



Article Variations in Sulfur and Nitrogen Oxidation Rates in Summer Aerosols from 2014 to 2020 in Wuhan, China

Jinhui Zhao^{1,*}, Chiyuan Ma^{1,*}, Chao He^{2,3}, Zhouxiang Zhang^{4,*}, Taotao Jiang¹, Rui Tang¹ and Qiang Chen¹

- ¹ Hubei Key Laboratory of Regional Development and Environmental Response, Hubei University, Wuhan 430062, China; 201931108010098@stu.hubu.cn (T.J.); 201822110810072@stu.hubu.cn (R.T.); 201931108010093@stu.hubu.cn (Q.C.)
- ² Hubei Key Laboratory of Petroleum Geochemistry and Environment, Wuhan 430100, China; hechao@yangtzeu.edu.cn
- ³ College of Resources and Environment, Yangtze University, Wuhan 430100, China
- ⁴ Hubei Ecological Environment Monitoring Center Station, Wuhan 430070, China
- * Correspondence: zhaojh2004@hubu.edu.cn (J.Z.); machiyuan52@163.com (C.M.); zhzhoux@126.com (Z.Z.); Tel.: +86-1807-1551-334 (J.Z. & Z.Z.)

Abstract: To date, research regarding the changes of the sulfur and nitrogen rates in Wuhan during the summer is limited. In this study, we analyzed the air quality in Wuhan, China, using water-soluble ion, gaseous precursor, and weather data. A Spearman correlation analysis was then performed to investigate the temporal changes in air quality characteristics and their driving factors to provide a reference for air pollution control in Wuhan. The results indicate that SO₂ in the atmosphere at Wuhan undergoes secondary conversion and photo-oxidation, and the conversion degree of SO₂ is higher than that of NO₂. During the summers of 2016 and 2017, secondary inorganic atmospheric pollution was more severe than during other years. The fewest oxidation days occurred in summer 2020 (11 days), followed by the summers of 2017 and 2014 (25 and 27 days, respectively). During the study period, ion neutralization was the strongest in summer 2015 and the weakest in August 2020. The aerosols in Wuhan were mostly acidic and NH₄⁺ was an important neutralizing component. The neutralization factors of all cations showed little change in 2015. K⁺, Mg²⁺, and Ca²⁺ level changes were the highest in 2017 and 2020. At low temperature, high humidity, and low wind speed conditions, SO₂ and NO₂ were more easily converted into SO₄²⁻ and NO₃⁻.

Keywords: air pollution; secondary oxidation; neutralization factor; meteorological factors; Wuhan

1. Introduction

In recent years, air pollution has become a common concern in China. Controlling air pollution is essential for sustainable development as well as for practicing low carbon environmental protection policies. The atmosphere of Wuhan is rich in SO₂ and NO₂, which causes harm to human health, and SO₂ and NO₂ will combine to form nitric acid, which is an important source of acid rain [1]. During the 2020 outbreak of COVID-19, studies have shown that air pollution can exacerbate the spread of severe acute respiratory syndrome coronavirus 2 (SARS-CoV-2) [2]. Furthermore, SO₂ specifically contributes to the spread of SARS-CoV-2 [3]. Therefore, air pollution problems should not be underestimated. These require our immediate attention for improving the overall air quality.

Previously, in order to study air pollution, many scholars have investigated the characteristics of air quality changes in different regions. For example, Liu [4] studied the characteristics of $PM_{2.5}$ pollution in the Wuhan urban circle. Guo [5] investigated the characteristics of the air quality index and its influencing factors from 2016 to 2019 in an urban agglomeration in the middle reaches of the Yangtze River. Yan [6] analyzed the characteristics of Zhengzhou's four-season sulfur oxidation rate (SOR) and nitrogen oxidation rate (NOR). Lei [7] analyzed the SOR and NOR of Heze City from November 2017



Citation: Zhao, J.; Ma, C.; He, C.; Zhang, Z.; Jiang, T.; Tang, R.; Chen, Q. Variations in Sulfur and Nitrogen Oxidation Rates in Summer Aerosols from 2014 to 2020 in Wuhan, China. *Atmosphere* 2022, *13*, 1199. https:// doi.org/10.3390/atmos13081199

Academic Editor: António Rodrigues Tomé

Received: 28 June 2022 Accepted: 26 July 2022 Published: 29 July 2022

Publisher's Note: MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.



Copyright: © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). to January 2018. Zong [8] analyzed the SOR and NOR of Hengyang City during January, April, July, and September of 2019. Wu [9] investigated the SOR and NOR in Baoding City from November to December 2017. Zhao [10] used a correlation econometric model to analyze the correlation between air quality index (AQI) and pollutants. In this study, we used the Spearman correlation analysis to investigate the correlation between AQI and meteorological factors in Wuhan.

