

# 1 Geostationary observations of atmospheric ammonia over East Asia: 2 spatio-temporal variations revealed by three years of FY-4B/GIIRS 3 measurements

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32 **Abstract.** Satellite observations play a crucial role in quantifying ammonia sources by capturing large-scale variations of  
33 atmospheric NH<sub>3</sub> concentrations. As the world's first geostationary hyperspectral infrared sounder, the Geostationary  
34 Interferometric Infrared Sounder (GIIRS) on board China's FengYun-4 satellite series provides a unique opportunity to  
35 monitor the diurnal cycle of NH<sub>3</sub>. Using NH<sub>3</sub> retrievals from July 2022 to June 2025, this study investigates the spatio-temporal  
36 variability of NH<sub>3</sub> columns over East Asia, with a focus on daytime variations (07:00–19:00 local time) in major agricultural  
37 regions. Inter-comparison with polar-orbiting IASI and CrIS data shows that GIIRS NH<sub>3</sub> retrievals are consistent in capturing  
38 spatial patterns and temporal dynamics. **The NH<sub>3</sub> peaks occur between March and July, with peak timing earlier in the south**

39 and later in the north, reflecting regional differences primarily driven by agricultural activities. Validation with ground-based  
40 FTIR measurements at Hefei in eastern China demonstrates the accuracy of GIIRS NH<sub>3</sub>, with a correlation coefficient of 0.77  
41 and an RMSE of  $9.67 \times 10^{15}$  molec/cm<sup>2</sup>, while reproducing daytime variations observed by FTIR. For major agricultural areas,  
42 the NH<sub>3</sub> columns generally increase from early morning to late afternoon, reaching 1.10–1.56 times morning levels in summer  
43 and spring. Compared with GEOS-CF model simulations, the results reveal pronounced discrepancies in spatial distributions  
44 over the Sichuan Basin in southwestern China and daytime variations over northern India. These findings highlight the valuable  
45 capability of FY-4B/GIIRS in identifying and tracking daytime dynamics of NH<sub>3</sub> sources over East Asia, offering new insights  
46 beyond current low-Earth orbit (LEO) instruments.

## 47 1 Introduction

48 Gaseous ammonia (NH<sub>3</sub>), the most abundant alkaline gas and a major reactive nitrogen compound in the atmosphere,  
49 plays a critical role in the global nitrogen cycle. NH<sub>3</sub> is an important precursor of secondary inorganic aerosols. It rapidly  
50 reacts with atmospheric acids such as sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) and nitric acid (HNO<sub>3</sub>) to form ammonium sulfate ((NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>)  
51 and ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>) (Seinfeld et al., 2016). These ammonium-containing aerosols are the primary components  
52 of fine particulate matter (PM<sub>2.5</sub>) and have been identified as key contributors to elevated PM<sub>2.5</sub> concentrations and haze  
53 pollution in both rural areas (Jang et al., 2025; Saraswati et al., 2019; Meng Z. et al., 2018) and megacities (Lin et al., 2020) in  
54 Asia, posing significant threats to air quality, visibility, and public health. NH<sub>3</sub> also exerts indirect influences on global climate  
55 change, for example, by driving nitrous oxide (N<sub>2</sub>O) formation via atmospheric oxidation (Pai et al., 2021) and by altering  
56 radiative forcing through aerosol-mediated processes (Gong et al., 2024). Moreover, excessive emissions and deposition of  
57 NH<sub>3</sub> have adverse effects on ecosystems, causing biodiversity loss, soil acidification, and water eutrophication (Bobbink et al.,  
58 2010). Anthropogenic NH<sub>3</sub> emissions primarily originate from agricultural activities, particularly from synthetic fertilizer  
59 application and livestock farming, with additional contributions from industrial processes and transportation (Behera et al.,  
60 2013). Numerous studies have indicated that a relatively strong reduction in PM<sub>2.5</sub> and N<sub>2</sub>O levels can be achieved by  
61 decreasing agricultural emissions (e.g., Luo et al., 2025). Therefore, comprehensive quantification and understanding of NH<sub>3</sub>  
62 emission sources are essential for developing effective air quality and climate mitigation strategies.

63 Asia is the world's most significant NH<sub>3</sub> emission regions, accounting for over 30% of global NH<sub>3</sub> emissions annually as  
64 reported by the Emissions Database for Global Atmospheric Research (EDGAR) inventory. Due to widespread use of urea-  
65 based nitrogen fertilizers and high livestock densities, cropland areas such as the Indo-Gangetic Plain in India and the North  
66 China Plain in China are recognized as major global NH<sub>3</sub> emission hot spots (Van Damme et al., 2018). NH<sub>3</sub> volatilization is  
67 strongly controlled by surface conditions, including temperature and moisture, and irrigation-driven NH<sub>4</sub><sup>+</sup> accumulation in  
68 soils (Behera et al., 2013). The resulting atmospheric NH<sub>3</sub> concentrations over a region are further influenced by meteorology-  
69 and topography-induced constraints on atmospheric mixing and transport, chemical reactions dependent on the concentrations  
70 of reactive acidic gases, and dry and wet deposition (Dammers et al., 2017a). In most regions, NH<sub>3</sub> concentrations in summer

71 are significantly higher than those in winter, driven by emissions from urea hydrolysis and livestock manure decomposition  
72 (Warner et al., 2016; Van Damme et al., 2015). Given its short atmospheric lifetime of a few hours to several days, NH<sub>3</sub> exhibits  
73 large intra-day variability in concentration from both agricultural and non-agricultural sources. Several studies have employed  
74 surface measurements to investigate the diurnal variability of NH<sub>3</sub> and its controlling mechanisms, revealing that diurnal  
75 patterns in urban and rural areas differ markedly across seasons (Gu et al., 2022; He et al., 2020; Chang et al., 2019; Huy et  
76 al., 2017; Meng Z. et al., 2011). These analyses are limited by the sparse spatial coverage and short temporal duration of  
77 ground-based monitoring networks. To complement observations, atmospheric chemistry models have been applied to simulate  
78 NH<sub>3</sub> at regional and global scales, and refinements in surface exchange schemes have improved NH<sub>3</sub> modeling (Jongenelen et  
79 al., 2025; Cao et al., 2022). However, the mechanisms underlying the diurnal variability of atmospheric NH<sub>3</sub> in different  
80 emission regions still remain poorly understood.

