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RESEARCH ARTICLE

THE INFLUENCING FACTORS OF SO_4^{2-} AND NO_3^- IN PM_{10} IN JINHUA, CHINA

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Abstract

Sulfate (SO_4^{2-}) and nitrate (NO_3^-) are the main secondary inorganic components in atmospheric particulates. It is of great significance to understand the formation and evolution of air pollution in the process of air pollution. In this study, samples of PM_{10} were collected in Jinhua City in eastern China from December 2019 to January 2020 and from June 2020 to August 2020, and also analyzed the influence of different meteorological conditions and gaseous pollutants on the formation of SO_4^{2-} and NO_3^- in PM_{10} . The results show that high relative humidity has a significant effect on the increase of sulfur oxidation rate (SOR) in winter, indicating that the winter liquid phase reaction is more conducive to the formation of SO_4^{2-} . SOR and nitrogen oxidation rate (NOR) decrease with increasing temperature in winter, and increase with increasing temperature in summer. The light intensity has an important promoting effect on both SOR and NOR, indicating that the photochemical reaction is beneficial to the formation of SO_4^{2-} and NO_3^- . Both the gaseous precursors SO_2 and NO_2 have significant promoting effects on the formation of SO_4^{2-} and NO_3^- in winter, and the promoting effect of O_3 on NOR is higher than that on SOR, indicating that atmospheric oxidation capacity has a greater effect on the formation of NO_3^- , and the effect of CO on SOR is higher than that on NOR, and there is a negative correlation with SOR.

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Introduction:-

In recent years, with the rapid economic development, the process of urbanization is gradually accelerating, energy consumption is increasing, and the pollutants discharged into the atmosphere are gradually increasing, resulting in the deterioration of atmospheric environmental quality (Gao et al., 2016; Hongbo et al., 2017; Liu et al., 2012; Wang et al., 2017a). Atmospheric particulate matter is considered to be one of the primary pollutants that affect air quality. Atmospheric particulate matter can not only participate in the physical changes of various clouds and fog in the atmosphere, but also continuously absorb and scatter solar short-wave radiation, thereby reduce the atmospheric visibility (Molnar et al., 2020), affect the transmission of atmospheric radiation (Zhu et al., 2018), also reduce the environmental air quality (Chan and Yao, 2008), affect global climate change (Ji et al., 2019), cause great harm to human health, such as respiratory and cardiovascular diseases, and even lead to death in severe cases (Bowe et al., 2020; Daiber et al., 2020; Kim et al., 2020; Liang et al., 2020). The composition of atmospheric particulate matter is very complicated, and its main components include trace elements, carbon-containing components, sulfates, nitrates, ammonium salts, organic matter, etc (Jun et al., 2013; Swap et al., 1992). Among them, sulfate(SO_4^{2-}), nitrate(NO_3^-), and ammonium salt(NH_4^+) are the main secondary inorganic aerosols in atmospheric particulate matter, referred to

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as SNA (Guo et al., 2014; Huang et al., 2021; Wang et al., 2017b). In fine particles, the ratio of SNA to the total mass can reach about 50%, and the mass of SO_4^{2-} and NO_3^- together account for 37~53% of the total mass of fine particles (Li et al., 2020; Pinder et al., 2007), and the ratio of SNA to water-soluble inorganic salts can be Achieved more than 70% (Shen et al., 2007; Wang et al., 2005). The concentration and the change of SNA have an important impact on the dissipation and formation of air pollution (Du et al., 2011).

As an important part of SNA, SO_4^{2-} and NO_3^- are mainly derived from the combustion of fossil fuels such as coal and the formation of NO_x and SO_2 from automobile exhaust emissions which through secondary conversion (Li et al., 2021). It's formation and evolution process is complicated, not only related to the oxidation of the atmosphere, the physicochemical property of particulate matter, and the concentration of gaseous precursors, but also related to meteorological conditions, such as temperature, humidity, and light intensity (Andreae, 2007). Studies have shown that SO_4^{2-} mainly has two generation mechanisms: gas-phase oxidation and liquid phase oxidation. Gas-phase oxidation is mainly the oxidation reaction between gaseous pollutants SO_2 and OH radicals; liquid-phase oxidation is mainly SO_2 generates SO_3 under the action of a catalyst, and then reacts with water on the surface of particles to generate SO_4^{2-} (Wang, 2016). The main formation path of NO_3^- is mainly gas-phase oxidation reaction, and the reaction path is different during the day and night. In the daytime, mainly under light conditions, NO_2 and the OH radicals in the atmosphere are oxidized to form HNO_3 , which further reacts with NH_3 to form NH_4NO_3 , and can also react heterogeneously on the surface of sea salt and other aerosols to form NO_3^- (Alexander et al., 2009; Martins-Costa et al., 2017). Among them, NH_4NO_3 is semi-volatile and easily exists in the form of gaseous HNO_3 at high temperatures (Elminir, 2005; Galindo et al., 2008).

