A critical review of the variation in rainwater acidity in 24 Chinese cities during 1982—2018

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Rainwater is an essential pathway to remove fine particulate matter and dissolved atmospheric pollutants (e.g., SO₂, HNO₃, and NH₃). Acid rain (pH < 5.6) has been a severe environmental issue in China since the 1970s, adversely impacting ecosystem health. This study focuses on the influence of anthropogenically induced anions (SO₄²⁻ and NO₃⁻) and alkaline cations (Ca²⁺ and NH₄⁺) on acid rain in Chinese cities. In this review, cities with high population density east of the Hu Huanyong Line that divides China geographically according to its uneven economic development were studied. Coastal and central areas of China to the east of the line are characterized by a much faster developing economy and rapid urbanization. The observed trends and spatial variability of acidity and chemical composition in rainwater are discussed in relation to industrialization and environmental changes in China. Over the past 3½ decades, the precipitation pH in the urban regions has exhibited reduced acidity. A mixed nitric–sulfuric acid rain type has become prominent due to the significant decrease in SO₄²⁻ via desulfurization. Ca²⁺ levels have decreased, while NH₄⁺ has increased slightly due to more vehicular transportation. In addition, the neutralization capacity of Ca²⁺ and NH₄⁺ has decreased from north to south. Overall, the acid rain problem in Chinese cities has been alleviated in recent years.

Keywords: Rainwater, Acid rain, Major ions, Neutralization capacity, Anthropogenic inputs, China

1. Introduction
1.1. Far-past review
Acid rain, defined as rainwater with a pH < 5.6, has become one of China’s most prominent environmental problems (Marion, 1998; Seinfeld, 1998; Aas et al., 2007; Niu et al., 2014). The harm caused by acid rain includes soil acidification, degradation of the soil agricultural ecosystem, forest decline, and adverse health impacts (Bian and Yu, 1992; Menz and Seip, 2004; Rice and Herman, 2012; Zhu et al., 2016). Severe acid rain is often caused by anthropogenic emission of SO₂ and NOx, arising from massive urbanization, industrialization, and transportation (Zhang et al., 2017a; Xu et al., 2020). With the acceleration of industrialization and increasing energy demand, China’s dependence on fossil fuels since the 1970s has resulted in frequent acid rain (Zhao et al., 1988; Han and Liu, 2006; Wu et al., 2012; Zhou et al., 2019). Over the past four decades, China has become the third largest region globally to suffer from acid rain, behind North America and Central Europe (Wang and Wang, 1995; Ito et al., 2002; Zhang et al., 2010).

Since 1990, acid rain has generally occurred in southwestern China, particularly in the southern Yangtze River, eastern Qinghai-Tibet Plateau, and the Sichuan Basin. From 1997 through 2002, acid rain prevailed over approximately 30% of the land area in northern, eastern, and southern China (Xu et al., 2009; Han et al., 2010b; Cheng et al., 2011; Lu et al., 2011; Zhang et al., 2012). Between 2003 and 2006, acid rain episodes spread to northern China, including Taiyuan city, Beijing, Liaoning province, and Jilin province (Tang et al., 2010; Wang and Xu, 2011; Luo et al., 2013). Due to rapid urbanization and industrialization, amid incomplete implementation of government control policies before 2006, acid rain was severe in developed cities (such as Guangzhou and Chengdu) in southern China.

1.2. Recent-past review
The severity of acid rain in China in recent decades has been attracting research attention, particularly in relation to the chemical composition of rainwater. Previous cases of acid rain were mainly caused by economic growth and human activities, while the recent alleviation is credited to Chinese government control policies (Huang et al., 2008; Cao et al., 2009; Han et al., 2010b; Liu et al., 2013; Bai and Wang, 2014; Rao et al., 2015; Xiao, 2016; Xing et al., 2017; Zhong et al., 2018; Zeng et al., 2019; Xu et al., 2020). In the mid-1990s, several policies, including energy structure changes, energy savings, and emission reduction, were first introduced but not fully implemented (e.g., Zheng et al., 2020).
The SO₂ emission in China has increased continuously since 1980, reaching >33 million tons in 2006 (Duan et al., 2016; Zheng et al., 2020). It began to decrease in 2006 due to the wide application of flue-gas desulfurization (FGD) in power plant units (Kurokawa et al., 2013; Duan et al., 2016). Still, the SO₂ emission load was approximately 28 million tons in 2010 (Li et al., 2017a), too high to be ignored (Kurokawa et al., 2013). Similarly, using selective catalytic reduction (SCR) in coal-fired power plants has lowered NOₓ emissions since 2012 (Wang et al., 2014b).