81 Satellite observations facilitate top-down inversion methods for monitoring NH<sub>3</sub> variations across multiple spatial scales.  
82 The hyperspectral infrared sounders onboard polar-orbiting satellites in low-Earth orbit (LEO), such as the Infrared  
83 Atmospheric Sounding Interferometer (IASI; Clarisse et al., 2009), the Tropospheric Emission Spectrometer (TES; Beer et al.,  
84 2008), the Atmospheric Infrared Sounder (AIRS; Warner et al., 2016), and the Cross-track Infrared Sounder (CrIS; Shephard  
85 et al., 2020), have provided global monitoring of atmospheric NH<sub>3</sub> column densities. Satellite-based NH<sub>3</sub> retrievals have been  
86 widely used to investigate the spatial distribution of NH<sub>3</sub> emission sources (e.g., Van Damme et al., 2018), to characterize  
87 spatiotemporal variations in atmospheric NH<sub>3</sub> concentrations (e.g., Van Damme et al., 2021; Shephard et al., 2020), and to  
88 constrain emission estimates (e.g., Cao et al., 2020; Dammers et al., 2019) and dry deposition estimates (e.g., Kharol et al.,  
89 2018). Van Damme et al. (2021) reported that NH<sub>3</sub> concentrations increased from 2008 to 2018 in major agricultural regions  
90 globally, including South Asia, China, the United States and some parts of Europe. Based on multi-year NH<sub>3</sub> maps, Van Damme  
91 et al. (2018) and Clarisse et al. (2019) identified and quantified major industrial and agricultural NH<sub>3</sub> point sources, revealing  
92 that NH<sub>3</sub> emissions from approximately two-thirds of these sources were underestimated by at least one order of magnitude in  
93 the EDGAR emission inventory. In addition, NH<sub>3</sub> emission estimates based on satellite observations and inverse modeling  
94 indicated that bottom-up emission inventories underestimate NH<sub>3</sub> emissions, especially during summer (e.g., Xu et al., 2023;  
95 Marais et al., 2021; Zhang et al., 2018).

96 Accurately monitoring the daily dynamics of NH<sub>3</sub> emissions imposes high sampling requirements in time and space on  
97 atmospheric NH<sub>3</sub> measurements. Polar-orbiting satellites typically measure a given location at most twice per day. For example,  
98 IASI and CrIS cross the equator at 09:30/21:30 and 01:30/13:30 local solar time (LST), respectively (Whitburn et al., 2016;  
99 Shephard et al., 2015). The above satellite-based studies mostly used morning observations from IASI and afternoon  
100 observations from CrIS, because of the lower uncertainties associated with a more favorable thermal state of the atmosphere  
101 for the remote sensing of its lowest layers. Investigations of the spatial and temporal variability of NH<sub>3</sub> are constrained by the  
102 limited coverage of satellite overpasses, leading to potential temporal representativeness biases and substantial uncertainties  
103 in daily NH<sub>3</sub> emission estimates (Clarisse et al., 2019; Van Damme et al., 2018). The Geostationary Interferometric Infrared  
104 Sounder (GIIRS) onboard the FY-4 series is the world's first hyperspectral infrared sounder in geostationary (GEO) orbit. It

105 was first launched as an experimental instrument on FY-4A in December 2016 and subsequently deployed as operational  
106 instruments with enhanced sensitivity on FY-4B in June 2021 (Yang et al., 2017) and on FY-4C in December 2025. Compared  
107 with current LEO instruments, GIIRS provides continuous day-and-night monitoring of atmospheric NH<sub>3</sub> over East Asia at  
108 hourly-scale temporal resolution, representing a breakthrough in monitoring the diurnal cycle of atmospheric NH<sub>3</sub> variations  
109 (Clarisse et al., 2021; Zeng et al., 2023a). Similarly, the Meteosat Third Generation sounding satellite (MTG-S1), launched in  
110 July 2025, carries the Infrared Sounder (IRS), another geostationary hyperspectral infrared instrument designed to provide  
111 higher spatio-temporal resolution observations (every 30 minutes) over Europe (Holmlund et al., 2021). A recent study by  
112 Guendouz et al. (2025) investigated the correlation between GIIRS NH<sub>3</sub> and surface skin temperature and showed the potential  
113 of discriminating sources between temperature-driven livestock/fertilizer volatilization and urea fertilizer application.

114 With over three years of FY-4B/GIIRS geostationary observations available from July 2022 to June 2025, unprecedented  
115 opportunities have emerged to monitor the spatial and temporal variations of NH<sub>3</sub> concentrations and their evolution  
116 throughout the day over East Asia. In this study, we highlighted the enhanced capability of GIIRS observations to identify  
117 emission sources and capture daytime variations associated with agricultural activities, surpassing those reported by polar-  
118 orbiting satellites. The data and methodology are described in Sect. 2. The spatial patterns and seasonal cycles of NH<sub>3</sub> observed  
119 by GIIRS are compared with observations from IASI and CrIS, as well as estimates from the mosaic Asian anthropogenic  
120 emission inventory (MIX) in Sect. 3.1. The data accuracy and temporal variations of satellite-derived NH<sub>3</sub> are validated against  
121 ground-based Fourier Transform Infrared (FTIR) measurements at the Hefei station in Sect. 3.2. The daytime variations of  
122 NH<sub>3</sub> from GIIRS observations and model simulations in major agricultural regions are discussed in Sect. 3.3. Conclusions are  
123 summarized in Sect. 4.