Jinhua is one of the important cities in the Yangtze River Delta region of China. It has rapid economic development and a large population. The increase in various emission sources such as motor vehicle exhaust, industrial exhaust, and construction site dust has caused serious air pollution problems and the factors are more complex. However, there are not many studies on Jinhua's water-soluble ions, and there are a few studies based on the high time resolution. Therefore, this study is based on sampling with a two-hour time resolution to study the effects of different meteorological factors and gaseous pollutants on the formation and evolution of SO_4^{2-} and NO_3^- which are in the main secondary inorganic ions of atmospheric particulate matter in Jinhua City. The purposes of this study are to clarify the source atmospheric particulate matter in urban areas, the law of change, and provide scientific basis for Jinhua's air pollution prevention and control.

Methods:-

Site description and sample collection

The study selected Zhejiang Normal University (119°64'62" E, 29°13'63"N) as the sampling point of atmospheric particulate matter. The sampling point is located in Wucheng District, Jinhua City, surrounded by residential areas and no major industries activity. Using Qingdao Laoying 2050 TSP atmospheric sampler to collect PM_{10} samples with a time resolution of every two hours. The winter sampling date is from December 2019 to January 2020, and the non-rainfall days were sampled for a total of 14 days, and the number of effective PM_{10} samples collected is 163. The summer sampling date is from June 2020 to August 2020, and the non-rainfall days were sampled for a total of 22 days, and the number of effective PM_{10} samples collected is 263. At the same time, the temperature and humidity recorder was used to synchronously record the meteorological data of relative humidity and temperature during the sampling period. The gaseous pollutant data came from the data released by the atmospheric monitoring point of the armed police detachment of Jinhua City. The location of the sampling point is shown in Figure 1.



Fig.1:- Schematic diagram of sampling point location.

Determination of water-soluble ions

The concentration of SO_4^{2-} and NO_3^- in PM_{10} sample was determined by ICS-900 ion chromatograph (Dion, USA). It is based on the separate determination of the ion-exchange liquid chromatography and suppressed conductivity detection mechanism ions technology. The analytical column at different retention times is used to identify the type of ion, the ion concentration is determined based on the peak height and area of the electrical conductivity, to achieve different analytical purposes by changing different analytical column and a detector. First, an appropriate amount cut filter to the sample cuvette was added 10 mL of ultrapure water and extracted ultrasound IH, followed by aqueous 0.22 μmL millipore syringe filter filtration volume. Then using a syringe needle through a 0.22 μm disposable filter is injected into the ion chromatography, then measure the SO_4^{2-} and NO_3^- in the solution.

Conversion rate of SO_4^{2-} and NO_3^-

In order to further study the contribution of secondary conversion to SO_4^{2-} and NO_3^- , this study calculated the degree of secondary conversion of gaseous SO_2 and NO_2 to particulate SO_4^{2-} and NO_3^- , namely sulfur oxidation rate (SOR) and nitrogen oxidation rate (NOR). SOR and NOR have been widely used to study the degree of secondary transformation of atmospheric particulate matter (Sun et al., 2006; Tian et al., 2016). The definitions of SOR and NOR can be described as follows, where n represents molar concentration.

$$\text{SOR} = \frac{n\text{SO}_4^{2-}}{n\text{SO}_4^{2-} + n\text{SO}_2} \quad \text{NOR} = \frac{n\text{NO}_3^-}{n\text{NO}_3^- + n\text{NO}_2}$$

When the value of SOR and NOR is greater than 0.1, it indicates that there is a secondary conversion process in the atmosphere (Ohta and Okita, 1990). The larger the SOR and NOR values are, the higher the probability that gaseous SO_2 and NO_2 will be converted into SO_4^{2-} and NO_3^- , so there are more secondary particles in the atmosphere.

Results And Discussion:-

The influence of relative humidity on the formation of SO_4^{2-} and NO_3^-

The variations of SOR and NOR in winter and summer day and night with the increase of relative humidity (RH) during the sampling periods are shown in Figure 2. It can be seen from the figure that for SOR, when the relative humidity is greater than 60% in winter, the SOR during the day and night shows a significant upward trend with the increase of RH, indicating that the high humidity in winter is more conducive to the formation of SO_4^{2-} from the gaseous precursor SO_2 through the liquid phase chemical reaction. In summer, it is found that with the increase of RH, the change trend of day and night SOR with the increase of RH is not very obvious. It may be because the SOR in summer is dominated by other factors, and the liquid phase reaction does not dominate the formation of SO_4^{2-} in summer. In winter and summer, the SOR is higher when the RH is lower during the day. The reason may be that the lower RH is usually in the period of strong solar radiation. At this time, the photochemical reaction is strong, and the efficiency of SO_2 forming SO_4^{2-} through photochemical reaction high. Comparing the day and night SOR in winter and summer under the same RH, it is found that when the RH is higher (RH>60% in winter; RH>50% in summer),

the SOR at night is usually higher than that in the day under the same RH. It shows that the humid atmosphere at night is more conducive to the conversion of SO_2 to SO_4^{2-} , and the liquid phase reaction makes a greater contribution to the formation of SO_4^{2-} at night.

