Satellite-Based Analysis of Spatial–Temporal Distributions of NH₃ and Factors of Influence in North China

Deng Zhi-li¹, Zhang Qian-qian²* and Zhang Xing-ying²,3*

¹Chinese Academy of Meteorological Sciences, Beijing, China, ²National Satellite Meteorological Center, Meteorological Administration, Beijing, China, ³FengYun Meteorological Satellite Innovation Center, Meteorological Administration, Beijing, China

NH₃ is an important part of the global nitrogen cycle as the most important atmospheric alkaline gas. NH₃ reacts rapidly with acidic substances and accelerates the generation of particulate matter (PM₂.₅), which has important effects on the atmosphere and climate change. In this study, satellite NH₃ column data were used to analyze spatial and temporal distributions of NH₃ in China, and because of high concentrations and rates of change, North China was selected for more detailed analysis. Qualitative analysis was conducted to understand the relations between concentrations of NH₃ and those of SO₂ and NO₂. Last, the random forest method was used to quantify relations between concentrations of atmospheric NH₃ and factors influencing those concentrations, such as meteorological factors, NH₃ self-emission, and concentrations of SO₂ and NO₂. Satellite-retrieved NH₃ column concentrations showed an increasing trend during the 11 years from 2008 to 2018, and the rate of increase in summer was faster than that in winter. In those 11 years, NH₃ self-emission had the greatest influence on NH₃ concentrations. Concentrations of SO₂ and NO₂ had some effect and were negatively correlated with NH₃ concentrations. The effect of SO₂ on NH₃ concentration was greater than that of NO₂. Atmospheric NH₃ concentration was also affected by meteorological conditions (temperature, relative humidity, pressure, and wind). In summer, temperature is the most important factors of meteorological conditions and relative humidity is the most important factors in winter. Therefore, to better control atmospheric NH₃ concentrations, it is particularly important to formulate practical NH₃ emission reduction policies and to consider the effects of SO₂ and NO₂ emission reduction policies.

Keywords: satellite observations, atmospheric ammonia, random forest model, spatial-temporal distributions, influencing factors

INTRODUCTION

On global and local scales, livestock emissions and extensive fertilizer use are major sources of atmospheric NH₃ (Galloway (2005). As one of the important precursors of reactive nitrogen (Nr) compounds, NH₃ is an important component of the global N cycle (Sutton et al., 2008). Atmospheric NH₃ is the most important alkaline gas and reacts rapidly with sulfuric and nitric acids, and it also accelerates the production of particulate matter (PM₂.₅) discharged into the atmosphere (Malm et al., 2004). NH₃-produced PM₂.₅ leads to 1.3 million premature deaths in China each year. In the
United States, the public health loss caused by NH₃ is estimated at $36 billion per year (Guo et al., 2020). NH₃ also affects the climate, because generated aerosols produce negative radiative forcing that affects the global radiation balance (Charlson et al., 2017). The surface deposition of NH₃ greatly alters the environment by causing problems such as soil acidification, eutrophication of water bodies, and decline in biodiversity. Therefore, it is particularly important to analyze NH₃ temporal and spatial trends and quantify the factors that influence NH₃ concentrations in order to better understand the sedimentation mechanism of NH₃ and formulate relevant emission reduction policies.