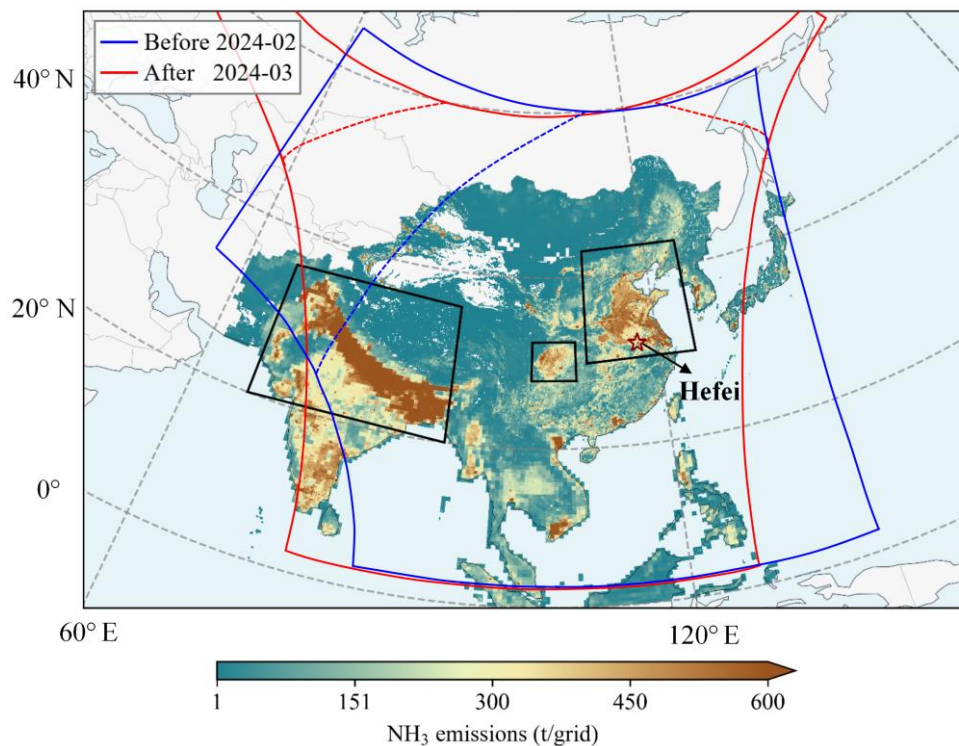
## 124 2 Data and Methodology

### 125 2.1 Satellite NH<sub>3</sub> retrievals

#### 126 2.1.1 GIIRS observations

127 FY-4B/GIIRS was successfully launched on 3 June 2021 and began operating at 133°E on 11 April 2022, providing  
128 observational data from 1 June 2022. As the FY-4A satellite approached fuel depletion, FY-4B was maneuvered from 133°E  
129 to 105°E between 1 February and 5 March 2024 to take over FY-4A's observation duties, with measurements temporarily  
130 suspended during this period. Since then, FY-4B/GIIRS has been operating nominally at 105°E. FY-4B/GIIRS observations  
131 primarily cover the regions from 3°N to 55°N and 60°E to 137°E, encompassing East Asia and parts of South and Southeast  
132 Asia (Fig. 1). The instrument performs measurements over the target region every 2 hours using a “step-stare” scanning mode.  
133 Observations are conducted along 12 scanning lines from north to south, with 27 fields of regard (FORs) arranged from west  
134 to east along each line. The FOR is a two-dimensional infrared detector array composed of 16 × 8 sparsely arranged pixels.  
135 Each pixel has a nadir-projected size of approximately 12 km × 12 km, with a spacing of 12 km in both along-track and cross-

136 track directions between adjacent pixels. Initially, 12 measurement cycles in a day started at 00:00, 02:00, 04:00, ..., 22:00  
137 UTC. After 6 September 2022, the start times were changed to 01:00, 03:00, 05:00, ..., 23:00 UTC, respectively.



138  
139 **Figure 1. Spatial domain of FY-4B/GIIRS observations and NH<sub>3</sub> retrievals over East Asia. Blue and red solid lines outline GIIRS**  
140 **observational coverage before and after the FY-4B orbital relocation, respectively. Blue and red dashed lines denote the**  
141 **corresponding NH<sub>3</sub> retrieval domains, with viewing zenith angles greater than 70° excluded. The background map shows total**  
142 **anthropogenic NH<sub>3</sub> emissions in 2017 from the MIXv2 emission inventory at a 0.1° grid resolution (Li et al., 2024). The red**  
143 **pentagram marks the location of the Hefei station, which is further described in Sect. 3.2. Black boxes outline major agricultural**  
144 **areas, including the North-Northeast China Plain, the Sichuan Basin, and the Indo-Gangetic Plain.**

145 Three years of NH<sub>3</sub> columns from July 2022 to June 2025 are retrieved from FY-4B/GIIRS spectra using the FengYun  
146 Geostationary satellite Atmospheric Infrared Retrieval (FY-GeoAIR) algorithm, which exploits the strong NH<sub>3</sub> absorption  
147 features in the 955–975 cm<sup>-1</sup> spectral region. FY-GeoAIR is a full-physics retrieval algorithm that integrates a forward radiative  
148 transfer model for simulating upwelling thermal radiation and an optimal estimation-based inverse model for retrieving trace  
149 gases and auxiliary parameters from the observed spectra (Zeng et al., 2023b). In this study, the retrieval configuration and  
150 parameters are identical to those described in Zeng et al. (2023a), except that a profile-scaling retrieval approach was adopted  
151 instead of the full-profile retrieval method. The full-profile retrieval method fully resolves the ammonia profile as 11 separate  
152 layers. In contrast, the profile-scaling retrieval approach only retrieves the total column of NH<sub>3</sub>. It applies a single  
153 representative a priori profile (identical for all scenes) and scales this profile in the retrieval algorithm to optimize the spectra  
154 fitting. In this case, there is only one state vector element for NH<sub>3</sub>, instead of 11. The reasons for this change of the retrieval

155 configuration are: (1) the degrees of freedom for signal (DOFS) for most NH<sub>3</sub> retrievals as shown in Zeng et al. (2023a) is less  
156 than one, indicating less than one piece of information is available from the spectra for NH<sub>3</sub> and not sufficient to constrain its  
157 vertical distribution; (2) using the profile-scaling retrieval approach speeds up the retrieval calculation. Similarly, only total  
158 column is retrieved for the interference gas H<sub>2</sub>O. The retrieval yields the NH<sub>3</sub> column and the posteriori error estimate. In  
159 addition, the column averaging kernel (AVK) is derived simultaneously in the optimal estimation framework. The column  
160 AVK value at a given atmospheric layer represents the change in the retrieved total NH<sub>3</sub> column with respect to a perturbation  
161 of the partial NH<sub>3</sub> column at that layer. It therefore reflects the height-dependent sensitivity determined by the temperature  
162 difference between the surface and the given atmospheric layer (Fig. A1 and Fig. S1). This definition of AVK is similar to that  
163 from the Total Carbon Column Observing Network (Wunch et al., 2011) and IASI (Clarisse et al., 2023), but differs from  
164 conventional matrix AVK from optimal estimation (Shephard et al., 2011, 2015). Further details on interpretation and  
165 calculation of the column AVK are provided in the Appendix.