Since 1982, nationwide surveys of environmental quality indicators (such as acid rain), sponsored by the Chinese Ministry of Environmental Protection Administration (re-organized as the Ministry of Ecology and Environment of the Peoples’ Republic of China after 2018), have been carried out annually (Wang and Wang, 1995; Fujita et al., 2000).

Acid rain frequency is the proportion of acid rain in the total number of rain events over a period. Regions with acid rain frequency >10% are regarded as acid rain areas. According to the State of the Environment Bulletin of China, the acid rain area covers approximately 530,000 km², accounting for 5.5% of China’s land area. Severe acid rain (pH < 4.5) areas account for approximately 0.6% of land area, mainly located in the southeast and coastal cities. Recently, acid rain in China has significantly improved; however, with ongoing urbanization and economic development, reducing acid rain and other chemical compositions in rainwater remain imperative.

Meanwhile, fine particles (PM₁₀) are primary pollutants of substantial public concern in urban cities (Wei et al., 2019; Wei et al., 2021b). Through wet precipitation, rainwater scavenges air pollutants from the air (Niu et al., 2014; Rao et al., 2016; Wei et al., 2021a). Thus, the chemical composition of rainwater can serve as a tracer for air pollution (Liu et al., 2013; Niu et al., 2014; Rao et al., 2015; Zheng et al., 2020) and monitors the impact of natural and anthropogenic sources on rainwater (Apko et al., 2015).

The primary chemical constituents of rainwater are divided into cations (K⁺, Na⁺, Ca²⁺, Mg²⁺, and NH₄⁺) and anions (F–, SO₄²⁻, NO₃⁻, and Cl⁻). Of the anions, SO₄²⁻ and NO₃⁻ originate from SO₂ and NOₓ and are the main contributors to rainwater acidification (Roy et al., 2016; Singh et al., 2016). Investigating the chemical composition of rainfall facilitates understanding of the air pollutant cycles by identifying sources such as local natural sources (Dordvic et al., 2005; Larsen et al., 2006), long-range transport, dust (Gioda et al., 2013; Li et al., 2016), and anthropogenic activities (Han et al., 2011; Xiao, 2016; Zhang et al., 2017a). For example, Ca²⁺ and Mg²⁺ may be attributed to soil dust, while SO₄²⁻ and NO₃⁻ may be derived from human activities such as fossil fuel combustion (Ciezka et al., 2016; Zeng et al., 2019).

Acid rain is reduced by Ca²⁺ and NH₄⁺ in a neutralization process (Zeng et al., 2020). Historically, acid rain occurred primarily in southern China, even though SO₂ levels were also high in northern China (Wang and Wang, 1995; Zhao et al., 1988; Fujita et al., 2000). However, the high SO₂ concentration in the north is not accompanied by acid rain because CaCO₃ and NH₃, which are essential neutralizing compounds, are prominent therein (Tang et al., 2005; Xu and Han, 2009; Wang et al., 2012; Wu et al., 2016). Between 1980 and 1996, NH₃ emissions initially increased, before decreasing, due to a reduction in the usage of ammonium bicarbonate fertilizer (Duan et al., 2016; Kang et al., 2016). Also, because northern China has more desert regions than the south, the former has abundant soil dust with high alkaline salts that neutralize acid rain (Zhao et al., 1988; Xu and Han, 2009; Xu et al., 2009; Han et al., 2010a; Xu et al., 2015; Wu et al., 2016). Therefore, understanding the influence of local environmental characteristics on the chemical composition of rainwater is essential. However, emissions of materials that are sources for alkaline ions in precipitation increased with the growth of urbanization from construction materials and vehicle emissions, resulting in neutralization of the acid rain (Han et al., 2011; Wu et al., 2012; Han et al., 2019; Zhou et al., 2019). These studies have indicated that the decreasing trend in acid rain in North and South China is due to ubiquitous urbanization.

