



Estimation of surface ammonia concentrations and emissions in China from the polar-orbiting Infrared Atmospheric Sounding Interferometer and the FY-4A Geostationary Interferometric Infrared Sounder

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Abstract. Ammonia (NH₃) is the most important alkaline gas in the atmosphere, which has negative effects on biodiversity, ecosystems, soil acidification and human health. China has the largest NH₃ emissions globally, mainly associated with agricultural sources including nitrogen fertilizer and livestock. However, there is still a limited number of ground monitoring sites in China, hindering our understanding of both surface NH₃ concentrations and emissions. In this study, using the polar-orbiting satellite (Infrared Atmospheric Sounding Interferometer – IASI) and Fengyun-4A Geostationary Interferometric Infrared Sounder (GIIRS), we analyzed the changes in hourly NH₃ concentrations and estimated surface NH₃ concentrations and NH₃ emissions in China. GIIRS-derived NH₃ concentrations in the daytime were generally higher than those at night, with high values during 10:00–16:00 local time. Satellite-derived surface NH₃ concentrations were generally consistent with the ground observations, with *R*-square at 0.72 and slope equal to 1.03. Satellite-based NH₃ emissions ranged from 12.17 to 17.77 Tg N yr^{−1} during 2008–2019. Spatially, high values of NH₃ emissions mainly occurred in the North China Plain, Northeast China and the Sichuan Basin, while low values were mainly distributed in West China (Qinghai–Tibet Plateau). Our study shows a high predictive power of using satellite data to estimate surface NH₃ concentrations and NH₃ emissions over multiple temporal and spatial scales, which provides an important reference for understanding NH₃ changes over China.

1 Introduction

Ammonia (NH₃) is a highly active gas in the atmosphere and the most important alkaline gas, playing an important role in atmospheric chemistry (Fowler et al., 2013). NH₃ reacts with acid pollutants (SO₂ and NO_x) to form fine particulate matter, such as PM_{2.5}, leading to haze pollution. In addition, the deposition of NH₃ and NH₄⁺ could also cause environmental problems such as water eutrophication, biodiversity loss and soil acidification (Paerl et al., 2014). China has become a major region for NH₃ emissions globally because of rapid

growth of population and agricultural production (Zhang et al., 2017; Liu et al., 2022). To provide a scientific basis for dealing with NH₃ pollution, it is urgent to accurately estimate both surface NH₃ concentrations and emissions in China.

Surface NH₃ concentrations can be estimated by ground measurements and model simulations. Ground measurements are considered to be the most accurate quantitative method. Current national NH₃ observation networks in China include the National Nitrogen Deposition Monitoring Network (NNDMN) established by China Agricultural University (Xu et al., 2015) and the Ammonia Monitoring

Network (AMoN-China) established based on the Chinese Ecosystem Research Network (CERN) (Pan et al., 2018). The NNDMN can measure ground NH_3 concentrations since 2010, while AMoN-China only makes the measurements in 2015–2016. The above two monitoring networks both monitor surface NH_3 concentrations on a monthly basis and lack monitoring of the hourly NH_3 changes. Some studies have conducted research on the intra-day/hourly changes in NH_3 concentrations based on ground observations. Werner et al. (2017) measured hourly NH_3 concentration in 2012 at the Harwell site in the UK and found that high NH_3 concentration usually occurred in the afternoon. Similarly, Kutzner et al. (2021) observed the hourly NH_3 concentration at the SIRTa Observatory in Paris and found that NH_3 concentration was highest in the late afternoon. Pandolfi et al. (2012) studied the day–night cycle of NH_3 concentration at the two stations in Barcelona in summer and found that the NH_3 concentration was highly associated with local meteorology and traffic emissions. However, there is still a limited number of monitoring sites on the hourly NH_3 changes in China.

Agricultural fertilizer and livestock production have led to a large amount of NH_3 emissions. China's cultivated land area accounts for less than 10 % of the world, but it consumes about 30 % of the world's nitrogen (N) fertilizer (Peng et al., 2002). Estimation of NH_3 emissions is mainly based on a bottom-up method using NH_3 source statistics (fertilization, animal husbandry, etc.) and emission factors. Zhou et al. (2016) calculated the annual farmland NH_3 emission ($3.96 \pm 0.76 \text{ Tg N yr}^{-1}$) over China in 2008 based on the bottom-up method, which was 40 % higher than the emission in the Intergovernmental Panel on Climate Change (IPCC) Tier-1 guidelines ($2.89 \text{ Tg N yr}^{-1}$). Zhang et al. (2017) reassessed China's NH_3 emissions based on the mass balance method and found that NH_3 emissions increased from 12.1 ± 0.8 in 2000 to $15.6 \pm 0.9 \text{ Tg N yr}^{-1}$ in 2015, with an annual growth rate of 1.9 %. Fu et al. (2020) estimated that China's NH_3 emissions increased from 4.7 in 1980 to 11 Tg N yr^{-1} in 2016. Although many studies regarding NH_3 emissions have been carried out in China, great uncertainties and large ranges ($7\text{--}16 \text{ Tg N yr}^{-1}$) still existed in the estimates of China's NH_3 emissions (Dong et al., 2010; Huang et al., 2012; Kang et al., 2016).

Besides the bottom-up estimates, some studies used data assimilation methods by ground monitoring data to constrain NH_3 emission estimates. Paulot et al. (2014) assimilated GEOS-Chem with ground observations of wet reactive Nitrogen (N_r) deposition and estimated China's NH_3 emission as 8.4 Tg N yr^{-1} in 2008, with seasonal NH_3 emission peaking in summer. Gilliland et al. (2003) used the data assimilation method by the Community Multiscale Air Quality (CMAQ) with the wet NH_4^+ concentration data from the USA National Atmospheric Deposition Program Network and found that obvious seasonal differences appeared in NH_3 emissions linked to N fertilizer and temperature. Kong et al. (2019) carried out inversion by assimilating surface AMoN NH_3 ob-

servation data and improved the accuracy of temporal and spatial patterns of NH_3 emissions in China.