166 In the NH<sub>3</sub> retrievals, regions with viewing zenith angles greater than 70° are excluded from the retrieval algorithm, as  
167 large zenith angles increase the atmospheric path length and scattering effects while reducing the signal-to-noise ratio and  
168 retrieval reliability. Observations that are cloud-contaminated or fail to converge within 10 iterations are also removed. To  
169 obtain high-quality GIRS NH<sub>3</sub> observations, we also applied the following filtering criteria: (1) a root mean square error  
170 (RMSE) of the fitting residual less than 0.2 K; (2) a reduced  $\chi^2$  less than 5; (3) retrieval error below 300%; (4) the absolute  
171 difference of the a priori and retrieved surface skin temperature less than 10 K; (5) the bottom layer (from surface to ~850 km)  
172 of column AVK greater than 0.1; (6) the thermal contrast (TC) greater than 0 K; (7) the retrieved NH<sub>3</sub> columns are positive.  
173 The retrieval error was calculated as the square root of the diagonal elements of the retrieval error covariance matrix for NH<sub>3</sub>,  
174 normalized by the ratio of the retrieved NH<sub>3</sub> column to the a priori NH<sub>3</sub> column. The TC is defined as the temperature difference  
175 between the surface and the lowest atmospheric layer. The strict quality filtering results in large data gaps over the Tibetan  
176 Plateau throughout the day, and over southern regions of China and India during 17:00–19:00 LST, where retrievals are  
177 frequently discarded due to low sensitivity or cloud screening.

178 In this study, NH<sub>3</sub> retrievals from individual observations over land during daytime (7:00–19:00 LST) were utilized to  
179 investigate spatial and temporal variations over East Asia (Fig. 2), whereas nighttime data were excluded due to insufficient  
180 thermal contrast. To facilitate regional analysis, individual retrievals were averaged onto a 0.5° × 0.5° grid. For the analysis of  
181 daytime NH<sub>3</sub> variations in Sect. 3.3, we further filtered the daytime observations by requiring TC > 5 K to ensure sufficient  
182 sensitivity to near-surface NH<sub>3</sub>. Figure S2 shows the number of data points before and after applying the TC > 5 K filtering  
183 criterion. The fractions of valid observations remaining in the North-Northeast China Plain, the Sichuan Basin, and the Indo-  
184 Gangetic Plain (as shown in Fig. 1) are about 83 %, 88 %, and 73 %, respectively.

## 185 2.1.2 IASI observations

186 The IASI instrument, jointly developed by the Centre National d'Études Spatiales (CNES) and the European Organisation  
187 for the Exploitation of Meteorological Satellites (EUMETSAT), was specifically designed for deployment aboard the Metop

188 series satellites. Three identical instruments were successively embarked on the Metop-A, Metop-B and Metop-C platforms,  
189 launched in 2006, 2012 and 2018, respectively. IASI operates in a sun-synchronous LEO at an altitude of approximately 800  
190 km, with a ground swath of  $\sim 2100$  km and a spatial resolution of 12 km at nadir. It measures the Earth's emitted infrared  
191 radiation across a spectral range of  $645\text{--}2760\text{ cm}^{-1}$  with an apodized resolution of  $0.5\text{ cm}^{-1}$  (Clerbaux et al., 2009). The near-  
192 real-time  $\text{NH}_3$  dataset is retrieved using the Artificial Neural Network for IASI (ANNI) framework. This method converts the  
193 hyperspectral signature of  $\text{NH}_3$ , quantified by the so-called hyperspectral range index (HRI), into total column amounts  
194 (Whitburn et al., 2016). Two independent validation studies demonstrated that IASI-retrieved  $\text{NH}_3$  columns show good  
195 consistency with both in situ and FTIR observations, exhibiting near-unity regression slopes and moderate biases (Guo et al.,  
196 2021; Wang et al., 2020). In this study, we utilized  $\text{NH}_3$  data from IASI observations on the Metop-B and Metop-C platforms  
197 from January 2017 to June 2025 for comparison with GIIRS observations. The data products provide total  $\text{NH}_3$  columns  
198 derived from the ANNI version 4 algorithm, which is about 15–20% larger than the previous version 3 due to the improved  
199 setup of HRI, but show very similar  $\text{NH}_3$  distribution (Clarisse et al., 2023). Here,  $\text{NH}_3$  columns marked as recommended by  
200 both pre-filter and post-retrieval quality flags in the data products were selected and then regridded onto a  $0.5^\circ$  spatial grid  
201 using arithmetic averaging. The quality flags allow some negative columns to be retained, as discussed by Clarisse et al. (2019)  
202 and Whitburn et al. (2016).

### 203 2.1.3 CrIS observations

204 The Cross-track Infrared Sounder (CrIS) is a Fourier transform spectrometer (FTS) jointly developed by the National  
205 Oceanic and Atmospheric Administration (NOAA) and the National Aeronautics and Space Administration (NASA), which  
206 provides hyperspectral infrared measurements at a spectral resolution of  $0.625\text{ cm}^{-1}$  with a nadir spatial resolution of  $\sim 14$  km.  
207 CrIS was first launched on the Suomi National Polar-orbiting Partnership (S-NPP) satellite on 28 October 2011 and  
208 subsequently deployed on NOAA-20 (JPSS-1) on 29 November 2017 and on JPSS-2 on 10 November 2022. The  $\text{NH}_3$  retrievals  
209 from CrIS observations on S-NPP and NOAA-20 have been generated using the CrIS Fast Physical Retrieval (CFPR) algorithm  
210 developed by Environment and Climate Change Canada (ECCC), which exploits spectral features near  $967\text{ cm}^{-1}$ . A detailed  
211 description of the CFPR algorithm can be found in Shephard et al. (2015, 2020). Independent validation studies against FTIR-  
212 derived total column measurements have demonstrated that CFPR (version 1.3) shows good overall consistency, with a  
213 correlation of approximately 0.8 and a slope of 1.02. For retrievals with total column values above  $1 \times 10^{16}\text{ molec/cm}^2$  (ranging  
214 from moderate to high levels), the relative bias is less than 5%. For smaller total column values ( $< 1 \times 10^{16}\text{ molec/cm}^2$ ), there  
215 are larger differences, with CrIS retrievals about 30% higher than FTIR values (Dammers et al., 2017a). However, these  
216 validations did not account for non-detects below the detection limit of the sensor ( $\sim 4 \times 10^{15}\text{ molec/cm}^2$  under typical remote  
217 sensing conditions) (Shephard et al., 2025; White et al., 2023), which could reduce potential high-biases over non-source  
218 conditions. In this study, we used CrIS  $\text{NH}_3$  data product from NOAA-20 (version 1.6.4), spanning the period from March  
219 2019 to April 2025. The product provides geolocation information, vertically integrated ammonia total columns, retrieval  
220 uncertainties, quality flags, and additional metadata. The dataset utilizes the information content of the satellite measurements

221 to explicitly identify and account for cloud-free observations below the sensor detection level (White et al., 2023). Our analysis  
222 focuses on NH<sub>3</sub> retrievals over land, characterized by land fraction > 0, quality flag ≥ 4, cloud flag ≠ 1, and DOFS ≥ 0.1.