The pH data are not enough to measure the improvement of atmospheric quality induced by wet precipitation, although pH directly reflects the acidity of rainwater. Therefore, the number and nature of ions in rainwater need to be considered. Previous studies have reported that Na⁺, K⁺, and Mg²⁺ account for a minor proportion of the total ions in urban areas (Xiao et al., 2013; Wang et al., 2018). These ions (Na⁺, K⁺, and Mg²⁺) are mainly from marine sources, while SO₄²⁻, NO₃⁻, Ca²⁺, and NH₄⁺ are primarily derived from crustal sources such as urban construction, agricultural activities, and vehicle emission (Moreira-Piñeiro et al., 2014; Fu et al., 2017; Warner et al., 2017; Han et al., 2019; Zeng et al., 2019).

In China, rainwater variations have impacted overall Pan-Pacific region’s atmospheric conditions and quality over several decades. The pH value reflects the acidity of rainwater, and an increasing pH value trend shows a decreasing acidity [H⁺] in rainwater. Therefore, this review focuses on SO₄²⁻, NO₃⁻, Ca²⁺, NH₄⁺, and pH values to investigate if and how the chemical constituents of rainwater influence severe atmospheric environmental problems in China, especially in urban metropolises.

2. Data and methods

All data in the figures and tables are volume-weighted mean (VWM) values from published scholarly articles (Table S1). The VWM was calculated as follows:

\[ X_{vwm} = \frac{X_1P_1 + X_2P_2 + \ldots + X_nP_n}{P_1 + P_2 + \ldots + P_n} \]

where \( X_n \) and \( P_n \) represent analyte (ion concentration or pH) value and rainwater volume collected in raining event \( n \), respectively.

The sampling and analytical methods for the rainwater are referenced accordingly (Han et al., 2019). The standard GBW08606 validated the analytical reliability of the ion concentrations (Tripathee et al., 2020). Further quality control of the gathered data was done according to the Environmental Protection Agency (EPA) statistical method (EPA, 2006; Tang et al., 2010). Even though researchers used different instruments and methods, their reported
analysis precision and reproducibility of results were consistently better than 5% (Han et al., 2011; Wu et al., 2012; Han et al., 2019).

Rainwater data from 24 Chinese cities (Figure 1) that span over 3½ decades were collected and compared in this study. As shown in Figure 1, the Hu Huanyong Line (Hu Line) divides China geographically into two regions based on the pattern of urbanization (Chen et al., 2016; Qi et al., 2016). The eastern area is characterized by faster urbanization, having a denser human population than the west (Chen et al., 2019). Based on the geographical and developmental characteristics, the 24 cities can be categorized into four groups: northern cities in northern provinces, southern cities in southern provinces, metropolises, and southeast coastal cities. The northern provinces include Henan province (Zhengzhou city and Anyang city), Shaanxi province (Xi’an city), Liaoning province (Dalian city and Anshan city), and Shandong province (Qingdao city). Rainwater data of Xi’an city and Anyang city from 2000 to 2015 were used, while data from before 2006 were used for other cities. The southern provinces include Sichuan province (Chengdu city, Leshan city, and Meishan city), Jiangsu province (Nanjing city, Changzhou city, and Taizhou city), Hunan province (Changsha city), and Chongqing city. For Chengdu city and Nanjing city, only rainwater data up to 2006 were used. The metropolises included Beijing, Guangzhou, and Shanghai, among which Guangzhou and Shanghai are southern coastal cities, while Beijing is in the north. These three cities are characterized by heavy industry, a strong economy, a high-density human population, and a sophisticated transportation network (Zhang et al., 2019). Guangzhou is the most developed city in the Pearl River Delta (Zeng and Han, 2020a; Zeng and Han, 2020b), and only data from before 2008 for Guangzhou were used. However, the data from Beijing and Shanghai spanned over 3.5 decades. The southern and southern coastal cities include Nanping city, Foshan city, Guilin city, Nanning city, Yingtan city, Nanchang city, and Nanchang city; rainwater data from before and after 2010 were used for these cities.