Studies have focused on fine particulate matter in the air but have largely ignored secondary ion oxidation rates. Those studies have used the secondary ion oxidation rate to compare polluted and clean days or to compare heating and non-heating seasons. This study focused on secondary ion oxidation rates and used a longer study period. Herein, summer data during seven years were compared and analyzed. In addition, we conducted a statistical analysis of the neutralization ratio of ions in order to understand the acid-base neutralization degree of atmospheric particles. The correlations between SOR, NOR, and meteorological factors were studied to explore their causal relationships. This study aimed to provide a clearer picture of the state of the atmosphere in Wuhan, such that appropriate measures can be taken to reduce air pollution.

2. Materials and Methods

2.1. Study Area

Wuhan (29°58′–31°22′ N, 113°41′–115°05′ E) covers an area of 8569.2 km² and has a subtropical monsoon climate with high summer temperatures and rain and low winter temperatures and rain. Wuhan is the capital of Hubei Province, located in the eastern part of Hubei Province and middle reaches of Yangtze River. Agricultural land in the city accounts for 564,609 hm², 66% of the total land area. Land under construction accounts for 139,699 hm², 16.3% of the total land area. Unused land accounts for 150,601 hm², 17.6% of the total land area. Wuhan is one of China's most economically developed cities. The seventh national census indicates that the permanent population of Wuhan is approximately 12.4 million people. The number of civil vehicles in 2020 was ~3.8 million. With the development of Wuhan's economy and rise in its population, SO₂ emission has increased. SO_2 has the characteristics of universality, and it seriously endangers the social economy and ecological environment [11]. Therefore, it is urgent to reduce SO₂ emissions. NO_2 in the air can promote O_3 formation, which, in turn, has several adverse environmental effects [12]. Data from Department of Ecology and Environment of Hubei Province indicates that since 2018-2020, SO₂ levels in Wuhan have reached the national ambient air quality level II. Although NO_2 compliance rate is not 100%, the trend is increasing annually. In 2018, the NO₂ compliance rate was 91.8%, while it was 93.7% in 2019 and 96.2% in 2020.

2.2. Datasets

The SO₂, NO₂, and AQI pollutant data used in this study were obtained from the Wuhan Ecological Environment Bureau (http://hbj.wuhan.gov.cn, accessed on 28 December 2020). The national secondary concentration limits of SO₂ and NO₂ are 150 μ g/m³ and 80 μ g/m³, respectively. Ion and meteorological data were obtained from the Hubei Provincial Environmental Monitoring Center station. The monitoring point is located at Hongshan District (30.54° N, 114.37° E) (Figure 1). Barring any missing data in some time periods, the data are quality controlled. The data processing in this paper mainly follows the mean method, which calculates the daily, monthly, and quarterly mean values of the ions of interest.



Figure 1. Map showing the location of the sampling point.

The sampling instrument used for ion data was the Monitor for Aerosols and Gasses in Ambient Air (MARGA) 1S (Metrohm AG, Herisau, Switzerland). It uses a unique sampling device to directly absorb particulate pollutants and soluble gases into the water phase, and then uses an ion analyzer to monitor their components. The process is fully automatic.

2.3. Methods

The time period of ion and gaseous precursor data selected in this study was sampled from 0:00 to 23:00 in June, July, and August 2014–2020. The time resolution of ion data and SO₂ was 1 h, and the time resolution of NO₂ was 1 d. The meteorological data used in this study were sampled from 0:00 to 23:00 in June, July, and August 2015–2020, with a time resolution of 1 h. During the sampling period, the wind speed ranged from 0.5 to 3 m s⁻¹, temperature ranged from 20 to 35 °C, humidity ranged from 50 to 100%, and air pressure ranged from 900 to 1100 hPa.

SOR and NOR are commonly used to express the degree of conversion of primary pollutants SO₂ and NO₂ to secondary ions SO₄^{2–} and NO₃[–] [13]. The calculation formulas for SOR and NOR are as follows:

$$SOR = \frac{SO_4^{2-}}{SO_2 + SO_4^{2-}}$$
(1)

$$NOR = \frac{NO_3^-}{NO_2 + NO_3^-} \tag{2}$$

The above factors refer to the molar concentrations of SO_2 , SO_4^{2-} , NO_2 , and NO_3^{-} . Generally, the value of SOR and NOR is 0.1 and functions as the boundary value for the secondary conversion of SO_2 and NO_2 in the atmosphere [14]. The larger the value, the more serious the secondary inorganic pollution [15]. Hence, the secondary reformer becomes evident [16], and more SO_2 and NO_2 concentrations are converted into SO_4^{2-} and NO_3^{-} [17].

The neutralization ratio (NR) represents the neutralization process between ions and can be used to determine the degree of acid-base neutralization of particulate matter. If

NR < 1, NH_4^+ cannot completely neutralize NO_3^- and SO_4^{2-} . Thus, atmospheric aerosols may be acidic [18]. The calculation formulas for NR is as follows:

$$NR = \frac{NH_4^+}{NO_3^- + SO_4^{2-}}$$
(3)

The concentration reported in the formula are equivalent concentration.