For NOR, the daytime NOR in winter and summer shows a clear downward trend with the increase of RH, which may be due to the strong solar radiation in the low humidity environment during the day, and the influence of solar radiation on NOR is dominant. However, at night, NOR in winter and summer seasons did not change significantly with the increase of RH, indicating that liquid phase reaction did not significantly promote the pathway of NO_2 to NO_3^- , and NOR may be more affected by other factors. By comparing the daytime and night NOR in winter and summer under the same RH, it is found that the daytime NOR is higher than the night NOR, which may be due to the abundant free radicals in the atmosphere in the daytime, which is more conducive to the conversion of NO_2 to NO_3^- under the light condition.

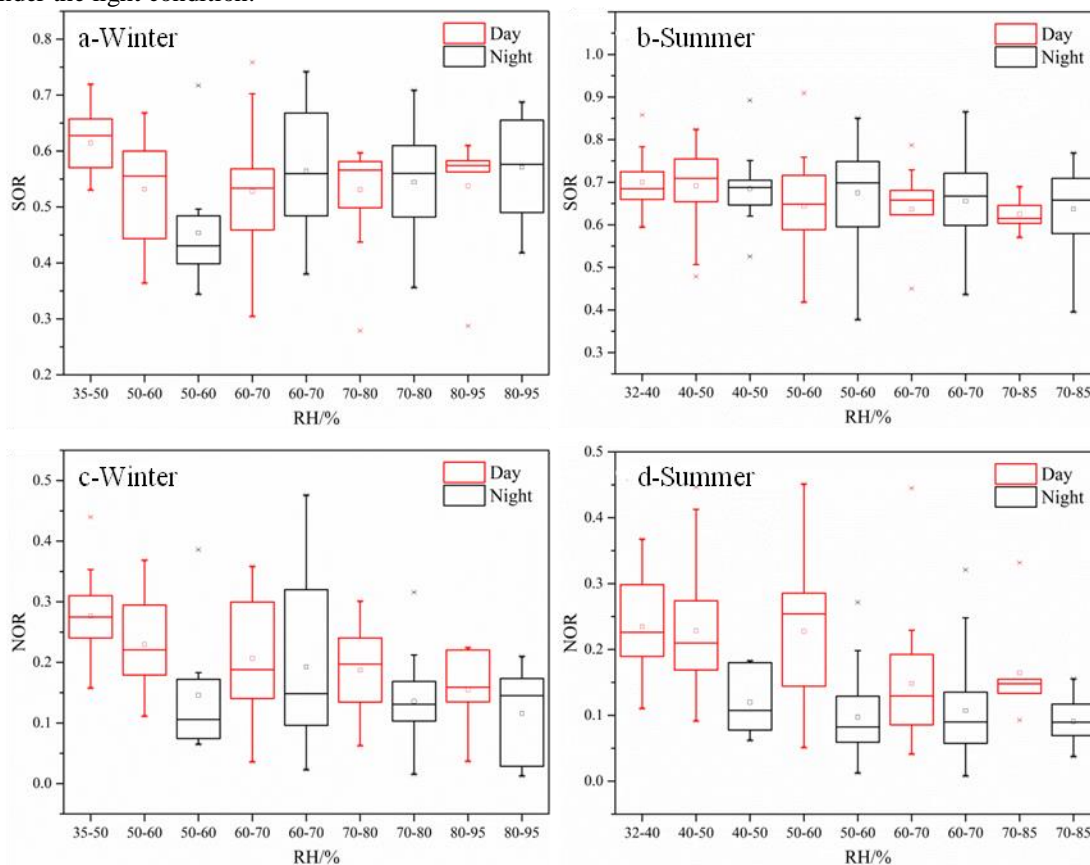


Fig.2:- Changes in SOR and NOR during the winter and summer day and night with increasing relative humidity.

The influence of temperature on the formation of SO_4^{2-} and NO_3^-

The changes of SOR and NOR with increasing temperature during the sampling period are shown in Figure 3. It can be seen from the figure that in winter when the temperature is lower than 25°C , the day and night SOR and NOR will decrease with the increase of temperature, and the value will be higher at low temperature. This may be due to the fact that the atmospheric reversal often occurs during the low temperature period in winter. The temperature phenomenon is conducive to the accumulation of atmospheric pollutants at low altitudes and is not easy to diffuse, so that the NO_2 and SO_2 in the atmosphere can more fully undergo chemical reactions to convert into NO_3^- and SO_4^{2-} . In summer, when the temperature is higher than 25°C , the day and night SOR and NOR generally show a significant upward trend with the increase of temperature. This is because the temperature's rise is conducive to the acceleration of the Brownian motion of atmospheric pollutants, indicating that high temperature in summer is beneficial to the increase of NOR and SOR. At the same temperature, it is found that the value of SOR during the day and night is not much different, but the value of NOR during the day is much higher than that at night, which further shows that the photochemical reaction during the day has a more obvious promotion effect on NOR.