NH₃ is a short-lived gas with a life span of only a few hours, and the concentration is generally low; for example, minimum concentrations are less than 1 ppb. These characteristics lead to large differences in temporal and spatial distributions of atmospheric NH₃. NH₃ emissions to the atmosphere are affected by local agricultural activities and weather factors such as temperature, pressure, and humidity (Bouwman et al., 1997). Studies show that SO₂ and NO₂ emissions can also affect atmospheric NH₃ concentrations (Pinder et al., 2008; Yu et al., 2018). Because of these factors, it is difficult for observation methods, such as those ground-based with low spatial resolution, to generate the data required for detailed analysis of NH₃ concentrations. However, with the development of satellite remote sensing technology, satellites can provide long-term high spatial resolution NH₃ concentration data on a global scale. Satellite instruments can fill observation gaps by providing daily global distributions and can be used to quantify spatial-temporal distributions and factors influencing NH₃ concentrations. Observations from the Infrared Atmospheric Sounding Interferometer (IASI) and the Tropospheric Emissions Spectrometer (TES) were used to obtain atmospheric NH₃ concentrations, respectively, and then, spatial and temporal distributions of NH₃ were analyzed (Shephard et al., 2011; Van Damme et al., 2014; Van Damme et al., 2015b; Damme et al., 2020). Satellite data have also been used in combination with ground-based observation and model simulation data to analyze temporal and spatial distribution trends in NH₃ concentrations in Colorado, USA, and NH₃ profiles in different seasons (Li et al., 2016).

At present, the effect of NH₃ concentration is primarily analyzed qualitatively from two aspects: meteorological conditions and SO₂ and NO₂ emissions. Schiferl et al. (2016) used satellite retrieval data and model simulation data to evaluate the NH₃ trend in the United States and concluded that meteorological factors and SO₂ and NO₂ emissions affected atmospheric NH₃ concentrations. Liu et al. (2018) used model simulation to analyze the influence of SO₂ and NO₂ emissions on NH₃ concentrations and concluded that NO₂ emissions had limited effect on increasing NH₃ concentrations. Warner et al. (2017) examined the temporal and spatial distributions of NH₃ and then conducted correlation analyses with several important influencing factors. However, although there are large differences in NH₃ concentrations between cold and warm seasons, previous studies did not consider the influence of different seasons on NH₃. Liu et al. (2017b) accounted for the influence of seasonal factors in seasonal trend analyses of NH₃ and NO₂ concentrations, but specific relations between them were not identified. Amount of NH₃ emissions is another important factor that affects NH₃ concentrations, but amounts are not easy to determine because of the different proportions from different emission sources in different regions. There is great uncertainty in estimated amounts of NH₃ emissions, and there are large errors in the current emission inventory data, with global emission inventory error reaching 50% (Galloway et al., 2008). Local error may be even greater. Thus, it remains very difficult to accurately quantify the effects of different emission sources on NH₃ concentrations. So in this study, Time stamp data and day_of_year data (the day of the year) were used to represent changes in NH₃ emissions between years and during a year separately. In this way, the relationship between NH₃ emissions and NH₃ concentration can be expressed, and the changes of NH₃ emissions can even be indirectly reversed through the observation of NH₃ concentration.

According to (Warner et al., 2017), different factors affect NH₃ concentrations in different regions. Therefore, when analyzing influencing factors, it is beneficial to narrow the study area to draw more accurate conclusions. Thus, in this study, following an analysis that included all of China, North China was selected as the research area. North China was selected because high levels of agricultural activities led to NH₃ concentrations that were higher than those in other regions in China. In addition, because North China has a developed economy, dense population, and abundant NH₃ emission sources, it is particularly important to analyze the key factors affecting NH₃ concentrations.

In this paper, NH₃ concentration data were obtained from the Infrared Atmospheric Sounding Interferometer (IASI), and the spatial distribution trend of NH₃ was analyzed for the 11 years from 2008 to 2018. North China (32°N–42°N, 110°E–120°E) was the focus of analysis because NH₃ concentrations were high and the changing trend was obvious. Time series analysis of NH₃ was also conducted in cold and warm seasons using annual average and monthly average data. On the basis of time series analysis, annual average concentration trends of SO₂ and NO₂ were analyzed in cold and warm seasons separately, and correlations were performed between concentrations of the two gases and those of NH₃. To further understand the influence of different factors, a random forest model was used to quantify the effects of each of the meteorological factors and NH₃ emissions and SO₂ and NO₂ concentrations on NH₃ concentrations in different seasons.