In recent years, atmospheric remote sensing has developed rapidly, which can monitor NH_3 at a global scale, including the polar-orbiting satellite instruments such as the Tropospheric Emission Spectrometer (TES), Infrared Atmospheric Sounding Interferometer (IASI), Cross-track Infrared Sounder (CrIS), Atmospheric Infrared Sounder (AIRS), and Greenhouse Gases Observing Satellite (GOSAT) (Someya et al., 2020). Many studies have reported the effectiveness of using satellite data to study NH_3 dynamics. Pinder et al. (2011) found that TES observation can capture spatial–temporal NH_3 patterns compared with surface measurement. Van Damme et al. (2014) studied the seasonal and annual NH_3 changes in the Northern Hemisphere and Southern Hemisphere using the IASI NH_3 column data and found that the seasonality in the Southern Hemisphere is mainly related to biomass burning. Shephard and Cady-Pereira (2015) developed the CrIS NH_3 inversion algorithms and found that CrIS can capture the global spatial distribution of NH_3 concentration. Warner et al. (2016) identified the main hotspots of agricultural NH_3 regions using AIRS, such as South Asia, China, the United States and some parts of Europe, and found that NH_3 concentrations had increased in these agricultural regions since 2003. In addition, some studies also used the satellite measurements to improve the estimates of NH_3 emissions. Zhang et al. (2017) developed a top-down inversion method using TES NH_3 observation to quantify China's NH_3 emission and obtained annual NH_3 emission as $11.7 \text{ Tg N yr}^{-1}$ in 2008. Marais et al. (2021) estimated NH_3 emissions in the UK based on the IASI and CrIS and found that the relative errors of IASI-derived NH_3 emissions were 11 %–36 % and 9 %–27 %, respectively. Van Damme et al. (2018) used the high-resolution IASI NH_3 maps to identify, classify and quantify NH_3 emission hotspots in the world, which was helpful for understanding the human point NH_3 sources. Dammers et al. (2019) identified global 249 NH_3 emission point sources based on CrIS, whose total emission was about 2.5 times higher than that reported in the HTAPv2 emissions.

Besides the polar-orbiting satellite, China's Geostationary Interferometric Infrared Sounder (GIIRS) on board the Chinese FY-4A satellite can measure hourly changes in atmospheric NH_3 in almost all of Asia per day, which provides great potential to study the diel cycle of NH_3 . In this study, GIIRS is used to study the NH_3 diel cycle (hourly changes), which is essential for understanding the differences between different times in a day by the polar-orbiting satellites (such as the IASI at 09:30 and CrIS at 13:30). Second, the surface NH_3 concentration in China is estimated based on both GIIRS and IASI, which is then compared with the NNDMN. Third, NH_3 emission in China is calculated based on satellite-derived surface NH_3 concentration and the feedback relationship between surface NH_3 concentration and emission by a chemistry transport model (GEOS-Chem). Fi-

nally, the spatial–temporal characteristics of satellite-derived surface NH_3 concentration and emission are analyzed, and the uncertainties are discussed.

2 Data and methods

2.1 Satellite GIIRS NH_3

The Geostationary Interferometric Infrared Sounder (GIIRS) on board the Fengyun-4A geostationary satellite (FY-4A) launched by China in 2016 is the world's first hyperspectral atmospheric infrared sounder (Cai et al., 2020). The FY-4A GIIRS-detected spectral range is $700\text{--}2250\text{ cm}^{-1}$, including 1648 spectral channels and 14 radiation-imaging channels, covering visible light, shortwave, medium and long-wave infrared bands. The spatial resolutions of the detector are 2.0 km in the visible bands and 16 km in the infrared bands. It covers almost the whole of Asia and scans 10 times a day (Zhang et al., 2019). The GIIRS can detect the temperature and humidity profiles and trace gases at high frequencies.

NH_3 has two large absorption characteristics in the long-wave infrared (about 930 and 965 cm^{-1}). The contribution of NH_3 to the brightness temperature of these two bands is between 2 and 4 K. The core of inversion algorithms is based on the so-called hyperspectral radiation index (HRI), which quantifies the spectral characteristics of NH_3 . The HRI depends on whether the satellite instrument detects the presence of NH_3 . The average value of the HRI is 0 with a standard deviation of 1, and the HRI range is $[-1, 1]$. The algorithm for estimating IASI NH_3 column concentration is to convert the HRI into a column using the so-called neural network (Clarisse et al., 2021).

In this study, we use hourly NH_3 concentrations during 2019–2020 (from November 2019 to October 2020) to study the NH_3 diel cycle with a resolution of 0.5° . The original data are in Hierarchical Data Format Version 5 (HDF5), and the unit of NH_3 is molec. cm^{-2} . The data are processed with MATLAB software. First, observations with considerable uncertainties (relative error exceeds 50 %) and high clouds (cloud cover exceeds 20 %) are removed (Fig. S1a in the Supplement). Secondly, the world standard time (UTC) by GIIRS is converted to local time (LT).

2.2 Satellite IASI NH_3

The IASI instrument is on board the polar solar-synchronous Metop-A platform. It has been running stably since 2006 to measure the infrared radiation emitted by the Earth (Van Damme et al., 2014). IASI can measure the infrared radiation emitted by the Earth's surface and atmosphere in the spectral range of $645\text{--}2760\text{ cm}^{-1}$. It can observe the world twice a day and cross the Equator at 09:30 and 21:30 local time, with a spatial resolution of 12 km at nadir. However, only daytime satellite measurements are used, because nighttime measure-

ments usually have greater uncertainties related to thermal contrasts (Van Damme et al., 2017).

