’s \( I \) statistic was characterized with the spatial weight matrices. The higher Moran’s \( I \) and the lower \( p \) value (\( p < 0.05 \)) suggested that significantly spatial relationships were observed in the neighboring cities (Anselin and Bera, 1998). The Moran’s \( I \) statistic is calculated as the following equation:

\[
I = \frac{n \sum \sum w_{ij} (y_i - \bar{y})(y_j - \bar{y})}{\sum \sum w_{ij} \sum (y_i - \bar{y})^2}
\]

where \( w_{ij} \) denotes the spatial weight value in the spatial weight matrix \( W \), and \( y \) represents the rainwater pH values in these cities. Generally, the Moran’s \( I \) ranges from -1 to 1. The Moran’s \( I \) between 0 and 1 represents a positive spatial autocorrelation of the rainwater pH values. In contrast, the negative Moran’s \( I \) represents a negative autocorrelation of the rainwater pH values. The Moran’s \( I \) close to 0 means the insignificant relationship between the neighboring cities.

2.2.2 Spatial econometric models

The spatial econometric models were regarded as the improvement of OLS regression through integrating the spatial weight matrix. The spatial econometric models can be classified into the spatial lag model (SLM) and the spatial error model (SEM). Both of these models were available to assess the effects of socioeconomic and natural factors on rainwater pH values in 336 cities.
The optimal model would be determined based on the robust LM lag and robust LM error. The detailed formulas are shown in supporting information (SI).

2.2.3 Geographical detector method (GDM)

As a novel and suitable spatial analysis toolbox, the GDM proposed by (Wang et al., 2010) has been widely used to explore the effects of influential factors on the environmental pollutants. This model did not require the linear hypothesis, and thus it could be applied to examine the nonlinear relationship between the independent and dependent variables. On the basis of the model, the remarkably spatially heterogeneity could be observed when the sum of the variances of sub-regions classified by the factors were less than the variance of the whole area. The \( q \)-statistic was an index to assess the effects of influential factors on the rainwater pH value. The detailed calculation equation is as follows:

\[
q = 1 - \sum_{j=1}^{m} \frac{n_{pH,j}}{n_{pH}} \] \( \text{(4)} \)

where \( q \) is the power of each influential factor \( I_j \); the \( q \) value generally ranges from 0 to 1. The \( q \) value closer to 1 indicates the stronger effect of factor \( I_j \) on the rainwater pH value. The whole area could be classified into three sub-regions (i.e., high-level, middle-level, and low-level) based on the influential factors. The threshold of the sub-regions are summarized in Tab. 1.

2.2.4 The GWR model

The spatial econometric models could not identify the relationship between the rainwater pH value and the explanatory variables (e.g., socioeconomic factors and meteorological condition) at a city level. Therefore, the GWR model was applied to estimate the determination coefficient (\( R^2 \))
and the local regression coefficients for all of the cities. The local regression coefficients were used to assess the contributions of influential factors to the rainwater pH values for all of the cities (Fang et al., 2015). The local regression coefficients are calculated as the following equation (Brunsdon et al., 1996):

$$\beta(u_i, v_i) = (X^T W(u_i, v_i) X)^{-1} X^T W(u_i, v_i) Y$$  \hspace{1cm} (5)$$

where $\beta(u_i, v_i)$ is the local regression coefficient at city $i$; $X$ denotes the influential factors; $Y$ represents the rainwater pH value in all of the cities; and $W(u_i, v_i)$ denotes the spatial weight matrix.

The spatial weight matrix was computed on the basis of the exponential distance decay form:

$$W(u_i, v_i) = \exp(-d^2(u_i, v_i) / b^2)$$  \hspace{1cm} (6)$$

where $d(u_i, v_i)$ represents the Euclidean distance between $i$ and $j$, and $b$ is the kernel bandwidth.

In the present study, various statistical methods have been applied to identify the key factors for the acid deposition in China. In order to facilitate the understanding of these methods, the flow chart of the methodology is depicted in Fig. 1.