DATA

Satellite Data

NH₃ Column Concentration

The IASI is an infrared Fourier transform spectrometer that observes the infrared radiation emitted by the atmosphere and the surface in the wave number range of 645 to 2,760 cm⁻¹ without gaps, with a spectral resolution of 0.5 cm⁻¹, and 8,461 total channels. Overpass times are 0930 and 2130 mean local solar time. Radiometric resolution is 0.1–0.5 k, and the spectral noise is...
are mole/cm². In spatial
according to
wind, these two parameters can be combined to get the speed and
which are
network algorithm (Dammers et al., 2017). In this paper, the data
cloud coverage less than 30% were selected for analysis. The cloud amount was screened, and the points with
time seasons from 2008 to 2018 were divided into cold (January to March and
and warm (April to September) seasons for analysis. The cloud amount was screened, and the points with
cloud coverage less than 30% were selected for analysis.

SO₂ and NO₂ Column Concentrations
Level-3 OMSO2e planetary boundary layer volume column density (VCD, unit is DU) data obtained from the Ozone Monitoring Instrument (OMI) were used to represent the SO₂ data. Data obtained were also from 2008 to 2018 and were used to analyze cold and warm seasons separately. The L3 data were obtained by excellent pixel screening of OMI L2OMSO2 data, which excluded the influence of clouds, observation angles, and ground conditions. The accuracy is higher than L2 data, and the spatial resolution is 0.25° × 0.25°.

The NO₂ data were obtained from the OMI L3 OMNO2d data, which are filtered by the cloud coverage rate of 30%, and the units are mole/cm². In spatial–temporal analyses, the daily data were processed into annual average data in cold and warm seasons separately, as NH₃ data.

Meteorological Data
Meteorological data were from the ERA5 land hourly data of the European Centre for Medium-Range Weather Forecasts. The dataset included data from 1981 to the present. The combination of model simulation data and observational data provides more accurate reanalysis data. Data at 0200 and 1400 UTC were selected as the meteorological data based on local overpass times of METOP at 0930 and 2130 and had a spatial resolution of 0.1° × 0.1°. Temperature, relative humidity, pressure, wind_u and wind_v were selected as the influencing factors of NH₃ concentration. Wind_u represents the eastward component of the wind, and wind_v represents the northward component of the wind, these two parameters can be combined to get the speed and direction of the horizontal wind.

METHODS
Time Series Analysis
Annual average and 7-days sliding average data were used to analyze the trend in NH₃ concentrations for 11 years in cold and warm seasons. Because the trend in NH₃ concentration was nonlinear, the annual average change rate in NH₃ concentration r was defined according to Equation 1. The equation accounts for the year-to-year change in NH₃ concentration (Liu et al., 2017a). Considering rates of change in NH₃ concentration before and after 2011, the 11-years dataset was divided into two parts.

\[ r = \sum_{i=2008}^{i=2017} \frac{Y_{i+1} - Y_i}{Y_i} \] (1)

Random Forest Method
The random forest method is an ensemble learning algorithm composed of many classifications. In the 1980s, Breiman invented the classification tree algorithm, and by repeatedly dichotomizing data for classification or regression, the amount of calculation is greatly reduced. Decision trees can be an efficient means to obtain nonlinear relations between variables. In 2001, classification trees were combined into a random forest (Breiman, 2001). In this process, variables (columns) and data (rows) are used randomly to generate many classification trees that are then aggregated. The random forest method is simple and fast with high prediction accuracy and is currently widely used in environmental studies to analyze factors influencing meteorological data (Wang et al., 2020; Zhang et al., 2020). A random forest model was also used with special conditions to quantify the effects of anthropogenic emissions on NH₃ concentrations during the epidemic (He et al., 2021).