The near-real-time dataset of the total NH_3 column (built by artificial neural network for IASI, ANNI; ANNI- $\text{NH}_3\text{-v3}$) was used here. The properties of IASI NH_3 data include NH_3 column concentration, longitude, latitude, measurement time, cloud cover, uncertainty, solar zenith angle and other parameters. The daily NH_3 column from 2008 to 2019 was used. The format of the original data is the Network Common Data Format, and the unit is mol m^{-2} . The observation data with cloud cover larger than 20 % and the uncertainty above 50 % were removed (Fig. S1b). We gridded the data to 0.1° by using the arithmetic average methods.

2.3 Ground NH_3 measurements

Surface NH_3 concentrations in the NNDMN were used to compare with the satellite estimates including 43 observation stations. The land types of the NNDMN sites cover cities, farmland, coastal areas, forests and grasslands. Measurements during the period from January 2010 to December 2015 by the NNDMN were used. Surface NH_3 concentrations were measured using an active DELTA (Denuder for Long-Term Atmospheric sampling) (Flechard et al., 2011). For the hourly measurements, we collected the data from the published papers, including five sites (Table S1 in the Supplement), Xianghe (39.75° N , 116.96° E ; December 2017–February 2018) (He et al., 2020), Fudan University (31.30° N , 121.50° E ; 1 July 2013–30 September 2014) (Wang et al., 2015), Dianshan Lake (31.09° N , 120.98° E ; 1 July 2013–30 June 2014) (Wang et al., 2015), Jinshan Chemical Industry Park (30.73° N , 121.27° E ; 6 January–30 June 2014) (Wang et al., 2015) and Gucheng (39.15° N , 115.73° E ; March 2016–May 2017) (Kuang et al., 2020). The Xianghe site in Hebei Province and the Dianshan Lake site in Shanghai represent rural environments. Jinshan Chemical Industry Park represents industrial environments. The Gucheng site in Hebei and the Fudan University site in Shanghai represent urban environments.

2.4 GEOS-Chem

The GEOS-Chem model version 12.3.0 is a three-dimensional chemistry transport model developed by Harvard University, which has been widely used in the field of atmospheric studies (Eastham et al., 2014). The nested regional model in Asia was used in this study driven by assimilated GEOS-5 meteorological data at a horizontal resolution of $1/2^\circ \times 2/3^\circ$. Dry deposition calculation in GEOS-Chem follows a standard resistance-in-series model (Wesely, 2007), while wet deposition includes both convective updraft and large-scale precipitation scavenging (Jacob, 1999). The GEOS-Chem model here does not consider land–atmosphere bidirectional NH_3 exchange, and the NH_3 flux was parameterized as uncoupled emission and dry deposition processes.

Anthropogenic emissions over China were from the Regional Emission in Asia (REAS-v2) inventory. The GEOS-Chem outputs of NH_3 concentrations include 47 layers from the ground to the top of the atmosphere, which were used to capture NH_3 vertical profiles. The feedback between surface NH_3 concentration and emissions was also calculated by GEOS-Chem.

2.5 Satellite-based surface NH_3 estimates and emissions

Surface NH_3 concentrations were estimated using the satellite NH_3 columns as well as NH_3 vertical profiles. To gain the continuous vertical NH_3 profile, the Gaussian function was used to fit the 47 layers' NH_3 concentrations. A three-parameter Gaussian function was used to fit NH_3 vertical profiles at each grid box from GEOS-Chem according to previous studies (Liu et al., 2019).

$$\rho(Z) = \sum_{i=1}^n \rho_{\max,i} e^{-\left(\frac{Z-Z_{0,i}}{\sigma_i}\right)^2}, \quad (1)$$

where Z is the height of a layer in an atmospheric chemical transport model (ACTM); ρ_{\max} , Z_0 and σ are the maximum of NH_3 concentration, the corresponding height with the maximum of NH_3 concentration and the thickness of the NH_3 concentration layer (1 standard error of the Gaussian function).

The satellite-derived NH_3 concentration at the height of h_G can be calculated as

$$S_{G\text{NH}_3} = S_{\text{trop}} \times \frac{\rho(h_G)}{\int_0^{h_{\text{trop}}} \rho(Z) \text{d}x} \times \frac{G_{\text{ACTM}}^{1-24}}{G_{\text{ACTM}}^{\text{overpass}}}, \quad (2)$$

where $\frac{\rho(h_G)}{\int_0^{h_{\text{trop}}} \rho(Z) \text{d}x}$ represents the ratio of NH_3 concentration at the height of h_G to total columns ($\int_0^{h_{\text{trop}}} \rho(Z) \text{d}x$), S_{trop} represents satellite-derived NH_3 columns, and $\frac{G_{\text{ACTM}}^{1-24}}{G_{\text{ACTM}}^{\text{overpass}}}$ is the ratio of average surface NH_3 concentration (G_{ACTM}^{1-24}) to that at satellite overpass time ($G_{\text{ACTM}}^{\text{overpass}}$) by an ACTM.

The mass balance method (Lamsal et al., 2011; Geddes and Martin, 2017; Cooper et al., 2017) was used to exploit the feedback ratio of surface NH_3 concentrations and NH_3 emissions (Marais et al., 2021):

$$E_s = S_{G\text{NH}_3} \times \left(\frac{E}{G_{G\text{NH}_3}} \right)_m, \quad (3)$$

where E_s is satellite-based NH_3 emissions, $S_{G\text{NH}_3}$ is satellite-derived surface NH_3 concentrations and $\left(\frac{E}{G_{G\text{NH}_3}} \right)_m$ is the ratio of surface NH_3 concentrations and NH_3 emissions simulated by GEOS-Chem.

3 Results and discussions

3.1 GIIRS-based hourly NH_3 concentrations during 2019–2020

Figure 1 shows the hourly NH_3 concentrations observed from GIIRS during 2019–2020. Daytime NH_3 columns were significantly higher than those at night. The intra-day hourly NH_3 columns showed an overall increase first and then a decrease, with high values during 10:00–16:00. The increase in temperature enhanced the volatilization of NH_3 , which may explain high values of NH_3 concentrations during the daytime.