3. Results and discussion

3.1 The spatiotemporal variation of the rainwater pH value in China

Figure 2 shows the temporal variations of the rainwater pH values in different climatic zones of China in 2010. The annual averaged pH value in the rainwater of China was 6.54 ± 0.72 at a national scale. The rainwater pH value in China displayed the inverted-U pattern, with the highest one in summer (6.74 ± 0.64), followed by ones in spring (6.71 ± 0.71), autumn (6.71 ± 0.69), and winter (6.01 ± 0.41), respectively. All of the climatic zones showed the lowest rainwater pH values
in winter, but they did not exhibit the highest rainwater pH values in the same season. The seasonal variations of rainwater pH values in Southeast China (5.93 ± 0.43), Northeast China (7.03 ± 0.21), and Inner Mongolia (7.11 ± 0.20) were in good agreement with those in the whole China. However, both of Northwest China (7.28 ± 0.33) and Southwest China (6.69 ± 0.59) presented the highest pH values in autumn (Fig. 2a-b). NCP displayed the highest rainwater pH value in spring (6.76 ± 0.17). The lowest rainwater pH values in winter was mainly attributed to the dense anthropogenic emissions such as the coal combustion for the domestic heating (Yang et al., 2016a). Tian et al. (2013) demonstrated that the NO\textsubscript{x} emission showed a notable peak in the cold season due to the increase of the domestic electricity demand North China. Besides, the stagnant meteorological conditions characterized with shallow mixing layers, scarce precipitation, weak solar radiation, and slow wind speed were frequently observed in winter, leading to the higher loadings of SO\textsubscript{2} and NO\textsubscript{2} in the atmosphere (Zhao et al., 2016). The highest rainwater pH value observed in spring of NCP might be associated with the dust events.

3.2 The spatial distribution of the rainwater pH values over China

The rainwater pH values across the whole China at the resolution of 0.25°× 0.25° and those at the city level are depicted in Fig. 3. Seinfeld (1986) estimated that the rainwater with pH lower than 5.60 was identified as acid rain based on that the pH value of natural water in equilibrium with atmospheric CO\textsubscript{2} was 5.60. In the present study, 57 prefecture-level cities over China were identified as the hot spot of acid deposition, which were mainly concentrated on the southeastern of China (e.g., Chizhou, Huangshan) (Fig. 3a). At the spatial scale, the annual mean rainwater pH exhibited the highest values in Northwest China (6.94 ± 0.36), followed by those in Inner Mongolia (6.82 ± 0.32), Northeast China (6.76 ± 0.28), NCP (6.51 ± 0.27), Southwest China (6.49
\[ \pm 0.61 \), and the lowest ones in Southeast China \((5.75 \pm 0.59)\) (Fig. 3b). The rainwater in
Northwest China and Inner Mongolia showed the higher pH values compared with other regions
because the rainwater acidity could be largely neutralized by some alkaline ions released from the
desert soils (Wang et al., 2016b). Although Northeast China and NCP were also faced of the
intrusion of dust storms, the higher SO\(_2\) and NO\(_x\) emissions might increase the sulfate and nitrate
levels in the atmosphere, thereby decreasing the rainwater pH values (Xu et al., 2015). The most
serious acid deposition was observed in Southeast China owing to the widespread distribution of
the iron-steel industries and cement industries especially in Jiangsu province (Hua et al., 2016;
Wang et al., 2016a). Hua et al. (2016) estimated that the SO\(_2\) emission from cement industries in
Jiangsu province reached 50 Gg/yr, which was significantly higher than that in other provinces.
Moreover, the typically acidic soil in Southeast China could even aggravate the acid deposition
during the soil dust resuspension. In addition, Sichuan Basin was also treated as a hot spot of acid
deposition because the relatively closed terrain coupled with the frequent air stagnation could
exacerbate the acid rain in the region (Zhang et al., 2019; Zhao et al., 2018).