3. Results and discussion

3.1. Chemical composition of rainwater in China

3.1.1. Rainwater chemistry of southern and northern cities

The chemical composition of rainwater in the southern and northern cities of China has changed significantly in recent decades (Figure 2). According to the China
Figure 2. Volume-weighted mean concentration of acid anions and pH value in several northern and southern cities in China. Data from Zhengzhou city (Sun, 1998; Guo et al., 2008); Anyang city (Zhao et al., 2017); Xi’an city (Song et al., 2013; Wang et al., 2018); Dalian city (Lu et al., 1987; Xie et al., 1998; Liu and Wang, 2011; Ji, 2012; Chen, 2013); Anshan city (Zhao, 2015); Qingdao city (Wei, 2008); Chongqing city (Zhao et al., 1988; Wei et al., 2005; Zhao et al., 2013; Song et al., 2017); Chengdu city (Lei et al., 1997; Mei et al., 2005; Wang and Han, 2011); Leshan city (Zheng, 2012; Li, 2015; Zhang, 2017); Nanjing city (Tu, 1999; Zheng et al., 2007); Changzhou city (Yang et al., 2009; Wang et al., 2014a); Taizhou city (Xu, 2017); Changsha city (Jiang et al., 2003; Liu and Chang, 2009; Yi et al., 2014). DOI: https://doi.org/10.1525/elementa.2021.00142.f2
Statistical Yearbook (National Bureau of Statistics of the People’s Republic of China, 2018), the cities showed significant SO$_2$ and NO$_x$ concentration variations, resulting in discrepancies in the chemical composition of rainwater. Still, we observed a general trend in the chemical composition throughout the decades. Generally, a decline in pH occurred mainly in the late 1990s to mid-2000s, resulting in acid rainwater across China. After that period, a trend of increasing pH ensued, presenting as the gradual alkalization of rainwater. Simultaneously, SO$_4^{2–}$ concentrations significantly declined, especially in recent years, whereas NO$_3^–$ levels were somewhat stable or increased slightly. These effects are attributed to the enforcement of some mitigation policies such as the Action Plan for Prevention and Control of Air Pollution in 2013 by the central government (Wang et al., 2018) and other environmental control measures by the local government. The SO$_4^{2–}$ concentrations of rainwater in the northern cities (e.g., Zhengzhou and Xi’an) were higher than those in southern cities. Such effects could be attributed to the large coal consumption in northern cities, where coal burning is their primary energy source (Shen et al., 2009; Zhang et al., 2015). A previous study reported that coal consumption in Qingdao city, as a primary energy source, reached 11.16 million tons in 2006, of which industrial coal burning accounted for 89% (Wei, 2008). Such dependence on coal energy likely contributed to the increase in SO$_4^{2–}$ levels in rainwater (Figure 2d).

The wide application of FGD and denitrification in Anyang cities achieved approximately 80% efficiency in eliminating acid rain from 2006 to 2016 (Zhao et al., 2017). As a result of coal-burning control in Xi’an city, SO$_2$ emissions decreased from 48 to 24 µg m$^{-3}$, leading to respective reductions in SO$_4^{2–}$ and NO$_3^–$ by 56% and 38% from 2009 to 2015 (Wang et al., 2018). As for southern Chinese cities, acid rain has been severer than that in northern cities, although the pH has been increasing gradually (Figure 2e). For instance, the pH increased from 4.4 to 5.1 between 2006 and 2008 in Chengdu city (Figure 2f), as the SO$_4^{2–}$ concentration decreased onefold, while NO$_3^–$ increased fivefold (Lei et al., 1997; Wang and Han, 2011). During 2009–2013, the pH range was 5.6–7.0, indicating that acid rain was not an issue in Meishan city (Li, 2011). During 2009–2013, the pH range was 5.6–7.0, indicating that acid rain was not an issue in Meishan city (Li, 2011). For example, the vehicular density increased fivefold in Nanning city during 2005–2014 (Guo et al., 2017), while the NO$_3^–$ only decreased by 20% during 2011–2016 (Meng et al., 2019). The reductions were ascribed to approximately 30% of the power plant equipped with SCR for NO$_x$ control and 80% with FGD for SO$_2$ control in 2011 (Fu et al., 2013). Nevertheless, increasing vehicular transportation has played an essential role in increasing NO$_x$ emissions in recent years, increasing the NO$_3^–$ concentration in rainwater (Liu et al., 2017).

### 3.1.2. pH and acidic ions of rainwater in metropolises

Figure 3 shows an increasing pH trend and decreasing SO$_4^{2–}$ and NO$_3^–$ trends in Chinese metropolises in recent years. The pH values in all the municipalities have increased since 2005. Because Beijing is located in northern China, while Guangzhou and Shanghai are southern coastal cities, the pH of rainwater in Beijing is higher than that of the other two metropolises. The rainwater pH and chemistry in Beijing changed significantly around 2000 (Tang et al., 2005; Xu and Han, 2009). Even though the pH decreased during 1980–2000, and the significant acid rain occurred after 2000, rain alkalization has occurred in recent years.

