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Analysis of the Spatio-temporal Changes in Acid Rain and Their Causes in China (1998–2018)

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Abstract: With the rapid development of the economy, acid rain has become one of the major environmental problems that endanger human health. Being the largest developing country, the environmental problems caused by acid rain are of increasing concern with the rapid industrialization and urbanization in China. Recently, many researchers have focused on acid rain. To better understand the temporal and spatial dynamics of acid rain in China, the monitoring data on acid rain from 1998 to 2018 were studied using ArcGIS 10.2. The results show that the proportion of acid rain cities, the frequency, and the area of acid rain were decreasing, however, the situation still remains serious. Overall, the chemical type of acid rain was mainly sulfuric acid rain. However, the concentration ratio of SO_4^{2-}/NO_3^- decreased by 81.90% in 2018 compared with 1998, and presented a decreasing trend, which indicates that the contribution of nitrate to precipitation acidity has been increasing year by year. This research will help us to understand the distribution characteristics and causes of acid rain in China, and it may provide an effective reference for the prevention and control of acid rain in China.

Key words: acid rain; pH; spatio-temporal change; chemical composition; trend

1　Introduction

With the rapid development of the economy, acid rain (AR) has attracted more and more attention in recent years (Xu et al., 2015). The increasing concentrations of nitrogen oxides (NO_X) and sulfur dioxide (SO_2) in the air are the main cause of AR. When released into the air, these oxides react quickly with water, after which nitric acid and sulfuric acid can form and fall as rain (Jalali and Naderi, 2012; Shu et al., 2019). Several widely-practiced human activities, such as the large-scale use of coal, construction of power plants, and emissions of automobile exhaust, have resulted in a gradual increase in the content of these oxides in the atmosphere (Mei et al., 2005; Liu et al., 2016). Precipitation with a pH lower than 5.6 is called AR. AR has serious impacts on the environment; it can acidify the soil and water, degrade forests, yellow leaves, reduce biodiversity, and damage buildings (Liu et al., 2001; Ramlall et al., 2015; Debnath et al., 2018; Shu et al., 2019). Studies have shown that China has become the third-largest acid rain region after Europe and North America. Currently, about 40% of the terrestrial area is suffering from AR (Dai et al., 2013; Shu et al., 2019). Although the region in China with the average precipitation pH value below 5.6 is mainly in the south of the Yangtze River, an expanding range of AR zones is obvious especial-

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ly in northern China (Zhao and Hou, 2008). Therefore, in recent years, more people are paying attention to the impact of AR on terrestrial ecosystems.

At the end of the 1970s, the Environmental Protection Department of China set up a series of AR monitoring stations throughout the country, and the Meteorological Department also successively established long-term AR monitoring networks (Ding, 2004). The monitoring results showed that the areas with average annual precipitation pH less than 5.6 were mainly in the southwest, south, and southeast in the mid-1980s (Dai, 1997). Since the 1990s, the area of AR has changed significantly compared with the 1980s. That is, the pollution level of the AR area in central China, which is centered on Nanchang and Changsha, is higher than that in the southwest. Although the intensity of AR has decreased, it remains at a severe level. The AR area in South China is mainly in the Pearl River Delta and eastern Guangxi. In addition, the pollution pattern in the AR area of East China, including the middle and lower reaches of the Yangtze River, fluctuates only on a small scale (Tian et al., 2001).