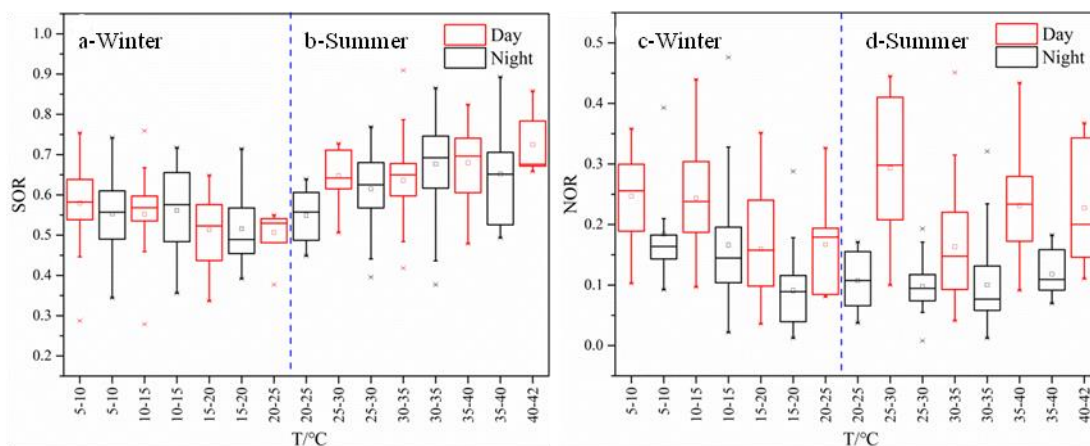


Fig.3:- SOR and NOR changes with temperature.

The influence of light on the formation of SO_4^{2-} and NO_3^-

The changes of SOR and NOR over time in winter and summer during the sampling period are shown in Figure 4. Divide 6 am to 18 pm into the light period, and 18 pm to 6 am of the next day as the non-light period. It can be seen from the figure that for SOR, the SOR in summer has a clear upward trend during the daylight hours from 8 am to 16 pm. Solar radiation will gradually increase during this period, and the photochemical reaction will be strong. In winter, the peak of SOR appears at 8 am. This may be due to the lower atmospheric boundary layer in the winter morning, weaker atmospheric oxidation, and lower solar radiation, so the SOR value is higher in the morning. During the strong period (from 12 pm to 16 pm), there is still a clear upward trend. The reason why the SOR of winter and summer is lower than 16 pm at 18 pm is that the sun at 18 pm is in the sunset period, and the solar radiation is further weakened, which further proves the strong solar radiation is conducive to the conversion of SO_2 to SO_4^{2-} .

For NOR, both winter and summer during the light period, there is an upward trend from 8 am to 14 pm, and the upward trend is more obvious in summer. This is because the solar radiation in summer is stronger than that in winter. The period when the solar radiation is gradually weakened (from 16 pm to 18 pm) also shows a downward trend. It can be seen that the intensity of solar radiation plays an important role in the conversion of NO_2 to NO_3^- . The stronger the solar radiation intensity, the higher the conversion efficiency of NO_2 to NO_3^- .

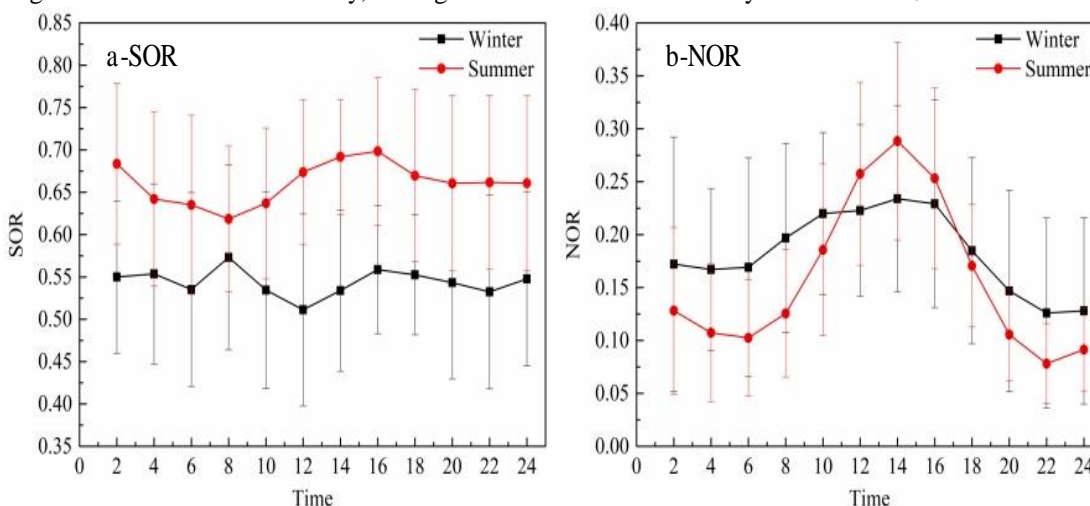


Fig.4:- Changes in SOR and NOR over time in winter and summer.