The rainwater pH value displayed the similarly spatial variation in all of the seasons as a
whole, whereas the spatial variability was not identical in four seasons (Fig. 4). In spring, summer,
and autumn (Fig. 4a-c), the rainwater pH values decreased significantly from Southeast China
(spring: 5.73 ± 0.58, summer: 5.93 ± 0.43, and autumn: 5.80 ± 0.33) to Northwest China (7.16 ±
0.31, 7.11 ± 0.22, and 7.28 ± 0.33). The rainwater pH values in nearly all of the cities of North
China were higher than 6.46. However, the notably spatial difference was not observed in winter
(Fig. 4d). Sichuan Basin encountered the severe acid deposition the same as Hunan and Jiangsu
provinces. Additionally, the rainwater pH values in NCP showed the rapid decrease in winter
because of the combustion of fossil fuels and the unfavorable meteorological conditions (Xu et al., 2015).

3.3 The effects of influential factors on the rainwater pH value at a national scale

To test the spatial autocorrelation of the rainwater pH value in China, the Moran’s I test has been performed. The test result suggested that Moran’s I reached 0.56 and the p value was lower than 1% (Fig. 5), implicating the significantly spatial correlation. Additionally, the Local Indicators of Spatial Association (LISA) map also has been drawn to clarify the spatial cluster of the rainwater pH values. The LISA map was characterized with the high-high, low-low, and insignificant cluster in China. However, the high-low and low-high cluster for the rainwater pH values were not observed in China. Northwest China, Inner Mongolia, and Northeast China exhibited the remarkable high-high cluster, suggesting the clear region lack of the acid deposition (Fig. 5). Southeast China showed the marked low-low cluster, suggesting that they can import/export acidic substances from/to other neighboring cities. NCP and Southwest China did not show significant spatial cluster though many cities of Southwest China such as Luzhou (pH = 5.78) and Leshan (5.88) encountered the severe acid rain.

In the present study, OLS, SLM, and SEM methods were applied to assess the effects of predictor variables on the rainwater pH value. The results of OLS and spatial regression analysis are summarized in Tab. 2 and Tab. 3, respectively. Some goodness-of-fit statistics including the $R^2$ value and the p value of OLS reached 0.75 and 0.00, suggesting that the data showed a good fitting performance using OLS method. However, the $R^2$ value of spatial regression model arrived at 0.76, which was slightly higher than that of OLS (Tab. 2). Moreover, other statistical indicators
including log likelihood and Akaike information criterion (AIC) also clearly reinforced the importance that the spatial autocorrelation cannot be ignored in the present study (Tab. 2). Therefore, spatial regression model should be applied to assess the effects of socioeconomic and natural factors on the rainwater pH values. Spatial regression model comprised of spatial error model (SEM) and spatial lag model (SLM). Both of SLM and SEM passed the Lagrange multiplier. However, the robust Lagrange multiplier of SLM ($p = 0.00$) showed the better performance than that of the robust Lagrange multiplier with SEM ($p = 0.04$). Hence, the SLM was selected as the final model to clarify the key factors for the rainwater pH value in China (Tab. 3).

Based on the SLM result, four socioeconomic indicators (e.g., per capita GIP and RBU) and two natural factors (e.g., AMP and AMRH) presented the significant correlation with the rainwater pH value (Tab. 3). Among the socioeconomic factors, the per capita GIP was denoted as the key factor to aggravate the acid deposition. The southeastern coastal cities with high per capita GIP generally showed the lower rainwater pH values. It was well documented that the industrial development was closely linked to the consumption of fossil fuels (McGlade and Ekins, 2015). Hao and Liu (2016) argued that the energy consumed by the secondary industry accounted for more than 70% of the total energy consumption of China, of which was linked to the production and emission of acidic pollutants (e.g., $\text{SO}_2$, and $\text{NO}_x$) (Dincer, 2000; Omer, 2008). Thus, the $\text{SO}_2$ emission also displayed a remarkable correlation with the rainwater pH value. In addition, the RBU was identified as the main deriver for acid deposition. It was well known that the urbanization level showed the remarkable relationship with air pollutants (e.g., $\text{PM}_{2.5}$ and $\text{SO}_2$) (Fang et al., 2015; Zhan et al., 2018). For instance, $\text{SO}_2$ in the atmosphere was inclined to be
transformed into secondary sulfates especially in cities with high AMRH, thereby worsening the acid deposition status (Zheng et al., 2015). It was interesting to note that the FDI played an important role on the rainwater pH value. With the development of economic globalization, the FDI has reached 126.3 billion dollars in 2015, which has become one of the most important factors for environmental pollution (Wang and Chen, 2014). Liu et al. (2018) revealed that the inflow of FDI exerted distinct effects on SO$_2$ pollution because the foreign enterprises were mainly constituted of the energy-intensive industries (Liu et al., 2018). Apart from the effects of socioeconomic factors, the meteorological factors were also responsible for the acid deposition. It was well documented that the precipitation generally decreased the pollutant levels in the atmosphere by wash-out process (Guo et al., 2016). However, the AMP exhibited the negative relationship with the rainwater pH because the rainfall was considered to be a major sink of air pollutants. It was assumed that the increase of the rainfall amount promoted the elevation of pollutant deposition fluxes (Liu et al., 2011). Besides, RH can alter the rainwater pH value by affecting the photochemical pathway of precursors in ambient air. Yang et al. (2016) have reported that high RH favored the heterogeneous transformation from SO$_2$ to sulfate (Yang et al., 2016b; Cheng et al., 2016).