From the Seventh Five-year Plan to the Ninth Five-year Plan, China had carried out extensive research on the mechanism of formation and transmission of AR, numerical simulations, control methods, and ecological effects. Research on the key problems of AR (1990) in the Seventh Five-year Plan focused on southwest and South China, where AR pollution was relatively serious at that time. The results showed that the AR pollution in Chongqing and Guizhou had reached a considerable degree, and the proportion of sulfate in the precipitation was very high, which characterized it as typical sulfuric acid pollution. During the Eighth Five-year Plan, the research area for AR extended to the eastern coast and central China. The Seventh Five-year Plan and the Eighth Five-year Plan conducted the program which assessed the degree of impact and economic loss of acid subsidence on crops and forests, put forward the principles, methods, and index system for the evaluation and screening of AR control technology, and obtained the control schemes and strategies based on the critical load of sulfur deposition (Hao et al., 2001). Also, in the study of the migratory process of atmospheric pollutants, the Seventh Five-year Plan and the Eighth Five-year Plan's researches accumulated certain experiences and a theoretical basis. Based on the establishment of a relatively reliable sulfur dioxide (SO_2) emission inventory, a sulfide migration model was established and used to calculate the provincial and cross-border transportation volumes (Yu et al., 2021). During the Ninth Five-year Plan, a study was carried out on the procedure for dividing the SO_2 pollution control zone and the AR control zone (The Two Control Zones). As a result, "The Two Control Zones" for controlling the pollution of AR and SO_2 were developed, with the SO_2 pollution control zone established in the north and the AR control zone estab-

lished in the south. During the Eleventh Five-year Plan, the Ministry of Science and Technology also established a project on the mechanism of AR formation and its prevention and control.

Determining the cause of AR is an important aspect of AR research. In terms of the composition of acidic substances in the AR in China, regional AR is mainly caused by a large amount of SO_2 emissions, also known as "soot type" or "sulfuric acid type" (Fan, 2002). In recent years, many large and medium-sized cities have increased nitrogen oxide emissions due to the rapid increase in the number of motor vehicles. As a result, therefore, the content of nitric acid in AR has been increasing year by year. Several studies suggested that the concentration of acid ions in northern China is higher than that in the south, but the acidic precipitation in the north is significantly lower than that in the south since the SO_2 content in the atmosphere in different regions is not positively correlated with the occurrence of AR or its acidity. That is, the precursor material source SO_2 is a necessary, but not sufficient condition, to form AR and the appropriate conditions also include a variety of other natural factors (Chen et al., 2020). For instance, important reasons for the distinctive features of AR in the north and south include differences in the concentration of ammonia in the atmosphere, the soil acid-base properties, and the concentration of alkaline substances in atmospheric particles (Feng, 2004). Zhang et al. (2010) found that the concentration of atmospheric particles (especially coarse particles) in China was higher than in some foreign countries, with significant differences between the north and south. That is, in the northern part of China, the content of alkaline substances was not only high in the soil but also in atmospheric particles. During rainfall, these atmospheric particles have a large neutralization capacity for buffering acidic precipitation. In addition, the content of alkaline ions and pH in the soil are the important factors affecting AR formation. The alkaline substances in precipitation mainly come from the soil, and the amount of alkaline particles in the soil increases from the south to the north, which leads to the main occurrence of AR in the south with a lower alkaline content and lower pH. In addition, Zhang et al. (2010) also found that the sources of AR in the eastern and southeastern coastal areas of China were more complex and affected by Western Europe, Japan, and South Korea in winter and spring. Taking Qingdao as an example, the large amount of acid-causing substances produced by modern industrial development and the natural emission of dimethyl sulfur from the ocean are the main reasons for the formation of AR, and the special laws of sidewinding and temperature fields are another important reason (Feng, 2004). On the contrary, although the concentrations of atmospheric pollutants (SO_2, NO_X, and inhalable particles) are not high in Xiamen, the concentrations of ozone and hydrogen peroxide are high, resulting in strong oxidation of the atmosphere. Therefore, the lack of alkaline

substances in the atmosphere is one of the main reasons for AR in Xiamen (Zhuang, 1998).

Generally, in the AR area of south China, the eluting of acidic substances by precipitation and the superposition of medium- and long-distance transmission are the decisive sources of AR (Guo et al., 2016). Besides, in small and medium-sized cities and rural areas with less pollution, the acidity of precipitation is mainly determined by intra-cloud processes (Huang et al., 1995).

To obtain a more comprehensive understanding of the status, and the temporal and spatial trends of acid rain in China, this paper examined 471 cities in China and evaluated their acid rain status based on their atmospheric environment indicators, such as precipitation pH and frequency, main cation contents, and the concentration ratio of SO_4^{2-}/NO_3^- from China Environment Bulletin issued by the Ministry of Ecology and Environment of the People's Republic of China. It then analyzed and evaluated the spatial and temporal distribution characteristics of acid rain in China.