## 223 2.2 Ground-based FTIR measurements at Hefei

224 The Hefei station (31.91°N, 117.17°E) is located in the northwestern rural area of Hefei city, China. The surrounding  
225 region is part of the typical Jianghuai agricultural region, where wetlands and croplands are interspersed in a distinctive  
226 landscape pattern. Since July 2015, a high-resolution ground-based Fourier transform infrared spectrometry (FTIR) has been  
227 operated at the station to record mid-infrared (MIR) solar absorption spectra (700–4000 cm<sup>-1</sup>) for the remote sensing of  
228 greenhouse gases and trace species (Wang et al., 2017). Previous study has demonstrated that FTIR measurements at Hefei  
229 effectively capture the spatiotemporal variability of NH<sub>3</sub> columns, and the retrievals are broadly consistent with IASI satellite  
230 data (Wang et al., 2022). In this study, FTIR-derived NH<sub>3</sub> column data from 2017 to 2024 were averaged to hourly means and  
231 used for the validation and cross-comparison of satellite-based NH<sub>3</sub> retrievals. It should be noted that the data primarily covers  
232 the period from 8:00 to 17:00 LST, with limited available observations between 17:00 and 18:00 LST.

## 233 2.3 NH<sub>3</sub> emission inventory

234 The MIXv2 Asian emission inventory was developed under the framework of the Model Inter-Comparison Study for Asia  
235 Phase IV (MICS-Asia IV) project by assembling a mosaic of up-to-date regional and national emission inventories (Li et al.,  
236 2024). It provides monthly estimates of NH<sub>3</sub> emissions for 2010–2017 at a spatial resolution of 0.1° × 0.1° grid across seven  
237 sectors. The inventory incorporated nine regional and two global emission inventories, covering 23 countries and regions in  
238 East, Southeast and South Asia. Anthropogenic NH<sub>3</sub> emissions were based on the best available emission inventories, including  
239 a process-based NH<sub>3</sub> emission inventory developed by Peking University (PKU-NH<sub>3</sub>) for China, the official Japan emission  
240 inventory (JPN), and the Clean Air Policy Support System (CAPSS) emissions for the Republic of Korea, and were gap-filled  
241 with REAS version 3 for Asia (REASv3). Studies have suggested that the spatial distribution of NH<sub>3</sub> emissions in the inventory  
242 is generally reliable, with differences compared to other emission inventories and satellite-constrained NH<sub>3</sub> emissions typically  
243 within 20–50% (Luo et al., 2022; Zhang et al., 2019; Kang et al., 2016).

## 244 2.4 Hourly NH<sub>3</sub> data from GEOS-CF model

245 The Goddard Earth Observing System composition forecast (GEOS-CF) system is a high-resolution global constituent  
246 prediction model developed by NASA's GMAO. It expands on the GEOS weather and aerosol modeling system by introducing  
247 the GEOS-Chem chemistry module, providing near-real-time, three-dimensional gridded information on atmospheric  
248 composition (Keller et al., 2021). The emission inputs of anthropogenic NH<sub>3</sub> emissions used by GEOS-CF are mainly from  
249 the Hemispheric Transport of Air Pollution emissions inventory (HTAP v2.2), which provides monthly data at a 0.1° spatial  
250 resolution. The model runs at hourly intervals with 0.25° × 0.25° horizontal resolution and 72 hybrid-eta levels from the surface

251 to 0.01 hPa. In this study, NH<sub>3</sub> vertical profiles were obtained from 1-day replay simulations (referred to as “hindcast”)  
252 constrained by pre-computed meteorological analysis fields, and NH<sub>3</sub> total columns were calculated by combining the model’s  
253 vertical pressure levels and surface pressures from the fifth-generation ECMWF reanalysis (ERA5) (Hersbach et al., 2023).

## 254 **2.5 The derivation of NH<sub>3</sub> variations**

### 255 **2.5.1. Curve-fitting Method for trend analysis**

256 To quantify the temporal changes in NH<sub>3</sub> columns, we applied a curve-fitting method to obtain daily time series of  
257 satellite-based NH<sub>3</sub> observations. This method is widely used to extract temporal variation characteristics of atmospheric  
258 composition, such as NH<sub>3</sub> (Wang et al., 2022; Van Damme et al., 2015). As shown in Eq. (1), it integrates a polynomial function  
259 and a sum of harmonic function, representing long-term linear trend and seasonal cycles, respectively.

$$260 \quad f(t) = a_0 + a_1 t + \sum_{n=1}^4 (\beta_n \sin(2n\pi t) + \gamma_n \cos(2n\pi t)) \quad (1)$$