The neutralization factor (NF_{xi}) is used to estimate the acid neutralization capacity of cations, and its calculation formula is as follows:

$$NF_{xi} = \frac{X_i}{NO_3^- + SO_4^{2-}}$$
(4)

In the formula, NF_{xi} is the neutralization factor of cation Xi, Xi is the cation concentration, and $NO_3^-+SO_4^{2-}$ is the sum of the concentrations of NO_3^- and SO_4^{2-} .

The Spearman correlation analysis method was used to analyze the correlation between different elements. The calculation formula for the correlation coefficient is as (5):

$$\mathbf{r} = l_{xy} / \sqrt{l_{xx} l_{yy}} \tag{5}$$

In the formula, r is a unitless value, -1 < r < 1, r > 0 is a positive correlation, and r < 0 is a negative correlation. The meaning of a correlation coefficient of -1 is the maximum value of negative correlation. The closer r is to 1, the stronger the correlation; the closer r to 0, the worse the correlation [19].

3. Results and Discussion

3.1. Analysis of the Time Change Law of SOR and NOR

The air condition in Wuhan is shown in Tables 1 and 2. SOR changed significantly in summer (except for in 2016 and 2017). SOR from 1 July to 19 July in 2015 was close to 1, indicating that almost all SO₂ in the air was converted into sulfuric acid salt during this period [20]. The values in the summer of 2016 and 2017 were generally high, ranging from 0.6 to 1.0 (except 0.4 on 1 July) and 0.7 to 1.0, respectively. This indicated that the secondary inorganic pollution in the atmosphere in the summer of these two years was very serious, the secondary conversion of SO₂ was high, and the conversion of SO₂ from gaseous to particulate matter increased [21]. Cheng also achieved similar results in 2016 and 2017 for SOR in Wuhan City. This is because strong photochemical reactions in summer weather conditions favor the secondary conversion of SO₂ [22].

Table 1. Seasonal SO₂, NO₂, PM_{2.5} and PM₁₀ data (μ g m⁻³).

	2014	2015	2016	2017	2018	2019	2020
SO ₂	5.35 ± 3.87	3.92 ± 3.71	2.39 ± 1.74	1.11 ± 0.97	5.17 ± 3.76	5.71 ± 4.40	2.14 ± 1.97
NO ₂	39.73 ± 11.62	59.91 ± 23.56	42.76 ± 10.17	41.93 ± 11.44	54.67 ± 12.09	36.98 ± 9.16	32.69 ± 7.67
PM _{2.5}	56.72 ± 33.63	39.32 ± 15.44	42.44 ± 17.20	37.32 ± 16.39	40.23 ± 13.27	39.39 ± 13.12	29.80 ± 12.01
PM_{10}	93.61 ± 49.05	77.03 ± 32.72	55.27 ± 18.29	50.30 ± 15.31	47.29 ± 12.27	46.70 ± 12.73	34.24 ± 12.96

Note: Values are expressed as mean \pm standard deviation.

The summers of 2014–2019 (seasonal averages for each year of 0.67, 0.76, 0.82, 0.84, 0.66, and 0.63, respectively), June 2020 (0.74), and July 2020 (0.62) had high SOR while August 2020 (0.27) had low SOR. The conversion of SO_2 to SO_4^{2-} is low in 2020, and the lowest in August. This is the result of the efforts of the state and people to protect the environment. For example, the country popularizes environmental education. Xi Jinping said that the conviction that lucid waters and lush mountains are invaluable assets. We should stick to the path of green and sustainable development. In addition, new energy vehicles have been widely used in 2020.

	2014	2015	2016	2017	2018	2019	2020
K ⁺	0.18 ± 0.14	0.47 ± 0.22	0.36 ± 0.15	0.26 ± 0.14	0.37 ± 0.14	0.33 ± 0.21	0.17 ± 0.11
Ca ²⁺	0.34 ± 0.18	0.14 ± 0.26	0.31 ± 0.16	1.28 ± 0.50	0.27 ± 0.14	0.49 ± 0.18	0.47 ± 0.29
Na ⁺	0.16 ± 0.08	0.13 ± 0.07	1.25 ± 0.10	0.37 ± 0.22	0.17 ± 0.05	0.28 ± 0.17	0.15 ± 0.12
Mg^{2+}	0.02 ± 0.02	0.01 ± 0.02	0.02 ± 0.01	0.10 ± 0.05	0.03 ± 0.01	0.12 ± 0.07	0.13 ± 0.10
CĨ-	1.34 ± 0.88	0.64 ± 0.54	1.05 ± 0.59	0.48 ± 0.20	0.41 ± 0.09	0.36 ± 0.24	0.27 ± 0.17
SO_4^{2-}	10.48 ± 5.41	8.69 ± 7.77	10.26 ± 5.66	6.14 ± 3.68	9.20 ± 3.31	8.20 ± 3.12	4.05 ± 2.62
NO_3^-	3.96 ± 4.09	7.55 ± 6.55	4.96 ± 4.06	3.49 ± 3.64	5.12 ± 2.59	5.30 ± 3.00	2.04 ± 1.82
$\mathrm{NH_4^+}$	5.44 ± 3.12	6.65 ± 3.47	5.98 ± 3.23	3.96 ± 2.93	5.97 ± 2.00	4.27 ± 2.21	2.16 ± 1.36

Table 2. Seasonal ion concentrations ($\mu g m^{-3}$).