Ground-based measurements of hourly NH_3 concentrations are very lacking, and the timespan may be different from GIIRS measurements. Here we only used them to show the hourly patterns of NH_3 concentrations (Fig. 2). The Xi'anqhe site in Hebei Province and Dianshan Lake site in Shanghai represent rural environments. Jinshan Chemical Industry Park represents industrial environments. The Gucheng site in Hebei and the Fudan University site in Shanghai represent urban environments.

NH_3 concentration in the rural environment basically shows a normal distribution, and high NH_3 concentration generally appears between 09:00 and 16:00, which may be related to agricultural activities and temperature. In the industrial environment (Jinshan Chemical Industry Park, JSP), NH_3 concentration fluctuates irregularly, and two peaks appear at 18:00–23:00 and 06:00–08:00, while NH_3 concentration tends to be stable at other times. In the urban environment, the changes in NH_3 concentration by satellite at the Gucheng site are consistent with ground monitoring, showing a clear peak around 09:00–14:00. NH_3 concentration at the Fudan University site gradually decreases from the morning peak to the afternoon. The evaporation of dew may drive the NH_3 increase from the morning to the noon (Wang et al., 2015). NH_3 concentration by ground monitoring in cities (Gucheng and Fudan University) has double peaks between 08:00–11:00 and 18:00–22:00, which may also be related to traffic emissions. In summary, except for industrial sites, hourly NH_3 in China has a large variability between day and night, and the hourly NH_3 patterns are affected by many factors, of which anthropogenic emissions and temperature seem to be the most important drivers.

Spatial distribution of GIIRS-based surface NH_3 concentrations across China had large variability (Fig. 3). High surface NH_3 concentration ($> 10 \mu\text{g N m}^{-3}$) is mainly concentrated in the North China Plain (NCP), followed by the Sichuan Basin, Northeast China and parts of Xinjiang, while low values ($< 4 \mu\text{g N m}^{-3}$) are mainly concentrated in the Qinghai–Tibet Plateau. High surface NH_3 concentration ($126.85 \mu\text{g N m}^{-3}$) appears in July ($6.78 \mu\text{g N m}^{-3}$ on average), and the lowest value ($0.23 \mu\text{g N m}^{-3}$) appears in November ($3.25 \mu\text{g N m}^{-3}$ on average). There are obvious seasonal changes in surface NH_3 concentrations in the NCP

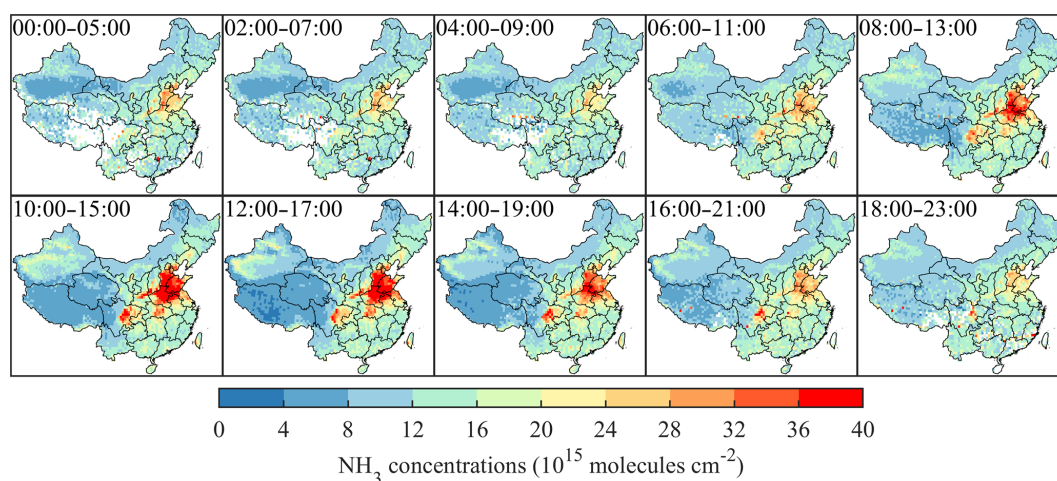


Figure 1. Monthly average NH_3 concentrations for each of the 10 GIIRS overpass time periods during 2019–2020.