The acid rain frequency during 2000–2005 was 16% (Yang et al., 2012), which increased to 27% during 2010–2013 (Zhu et al., 2016). The pH of 97% of the rainwater samples during 2017–2018 ranged from 6.3 to 7.5 in Beijing’s rural area (Xu et al., 2020). The government has carried out a series of air pollution control measures since 1998 (Yang et al., 2012). However, the VWM SO$_4^{2–}$ concentration decreased by 13% during 1998–2005, much less compared to the 58% annual reduction in SO$_2$. In the same period, the NO$_3^–$ concentration doubled, whereas NO$_2$ saw a 5% reduction (Yang et al., 2012). Nevertheless, after implementing the 13th Five Year Plan (2016–2020), with a 15% decrease in SO$_2$ emissions in 2020 compared to 2015, the rainwater pH in China increased. Pu et al. (2017) also reported that the rainwater pH increased significantly during 2008–2014 in Beijing’s remote area.

Elsewhere, Zhu et al. (2016) reported that the anthropogenic source of SO$_4^{2–}$ and NO$_3^–$ in rainwater accounted for 97% and 99%, respectively. The acid ion concentrations in the northern cities, including Beijing, are generally higher than that in southern cities. Similarly, the rainwater pH is also higher in the northern cities due to Ca$^{2+}$ and NH$_4^+$ (Section 3.2).

Shanghai’s rainwater has been acidic for three decades (Figure 3c). From 1986 to 2016, three pH variation trends have been evident. From 1992 to 1997, the pH increased from 4.82 to 5.72, reducing acid rain frequency from 28% to 11%. However, the pH decreased from 5.72 to 4.92, with acid rain frequency increasing to 32% during 1997–2004 (Sha et al., 2007). Wang et al. (2016) reported an apparent decline in SO$_4^{2–}$ and NO$_3^–$ concentrations in Shanghai, consistent with Figure 3c. Such an achievement is attributed to the Shanghai Government’s control policies during the 12th Five-Year Plan period, that is, 2011–2015 (Lin et al., 2013; Li et al., 2017b). In 2005, the SO$_4^{2–}$ concentration decreased by >60% in Shanghai, while the NO$_3^–$ only decreased by 20% during 2011–2016 (Meng et al., 2019). The reductions were ascribed to approximately 30% of the power plant equipped with SCR for NO$_x$ control and 80% with FGD for SO$_2$ control in 2011 (Fu et al., 2013). Nevertheless, increasing vehicular transportation has played an essential role in increasing NO$_x$ emissions in recent years, increasing the NO$_3^–$ concentration in rainwater (Liu et al., 2017).

### 3.1.3. pH and acidic ions of rainwater in southeast and coastal cities

The southeast and coastal areas of China are traditional acid rain areas (Figures 3b and 4). Some areas therein continue to suffer from acid rain despite the recent improvements. The SO$_4^{2–}$ and NO$_3^–$ concentration variations were similar to those of other regions in China, that is, control of SO$_2$ and increasing NO$_x$ levels in recent years. For example, the vehicular density increased fivefold in Nanning city during 2005–2014 (Guo et al., 2017), while a 87% increase was recorded in Foshan city during 2006–2010 (Yao and Liang, 2012). According to Huang et al. (2009), acid rain frequency showed a strong correlation
with the emission of NO\textsubscript{x} and SO\textsubscript{2} in Guangzhou. Although the pH has increased in these areas, the severe acid rain situation has not seen a significant change. Therefore, the southeast coastal cities are the few remaining areas experiencing acid rain nowadays.