Note: Values are expressed as mean \pm standard deviation.

Oxidation in 2014 mainly occurred in June and there were only a few days of oxidation in July and August. The oxidation in 2015 mainly occurred from 16 June to 26 July; oxidation in 2016 mainly occurred from 3 June to 13 June, as well as August. Meanwhile, oxidation in 2017 mainly occurred from 1 June to 20 June. In 2019, oxidation mainly occurred from 2 July to 20 July and from 16 August to 31 August. The number of oxidation days in summer was the lowest in 2020 (11 d), which mainly occurred from June to July. The summer of 2017 had the second lowest oxidation days (25 d) and the summer of 2014 had only 27 oxidation days.

Nitrogen oxidation in 2014, 2015, 2016, 2017, and 2019 mainly occurred in June (0.13), June (0.1) and July (0.17), June (0.12) and August (0.11), June (0.12), and July (0.12) and August (0.12), respectively. From a macro point of view, the degree of nitrogen oxidation in the summer of 2020 was the lowest (June: 0.08, July: 0.06, August: 0.03), followed by that in the summers of 2014 (June: 0.13, July: 0.06, August: 0.07) and 2017 (June: 0.12, July: 0.04, August: 0.04).

NOR from June 3 to June 10 in 2014 was higher than that in other years for the same period. In 2015, NOR in July was the highest of the summer and much higher than that in other years for the same period; NOR in 2014 was 6.3–6.10 and the NO₂ content in the air was relatively low in recent years. Generally, NOR in August was lower than in June and July, indicating that the number of oxidized days in August was less than that in June and July.

When the SOR or NOR is greater than 0.1, it indicates oxidation. In the summers of 2014–2020, SOR was greater than 0.1, indicating that the SO_2 in the atmosphere underwent secondary conversion, and the secondary generation rate of sulfur is relatively high [23]. Lv studied the atmosphere of Shijiazhuang City from September 2017 to February 2018 and found similar results [24]. In addition, studies have shown that this situation indicates that SO_2 undergoes photo-oxidation and the conversion of SO_2 is relatively high [25]. This phenomenon requires attention, as secondary conversion contributes more than primary emissions during heavy air pollution events [26]. In this study, SOR was always greater than NOR in the summers of 2014–2020 (Table 3), indicating that the oxidation degree of SO₂ is much higher than that of NO₂. The degree of conversion of SO₂ to SO₄²⁻ is generally more severe than the conversion degree of NO_2 to NO_3^- . Studies have shown that the SO_4^{2-} concentration is the highest in summer in Wuhan and that the secondary conversion of SO_2 is stronger [27], which reflects the findings of this study. In addition, Zhao [28] research on the atmospheric pollution characteristics of Handan City, Zhang [29] atmospheric research on Beijing's late summer and early autumn, Zhao [30] research on Xiamen's atmosphere, and Zhao [31] research on Zhengzhou's summer atmosphere drew similar conclusions to this study, indicating that other cities also have secondary conversion of SO₂ to NO₂.

	2014	2015	2016	2017	2018	2019	2020
SOR	0.67 ± 0.15	0.76 ± 0.19	0.82 ± 0.1	0.83 ± 0.07	0.66 ± 0.15	0.63 ± 0.16	0.51 ± 0.23
NOR	0.09 ± 0.07	0.11 ± 0.09	0.1 ± 0.06	0.07 ± 0.06	0.09 ± 0.05	0.12 ± 0.06	0.05 ± 0.04
NO_2 oxidation days	27	40	30	25	4	33	11

Table 3. Seasonal sulfur oxidation rate (SOR) and nitrogen oxidation rate (NOR).

Note: Values are expressed as mean \pm standard deviation.

The average NOR greater than 0.1 in the summers of two years (2015 and 2019), indicating that the conversion rate of NO_3^- is low, and conversion conditions are not sufficient in most years to cause serious problems [32].

3.2. Analysis of Interannual Variation of Neutralization Ratio

According to statistical analysis, the neutralization ratios of SNA (SO_4^{2-} , NO_3^{-} , NH_4^+) in June, July, and August of 2014–2020 were all less than 1.5. Furthermore, in August 2015 and June 2020, majority of the days exhibited a neutralization ratio of less than 1, which indicates that NH_4^+ cannot completely neutralize NO_3^- and SO_4^{2-} and must pass other cations to neutralize NO_3^- and SO_4^{2-} .