Time stamp data were used to represent changes in NH₃ emissions between years, and day_of_year data were used to represent changes in NH₃ emissions during a year. Meteorological elements (temperature, pressure, wind speed, wind direction and relative humidity) and SO₂ and NO₂ concentrations were selected as factors influencing NH₃ concentrations. These influencing factors combined with NH₃ concentrations formed a dataset. Data for the 11 years were first divided into cold and warm seasons and then processed using a 7-day sliding average. Data were then divided into training and validation sets randomly according to the ratio of seven to three. The training set as the input of the random-forest model was used to train the random forest model, and the validation set was used to test the accuracy. R² was calculated to indicate the significance of the model. For verification, the model qualification standard was 0.85. Only if R² is greater than 0.85, the trained model can be used for the prediction of NH₃ concentration. Finally, the trained model can be used to simulate and predict NH₃ concentrations in any artificially settings.

After the model was established, data on influencing factors could be changed and NH₃ concentrations predicted in the simulated environment. The 2008 data were used as the baseline, assuming that each kind of meteorological data, SO₂ and NO₂ concentrations and emission data were always in the state of 2008, respectively. Different meteorological data were also discussed separately. Other data were not processed, and then, the NH₃ concentration was predicted in the changed conditions. The purpose of ensuring that data remained unchanged in 2008 was to verify the influence of various factors on inter-annual changes, while ignoring the changes caused by changes during a year. Predicted results were compared with NH₃ concentration data predicted by real data in order to eliminate systematic errors in the random forest model prediction process to the greatest extent. Differences represented the changes in NH₃.
concentration caused by fixing various influencing factors. Eq. 2 defines the variable \(\text{contribute}\), which represents the influence of changes in various influencing factors on NH\(_3\) concentration. The value of \(\text{contribute}\) is equal to the predicted rate of change (PRC) divided by the true rate of change (TRC). With fixed meteorological data, results represented the effects of meteorological conditions. With fixed unix data, results represented the effect of NH\(_3\) emission, and with fixed SO\(_2\) and NO\(_2\) data, results represented the effects of SO\(_2\) and NO\(_2\). Because the calculated influences of the various influencing factors are relative values, it is necessary in a final step to normalize the percentages of calculated influences on NH\(_3\) concentrations. Thus, the sum of influencing factors contributions is 1.

\[
\text{contribute} = \sum_{i=2008}^{i=2018} \left( \frac{\text{PRC}_i}{\text{TRC}_i} \right) \frac{1}{n}
\]  

(2)

**RESULTS AND DISCUSSION**

**NH\(_3\) Trend in North China**

Annual average values from 2008 to 2018 in each grid were used to obtain Figure 1, which shows the spatial distribution of NH\(_3\) concentration in mole/cm\(^2\) in the east of China during cold and warm seasons. Figure 2 is a map of the rate of change in NH\(_3\) volume mixing ratio computed using linear regression in each 0.25° × 0.25° latitude–longitude grid cell, and the point with significant linear trend was marked with a red cross. NH\(_3\) concentration (Figure 1) and rate of increase in NH\(_3\) concentration (Figure 2) were higher in North China than in other regions. These results are consistent with high NH\(_3\) emissions due to intensive fertilizer use and livestock activity in North China (Huang et al., 2012). The eastern part of Sichuan and central parts of Shaanxi and Hubei were also areas with highest NH\(_3\) concentrations as well as high rates of change, which might be primarily associated with NH\(_3\) emissions from rice cultivation. There was also a large area of relatively high NH\(_3\) concentration in Xinjiang, which was likely associated with management of sheep manure (Huang et al., 2012). Some other regions of China also had sporadic high NH\(_3\) concentrations, although the rate of change in NH\(_3\) concentration was low. The Qinghai–Tibet Plateau had the lowest NH\(_3\) concentrations, which also has a small amount of arable lands and low nitrogen fertilizer use. NH\(_3\) concentrations and rates of increase were generally higher in the warm season than in the cold season, and compared with other regions in China, the contrast between cold and warm seasons was most obvious in North China. Maximum NH\(_3\) concentration in
summer during the 11 years was 4.5 e^{16} mole/cm^2, whereas the maximum in winter during the same period was 3.5 e^{16} mole/cm^2. Both maximums were in North China. The maximum change rate in NH₃ concentration in summer was 3.6 e^{15} mole/cm^2 yr, whereas the maximum change rate in winter was 3.2 e^{15} mole/cm^2 yr. Both maximums also occurred in North China. High concentrations and rates of change occurred primarily in North China because of the developed economy, dense population, and many types of NH₃ emission sources, especially large amounts of agricultural emissions. Therefore, because of high NH₃ concentrations, high change rates, and obvious differences between cold and warm seasons, the North China region was selected as the main area of research focus. Time series analysis of NH₃ concentrations was performed, and on that basis, qualitative and quantitative analyses of NH₃ concentrations were conducted.