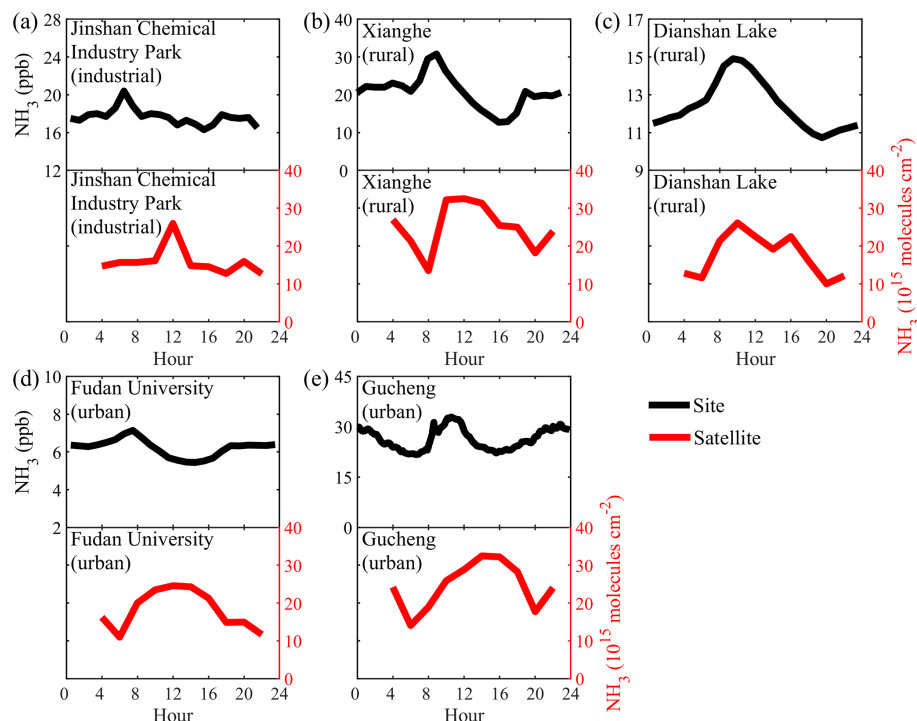


Figure 2. GIIRS-based and measured hourly NH_3 concentrations at five sites: Jinshan Chemical Industry Park (JSP, **a**), Xianghe (XH, **b**), Dianshan Lake (DSL, **c**), Fudan University (FDU, **d**) and Gucheng (GC, **e**).

with high values in summer and low values in winter, related to both agricultural N fertilizer and higher temperature.

3.2 IASI-based NH_3 surface concentrations

The observation data of the NNDMN in China were collected to compare with the IASI-derived surface NH_3 concentration. In general, a good consistency was found between measurements and satellite estimates, with the regression R^2 as

0.72 and the RMSE as $2.24 \mu\text{g N m}^{-3}$. The coefficient of the fitted line was $1.03 \approx 1$, and the bias was 2.59 % (Fig. 4).

Monthly regression R^2 between the satellite-derived NH_3 concentration and the measured NH_3 was 0.38–0.84 (Fig. 5). The regression R^2 reached the higher value (> 0.80) in July and August. The RMSE ranged from 2.29 to $3.36 \mu\text{g N m}^{-3}$, which reached the maximum value of $3.36 \mu\text{g N m}^{-3}$ in July and reached the smallest value in March ($2.29 \mu\text{g N m}^{-3}$). The bias is basically less than 31 % for all months and reached the minimum value of 0.67 % in February, indicat-

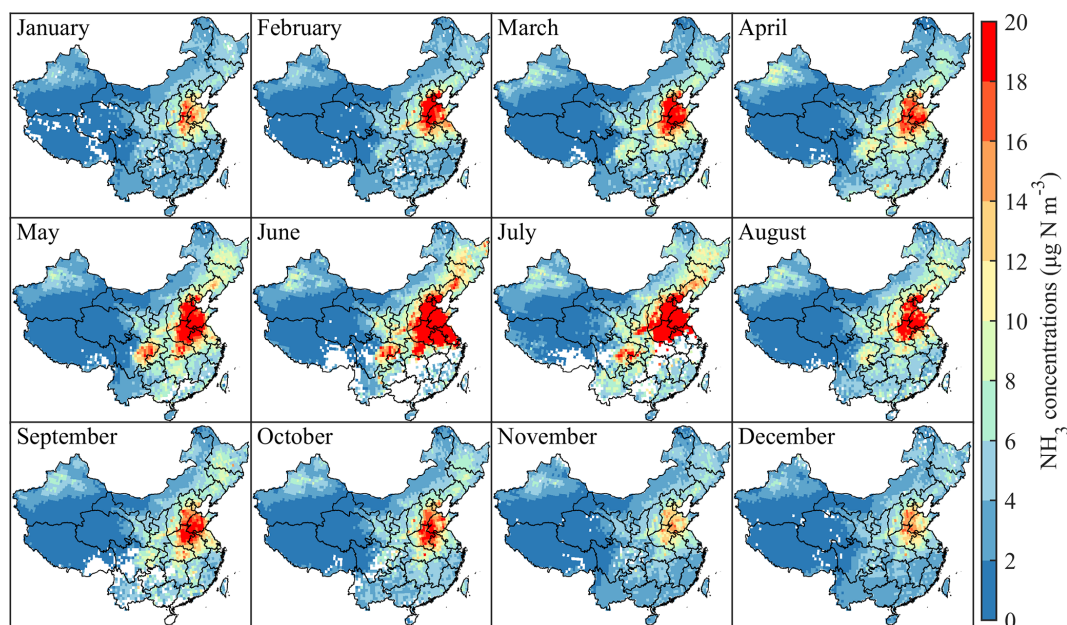


Figure 3. Spatial distribution of monthly surface NH_3 concentrations in China by GIIRS in 2019–2020.

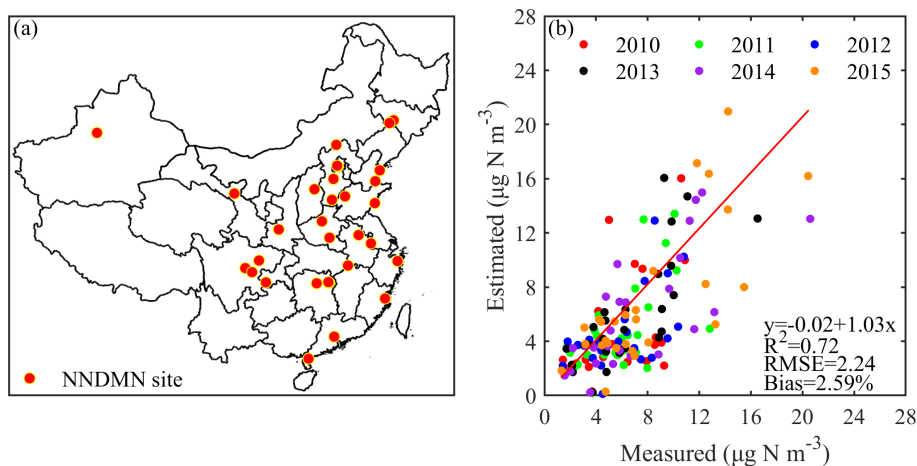


Figure 4. Comparison of IASI surface NH_3 concentrations with NNDMN measurements. (a) The locations of NNDMN; (b) the regression results between satellite estimates and measurements.

ing that the monthly IASI-derived surface concentrations obtained are consistent with measurements.