2 Evaluation methods

In this study, a precipitation pH below 5.6 was used as the AR criterion. pH between 5.0 and 5.6 was considered as light AR, pH between 4.5 and 5.0 was considered as medium AR, and pH below 4.5 was considered as heavy AR. The AR occurrence situation was evaluated by the average annual precipitation pH and the frequency of AR. Additionally, the "AR cities" in this study refer to cities with an average annual precipitation pH of less than 5.6. In addition, the current situation of AR in 2018 was analyzed using monitoring data from all 471 cities. From 1998 to 2018, the changing trends of average annual precipitation pH and frequency of AR were analyzed using monitoring data from 252 comparable cities, and the changing trends of nine ion components were analyzed using monitoring data from 202 comparable cities. All the data were analyzed via Microsoft Office Excel 2007, and all the figures were made using Origin 8.0 and ArcGIS 10.2.

The relevant data used in this study were mainly from the "Chinese Ecological Environment Bulletin" (1998–2018) published by the Ministry of Ecology and Environment of the People's Republic of China. The statistical data mainly included the pH of precipitation, the frequency of AR, the emissions of sulfur dioxide and nitrogen oxides, and the concentration ratio of SO_4^{2-}/NO_3^-.

3 Results

3.1 Characteristics of the variation of precipitation acidity (pH value) and AR frequency

From 1998 to 2018, the pH of precipitation in China exhibited an increasing trend (Fig. 1), and since 2008, the pH of precipitation across the country has maintained values between 5.0 and 5.6, belonging to the light AR category. The AR frequency showed a "low-high-low" downward trend from 1998 to 2018, rising from 25.9% in 1998 to 41.8% in 2005, and then dropping to 10.5% in 2018.

In 1998, the average annual precipitation pH values of 52.8% of the statistical cities with records were less than 5.6 (Fig. 2), and 73.03% of the southern cities had an average annual precipitation pH below 5.6, among which the pH values of Lin'an, Zhuzhou, Yiyang, Shaoguan, Qingyuan, Nanchang, Yingtan and Changsha were less than 4.5. Also, some northern cities, such as Tumen, Qingdao, Xi'an, and Tongchuan, had an average annual precipitation pH below 5.6. By 2018, the AR area was about 5.3×10^5 km² (Fig. 2), accounting for 5.5% of the terrestrial area, of which the light AR area accounted for 0.6%. Spatially, the AR pollution was mainly in the south of the Yangtze River to the east of the Yunnan-Guizhou Plateau, including Shanghai, Zhejiang, central Jiangxi, northern Fujian, central and eastern Hunan, southern Chongqing, and central Guangdong.

To sum up, the pH of precipitation in China has been increasing year by year, and the frequency of AR has been decreasing significantly, which indicates that the quality of acid precipitation in China has been improving.
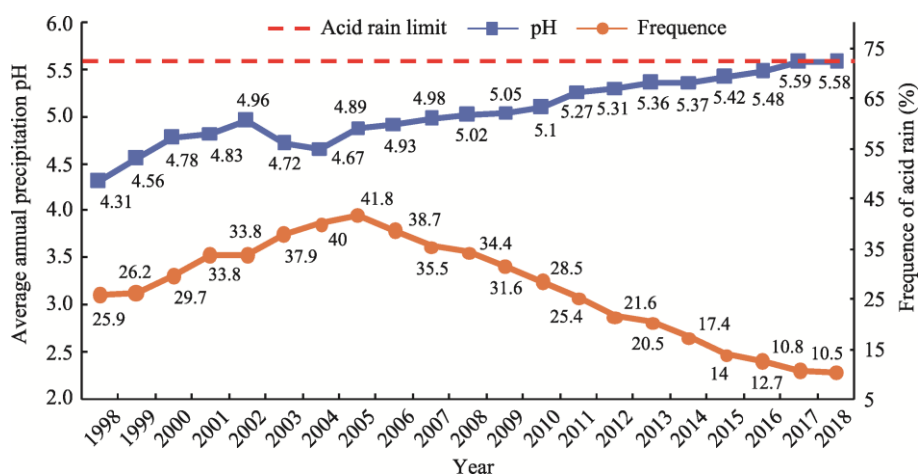