Based on the change trend chart of the neutralization ratio in the summers of 2014–2020 (Figure 2), the greatest changes were observed in 2015 and 2020. The neutralization ratio increased to 0.8 and 0.7 on 19 and 20 June in 2014, respectively, which is relatively stable. June 2014 and August 2015 had the highest mid-month averages in their summers. The monthly means were 0.43 and 0.79, respectively. The monthly means of the mid-August 2020 ratio are lower than those of June and July of the same year. From a macro point of view, the neutralization ratio in summer 2015 was greater than in other years. The neutralization ratio in August 2020 was the lowest in the study period, as the concentration of NH_4^+ in the air was the lowest at this time.



Figure 2. Changes in the neutralization ratio (NR) during the summers of 2014–2020.

3.3. Analysis of Interannual Variation of Neutralization Factor

After calculation, the neutralization factors of Na⁺, NH₄⁺, K⁺, Mg²⁺, and Ca²⁺ were all less than 1, indicating that the aerosols in Wuhan were mostly acidic. These aerosols can easily aggravate acidification of the atmosphere and the formation of acid rain [33]. Among them, the neutralization factor of NH₄⁺ is the largest, indicating that NH₄⁺ is an important neutralization component.

Based on Figure 3 and data analysis, it can be observed that the neutralization factor of Na⁺ in the summers of 2014, 2015, 2018, and 2019 did not change much, and that the values are generally small (almost all range between 0.0–0.1). The changes in July and August are phased. The watershed moment is 19 July. The range of changes from 1 June to 19 July was

small, ranging from 0 to 0.05. After 19 July, there was a sharp increase above 0.1, and the range of changes from 19 July to 31 August was large. The neutralization factor of Na⁺ in July 2017 was higher than that in June and August. The neutralization factors of Na⁺ on 22 July (0.1) and 7 August (0.01) in 2020 increased to 0.3 from the previous day, while the others did not change much. In June, the difference in the value of the neutralization factor of Na⁺ between 2014 and 2020 was not obvious. However, this difference was significantly higher in July and August 2016 than in other years.



Figure 3. Changes in the neutralization factor (NF_{xi}) during the summers of 2014–2020.

The NH₄⁺ neutralization factor fluctuated little in summer 2015, 2016, and 2017. In 2014, except for a sharp increase on 20 June to 0.8, the remaining days exhibited relatively stable values that fluctuated around 0.4. In August 2019 and 2020, the NH₄⁺ neutralization factor fluctuated. The NH₄⁺ neutralization factor in June 2014 was larger than that in July and August. The NH₄⁺ neutralization factor in August 2020 was smaller than that in June and July. The NH₄⁺ neutralization factor in June and July 2020 was larger than that of other years in the same period, but that in August was smaller than that of other years in the same period.

Except from 17 July to 19 July 2017 and 19 July to 22 July 2020, K⁺ had a neutralization factor greater than 0.1; the remaining days exhibited values between 0.0–0.1. The value of Mg^{2+} neutralization factor in July 2017 was higher than that in June and August, and the value of Mg^{2+} neutralization factor in June and July 2020 was higher than that in August and in other years during the same period. The neutralization factor of Ca^{2+} varied greatly in 2017 and 2020; it was the highest in July 2017, and the values in July and August 2017 were the highest of all years in the same period. Large amounts of Ca^{2+} in the atmosphere could be due to its high concentration in local soil [34]. Only the value on 12 June 2015 increased from 0.02 to 0.16; other years exhibited relatively stable and low values. Combining the above neutralization factors, the neutralization factors of all cations showed little change in 2015. The changes in K⁺, Mg²⁺, and Ca²⁺ were relatively large in 2017 and 2020.

3.4. Correlation Analysis between Meteorological Factors and Pollution Factors

Using Spearman correlation analysis of pollution and meteorological factors for the summers of 2015 to 2020 (Figure 4) we concluded that SOR and humidity were positively correlated, which is similar to Liu's [35] observations in the northern suburbs of Nanning.

The correlation coefficients for the summers of 2015, 2016, 2017, 2018, 2019, and 2020 were 0.37, 0.42, 0.44, 0.27, 0.38, and 0.78, respectively. There is no strong correlation between wind direction and SOR. During the 2015–2020 period (except for 2019), SOR was negatively correlated with temperature, and the correlation coefficients were -0.42, -0.32, -0.42, -0.48, -0.76, for 2015, 2016, 2017, 2018, and 2020, respectively. SOR was also negatively correlated with wind speed (except for 2018) and the correlation coefficients were -0.14, -0.33, -0.23, -0.06, -0.48 for 2015, 2016, 2017, 2019, and 2020. These findings are similar to those obtained in He's research [36]. The correlation being most significant in 2020 indicates that sulfate is easier to convert in an environment of low temperature, high humidity, and low wind speed. SO₂ is more easily converted to SO₄^{2–} in these conditions because the low-temperature environment is not conducive to the volatilization of secondary inorganic ions [37], which is beneficial for the heterogeneous reaction of existing particulate matter in an environment with high relative humidity [38] and the accumulation of pollutants in an environment with low wind speed [39].