Figure 6 shows the monthly changes in surface NH_3 concentrations in Huinong County in Ningxia from 2010 to 2015 for a total of 72 months. Surface NH_3 concentrations retrieved by the IASI were compared with the observation data at Huinong. The highest value of each year basically appeared from June to August, and the lowest values appeared from December to January. In the past 6 years, the maximum measured NH_3 concentration appeared in June 2015 ($18.9 \mu\text{g N m}^{-3}$), and the minimum appeared in November 2012 ($0.6 \mu\text{g N m}^{-3}$). The observation data and satellite data have the same seasonal changes.

Figures 3 and 7 show the spatial distribution of GIIRS-derived and IASI-derived surface NH_3 concentrations in 2019. The spatial distribution and gradients of surface NH_3 concentrations by the GIIRS and IASI have the same gradients from the eastern to western regions. One notable difference occurred in the middle and lower reaches of the Yangtze River in June and July since the GIIRS observations are affected by clouds and had missing data.

3.3 IASI-derived NH_3 emissions

Based on the top-down estimates, China's NH_3 emissions ranged from 12.17 to $17.77 \text{ Tg N yr}^{-1}$ during 2008–2019.

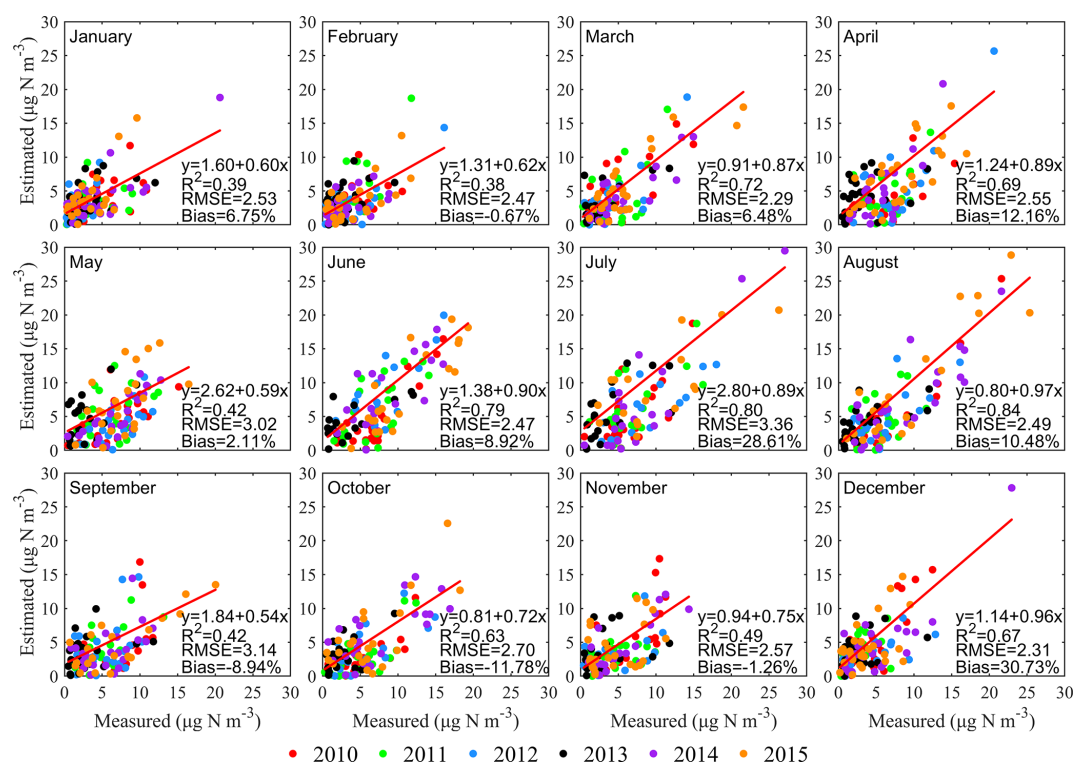


Figure 5. Comparison of monthly average values of ground measured and IASI-derived NH_3 surface concentrations in 2010–2015.

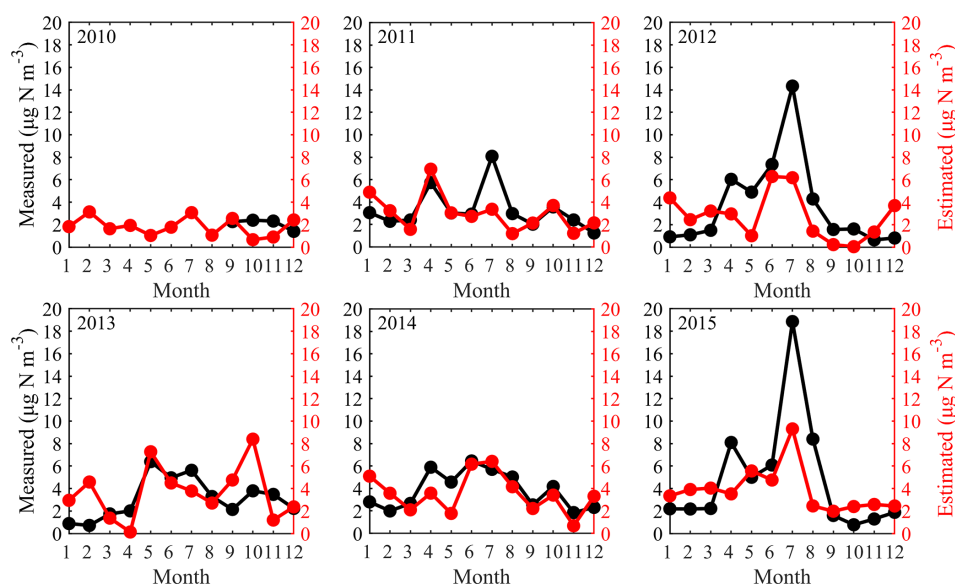


Figure 6. Monthly changes in NH_3 concentrations in Huinong County in Ningxia from 2010 to 2015 for a total of 72 months during 2010–2015.