The influence of SO_2 and NO_2 on the formation of SO_4^{2-} and NO_3^-

SO_2 and NO_2 are important gaseous precursors for the formation of SO_4^{2-} and NO_3^- . The scatter plots of SO_2 and SO_4^{2-} as well as NO_2 and NO_3^- during the sampling periods in winter and summer are shown in Figure 5. It can be seen from the figure that the slope of the scatter plot of the SO_4^{2-} concentration in winter with the increase of SO_2

concentration is 0.3314 higher than the slope in summer (0.141), and the slope of the scatter plot of the NO_3^- concentration in winter with the increase in NO_2 concentration is 0.1304, which is also higher than the slope in summer (0.0339). It can be clearly seen that with the increase in the concentration of gaseous precursors SO_2 and NO_2 in winter, the concentration of SO_4^{2-} and NO_3^- also has a clear upward trend. These may be due to the low temperature in winter, which is prone to inversion effect, which causes SO_2 and NO_2 to gather in the low altitude, and the diffusion capacity of atmospheric is not strong. So that SO_2 and NO_2 can react more continuously to produce SO_4^{2-} and NO_3^- . In summer, due to the higher boundary layer and the strong atmospheric diffusion capacity, it is conducive to the diffusion of SO_2 and NO_2 . Therefore, the slope of the correlation change between SO_2 and SO_4^{2-} as well as NO_2 and NO_3^- in summer is not high.

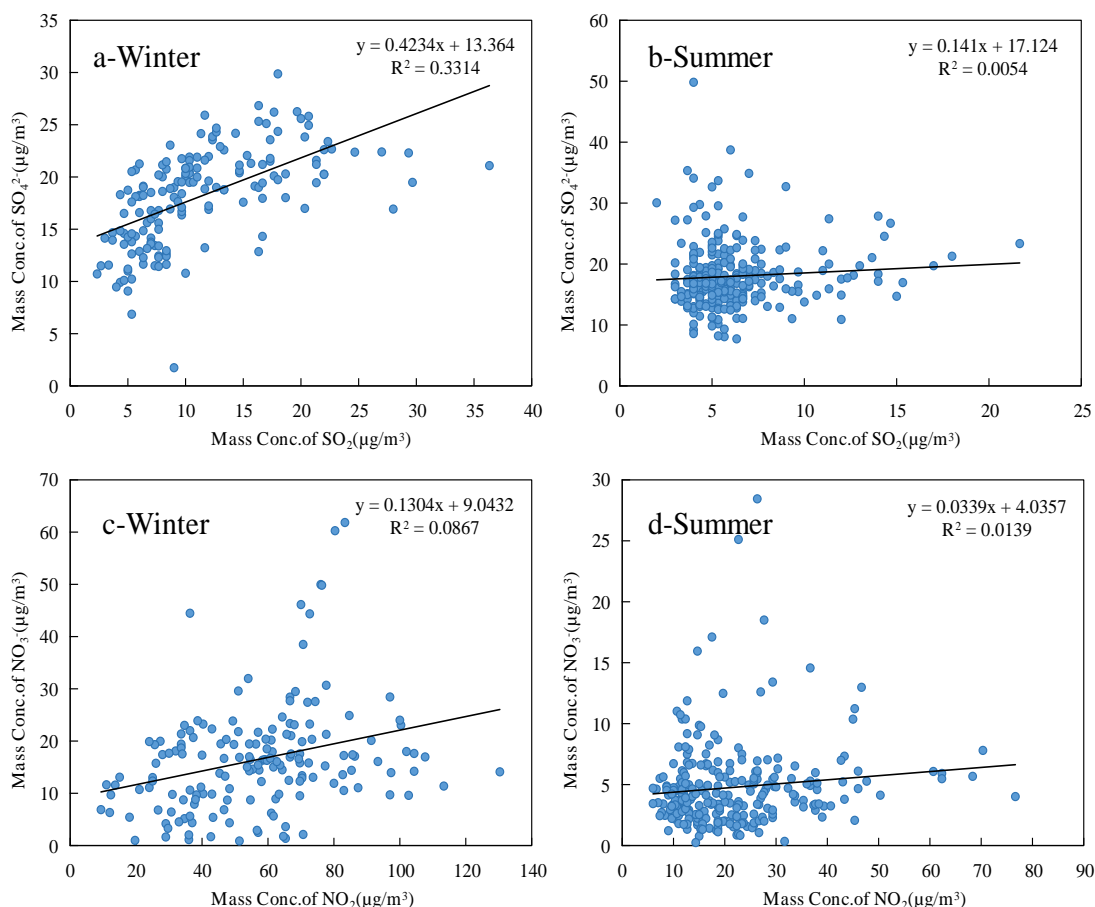


Fig.5 Scatter plots of SO_2 and SO_4^{2-} as well as NO_2 and NO_3^- in winter and summer

The influence of O_3 on the formation of SO_4^{2-} and NO_3^-

The O_3 concentration level in the atmosphere can not only represent the oxidation capacity of the atmosphere, but also can be used as an ideal tracer in the reaction process of atmospheric photochemical reactions (Herndon et al., 2008). The scatter plots of SOR and NOR changes with O_3 concentration in winter and summer during the sampling period are shown in Figure 6. It can be seen from the figure that the slopes of the scatter plots of SOR and O_3 concentration in winter and summer are both positive. Which shows that the increase of O_3 concentration has a certain promoting effect on the conversion of SO_2 to SO_4^{2-} , and its R^2 is lower than the R^2 of NOR with the increase of O_3 concentration. This is because the reaction rate of OH radicals produced by O_3 photochemical reaction with NO_2 is far Higher than the reaction rate with SO_2 (Wang et al., 2013). For NOR, the R^2 (0.4501) of the scatter plot with increasing O_3 concentration in summer is higher than R^2 (0.1812) in winter, indicating that the increase in O_3 concentration in summer has a stronger correlation with the conversion of NO_2 to NO_3^- , and has a greater promotion effect. Obviously, these may be due to the strong solar radiation in summer, and the OH radicals produced by O_3 photochemical reactions are more likely to react with NO_2 in the gas phase to produce NO_3^- .