In the region of North China, It can easily find that the NH₃ concentration (Figure 1) presents a decreasing trend distribution from the center to the surroundings. The high-value center is located at the junction of Hebei, Henan, and Shandong province. No matter in both cold and warm seasons, maximum NH₃ concentration all occur in Henan province, The NH₃ concentration in warm season is higher than that in cold season. The change rate of NH₃ concentration (Figure 2) has a similar distribution characteristics. It can easily find that the NH₃ concentration has increased significantly. However, the linear trend of growth is not significant, especially in warm season. Only a few points (red cross) have a p-value greater than 0.85. So we have defined a new way (Eq. 1) to express the rate of change in time series analysis. The unit of this is not mole/cm^2 yr any more, the unit should be “%”.

**NH₃ Trends and Their Driving Mechanisms in North China**

Figure 3A shows the yearly mean IASI NH₃ Volume Mixing Ratio (VMRs) in North China from 2008 to 2018 separately for warm and cold seasons. Pink and gray solid lines represent 7-days moving average data to show inter-annual changes in NH₃ in warm and cold seasons, respectively. Seasonal changes were apparent, and especially in the warm season, there were greater fluctuations in NH₃ concentrations. The maximum NH₃ concentrations occurred in June and July each year, which might be related to the meteorological conditions during those months. The orange solid line represents annual average data indicating the trend in NH₃ concentration in the warm season, whereas the blue solid line represents the trend in the cold season. NH₃ concentrations and rates of increase in summer were significantly higher than those in winter. The change rate over 11 years in summer was 106.7% (change_warm), whereas that in winter was approximately half of that in summer, at 54.7% (change_cold). After 2011, the rate of increase in NH₃ concentration accelerated, especially in summer, with the increase almost exponential. Before 2011, the NH₃ concentration in the warm season increased by 5.38% (r_warm08-11) per year, whereas there was a slight downward
trend in the cold season, with a change rate of $-0.59\%$ ($r_{\text{cold08-11}}$) per year. After 2011, NH$_3$ concentration increased by 8.93\% ($r_{\text{warm11-18}}$) per year in the warm season and by 6.9\% ($r_{\text{cold11-18}}$) per year in the cold season, the trend of ammonia is similar to other research (Liu et al., 2017a; Warner et al., 2017). As an important atmospheric alkaline gas, NH$_3$ reacts with atmospheric SO$_2$ and NO$_2$, and therefore, increases in NH$_3$ concentration may be related to decreases in SO$_2$ and NO$_2$ concentrations. An inflection point in the trend was apparent, which might be related to policies limiting SO$_2$ and NO$_2$ emissions promulgated around 2011. Therefore, the trends in atmospheric SO$_2$ and NO$_2$ concentrations were analyzed further.