### 3.1.4. Overview of pH and acidic ions trends in cities of China

Figure 5 presents the change in the concentration ratio of SO\textsubscript{4}\textsuperscript{2–} to NO\textsubscript{3}– in rainwater, which affects the acid rain type, for more than three decades in China. In the early stages, there was a decline in the [SO\textsubscript{4}\textsuperscript{2–}]/[NO\textsubscript{3}–] ratio in northern and southern China, indicating the rainwater was predominantly polluted with sulfuric acid. Gradually, a mixture of sulfuric acid and nitric acid became the main pollutants. The decrease in [SO\textsubscript{4}\textsuperscript{2–}] and slight increase in [NO\textsubscript{3}–] resulted in a decline in the [SO\textsubscript{4}\textsuperscript{2–}]/[NO\textsubscript{3}–] ratio, ascribed to the effective environmental management of SO\textsubscript{2} emissions and the rise in vehicular transportation. For example, according to the Shanghai Statistical Annual Report, the number of vehicles increased by 43% from 2011 to 2016. SO\textsubscript{2} emissions originate primarily from coal combustion, while NO\textsubscript{x} emissions are from oil fuel combustion (Larssen et al., 2011; Zhang et al., 2017b). Such observations are consistent with the high SO\textsubscript{4}\textsuperscript{2–}/NO\textsubscript{3}– ratio in Shanghai compared to other areas (Figure 5), due to the concentrated coal resources in northern China.

To evaluate the degree of acid neutralization, \(\Delta p\text{H}\) is defined as Equation 1:

\[
\Delta p\text{H} = p\text{H} - p\text{Ai},
\]

where \(p\text{Ai}\) is the estimated pH without neutralization (Hara et al., 1995). The \(p\text{Ai}\) is computed using Equation 2:

\[
p\text{Ai} = -\log([\text{nss-SO}_4^{2–} + \text{NO}_3^{–}]),
\]

where nss-SO\textsubscript{4}\textsuperscript{2–} is referred to as the non-sea salt (nss) values of the SO\textsubscript{4}\textsuperscript{2–} concentration.

Using Na\textsuperscript{+} as the reference cation and assuming that all the Na\textsuperscript{+} came from marine sources (Keene et al., 1986), the nss was calculated using Equation 3:

\[
\text{nss-SO}_4^{2–}/C_0 \times \frac{X}{C_0} = \frac{X}{C_0} \frac{\text{Na}_\text{rainwater}}{C_0} \times \frac{\text{Na}_\text{sea}}{C_0},
\]

where X denotes the estimated ion.

Therefore, differences in \(\Delta p\text{H}\) among the Chinese cities can indicate the acid neutralization capacity. As shown in Figure 6, the rainwater pH in southeast and coastal cities was generally lower than in other cities. Similarly, the \(\Delta p\text{H}\) was also lower in southeast and coastal cities.
3.2. Variation in alkaline abundances and neutralizing capacity

3.2.1. Alkaline constituent variation of rainwater in southern and northern cities

The significant, positive correlation between pH and ΔpH across China (Figure 7) reflects how the neutralizing capacity of the alkaline composition impacts the rainwater pH (Zhou et al., 2019). Previous studies have shown that the alkaline composition of soil dust from desert and semiarid areas was the primary contributor to the Ca²⁺ in rainwater in northern China (Xu and Han, 2009; Wang et al., 2012; Wu et al., 2013). Nevertheless, it is challenging for the relatively wet surface dust to enter into the southern China air due to the humid climate (Zhou et al., 2019). Therefore, alkaline compositions of soil dust in northern China have a greater impact on neutralization than in southern China (Huang et al., 2009), especially in southeast and coastal areas.

Moreover, high NH₄⁺ concentrations in rainwater has been attributed to gaseous ammonia, primarily emitted from agricultural fertilizer (17%), cattle breeding (80%), and industrial processes (Flues et al., 2002). The highest NH₃ concentration was observed in South Asia and Northeast China, where heavy fertilizer application is common (Huang et al., 2012). As for the Midwestern United States, the increase in fertilizer usage is approximately 1.3% yr⁻¹, resulting in a significant increase in NH₃ levels (Warner et al., 2017). Therefore, the influence of NH₃ on rainwater in China may be referential for atmospheric changes in the Pan-Pacific region. Along with Ca²⁺, NH₄⁺ is a principal alkaline cation that neutralizes rainwater acidity (Larssen et al., 2006; Wang et al., 2012; Wu et al., 2016; Han et al., 2019).