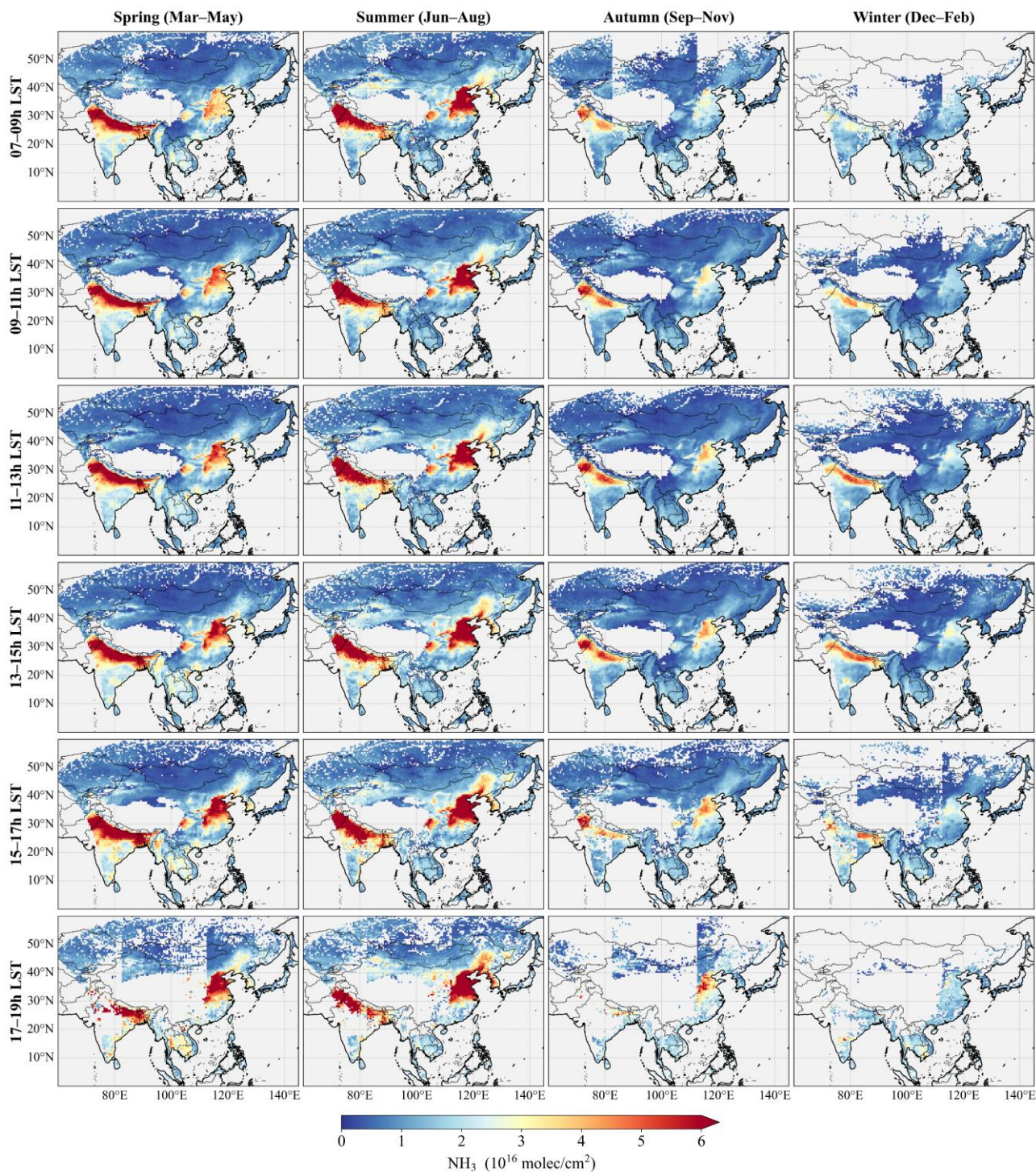
261 where  $f$  is the daily fitting result,  $t$  is the time in decimal years, the coefficients ( $a_0$ ,  $a_1$ ,  $\beta_n$ ,  $\gamma_n$ ) are determined by the least  
262 squares regression. In Sect. 3.1, the fitting analysis was performed separately for each  $0.5^\circ \times 0.5^\circ$  grid cell, with more than  
263 500 data points and observation dates spanning over 100 days. Extreme values and minima of the fitted curves were then  
264 calculated to identify temporal inflection points and determine the months with peak NH<sub>3</sub> concentrations.

### 265 **2.5.2. Gridded NH<sub>3</sub> enhancements**

266 Based on the spatial distribution of NH<sub>3</sub> emissions and concentrations, the study domain was divided into 14 subregions  
267 (Fig. 6). Monthly variations of NH<sub>3</sub> columns for each subregion were obtained by aggregating the daily fitted values calculated  
268 in Sect. 2.5.1, which represent the regional background levels. NH<sub>3</sub> anomalies from individual measurements were then  
269 calculated by subtracting NH<sub>3</sub> backgrounds, as defined in Eq. (2).

$$270 \quad A_i = X_i - BK_{r,m} \quad (2)$$

271 where  $A_i$  is the NH<sub>3</sub> anomaly for the individual  $i$ -th retrieval,  $X_i$  is the observed NH<sub>3</sub> column, and  $BK_{r,m}$  represents the NH<sub>3</sub>  
272 background for the region  $r$  and month  $m$  corresponding to the spatial location and observation time of retrieval  $i$ . For each  
273 grid cell, positive anomalies with occurrences of elevated concentrations ( $> 1.0 \times 10^{16}$  molec/cm<sup>2</sup>) exceeding 30 days were  
274 classified as NH<sub>3</sub> enhancements. The frequency of such occurrences, expressed in number of days, serves as a useful indicator  
275 of enhanced emissions, such as recurring agricultural activities (Shephard et al., 2020; Warner et al., 2016).



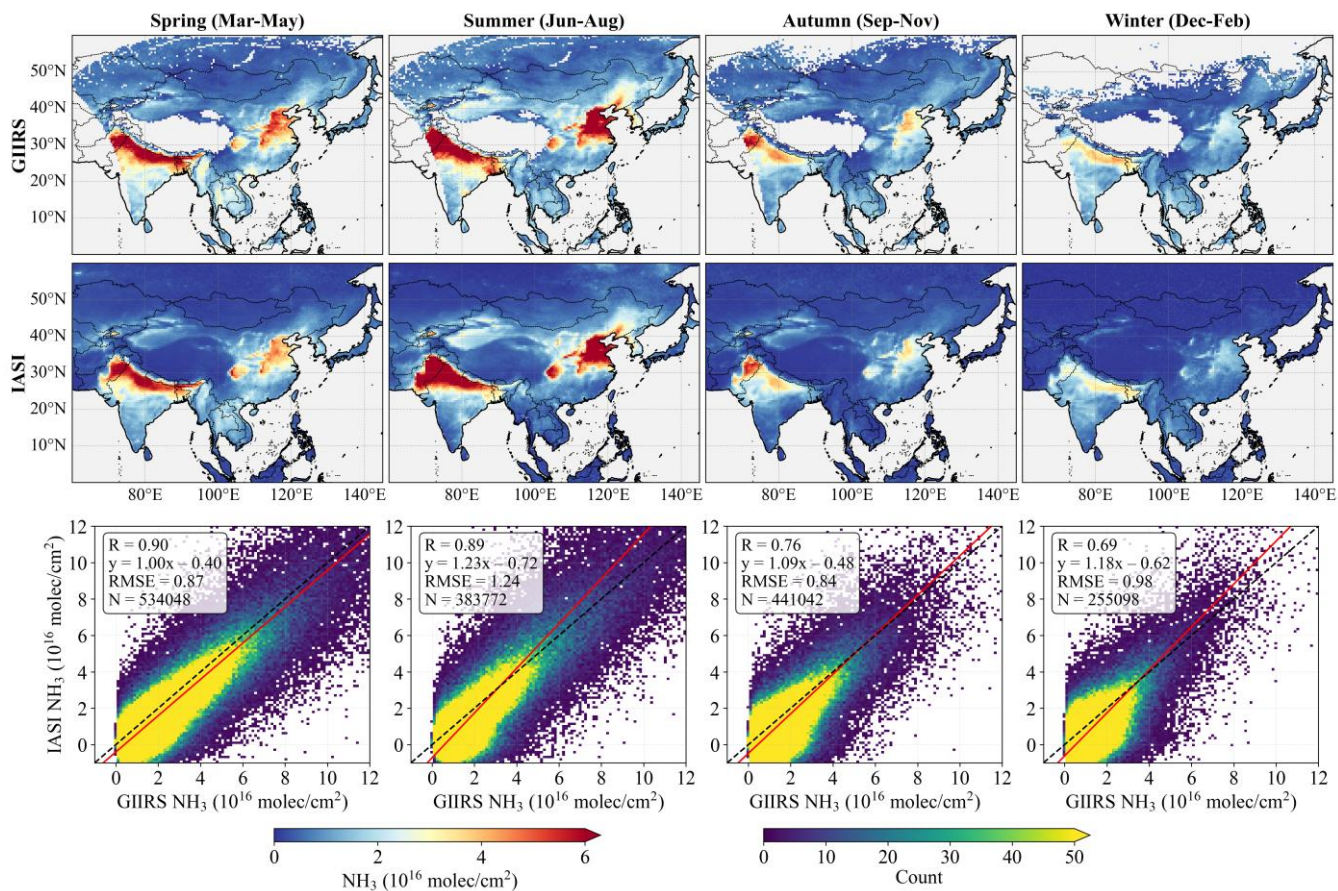
276  
 277 **Figure 2. Seasonal maps of NH<sub>3</sub> columns from FY-4B/GHRS observations from July 2022 to June 2025 averaged for every 2 hours**  
 278 **according to local solar time (LST) on a 0.5° by 0.5° grid and for different seasons (spring: March-April-May; summer: June-July-**  
 279 **August; autumn: September-October-November; winter: December-January-February).**