Trend analysis was performed on SO$_2$ and NO$_2$ concentrations, and yearly mean VMRs of SO$_2$ (Figure 3B) and NO$_2$ (Figure 3C) were plotted under the same space–time conditions. Overall, the trends in SO$_2$ and NO$_2$ concentrations were downward, in contrast to the trend in NH$_3$ concentrations. Both gases also exhibited inflection points in 2011. During the 11 years, SO$_2$ concentrations decreased by 45.8\% in the warm season and by 36.1\% in the cold season, whereas NO$_2$ concentrations decreased by 11.2\% in the warm season and by 5.56\% in the cold season. Thus, the decreases were greater in the warm season than in the cold season. However, SO$_2$ and NO$_2$ concentrations in the cold season were higher than those in the warm season, which was the opposite of lower NH$_3$ concentrations in winter than in summer. In addition, before the inflection point in 2011, there was a trend of increase in NO$_2$ concentration. Before 2011, the rate of increase in NO$_2$ concentration in cold and warm seasons was 4.69 and 5.17\%, respectively, and the changing trends in SO$_2$ and NO$_2$ concentrations were not consistent. After 2011, the trend in NO$_2$ concentrations was downward, with rates of decline in cold and warm seasons of $-2.53\%$ and $-3.72\%$, respectively. The concentration of SO$_2$ also showed a downward trend.

China’s pollution control regulations are issued in 5-year increments, and during the 11th Chinese Five Year Plan (FYP) period (2006–2010); the Chinese government implemented a series of strategies to reduce SO$_2$ emissions. During the 12th Chinese FYP period (2011–2015) more stringent strategies were implemented to control both SO$_2$ and NO$_2$ (China Statistical Yearbook, http://www.stats.gov.cn/). Therefore, SO$_2$ concentrations always showed a trend of decrease, whereas NO$_2$ concentrations only started to decrease after 2011. The vertical dashed line drawn from 2011 point on the x-axis intersects the trend line at two points and they were marked on the chart. This decrease may be one reason for the increased rate of increase in NH$_3$ concentration after 2011. In the two time periods before and after the inflection point in 2011, there were no significant differences in the rates of increase and decrease in SO$_2$ and NO$_2$ in cold and warm seasons. In the absence of differences, it is difficult to explain why NH$_3$ increased significantly faster in summer than in winter after 2012. Thus, further quantitative analysis and consideration of the influence of meteorological conditions on NH$_3$ concentrations are needed.

Factors Influencing Concentrations of NH$_3$

Based on qualitative analysis, the random forest algorithm was used to quantify the effects of influencing factors on atmospheric NH$_3$ concentrations in cold and warm seasons. Those factors included meteorological factors, NH$_3$ emissions, and concentrations of SO$_2$ and NO$_2$. In Figures 4, 5, blue columns represent annual average NH$_3$ concentration under real conditions, and the NH$_3$ trend was consistent with that in the time series analysis. Red bars represent the predicted NH$_3$ concentration when an influencing factor was changed and fixed at the state of 2008. The difference between the two represents the effect of a particular influencing factor on NH$_3$ concentrations. Dotted and dashed lines in Figures below indicate trends in NH$_3$ concentration under actual and predicted scenarios, respectively. Because linear trends in NH$_3$ concentrations were not obvious, difference in slope was not a good representation of the effect of an influencing factor on NH$_3$ concentration. The contribute shown in Figures was calculated according to Eq. 2. The normalized results shown in the Table 1 can represent the relative influence of each influencing factors on NH$_3$ concentration. Figure 4 presents the result of fixing different meteorological factors. Figure 5 present the result of fixing other factors expect for meteorological.