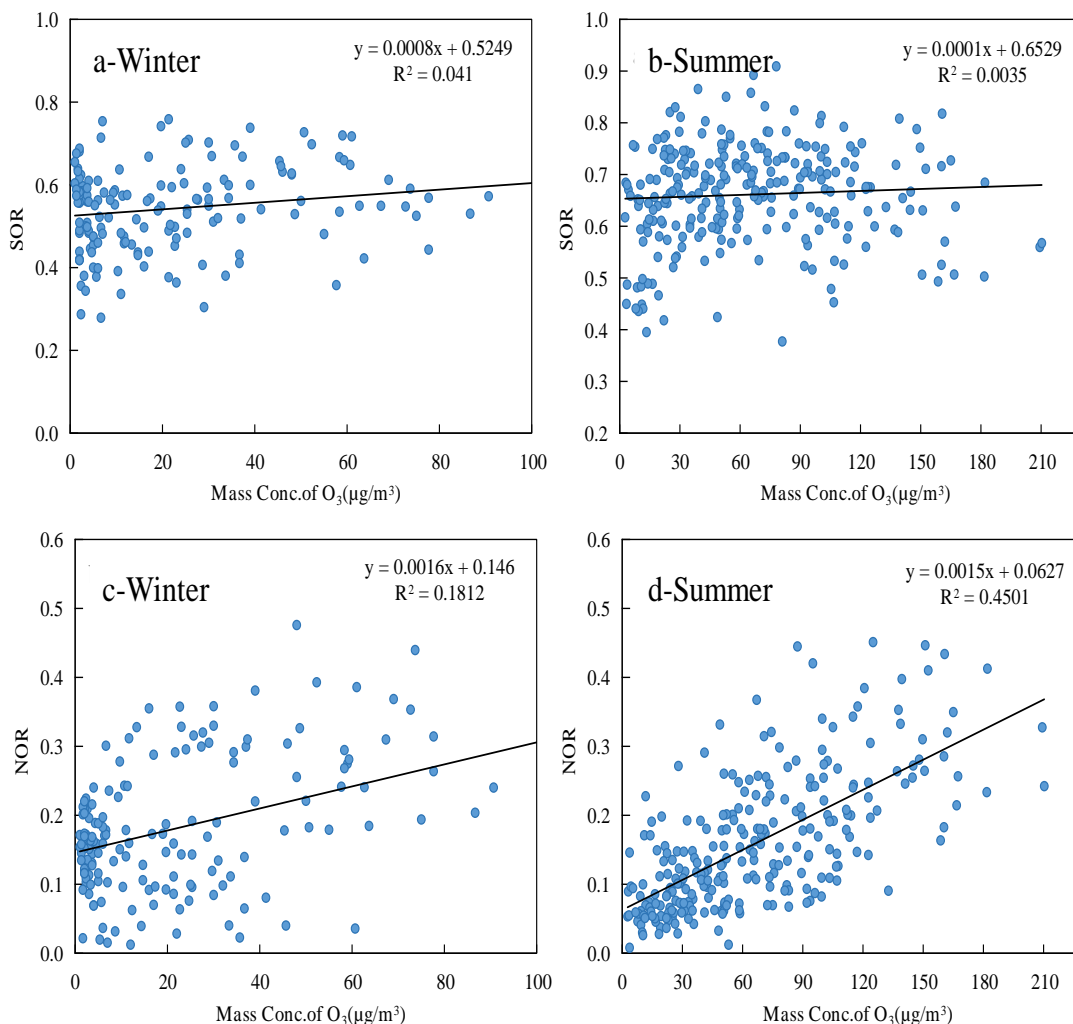


Fig.6 Scatter plot of SOR and NOR changes with O₃ concentration in winter and summer

The influence of CO on the formation of SO₄²⁻ and NO₃⁻

CO is also an important pollutant in the urban atmosphere, and its concentration in the atmosphere is low. Its main source in cities is the emission of car exhaust, and it can participate in photochemical reactions. The scatter plot of SOR and NOR with CO concentration in winter and summer during the sampling period is shown in Figure 7. It can be seen from the figure that the R² of the scatter plot of SOR with CO concentration in winter and summer is significantly higher than that of NOR with NOR. The R² of the scatter diagram of CO concentration changes indicates that the impact of CO on SOR is higher than the impact on NOR, and that SOR has a decreasing trend with the increase of CO concentration. It is speculated that the CO in the atmosphere consumes the OH radicals in the atmosphere, which indirectly affects the conversion efficiency of SO₂ to SO₄²⁻, so the value of SOR is lower at high concentrations of CO. Since the reaction rate of OH radicals and NO₂ is relatively fast, it is possible that the amount of OH radicals consumed by CO has little effect on the reaction between OH radicals and NO₂.