## 280 **3 Result and discussion**

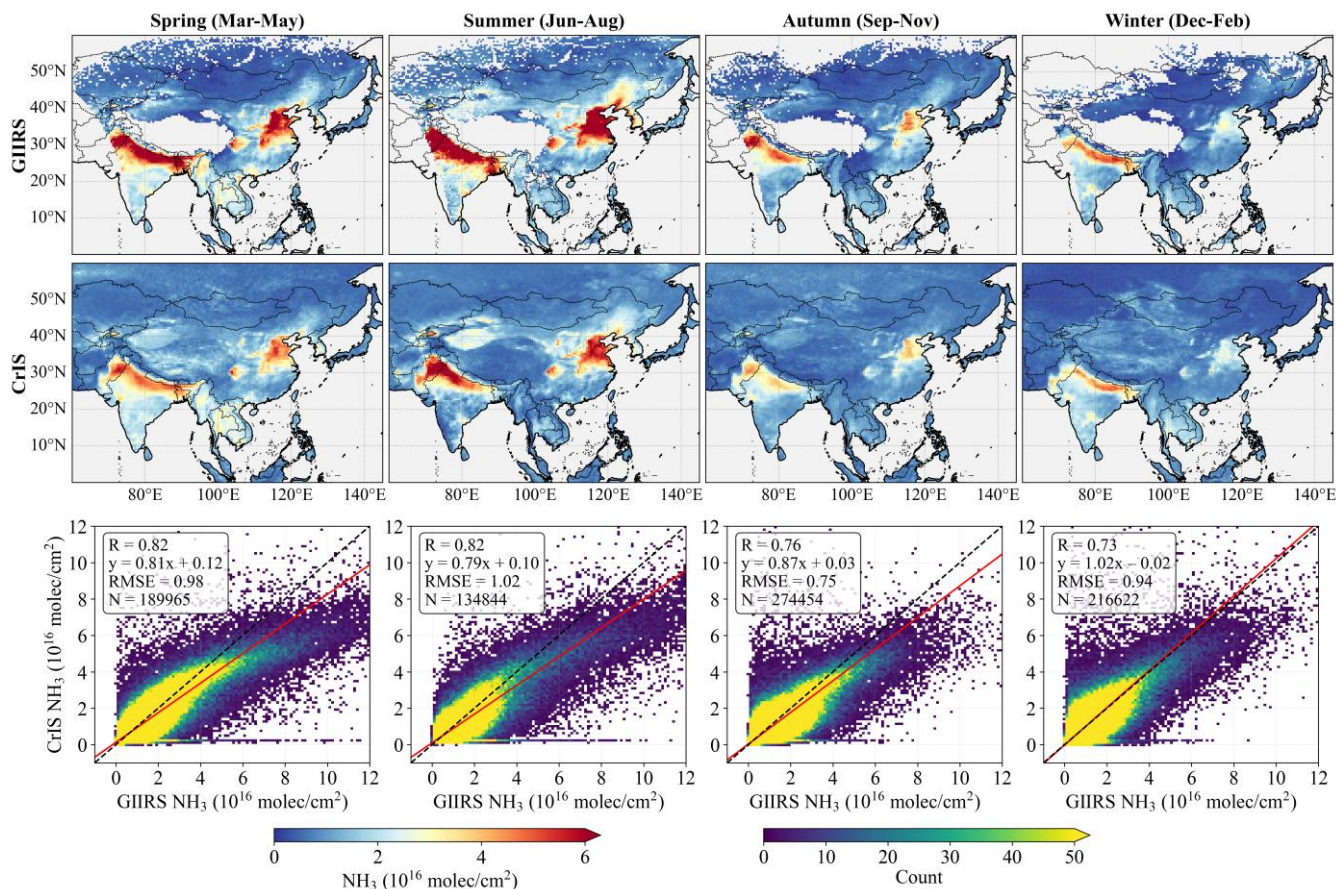
### 281 **3.1 Spatial and temporal NH<sub>3</sub> distribution over East Asia**