The relationship between acidic and alkaline constituents evaluates the neutralization capacity of the latter. Fujita et al. (2000) proposed that Equation 4 could describe the interrelationship between the neutralizing potential (NP) and acidifying potential (AP):

\[
NP/AP = \frac{\text{[nss-Ca}^{2+} + \text{NH}_4^+]}{\text{[nss-\text{SO}_4^{2-} + \text{NO}_3^-}}}.
\]

(4)

where NP is [nss-Ca²⁺ + NH₄⁺] and AP is [nss-SO₄²⁻ + NO₃⁻]

The changes in Ca²⁺, NH₄⁺, pH, and NP/AP over the decades in China are shown in Figure 8. In general, the trends of Ca²⁺ and NH₄⁺ are significantly different from...
those of $\text{SO}_4^{2-}$ and $\text{NO}_3^-$, showing a significant decline in recent years due to the control measures implemented (Section 3.1). The NP/AP shows a similar tendency with pH, indicating that the alkaline ions are essential to neutralizing rainwater acidity (Zhou et al., 2019). In Zhengzhou city for instance, $\text{Ca}^{2+}$ and $\text{NH}_4^+$ decreased by 76% and 55% during 1996–2002, respectively, while $\text{SO}_4^{2-}$ decreased by 73% and $\text{NO}_3^-$ remained relatively stable. Then, during 2002–2005, $\text{Ca}^{2+}$ increased from 315 to 502 $\mu$eq/L, and $\text{NO}_3^-$ increased from 43.4 to 86.3 $\mu$eq/L, while $\text{SO}_4^{2-}$ and $\text{NH}_4^+$ remained relatively unchanged. Meanwhile, the pH decreased during 1994–2002 but increased mildly during 2003–2005 with increasing NP/AP. An increase in $\text{Ca}^{2+}$ concentration probably improved the pH in rainwater during 2002–2005 in Zhengzhou city.

Figure 5. The ratio of $\text{SO}_4^{2-}$ and $\text{NO}_3^-$ concentration in rainwater around China during 1990–2016. DOI: https://doi.org/10.1525/elementa.2021.00142.f5
As for Anyang city, the NP/AP showed a significant increasing trend with relatively high Ca$^{2+}$ concentrations during 2006–2015, confirming that rainwater alkalization increased as the pH increased. Moreover, the NP/AP exhibited a similar trend to pH in Xi’an city, showing that the alkaline ions were predominant in the rainwater despite the decline in cation concentration in recent years. Furthermore, higher levels of NH$_4^+$ were observed in the southern cities than in the north (Figure 8), confirming that Ca$^{2+}$ resources were affected by the northern desert and semiarid areas. Regarding the rainwater in Chengdu city (Mei et al., 2005; Wang and Han, 2011), the main sources of Ca$^{2+}$ were soil dust and industrial dust, primarily attributed to urban construction. This observation is consistent with other rainwater studies in a southern city (Han et al., 2011).

In another study, Sichuan was reported as an intense agricultural production area, exhibiting a high concentration of NH$_4^+$ in rainwater due to the high emission of NH$_3$ in the region (Wang and Han, 2011). As for Meishan city, the frequency of alkaline rain (pH > 7.0) increased from 5% to 44% during 2009–2013, and the correlation between Ca$^{2+}$ and Mg$^{2+}$ indicated that the alkaline ions originated from urban construction dust (Li, 2015).

### 3.2.2. Variation in the alkaline constituents of rainwater in metropolises, southeast, and coastal cities

The NP/AP values in Beijing were generally higher than those in Guangzhou and Shanghai, and correlated more closely with trends in pH, because Beijing is a northern city. The decrease of Ca$^{2+}$ directly improved air quality, resulting in a significant reduction of sandstorm and dust weather in Beijing, especially since the 2008 Olympic Games (Yang et al., 2011; Xu et al., 2012). However, the proportion of NH$_4^+$ increased despite the reduction in NH$_4^+$ in the rainwater. In the urban areas of Beijing, the high NH$_4^+$ originating primarily from fertilizer...
contributed 45% of the total NH$_4^+$ (Pan et al., 2018). Since 2006, NH$_4^+$ has become the dominant cation in rainwater (Figure 9c), accounting for 48% of total cations during 2011–2016 (Meng et al., 2019).

Because Shanghai is surrounded by developed agricultural provinces (such as Zhejiang and Jiangsu), the city has been affected by prevailing southeastern winds, bearing high NH$_3$ levels from agriculture (Huang et al., 2013; Wang et al., 2015). Furthermore, the increasing industrial and vehicular emissions also contributed majorly to the atmospheric NH$_3$ levels (Liu et al., 2014; Wang et al., 2015).