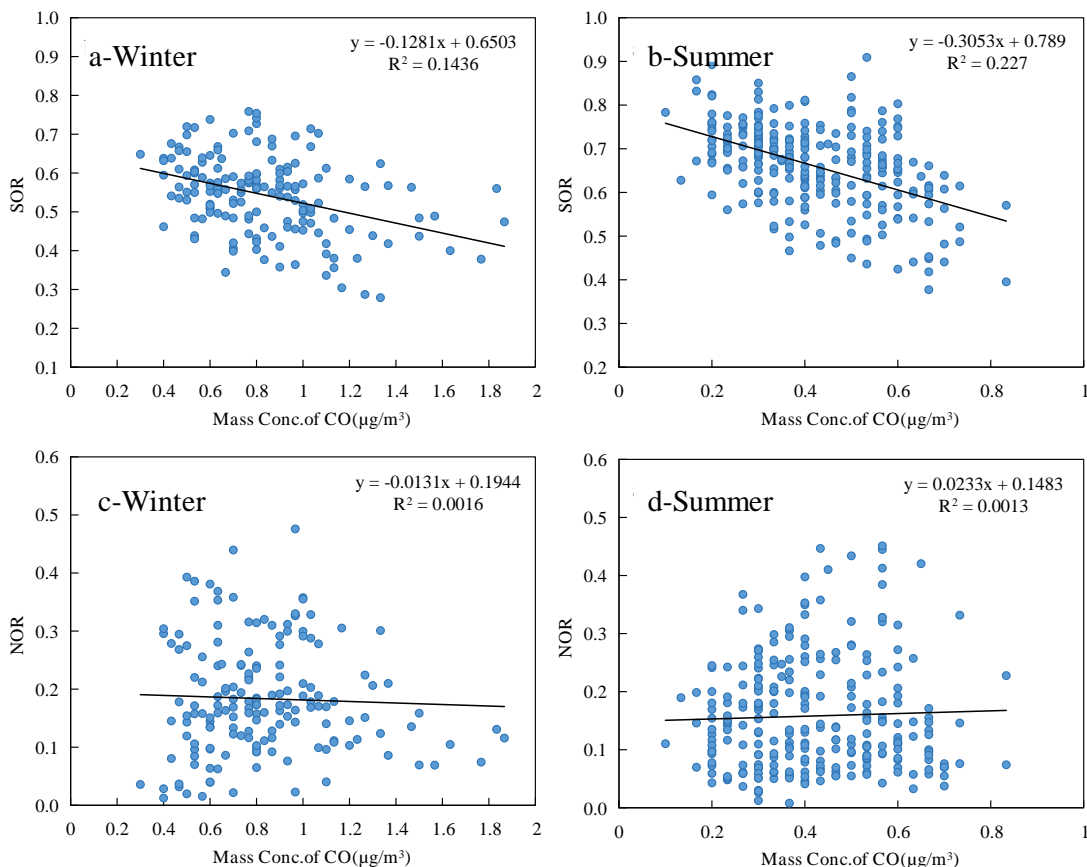


Fig.7:- Scatter plot of SOR and NOR changes with CO concentration in winter and summer.

Multiple linear regression equation

To further explore the influence of meteorological conditions and gaseous precursor emissions on the formation of SO_4^{2-} and NO_3^- , SPSS multiple linear regression was used to analyze the influence of the changes of these factors on the formation of SO_4^{2-} and NO_3^- in winter and summer. The results of multiple regression are shown in Table 1.

Table 1:- SO_4^{2-} and NO_3^- linear regression equations in winter and summer.

Season	ion	Multiple linear regression equation	R
winter	SO_4^{2-}	$\text{SO}_4^{2-} = 17.53 + 1.71\text{CO} - 0.02\text{O}_3 + 0.35\text{SO}_2 + 0.10\text{T} - 0.08\text{RH}$	0.60
	NO_3^-	$\text{NO}_3^- = -3.48 + 5.94\text{CO} + 0.21\text{O}_3 + 0.23\text{NO}_2 - 0.86\text{T} + 0.12\text{RH}$	0.45
summer	SO_4^{2-}	$\text{SO}_4^{2-} = 9.72 - 0.24\text{CO} - 0.02\text{O}_3 + 0.17\text{SO}_2 + 0.27\text{T} - 0.01\text{RH}$	0.19
	NO_3^-	$\text{NO}_3^- = 22.28 + 11.90\text{CO} + 0.04\text{O}_3 - 0.04\text{NO}_2 - 0.53\text{T} - 0.12\text{RH}$	0.70