NH$_3$ emissions had the greatest effect on NH$_3$ concentrations in both cold and warm seasons, contributing 49.85 and 49.69\% to the NH$_3$ concentration in cold and warm seasons, respectively. Atmospheric NH$_3$ was also influenced by meteorological conditions (temperature, pressure, and wind). In warm season, the most important factor is temperature, it can lead to a change of NH$_3$ concentration of 10.02%, however, in cold season, it only cause 6.73\% change of NH$_3$ concentration. In cold season, the most important factor is relative humidity. It can cause 13.28\% change of NH$_3$ concentration, and in warm season, it only change 5.28\%. There is no obvious contrast between other factors in the cold and warm seasons. Concentrations of SO$_2$ and NO$_2$ also affect NH$_3$ concentration, to some extent. Their concentrations were negatively correlated with those of NH$_3$, and the effect of SO$_2$ on NH$_3$ was greater than that of NO$_2$. In this study, concentrations of SO$_2$ caused a change in NH$_3$ concentration of 19.65\% in the warm season and 11.91\% in the cold season. Concentrations of NO$_2$ caused a change in NH$_3$ concentration of 14.23\% in the warm season and 5.72\% in the cold season, (Liu et al., 2018) shows that SO$_2$ has a greater impact on NH$_3$ concentration than NO$_2$ when the emission of NH$_3$ is small, which maybe related to ozone photochemical reaction. Besides, summer has greater influence than other seasons. Compared with the result of this study, without the limitation of NH$_3$ emission, this study also gets similar results. Yu et al. (2018) only considered the impact of acid gases on NH$_3$ concentration, and it got a conclusion that SO$_2$ affects two thirds of the NH$_3$ concentration and NO$_2$ affects one third. Those results could explain why before 2011, the overall trend in NH$_3$ concentration depended on the trend in SO$_2$ and was opposite to that trend. The results could also explain why the change rate in NH$_3$ before 2011 was lower than that after 2011.
FIGURE 4 | Results of random forest models on meteorological factors that influence NH3 concentrations in North China. Blue columns represent annual average NH3 concentration under real conditions. Red bars represent the predicted NH3 concentration when an influencing factor was changed and fixed at the state of 2008. Dotted and dashed lines represent indicate trends in NH3 concentration under actual and predicted scenarios, respectively.
What's more, the effect of these two gases is greater in the warm season than in the cold season. This may be related to the rapid rate of chemical reaction caused by the high temperature in summer. Schiferl et al. (2016) used Geos-Chem model to analyze the impact of ammonia in the U.S., the results show that meteorological factors have a greater impact on NH₃ concentration than acid gases, however, NH₃ emission have little effect. It is different from this article, which maybe caused by regional differences (Warner et al., 2017). Warner et al. (2017) carried out key analysis on several places with high NH₃ concentration. It indicated that over the U.S., the increase results from a combination of acid gases and temperatures. Over China, acid gases, temperatures, and fertilizer use all play a role.

The random forest results in Figure 4 show the relative magnitude of the effect of each influencing factor on NH₃ concentration. The contribution of each influencing factor to NH₃ concentration was determined separately in cold and warm seasons, because there was no mutual relation between

| TABLE 1 | The degree of influence of acid gas emissions, NH₃ emissions and meteorological conditions on NH₃ concentration after normalized. |
|-----------------------------------------------|-----------------|-----------------|
| Influencing factors                           | Cold (%)        | Warm (%)        |
| NH₃ emissions                                 | 49.85           | 49.69           |
| SO₂ concentration                             | 11.91           | 19.65           |
| NO₂ concentration                             | 5.72            | 14.23           |
| Meteorological conditions                     |                 |                 |
| Temperature                                   | 6.73            | 10.02           |
| Pressure                                      | 8.87            | 6.26            |
| Wind_u                                        | 6.28            | 2.42            |
| Wind_v                                        | 9.27            | 2.48            |
| Relative-humidity                             | 13.28           | 5.28            |
the two seasons. Whether cold or warm season, the most important factor influencing atmospheric NH$_3$ concentrations was the emission of NH$_3$. After 2011, when there were no significant differences in the rates of change in SO$_2$ and NO$_2$ concentrations, the rate of increase in NH$_3$ concentration was clearly higher than that in winter, indicating temperature might be an important factor. In addition, NH$_3$ emissions in the cold and warm seasons will affect half part of the NH$_3$ concentration, playing a decisive role in NH$_3$ concentration. However, there are many types of NH$_3$ emissions, and the types of emission sources in cold and warm seasons are not the same, so it was difficult to establish the emission inventory, and the effects of different emission sources on NH$_3$ concentrations were not easy to determine. Emissions of NH$_3$ had the greatest effect on NH$_3$ concentrations. Therefore, a more detailed understanding of the effects of different emission sources on atmospheric NH$_3$ is necessary to increase understanding of changes in NH$_3$ concentrations and help formulate relevant emission reduction policies.