Fig. 1   Average annual precipitation pH and frequency of AR in China from 1998 to 2018
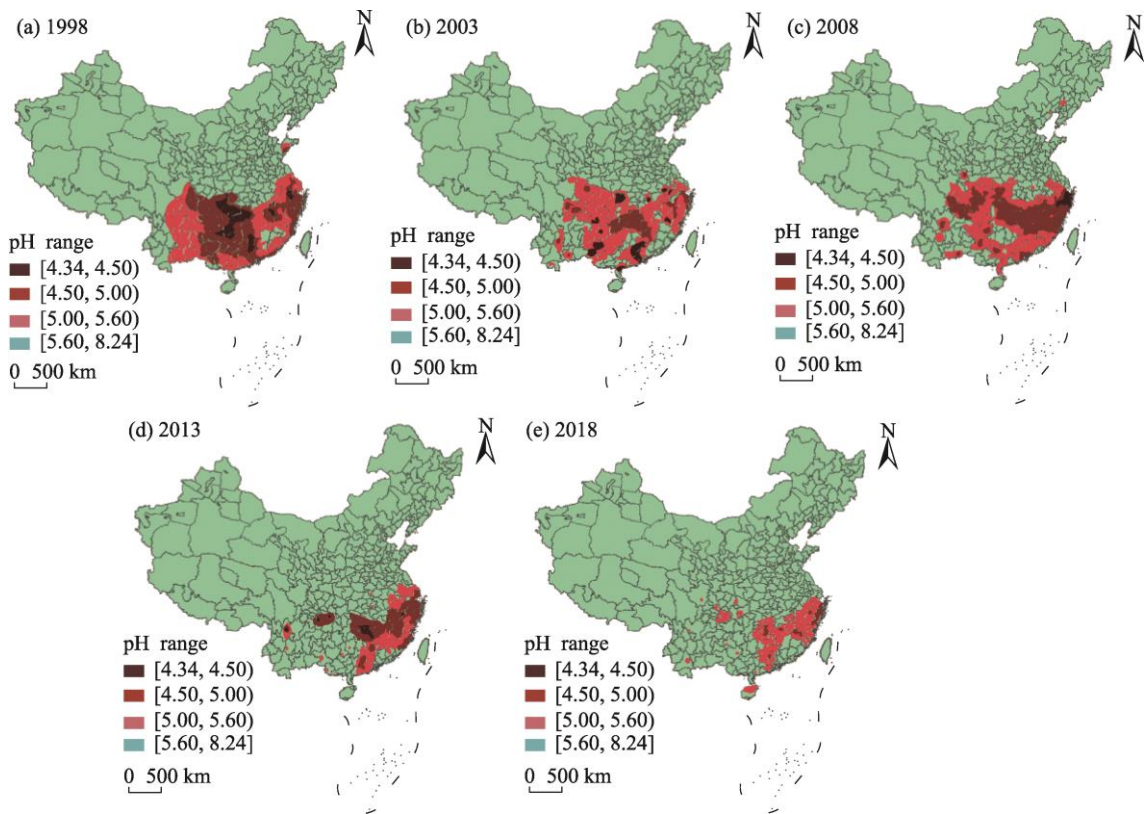
Fig. 2   Regional distribution of AR in China from 1998 to 2018

3.2   Regional distribution of AR

From 1998 to 2018, the distribution pattern of AR remained stable, and the total area of AR showed a downward trend (Fig. 2). Among the regions, the areas of AR in central Guangxi and central Guizhou region decreased, and the acidity of precipitation in the Yangtze River Delta and northern Hunan province dropped. However, the acidity of precipitation in southwest Chongqing and central Fujian increased. Since 1998, the area of AR in China has been on a downward trend. Among the two acidity levels, the area of light AR has been decreasing year by year, and the area of heavy AR has remained stable. In addition, comparing the average annual precipitation pH in 1998 and 2018, the result shows that the AR conditions had been improved in most areas, with the most significant improvement in the southern regions.