The results for NOR are similar to those for SOR. From 2015–2020 (except 2018), there was a positive correlation between NOR and relative humidity, with correlation coefficients of 0.25, 0.28, 0.55, 0.25, and 0.44 for 2015, 2016, 2017, 2019, and 2020, respectively. The correlation with temperature was negative with coefficients of -0.2, -0.34, -0.53, -0.49, and -0.51 for 2015, 2016, 2017, 2018, 2019, and 2020, respectively. The correlation with wind speed was also negative with correlation coefficients of -0.37, -0.4, -0.1, -0.26, -0.15, and -0.22 for 2015, 2016, 2017, 2018, 2019, and 2020, respectively. This indicated that low temperature, high humidity, and low wind speed are ideal conditions for nitrate conversion and for NO₂ to convert to NO₃⁻.

With warming caused by global climate change, the humidity in subtropical regions of the Northern Hemisphere (10–30° N) has decreased by 2–3%. Wuhan is located in the subtropics and has a subtropical monsoon climate that is hot and rainy during the summer, which has an inhibitory effect on the secondary conversion of SO₂ and NO₂.

Except for the insignificant positive correlation between AQI and wind speed in 2020 (correlation coefficient is 0.17), AQI was negatively correlated with both wind speed and humidity. That is, low wind speed and low relative humidity are likely to increase the air pollution index. This is because low wind speed is not conducive to pollutant diffusion and subsequent air purification and low relative humidity are not conducive to pollutant settlement.



Figure 4. Cont.



Figure 4. Spearman correlation analysis results for each year during the study period.

4. Conclusions

The oxidation rate results show that the SO₂ in the atmosphere in Wuhan underwent secondary conversion and photo-oxidation, and the conversion degree of SO₂ was higher than that of NO₂. The conversion rate of NO₃⁻ was low, implying that the conversion conditions were insufficient in most years. The secondary inorganic pollution in the atmosphere in the summers of 2016 and 2017 was very serious, and the secondary conversion of SO₂ was obvious. Accordingly, the conversion of SO₂ mas observed than that in other months. The number of oxidation days in the summer of 2020 was the lowest in recent years (11 days), which was followed by that in the summers of 2017 (25 days) and 2014 (27 days). From a macro point of view, the number of oxidation days in August was less than that in June and July. During the study period, NH₄⁺ in the air of Wuhan could not completely neutralize NO₃⁻ and SO₄²⁻. Therefore, other cations were needed to neutralize NO₃⁻ and SO₄²⁻.

2015 and the weakest in August 2020. The aerosols in Wuhan are mostly acidic, which can easily aggravate acidification of the atmosphere and the formation of acid rain. NH_4^+ is an important component of the neutralization process. Compared with other years, the neutralization factor of all cations showed little change in 2015. In 2017 and 2020, K⁺, Mg²⁺, and Ca²⁺ showed significant change. In this study, under the conditions of low temperature, high humidity, and low wind speed, SO₂ and NO₂ were more easily converted into SO₄^{2–} and NO₃⁻.

Author Contributions: Methodology, J.Z.; provision of data, Z.Z., T.J.; writing—original draft preparation, J.Z. and C.M.; writing—review and editing, C.M., C.H. and J.Z.; supervision: Z.Z., R.T. and Q.C. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the National Natural Science Foundation of China (No. 41401559), the national large-scale innovation project (No. 201910512028), and the school-level large-scale innovation project (No. X202010512077).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Data used in this paper can be obtained from Chiyuan Ma (machiyuan52 @163.com) upon request.

Conflicts of Interest: The authors declare no conflict of interest.