**CONCLUSION**

In this paper, the spatial distribution of NH$_3$ concentration in China was analyzed, and then, North China was selected as the area of focus because of its high and clearly increasing NH$_3$ concentrations. In addition, there were clear differences in NH$_3$ concentrations between cold and warm seasons. Temporal distributions of atmospheric NH$_3$ concentrations and the influencing factors were analyzed, which included NH$_3$ emissions, meteorological conditions (temperature, pressure, and wind), and SO$_2$ and NO$_2$ concentrations. The conclusions of the study were the following:

1. Within China, NH$_3$ concentrations in North China were generally higher than those in surrounding areas, and the rate of change was rapid. The eastern part of Sichuan and central parts of Shanxi and Hubei were also areas with maximum NH$_3$ concentrations. Sporadic high NH$_3$ concentrations were also detected in other regions of China. Overall, NH$_3$ concentration and rate of change in concentration were higher in summer than in winter.
2. Atmospheric NH$_3$ concentrations increased year by year, and the change rate was faster in summer than in winter. The change rate in NH$_3$ concentrations was related to the concentrations of SO$_2$ and NO$_2$.
3. NH$_3$ emissions had the greatest effect on atmospheric NH$_3$ concentrations, it will determine the change in NH$_3$ concentration by half. In the cold season, emissions changed average NH$_3$ concentration by 49.85% every year. In the warm season, the change was 49.69%. Concentrations of SO$_2$ and NO$_2$ affected NH$_3$ concentration to a certain extent, and their concentrations were negatively correlated with those of NH$_3$. The effect of SO$_2$ on NH$_3$ was greater than that of NO$_2$. Concentrations of SO$_2$ led to changes in NH$_3$ concentration of 19.65% in the warm season and 11.91% in the cold season. Concentration of NO$_2$ caused changes in NH$_3$ concentration of 5.72% in the cold season and 14.23% in the warm seasons. Decreases in atmospheric SO$_2$ and NO$_2$ concentrations lead to increases NH$_3$ concentration. Meteorological conditions (atmospheric pressure and wind) also affected atmospheric NH$_3$ concentration. In warm season, the most important factor is temperature, and in cold season, the most important factor is relative humidity. On the other hand, in addition to NH$_3$ emissions, which is the most important influencing factor, the role of acid gases in summer if greater than that of meteorological factors, and it is contrast in the cold season.

To further understand the trend in NH$_3$ concentration, more accurate emission inventories are needed. Moreover, because reductions in atmospheric SO$_2$ and NO$_2$ concentrations can lead to increases in NH$_3$ concentrations, the implementation of emission reduction policies such as desulfurization and deacidification in air pollution control actions could increase atmospheric NH$_3$ concentrations. Therefore, in addition to improving emission inventories, the effects of SO$_2$ and NO$_2$ emissions on atmospheric NH$_3$ should be considered (Heald et al., 2016).

**DATA AVAILABILITY STATEMENT**

The original contributions presented in the study are included in the article/Supplementary Material, further inquiries can be directed to the corresponding authors.

**AUTHOR CONTRIBUTIONS**

Methodology: DZ-l, ZQ-q Investigation: DZ-l, ZQ-q Writing-original draft: DZ-l Writing-review and editing: DZ-l, ZQ-q, ZX-y Funding acquisition: ZX-y, ZQ-q.

**FUNDING**

This work is funded by the National Natural Science Foundation of China (No.: 41775028, 41805098), National Key Research and Development Program Earth Observation and Navigation Key Project (No. 2017YFB0504001, No.2016YFB0500705).

**ACKNOWLEDGMENTS**

The authors acknowledge the AERIS data infrastructure for providing access to the IASI data in this study and ULB-LATMOS for development of the retrieval algorithms.