3.3   Chemical composition of precipitation

In 2018, the main cations in precipitation in China were calcium ions and ammonium ions, with equivalent concentration ratios of 26.6% and 15%, respectively (Fig. 3). The main anion was sulfate, with an equivalent concentration ratio of 19.9%, and the nitrate equivalent concentration ratio was 9.5%. Since 1998, the proportion of sulfate ions in precipitation has been declining (Fig. 4). On the contrary, the ratio of nitrate ions has increased slightly, while the ratios of chloride and fluorine ions remained stable. Furthermore, for the main cations, the proportion of calcium ions increased
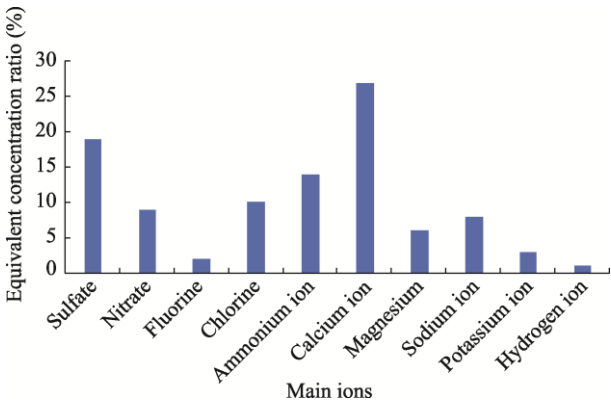

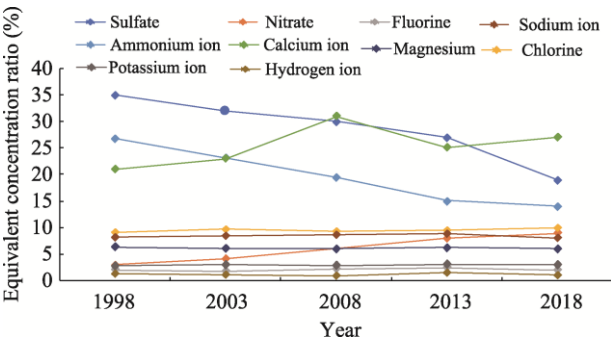Fig. 3   Equivalent concentration ratios of the main ions in China in 2018

Fig. 4   Equivalent concentration ratios of the main ions in China from 1998 to 2018

in 2008, but then declined, and increased again in 2018. The proportion of ammonium ions presented a decreasing trend over this period. Meanwhile, the proportions of other cations did not change significantly.

3.4 Emissions of sulfur dioxide and nitrogen oxides

From 1998 to 2013, the emission of sulfur dioxide presented a slowly increasing trend, and compared with 1998, the emission of sulfur dioxide increased by 20.24% in 2013. But since 2013, it began to decrease rapidly (Fig. 5). In contrast, the emission of nitrogen oxides showed a rapid growth trend from 1998 to 2013. In addition, compared to 1998, the emission of nitrogen oxides increased by 38.62% in 2013, and then it showed a slow decline thereafter (Fig. 5).
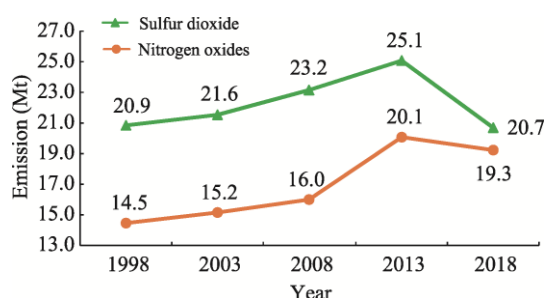

Fig. 5   Emissions of sulfur dioxide and nitrogen oxides in China from 1998 to 2018