References

- 1. Zhang, J.H.; Yan, X.; Zhang, J. The relationship between air pollution and human health. Shanxi Med. J. 2021, 50, 3339–3341.
- Chen, Z.; Chen, K.Q.; Li, J. Does Air Pollution Affect the Transmission of COVID-19? Evidence from China. *China J. Econ.* 2021, 8, 224–258.
- Shim, S.R.; Kim, H.J.; Hong, M.; Kwon, S.K.; Kim, J.H.; Lee, S.J.; Lee, S.W.; Han, H.W. Effects of meteorological factors and air pollutants on the incidence of COVID-19 in South Korea. *Environ. Res.* 2022, 212, 113392. [CrossRef] [PubMed]
- Liu, Z.H.; Hang, J.W.; Kong, D.Y. The spatial—Temporal characteristics and influencing factors of PM_{2.5} in Wuhan metropolitan area. *Environ. Prot. Sci.* 2019, 45, 51–59.
- 5. Guo, W.W.; Chen, Y.J.; Liu, G.; Song, K.S.; Tao, B.X. Analysis on the characteristics and influencing factors of air quality of urban agglomeration in the middle reaches of the Yangtze River in 2016 to 2019. *Ecol. Environ. Sci.* **2020**, *29*, 2034–2044.
- Yan, G.-X.; Zhang, J.-W.; Lei, H.-J.; Huang, H.-Y.; Tang, M.-S.; Cao, Z.-G.; Li, Y.-B.; Fan, J.; Wang, Y.-S.; Li, H.-G. Seasonal Variation and Source Analysis of Water-soluble Inorganic Ions in Fine Particulate Matter in Zhengzhou. *Huanjing Kexue* 2019, 40, 1545–1552.
- Lei, T.Y.; Zang, Y.; Gao, Y.G.; Li, G.; Wang, W.; Miao, Y.; Ren, L. Chemical characteristics of water-soluble ions of PM_{2.5} in autumn and winter in Heze City. *Res. Environ. Sci.* 2020, 33, 831–840.
- 8. Zong, L. *Pollution Characteristics of Water-Soluble Ion in PM*_{2.5} *in Main Urban Area of Hengyang City;* University of South China: Hengyang, China, 2020.
- Wu, Z.; Sun, S.; Wu, G.; Jiang, J.; Liu, S.; Zhao, W. The pollution pattern and source analysis of water-soluble ions of PM_{2.5} in Baoding City. *Environ. Chem.* 2021, 40, 1421–1430.
- Zhao, D.F. Research on Spatial Characteristics of Air Pollution in Yulin Industrial District Based on Correlation Measurement Model. *Environ. Sci. Manag.* 2021, 46, 119–122.
- 11. Wang, S.; Nie, S.-S.; Feng, Y.-P.; Cui, J.-S.; Chen, J.; Liu, D.-X.; Shi, W.-Y. Spatio-Temporal Evolution Characteristics and Source Apportionment of O₃ and NO₂ in Shijiazhuang. *Huanjing Kexue* **2021**, *42*, 2679–2690.
- 12. Jiang, L.; He, S.X.; Cui, Y.Z. Analysis of sulphur dioxide control in China: An empirical study based on satellite-observed data and spatial econometric models. *Acta Sci. Circumstantiae* **2021**, *41*, 153–1164.
- 13. Wen, T.X.; Wang, Y.S.; Zhang, K. Study on sulfate and sulfur oxidation ratio in PM₁₀ during heating season in Beijing. *J. Grad. Sch. Chin. Acad. Sci.* **2007**, *24*, 584–589.
- 14. Li, P.; Gao, J.Y.; Xiao, Z.M.; Bi, W.K.; Li, Y. Characteristics of water-soluble ions of PM_{2.5} in Tianjin. *Environ. Sci. Manag.* **2021**, *46*, 95–99.
- Zhang, J.Q.; Luo, D.T.; Wang, S.B.; Wang, H.; Hu, W.Z.; Li, H.; Liu, R.Z.; Wang, S.L. Characterization and source analysis of water-soluble ions in PM_{2.5} during autumn in Liaocheng City. *J. Environ. Eng. Technol.* 2021, *11*, 617–623.
- 16. An, X.W.; Ni, S.Y.; Zhao, W.F.; Wang, H.H.; Meng, C.C.; Su, W.K. Analysis of PM_{2.5} water-soluble ionic pollution characteristics in Shijiazhuang based on high resolution MARGA. *Coal Chem. Ind.* **2021**, *44*, 145–151.
- 17. Zhou, J.N. Study on Pollution Status and Causes of Water Soluble Components with Different Particle Sizes in Beijing; Beijing University of Technology: Beijing, China, 2020.
- 18. Feng, J.L.; Hu, X.L.; Guan, J.J.; Zhao, W. Acidity of PM_{2.5} in Shanghai and its correlation with chemical composition of the particles. *J. Shanghai Univ. Nat. Sci.* **2010**, *16*, 541–546.