From 2008 to 2015, NH_3 emissions increased from 13.00 to 17.06 Tg N yr^{-1} . Since 2008, the temperature in China has risen steadily (Ding et al., 2007), promoting the volatilization of NH_3 , which partly explains the increase in NH_3 emissions from 2008 to 2015. After 2015, NH_3 emissions fluctuated and changed slightly (16.08–17.77 Tg N yr^{-1}). Com-

pared with other studies, the change in NH_3 emissions from 2008 to 2015 is consistent with previous estimates, and the overall NH_3 emissions show an upward trend (Kang et al., 2016; Zhang et al., 2018; Ma, 2020; Fu et al., 2020; Zhang et al., 2021). Our estimates are on the rise as a whole, but the calculated values are generally lower than those by Zhang

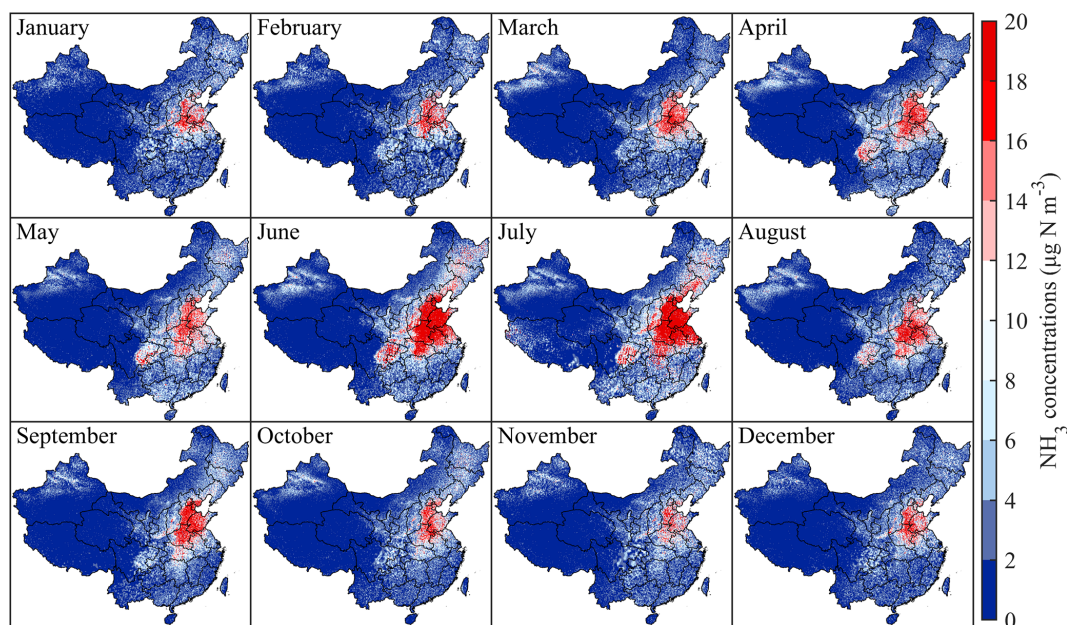


Figure 7. Spatial distribution of monthly surface NH_3 concentrations in China by the IASI in 2019.

et al. (2017) (around 15 Tg N yr^{-1}) but larger than those by EDGAR and Kang et al. (2016).

In terms of spatial distribution, high NH_3 emissions are generally distributed in the North China Plain, Sichuan Basin and Northeast China, while the low values are mainly distributed in Southwest China (especially the Qinghai–Tibet Plateau). The North China Plain is China’s granary, with developed agriculture and animal husbandry, high population densities and strong human activities (including vehicle emissions) (Zhang et al., 2006; Wang et al., 2020). In contrast, South China is rich in rainfall, which promotes the deposition of NH_3 and suppresses its volatilization to a certain extent.

The spatial distributions of NH_3 emissions in January, April, July and October were selected to characterize the seasonal variations. The average emissions for the four months were 1.15 , 1.31 , 2.31 and $1.16 \text{ kg N ha}^{-1}$ in January, April, July and October, indicating that NH_3 emission is highest in summer and lowest in winter. The annual average emission intensity of 2019 is $16.53 \text{ kg N ha}^{-1}$ (0.09 – $313.47 \text{ kg N ha}^{-1}$). Figure 8b shows the monthly changes in NH_3 emissions, which basically shows a normal distribution. High values are generally distributed in June and July, and low values are generally distributed in November and December. They reached the maximum monthly emission of 2.6 Tg N m^{-1} in July 2019 and reached the minimum monthly emission in November and December (1.1 Tg N m^{-1}).

4 Limitations and outlook

This study developed satellite-based surface NH_3 concentrations and emissions in China based on remote sensing data (IASI and GIIIRS). However, several limitations have been identified in this study. First, the Fengyun geostationary satellite used in this study can achieve hourly NH_3 concentrations, but the time series is still very short (November 2019–October 2020), and satellite observations are affected by clouds and meteorological conditions, resulting in missing values in parts of the Yangtze River basin.

Second, we used a relatively fixed average conversion ratio (Fig. S2) to estimate surface NH_3 concentrations and NH_3 emissions in China, ignoring the time-series variation of the ratio, due to the temporal constraint of the emission inventory. In this case, non-emission factors led to a higher satellite-observed NH_3 column; for example, emission reductions of SO_2 and NO_2 led to an increased NH_3 column (Lachatre et al., 2019; Fu et al., 2020), which can introduce uncertainty into NH_3 emission calculations using concentration as the main parameter.