3.5 Changes of $C_{SO_4^{2-}} / C_{NO_3^-}$ in precipitation

The data in Fig. 6 show that compared with 1998, the concentration ratio of SO_4^{2-}/NO_3^- decreased by 81.90% in 2018 and presented a decreasing trend, which indicates that the concentration ratio of SO_4^{2-}/NO_3^- in precipitation has been decreasing year by year since 1998. In sum, as can be seen in Fig. 5 and Fig. 6, the contribution of nitrogen oxides to rainfall acidity has been increasing. This indicates that acid rain gradually changed during this period in China, from the previous sulfuric acid-dominant status to one of the combined contributions of sulfuric acid and nitric acid.
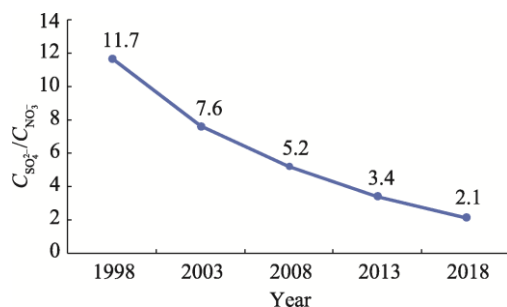

Fig. 6   Variation of $C_{SO_4^{2-}} / C_{NO_3^-}$ in precipitation in China from 1998 to 2018

4   Discussion

This work shows that industrial production and civilian life need to consume a large amount of coal and other forms of energy. However, while using them, coal and petroleum combustion can produce sulfur oxides, and automobile exhaust can release nitrogen oxides. After a series of complex chemical reactions, these acidic compounds form sulfuric acid or nitric acid aerosols which can migrate to the ground in the form of AR through processes such as rain, snow, and fog (Xie et al., 2012). Therefore, the sulfur dioxide and nitrogen oxides produced by energy consumption are the main sources of the acidic substances in precipitation.

In the early 1980s, China was in the early stage of reform and opening up, during which most regions advanced industrialization at a high rate and accelerated the process of urbanization. In the process of urbanization, rural industrialization developed rapidly, and the establishment of township enterprises in rural areas became the most common phenomenon. However, while driving the development of the rural economy, township enterprises caused serious pollution to the environment due to the decentralization of layout and extensive operation mode, which not only produced a wide area of pollution, but it was also difficult to control (Chen et al., 2020). With the rapid development of industrialization, the energy consumption of steel, refining, coking, and chemical industries also gradually increased, and the energy structure of related industries was mostly coal. Therefore, the type of acid rain in the 1980s was mainly sulfuric acid rain. By the late 1980s, China had banned the construction of new coal-fired thermal power plants in urban and suburban areas of large and medium-sized cities. By these standards, desulphurization facilities should be constructed for renewing the coal-fired power plants with a sulfur content of more than 1% (Wu et al., 2006).

In recent years, China has paid more and more attention to controlling atmospheric pollution. It has not only promulgated the "Eleventh Five-year Plan for the Prevention and Control of AR and sulfur dioxide pollution", but also implemented the "Action Plan for the Prevention and Control of Air pollution". During the "Eleventh Five-year Plan", the Chinese government proposed a strategic decision to reduce the total amount of major pollutants, which required sulfur dioxide emissions reduction by 10% as one of the binding indicators. Therefore, through the effective measures in the intervening years, the equivalent concentration of sulfate ion in precipitation has generally declined since 1998 (Zhao and Hou, 2008), which indicates that China has completed the task of reducing emissions.

Although the area and acidity of acid rain in China have decreased, the total amount of nitrogen oxide emissions in the air has been increasing due to the continuous increase in energy consumption and vehicle ownership. As a result, more and more nitrate aerosols are dissolved in the process of precipitation, increasing the concentration of nitrate (Cai, 2018). In other words, throughout the country, the gradual increase in nitrogen oxide content under the condition of the effective control of industrial sulfur dioxide emissions has

led to a gradual decrease in the concentration ratio of SO_4^{2-}/NO_3^- in precipitation, which indicates that the contribution of nitrogen oxides to the acidity of precipitation is increasing (Zhang and Li, 2011).