- 19. Zhang, W.T. SPSS Statistical Analysis Basic Course, 3rd ed.; Higher Education Press: Beijing, China, 2017; pp. 285–295.
- 20. Lin, C.-A.; Chen, Y.-C.; Liu, C.-Y.; Chen, W.-T.; Seinfeld, J.H.; Chou, C.C.-K. Satellite-Derived Correlation of SO₂, NO₂, and Aerosol Optical Depth with Meteorological Conditions over East Asia from 2005 to 2015. *Remote Sens.* **2019**, *11*, 1738. [CrossRef]
- Xue, F.L. Pollution Characteristics of PM_{2.5} and Its Secondary Components in Handan City from 2016 to 2018; Hebei University of Engineering: Handan, China, 2020.
- Cheng, Y.; Wu, J.H.; Bi, X.H.; Yang, J.; Liu, B.; Dai, Q.; Li, P.; Yu, J. Characteristics and source apportionment of water-soluble ions in ambient PM_{2.5} in Wuhan, China. Acta Sci. Circumstantiae 2019, 39, 189–196.
- Qiu, C.C.; Gong, H.X.; Yu, X.N.; Ding, C.; Huo, S.; Zhang, R.; Huo, X. Seasonal characteristics and source apportionment of water-soluble ions in PM_{2.5} of Nanjing Jiangbei New Area. *Acta Sci. Circumstantiae* 2021, 41, 1718–1726.
- 24. Lv, Z. Chemical Characteristics and Source Apportionment of Water-Soluble Ions in PM_{2.5} in Shijiazhuang; East China University of Technology: Nanchang, China, 2019.
- Huang, F.; Zhou, J.; Chen, N.; Li, Y.; Li, K.; Wu, S. Chemical characteristics and source apportionment of PM_{2.5} in Wuhan, China. J. Atmos. Chem. 2019, 76, 245–262. [CrossRef]
- Yang, J.P.; Huang, B.J.; Zhang, M.R.; Wang, Y.J. Analysis of heavy air pollution periods and the secondary transformation of pollutants in Wuhan from 2017 to 2019. J. Green Sci. Technol. 2021, 23, 57–60.
- Zhang, H.-T.; Tian, Y.-Z.; Liu, B.-S.; Yang, J.-M.; Yu, J.; Gong, P.; Wu, J.-H.; Zhang, Y.-F. Spatial Temporal Characteristics and Cluster Analysis of Chemical Components for Ambient PM_{2.5} in Wuhan. *Environ. Sci.* 2019, 40, 4764–4773.
- 28. Zhao, L. The Change of Air Pollution Characteristics in Recent Years in Handan; Hebei University of Engineering: Handan, China, 2018.
- Zhang, K.; Wang, Y.S.; Wen, T.X.; Liu, G.R.; Xu, H. On-line analysis the water soluble chemical of PM_{2.5} in late summer and early autumn in Beijing. *Acta Sci. Circumstantiae* 2007, 4, 459–465.
- Zhao, J.P.; Zhang, F.W.; Xu, Y.; Chen, J.S. Distribution characteristics of water-soluble ions in atmospheric particles with different sizes in coastal city. *Ecol. Environ. Sci.* 2010, 19, 300–306.
- Zhao, Q.-Y.; Jiang, N.; Yan, Q.-S.; Wang, S.; Han, S.-J.; Yang, L.-M.; Zhang, R.-Q. Size Distribution Characteristics of Water-Soluble Inorganic Ions During Summer and Autumn in Zhengzhou. *Huanjing Kexue* 2018, 39, 4866–4875.
- Long, C.K. Variation Characteristics and Source Analysis of Water-Soluble Ions in PM_{2.5} of Nanning City; Guangxi University: Nanning, China, 2020.
- 33. Zhang, T.; Cao, J.J.; Wu, F.; Liu, S.-X.; Zhu, C.-S.; Du, N. Characterization of gases and water soluble ion of PM_{2.5} during spring and summer of 2006 in Xi'an. J. Grad. Sch. Chin. Acad. Sci. 2007, 4, 641–647.
- 34. Aparna, S.; Tripti, P.; Vyoma, S.; Lakhani, A.; Maharaj Kumari, A. Water Soluble Ionic Species in Atmospheric Aerosols: Concentrations and Sources at Agra in the Indo-Gangetic Plain (IGP). *Aerosol Air Qual. Res.* **2013**, *13*, 1877–1889.
- Liu, S.D.; Zhang, L.; Zhang, Y.Y.; Lin, X.; Fan, M.Y.; Zhao, X.; Cao, F.; Zhang, Y.L. Influences of temperature and humidity on formation and evolution of secondary aerosol inorganic ions of PM_{2.5} at Northern Suburban Nanjing. *Ecol. Environ. Sci.* 2018, 27, 714–721.
- He, C.; Hong, S.; Mu, H.; Tu, P.; Yang, L.; Ke, B.; Huang, J. Characteristics and Meteorological Factors of Severe Haze Pollution in China. *Adv. Meteorol.* 2021, 2021, 6680564. [CrossRef]
- 37. Guo, W.; Long, C.; Zhang, Z.; Zheng, N.; Xiao, H.; Xiao, H. Seasonal Control of Water-Soluble Inorganic Ions in PM_{2.5} from Nanning, a Subtropical Monsoon Climate City in Southwestern China. *Atmosphere* **2019**, *11*, 5. [CrossRef]
- Fan, J.; Yue, X.; Jing, Y.; Chen, Q.; Wang, S. Online monitoring of water-soluble ionic composition of PM₁₀ during early summer over Lanzhou City. J. Environ. Sci. 2014, 26, 353–361. [CrossRef]
- 39. Zhou, B.; Yu, L.; Zhong, S.; Bian, X. The spatiotemporal inhomogeneity of pollutant concentrations and its dependence on regional weather conditions in a coastal city of China. *Environ. Monit. Assess.* **2018**, *190*, 261. [CrossRef] [PubMed]