It should be noted that GDP was not closely linked to the rainwater pH value and such scene seemed to be in contrast to the result reported by Wang et al. (2018). It was well documented that GDP showed the positive or negative correlation with the environmental pollutants depending on EKC. The insignificant relevance between GDP and the rainwater pH was caused by the spatial uneven of economic development in China. The developed regions such as Southeast China has crossed the inflection point of the EKC, and realized the efficient production mode with low
energy consumption (Naminse and Zhuang, 2018). In stark contrast to the developed regions, the undeveloped areas in Northwest China still implemented the extensive production mode (Wu et al., 2016), leading to the higher energy intensity and pollutant emission.

Although SLM can clarify the key factors for the rainwater pH value in China, the method cannot evaluate the power of determinants on the rainwater pH value. Therefore, the GDM has been applied to determine the strength of influential factors for the rainwater pH value. First of all, all of the prefecture-level cities have been classified into three sub-regions based on the original value of each factor. The thresholds of the sub-regions for these key factors are summarized in Tab. S1. After the region division, we employed the GDM to quantify the strength (the $q$ value) of each key factor. As shown in Fig. 6, the power of determinants was in the order of AMRH (10.00%) = AMP (10.00%) > $\text{SO}_2$ emission (8.51%) > FDI (8.32%) > RBU (7.64%) > per capita GIP (7.00%), suggesting that the meteorological condition can account predominantly for the spatial variability of the rainwater pH value in China. The effects of socioeconomic factors were still weaker than those of natural conditions for the acid deposition.

3.4 The effects of socioeconomic and natural factors on the rainwater pH value at a regional scale

Both of the SLM and GDM cannot evaluate the local effects of influential factors on the rainwater pH value at a region or city level. Thus, the GWR model was applied to determine the relationship between influential factors and the dependent variable in depth. All of the predictor variables were integrated into the original model, but many independent variables did not show significant relevance with the rainwater pH values. To assure the good performance of GWR, many predictor variables were excluded from the original model. In the final model, the local $R^2$
value and spatial variability of local regression coefficients are depicted in Fig. S1 and Fig. 7, respectively. The local $R^2$ in all of the cities were higher than 0.55, and the p values were lower than 0.05, indicating that GWR model was suitable to be performed.

The local regression coefficient of AMP on the rainwater pH value ranged from -1.21 to 2.01 (Fig. 7a). As shown in Fig. 7a, the highest negative correlation coefficient of AMP and the rainwater pH value was observed in NCP and some cities of Southeast China. It was supposed that high rainfall amount in these regions made the wet deposition as an important sink of acidic pollutants. In Fig. 7b, a negative relationship was identified between AMRH and the rainwater pH value across the whole China. The highest correlation coefficients were observed in Southeast China and the western of Xinjiang and Tibet autonomous region. It was widely shared that the strong pressure gradient (summer monsoon) between Pacific and Mainland China resulted in the deep convection, and thus increased the rainfall amount and moisture content (Yihui and Chan, 2005; Yu et al., 2004), which could accelerate the sulfate and nitrate formation, thereby changing the rainwater pH value. The significantly negative relationship between the rainwater pH value and RH in the western of Xinjiang and Tibet autonomous region because the low RH was not favorable to the photochemical transformation of acidic precursors in these arid regions (Xiao et al., 2015).