In view of the present situation of acid rain in China, the following prevention and control measures are put forward.

4.1 Optimizing industrial structure and rational use of clean energy

Controlling and reducing SO_2 emission from coal combustion is the most direct and effective method for the comprehensive control of acid rain (Yang et al., 2005). Therefore, first of all, we need to study and establish the early warning mechanisms for the carrying capacity of resources and the environment. Second, it is necessary to accelerate the development of clean energy such as hydropower, nuclear power, and wind power, while promoting the development of distributed energy and giving priority to the use of renewable energy. Third, formulating plans for controlling total coal consumption in key regions are needed. Fourth, improving the quality of oil products should be considered as well.

4.2 Developing and promoting of environmentally friendly and efficient energy-saving technologies

At present, acid rain in China has basically changed from the sulfuric acid type to a sulfuric acid-nitric acid mixed type, so it is necessary to reduce the emissions of both sulfur dioxide and nitrogen oxides into the atmosphere. In China, the main energy source is coal, and this energy structure will not be changed in a short period. To this end, it is necessary to actively control the emission of sulfur dioxide through the development of efficient and clean energy-saving technologies, such as energy cascade utilization, efficient power generation, transmission, and storage technology, and terminal energy-saving technology (Wan and Wang, 2010). The emission of nitrogen oxides into the air in China mainly comes from the exhaust pollution of motor vehicles. Therefore, first of all, the government should formulate emission standards. Secondly, the major automobile manufacturers should introduce new technologies, such as the engine structure, anti-fouling devices, more effective automobile exhaust treatment systems, and new energy vehicles.

4.3 Strengthening environmental management and environmental education

To strengthen the control of acid rain, the government should enhance the supervision of environmental management and improve the awareness and education of environmental protection. As long as our environmental protection departments at all levels strengthen environmental law enforcement and make the law play a full role, the national policy of environmental protection can be truly implemented.

Acid rain is a global problem (Li et al., 2017). People around the world should take joint action to achieve effective international cooperation with a sincere attitude, actively exchange experiences, information, and research results in environmental protection, and jointly control acid rain pollution.

5 Conclusions

In 1998, the proportion of AR cities in China was 52.8%, and by 2018, it had fallen to 37.6%. The area of AR showed a downward trend, indicating that the air quality had been improved in China. In 2018, AR pollution was mainly in the south of the Yangtze River to the east of the Yunnan-Guizhou Plateau, including Shanghai, Zhejiang, central Jiangxi, northern Fujian, central and eastern Hunan, southern Chongqing, and central Guangdong. In precipitation, the main cations were calcium and ammonium ions, and the main anion was sulfate. But since 1998, the proportion of sulfate ions in precipitation has been decreasing, and the proportion of nitrate ions has increased slightly, resulting in a decrease in the concentration ratio of SO_4^{2-}/NO_3^- in precipitation, which indicates that the contribution of nitrogen oxides to precipitation acidity has been increasing. Even so, in the current situation (as of 2018), the type of acid rain is mainly sulfuric acid rain.

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1998—2018 年我国酸雨的时空变化及其原因分析

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　　摘　要：随着经济的快速发展，酸雨已成为危害人类健康的生态环境问题之一。作为最大的发展中国家，中国的工业化和城市化进程加快，酸雨引起的环境问题日益受到各界人士的广泛关注。为了更好地了解酸雨的时空变化动态，本文使用 ArcGIS 10.2 对 1998–2018 年中国酸雨的监测数据进行了研究分析。结果表明，我国酸雨城市的比例、酸雨频率以及酸雨面积均有所下降，但形势依然严峻。中国酸雨类型以硫酸型酸雨为主，但与 1998 年相比，2018 年 SO_4^{2-}/NO_3^- 的浓度比从 11.7 下降到 2.1，整体呈下降趋势，表明硝酸盐对降水酸度的贡献逐年增加。本研究有助于我们了解我国酸雨的分布特征和成因，可为我国酸雨的防治提供参考。

　　关键词：酸雨；pH；时空变化；化学成分；趋势