The levels of Ca$^{2+}$ and NH$_4^+$ in southeast and coastal city rainwaters were relatively lower than those in other Chinese cities (Figure 10). NH$_4^+$ was the predominant cation in the rainwaters of southeastern and coastal cities (such as Nanping and Yingtan city). Although Ca$^{2+}$ originated mainly from local sources (Huang et al., 2009), the rare desert dust in southeast coastal cities resulted in a lower Ca$^{2+}$ in their rainwaters.

To assess the neutralization ability of basic cations in rainwater, the neutralization factor (NF) was calculated as follows (Possanzini et al., 1988):

$$\text{NF} = \frac{X}{([\text{nss-SO}_4^{2-}] + [\text{NO}_3^-])},$$

where X represents the [Ca$^{2+}$] and [NH$_4^+$] in μeq/L.

The NF of Ca$^{2+}$ and NH$_4^+$ in different cities of China over the study period is shown in Figure 11. In general, the neutralization capacity of alkaline cations showed a north–south decreasing trend in China. The NF values in southeast and coastal cities were significantly lower than those in China’s other two regions. This observation implies that the weak neutralization in rainwater was due to inadequate alkaline constituents (Huang et al., 2009; Zhou et al., 2019). We attributed the higher NF value to stricter emission controls and faster urban construction in southeast and coastal cities (especially Guangzhou and Shanghai).

Finally, Ca$^{2+}$ was the dominant neutralization cation in most cities, especially in the past few decades. However, with the rapid increase in vehicular use, the influence of NH$_4^+$ in the neutralization reaction has increased gradually, especially in the metropolises. The neutralization capacity of Ca$^{2+}$ and NH$_4^+$ variation in China indicates that economic development has caused a change in the alkaline constituents over the years. Thus, this inference strengthens our understanding of acid rain pollution control.

4. Conclusions

The acidity and chemical constituents of rainwater in China have undergone significant changes since the 1970s. However, rapid economic development and environmental policies have significantly impacted acid rain episodes in China over the decades. From these changes, we conclude as follows:

1. SO$_4^{2-}$ and NO$_3^-$, as the dominant precursors for acid rain, have significantly
decreased in concentration due to mitigation measures. Since these measures have reduced $SO_4^{2-}$ considerably, the type of acid rain in China has changed from sulfuric acid to a mixture of nitric acid and sulfuric acid.

2. The $Ca^{2+}$ in rainwater has decreased in recent years, although not as much as acidic ions.
Figure 9. Volume-weighted mean concentration of basic ions, pH value, neutralizing potential/acidifying potential value in the metropolis in China. DOI: https://doi.org/10.1525/elementa.2021.00142.f9

Figure 10. Volume-weighted mean concentration of basic ions, pH value, neutralizing potential/acidifying potential value in several southeast and coastal cities in China. DOI: https://doi.org/10.1525/elementa.2021.00142.f10
Simultaneously, the $\text{NH}_4^+$ remained relatively stable, with a mild increase observed due to the increasing number of vehicles in China.

3. $\text{Ca}^{2+}$ was the primary neutralizing cation in rainwater, and the neutralization capacity of $\text{NH}_4^+$ in China has gradually increased in
recent years. The neutralization capacity of alkaline cations has decreased from north to south, with the lowest in the southeast and coastal cities, excluding metropolises (due to rapid urbanization).

The review of 3.5 decades of the precipitation pH data from urban regions in China shows an overall trend toward reduced acidity. The further elucidation of these trends is hampered by a relative scarcity of rainwater alkalinity data. Such records are particularly required to evaluate the effects of emission changes from the remarkable growth of industrialization and the number of vehicles in China on the rainwater acidity and ion chemistry.

Data accessibility statement
Data summarized in this review are available online as supporting information.

Supplemental file
The supplemental file for this article can be found as follows:

Table S1. Rainwater data of different cities in China (pH in unit and volume-weighted mean concentration of ion in μeq/L).

Acknowledgment
The authors appreciate our colleagues, Jie Zeng and Kunhua Yang, for providing help with the stimulating discussions.

Funding
This work was supported by the National Natural Science Foundation of China (41661144029; 41325010).

Competing interests
The authors declared no competing interests.

Author contributions
Contributed to conception and design: RQ, GH.
Contributed to the acquisition of data: RQ, GH.
Contributed to analysis and interpretation of data: RQ, GH.
Drafted and/or revised the article: RQ, GH.
Approved the submitted version for publication: RQ, GH.

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