In Fig. 7c, the highest negative effects of GIP on the rainwater pH value focused on NCP and the western of Sichuan and Yunnan provinces. It was assumed that the iron and steel industries, power plants, cement industries, and the non-ferrous smelting industries in these regions might be the major source of gaseous pollutants (Qi et al., 2017). In contrast, the higher GIP in Kashi did not contribute to the severe acid deposition due to the strong neutralization capacity of alkaline
ions (Ma et al., 2017). As shown in Fig. 7d, the RBU played negative effect on the rainwater pH in most of the cities across the whole China. It was generally believed that the requirements of transportation and infrastructure could increase with the expansion of the built-upon land, and thus increased the energy consumption (Xu et al., 2018b). Moreover, the urbanization generally promoted the increase of human activity instead of concentrating on an isolated region, which was favorable to the release of gaseous pollutants (Lou et al., 2016). In the western of China, the low urbanization level restricted the energy demand, and thus decrease the hazards of acid deposition. It should be noted that the higher effect of FDI on acid deposition focused on the East China (Fig. 7e) because most of the foreign enterprises were mainly concentrated on the coastal cities of East China (e.g., Shanghai, Tianjin, and Hangzhou) since economic reform in China (Elliott and Zhou, 2015). Moreover, many energy-intensive foreign enterprises imported from developed country resulted in the air pollution. Lan et al. (2012) also verified that FDI significantly aggravated the environmental pollution using empirical analysis. As depicted in Fig. 7f, the effect of SO$_2$ emission on the acid deposition was in coincident with the SO$_2$ emission intensity, which centered on NCP and some cities of Southwest China. It was well recognized that the acidic substances in the precipitation were mainly originated from the anthropogenic SO$_2$ emission, and thus the restriction of SO$_2$ emission might be an efficient pathway to mitigate the acid rain in China.

4. Conclusion and policy implications

The rainwater samples at 407 monitoring sites over China were collected to determine the rainwater pH values. The annual mean pH value in the rainwater of China was 6.54 ± 0.72. The rainwater pH value exhibited the remarkably seasonal evolution with the highest one in summer (6.74 ± 0.64), followed by ones in spring (6.71 ± 0.71) and autumn (6.71 ± 0.69), and the lowest
Based on the spatial regression analysis, four socioeconomic indicators including GIP, RBU, FDI, and SO$_2$ emission, and two natural factors including AMP and AMRH were identified as the major deriver for the acid deposition. Amongst these key factors, the effects of meteorological factors on the rainwater pH value were stronger than those of socioeconomic indicators, indicating that the socioeconomic factors played the important roles on the acid deposition coupled with the adverse meteorological factors. At a regional scale, the higher effects of GIP, FDI, and SO$_2$ emission on the acid deposition focused on East China because of the frequent human activity and substantial energy consumption. Besides, the higher rainfall amount and RH in Southeast China increased the pollutant deposition fluxes and promoted the photochemical transformations of gaseous pollutants, respectively. The joint effects of socioeconomic and natural factors caused the severe acid rain in Southeast China.