According to Table 1, it is found that for SO_4^{2-} , the regression coefficients of SO_2 concentration in winter and summer are both positive, indicating that SO_2 as a gaseous precursor to form SO_4^{2-} has an important role in promoting the formation of SO_4^{2-} . O_3 is negative in the regression coefficient of SO_4^{2-} concentration. The previous chapter also shows that the promotion of O_3 is not as good as that of NOR, so it is possible that more OH radicals produced by O_3 participate in the gas phase conversion of NO_2 to NO_3^- Reaction. It is found that the regression coefficients of temperature in SO_4^{2-} concentration are all positive, indicating that high temperature is conducive to the increase of SO_4^{2-} concentration, and the regression coefficients of relative humidity in SO_4^{2-} concentration are all negative values. The previous chapter also discussed the situation of high humidity only in winter. The promotion of the conversion of SO_2 to SO_4^{2-} is obvious. Among them, the winter correlation coefficient ($R=0.60$) of the SO_4^{2-} multiple linear regression equation is greater than the summer correlation coefficient ($R=0.19$), and the SO_4^{2-} multiple linear regression equation has a stronger ability to explain winter data.

For NO_3^- , it is found that the regression coefficient of NO_3^- concentration as a gaseous precursor of NO_2 shows a positive value in winter and a negative value in summer, indicating that the increase of NO_2 concentration in winter has a significant promotion effect on the increase of NO_3^- concentration. It is also found that the regression coefficients of NO_3^- concentration in winter and summer O_3 are both positive, indicating that the stronger the atmospheric oxidation capacity, the more favorable the formation of NO_3^- . CO has a certain similarity with NO_2 in source, so its regression coefficient of NO_3^- concentration is positive. The regression coefficients of the temperature in winter and summer in the NO_3^- concentration are both negative, which further confirms the NO_3^- gas-particle conversion effect, indicating that it is more conducive to the existence of NO_3^- in the form of solid under low temperature conditions. Among them, the correlation coefficient of NO_3^- multiple linear regression equation in summer ($R=0.70$) is greater than that in winter ($R=0.45$), and the NO_3^- multiple linear regression equation has a stronger ability to explain data in summer.

In order to further find the main prominent factors of different meteorological conditions and gaseous pollutants for the formation of SO_4^{2-} and NO_3^- in winter and summer, the associated probability value P, namely Sig, of each factor in each regression equation is further discussed on the basis of multiple linear regression equation. When $\text{Sig}<0.05$, it means that this factor has important significance in the statistics of the model and is the main influencing factor. The statistical results of the associated probability values are shown in Table 2.

Table 2:- The associated probability value P of SO_4^{2-} and NO_3^- linear regression equation.

season	ion	CO	O_3	NO_2	SO_2	T	RH
winter	SO_4^{2-}	0.149	0.378	\	0.000	0.245	0.030
	NO_3^-	0.052	0.001	0.000	\	0.001	0.261
summer	SO_4^{2-}	0.946	0.152	\	0.240	0.099	0.849
	NO_3^-	0.000	0.000	0.040	\	0.000	0.000

According to Table 2, the main influencing factors on the formation of SO_4^{2-} in winter are SO_2 and RH, and the main influencing factors on the formation of NO_3^- are O_3 , NO_2 , and T. The main influencing factors on the formation of SO_4^{2-} in summer are not obvious, and the main influencing factors on the formation of NO_3^- are CO, O_3 , NO_2 , T, RH. It can be seen that meteorological conditions (RH, T) and gaseous pollutants (SO_2 , NO_2 , CO, O_3) play an important role in the formation of SO_4^{2-} and NO_3^- in the secondary inorganic components.

Conclusions:-

Through the above research, we can draw the conclusion that the higher relative humidity has a significant effect on the increase of SOR in winter, but has no obvious effect on the increase of SOR in summer, indicating that the liquid phase reaction in winter is more conducive to the formation of SO_4^{2-} . The increase of relative humidity in winter and summer did not significantly increase the NOR, indicating that the liquid phase reaction did not greatly promote the conversion of NO_2 to NO_3^- . The higher SOR and NOR in winter low temperature period may be caused by atmospheric inversion in winter low temperature period, while the higher SOR and NOR in summer high temperature period may be caused by accelerated Brownian motion between molecules in summer high temperature period. SOR and NOR are both generally higher in winter and summer when the light intensity is strong, indicating that the photochemical reaction has an important role in promoting the formation of SO_4^{2-} and NO_3^- .

The gaseous precursors SO_2 and NO_2 have an important promotion effect on the formation of SO_4^{2-} and NO_3^- , and the direct promotion effect is more obvious in winter than in summer. The correlation between O_3 concentration and NOR in winter and summer is much higher than the correlation between O_3 concentration and SOR, indicating that the atmospheric oxidation capacity has a more significant influence on the conversion of NO_2 to NO_3^- . The correlation between CO concentration and SOR in winter and summer is higher than the correlation between CO concentration and NOR, indicating that CO concentration has a significant impact on the conversion of SO_2 to SO_4^{2-} .

The results of multiple linear regression showed that the main positive effects on the increase of SO_4^{2-} concentration are SO_2 and T, and the negative effects are O_3 and RH. NO_2 and O_3 generally have positive effects on the increase of NO_3^- concentration, while T has certain negative effects.

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