Third, the spatial resolution of the NH_3 vertical profile simulated by the atmospheric model is relatively coarse (0.5°). In order to make it consistent with the spatial resolution of the remote sensing data, the outputs of GEOS-Chem (vertical profiles and feedback ratio between emissions and surface NH_3 concentrations) were adjusted through resampling methods. Owing to the resolution limit, the ratio-based mass balance approach to estimate NH_3 emissions neglected the effects of internal transport of NH_3 and displacement of emission sources within the fine grid.

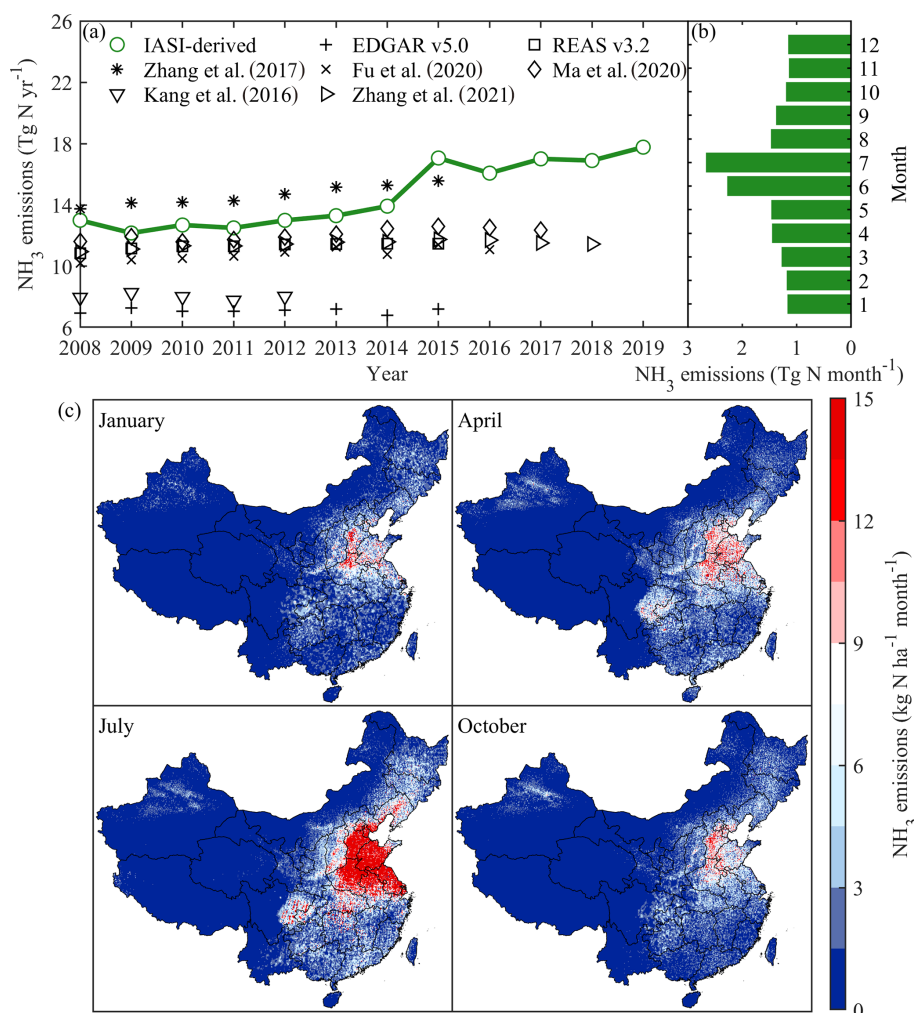


Figure 8. Annual changes in NH₃ emissions (a), monthly changes in NH₃ emissions in 2019 (b) and spatial distribution of NH₃ emissions by month in 2019 (c).

Finally, there are some uncertainties and biases in the observed NH₃ column by satellite. Earlier versions of the IASI NH₃ column product were 25 %–50 % lower than ground-based measurements (Whitburn et al., 2016; Damers et al., 2017). However, version 3 of IASI–NH₃ lacks a comprehensive ground-based measurement assessment, which has only been compared with limited aircraft observations (Guo et al., 2021). Comparing IASI-derived surface NH₃ concentrations with measurements of ground sites (NNDMN) generally shows consistency in this study. Further work is needed for the complete assessment and error analysis.

At present, there are more and more satellite sensors (GOSAT, CrIS, AIRS, etc.) that can monitor NH₃ concentrations. This study only used IASI and GIIRS, and in the future, data from different satellites can be merged to analyze NH₃ changes on multiple temporal and spatial scales.

5 Conclusion

We use GIIRS to study the NH₃ diel cycle and estimate surface NH₃ concentrations and emissions based on the IASI in China. There are obvious hourly changes in NH₃ concentration in China using GIIRS. Overall, NH₃ concentrations are larger in the daytime than at nighttime in China. Hourly NH₃ concentrations at different regions show different patterns, but high values generally appear at 10:00–16:00. Comparing ground measured and IASI-derived NH₃ surface concentrations by NNDMN from 2010 to 2015, the coefficient of the fitted line is $1.03 \approx 1$ and the low bias is 2.59 %, indicating satellite estimates have good consistency with the measurements. IASI-derived Chinese NH₃ emissions range from 12.17 to 17.77 Tg N yr⁻¹ during 2008–2019, among which NH₃ emissions increase from 2008 to 2015. The emission intensity of NH₃ in China presents a strong spatial heterogeneity, being high in the east and low in the west. The high val-

ues are mainly distributed in the North China Plain, Sichuan Basin and Northeast China. High values are generally distributed in summer, and low values generally occur in winter. This study provides an important reference basis for the formulation of NH_3 pollution prevention and control policy in China.

Data availability. IASI NH_3 data were obtained from https://iasi.aeris-data.fr/nh3_iasi_a_data (Clarisse et al., 2022). GHIRS NH_3 data were obtained from <https://doi.org/10.5281/zenodo.5051433> (Clarisse, 2021).

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