To mitigate the severe acid deposition in China, some policy implications should be considered in the future. First of all, the structure and layout of the energy-intensive industries should be urgently adjusted and optimized especially in NCP and YRD to decrease the gaseous pollutant emissions (e.g., SO$_2$ emission). The proportion of secondary industry in the hot spot (e.g., South China) of acid deposition should be reduced to a large extent, and the proportion of tertiary industry should be increased. In addition, the energy-intensive industries cannot be concentrated on NCP and YRD and some of them should be migrated to the cities of Northwest China with low RH. Secondly, the inappropriate energy consumption structure and low energy efficiency brought about the severe acid deposition in Southeast China. Consequently, all of the newly built power plants and most of the in-use plants must be equipped with the high-efficient flue gas desulfurization (FGD), NO$_x$ treatment projects, and selective catalytic reduction (SCR). Besides,
yellow-sticker vehicle elimination projects should be also performed simultaneously. Moreover, the coal should be gradually replaced by natural gas in the residential usage because coal generally showed the higher S content and lower combustion efficiency compared with natural gas. Thirdly, FDI played a negative role on the acid rain especially in East China. Hence, Chinese government should formulate and implement the environmental protection standards as soon as possible and restrict the foreign investment with high-pollution enterprises. China should introduce more high-technology industries instead of the energy-intensive industries. Fourth, the expansion of built-up area especially the industrial district should be restrained to a certain extent because over-urbanization can result in environmental pollution alongside with a series of social problem. More importantly, the effects of socioeconomic factors on acid deposition were significantly modulated by the meteorological conditions. Thus, the high-pollution enterprises in the acid rain region should be migrated to the region with favorable diffusion condition (e.g., low RH and high WS).

Acknowledgements

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Table and figure captions

**Fig. 1** The flow chart of methodology for analyzing the relationship between acid deposition and influential factors.

**Fig. 2** The seasonal variation of the rainwater pH value in six climatic zones of China.

**Fig. 3** The spatial distribution of the annual mean pH value in the rainfall of China. (a) displays the high-resolution (0.25° × 0.25°) rainwater pH distribution. (b) exhibits the spatial variation of the rainwater pH value at a city level.

**Fig. 4** The spatial variation of the rainwater pH value in different seasons.

**Fig. 5** The spatial cluster of the rainwater pH value at a city level.

**Fig. 6** The power of six factors responsible for the rainwater pH value.

**Fig. 7** The spatial distribution of the local regression coefficients for the six factors based on the GWR model.

**Tab. 1** The threshold of the sub-regions for six influential factors.

**Tab. 2** The results of OLS regression model for the estimation of rainfall pH value.

**Tab. 3** The results of SLM for the estimation of rainfall pH value.
Fig. 2

(a) The rainfall pH value over different seasons in Northeast China, Inner Mongolia, and NCP.

(b) The rainfall pH value over different seasons in Northwest China, Southwest China, Southeast China, and China.
Fig. 3
Fig. 5
Fig. 6
Fig. 7
<table>
<thead>
<tr>
<th>Threshold</th>
<th>Per capita GIP ($10^4$ yuan)</th>
<th>RBU (%)</th>
<th>FDI ($10^4$ dollars)</th>
<th>SO$_2$ emission (t)</th>
<th>Mean precipitation (mm)</th>
<th>Mean RH (%)</th>
</tr>
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<tr>
<td>Low level</td>
<td>&lt; 2.10</td>
<td>&lt; 1.39</td>
<td>&lt; 2167</td>
<td>&lt; 15852</td>
<td>&lt; 527.8</td>
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<td>1.39-6.41</td>
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<td>15852-54882</td>
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<td>High level</td>
<td>&gt; 4.69</td>
<td>&gt; 6.41</td>
<td>&gt; 20100</td>
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<td></td>
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<td>Standard error</td>
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<td>0.00b</td>
<td></td>
<td></td>
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<td>0.010</td>
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<td>AMP</td>
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R-squared: 0.75, adjusted R²: 0.72, F statistics: 90.83, p value: 0.00

a: p < 0.05    b: p < 0.01
<table>
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<tr>
<th></th>
<th>Regression coefficient</th>
<th>Standard error</th>
<th>z value</th>
<th>p value</th>
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</table>

R-squared: 0.76; log likelihood: −200.19; AIC: 430.39; lambda: 0.52; Breusch-Pagan test: 262.54; P-value: 0.00

a: p < 0.05  b: p < 0.01
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1. The spatial econometric models were used to identify key factors for acid deposition in China.

2. GIP, RBU, FDI, and SO$_2$ emission are main factors to aggravate acid deposition over China.

3. These socioeconomic factors played the most significant roles on the acid deposition of East China.
Lulu Cui wrote and revised the manuscript; Jianhong Liang processed the data and performed the statistical analysis; Hongbo Fu and Liwu Zhang revised the manuscript.
Conflict of interest statement

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.