282 [Figure 2](#) illustrates the three-year averaged seasonal NH<sub>3</sub> column concentrations retrieved from FY-4B/GIIRS  
283 observations over East Asia from July 2022 to June 2025, presented on a 0.5°×0.5° grid at two-hour intervals. **The observations**  
284 **are aggregated according to local solar time (LST) to facilitate the analysis of NH<sub>3</sub> diurnal variations and consistent**  
285 **comparisons. The study region spans multiple time zones and data availability during the 7–9 h and 17–19 h periods is**  
286 **substantially reduced due to retrieval filtering, resulting in noticeable boundaries across longitudes.** Elevated NH<sub>3</sub> columns,  
287 significantly above background levels, are observed over the Indo-Gangetic Plain, the North China Plain, the Sichuan Basin,  
288 and the Northeast China Plain (as shown by the black boxes in [Fig. 1](#)). The daytime NH<sub>3</sub> columns reach values exceeding  
289 4×10<sup>16</sup> molec/cm<sup>2</sup>, primarily driven by intensive agricultural activities and supplemented by non-negligible contributions from  
290 urban and industrial emissions. Localized weaker enhancements of NH<sub>3</sub> column concentrations are evident over agricultural  
291 core areas in China (including the Ningxia Irrigation Plain, the Wei River Plain, the Jiangnan Plain, and oasis agriculture in  
292 the arid regions of Xinjiang), as well as the central Deccan Plateau in India, the Mekong Delta in Vietnam, the Chao Phraya  
293 River Plain in Thailand, and the Fergana Valley in Uzbekistan (as shown by the dashed boxes in [Fig. S4](#)). Most of these regions  
294 exhibit higher NH<sub>3</sub> columns in summer and considerable daytime variability, with large differences between morning and  
295 afternoon observations. A detailed analysis of diurnal patterns over major source regions is provided in [Sect. 3.3](#).

296 Comparisons with the spatial distribution of NH<sub>3</sub> columns observed by polar-orbiting infrared sounders of IASI and CrIS  
297 show reasonable agreement, especially in regions with high NH<sub>3</sub> columns above 3×10<sup>16</sup> molec/cm<sup>2</sup> ([Fig. 3](#) and [Fig. 4](#)). The  
298 seasonal increases in atmospheric NH<sub>3</sub> are most pronounced in spring and summer. Despite generally similar seasonal and  
299 spatial patterns, inter-satellite NH<sub>3</sub> observations exhibit noticeable differences. Relative to the IASI observations, GIIRS  
300 typically shows very small systematic differences with fitted slopes close to unity. In contrast, GIIRS observations tend to  
301 show higher NH<sub>3</sub> columns than CrIS in most high-concentration regions, with mean differences of approximately 25–50%  
302 relative to the corresponding GIIRS values in summer. The data point scattering in the comparison plots primarily results from  
303 spectral noise. Overall, there is good agreement between GIIRS-derived NH<sub>3</sub> columns and those from IASI and CrIS across  
304 different seasons.



305  
 306 **Figure 3. Comparison of  $\text{NH}_3$  columns observed by GHIRS and IASI during morning overpasses ( $\sim 09:30$  LST) from July 2022 to**  
 307 **June 2025. Seasonal maps are presented on a  $0.5^\circ \times 0.5^\circ$  grid. Scatter plots compare GHIRS and IASI data matched by overpass**  
 308 **times within 1 hour and spatial locations within the same  $0.5^\circ \times 0.5^\circ$  grid cells.**



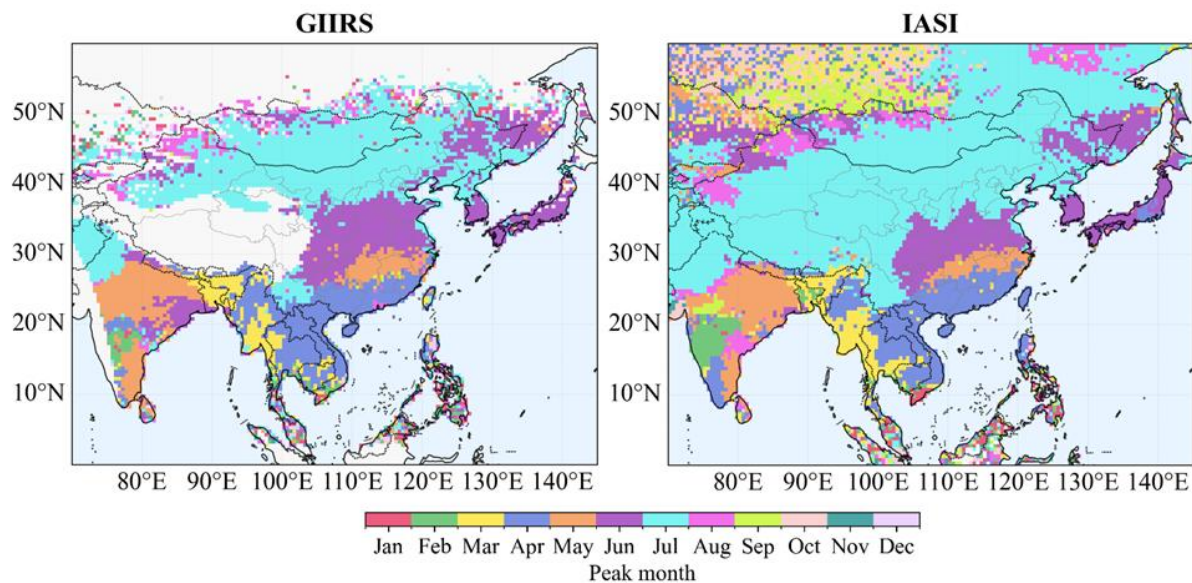
309  
 310 **Figure 4. Comparison of NH<sub>3</sub> columns observed by GIIRS and CrIS during afternoon overpasses (~13:30 LST) from July 2022 to**  
 311 **April 2025. Seasonal maps are presented on a 0.5° × 0.5° grid. Scatter plots compare GIIRS and CrIS data matched by overpass**  
 312 **times within 1 hour and spatial locations within the same 0.5° × 0.5° grid cells.**

313 To account for the effects of differing vertical sensitivities, as reflected by the AVKs, and a priori profiles among these  
 314 satellites (see Fig. S1 for example), we conducted experiments using model profile data and satellite AVKs over the North-  
 315 Northeast China Plain (30°–43°N, 110°–125°E) in June 2024 (Text S1). Model simulations of NH<sub>3</sub> profiles were convolved  
 316 with different satellite AVKs to generate AVK-smoothed NH<sub>3</sub> columns, representing the NH<sub>3</sub> columns that a certain satellite  
 317 would retrieve if the model simulations were the “truth”. We then compared AVK-smoothed NH<sub>3</sub> column datasets generated  
 318 using different satellite AVKs to evaluate the effects of different satellite vertical sensitivities. The comparison results (Fig. S5)  
 319 show high correlations ( $R > 0.9$ ) and slopes close to 1. This indicates consistency in the detection of NH<sub>3</sub> across different  
 320 sensors, and that the effects of different AVKs and a priori profiles are small in terms of total column retrieval when comparing  
 321 different satellite retrievals.

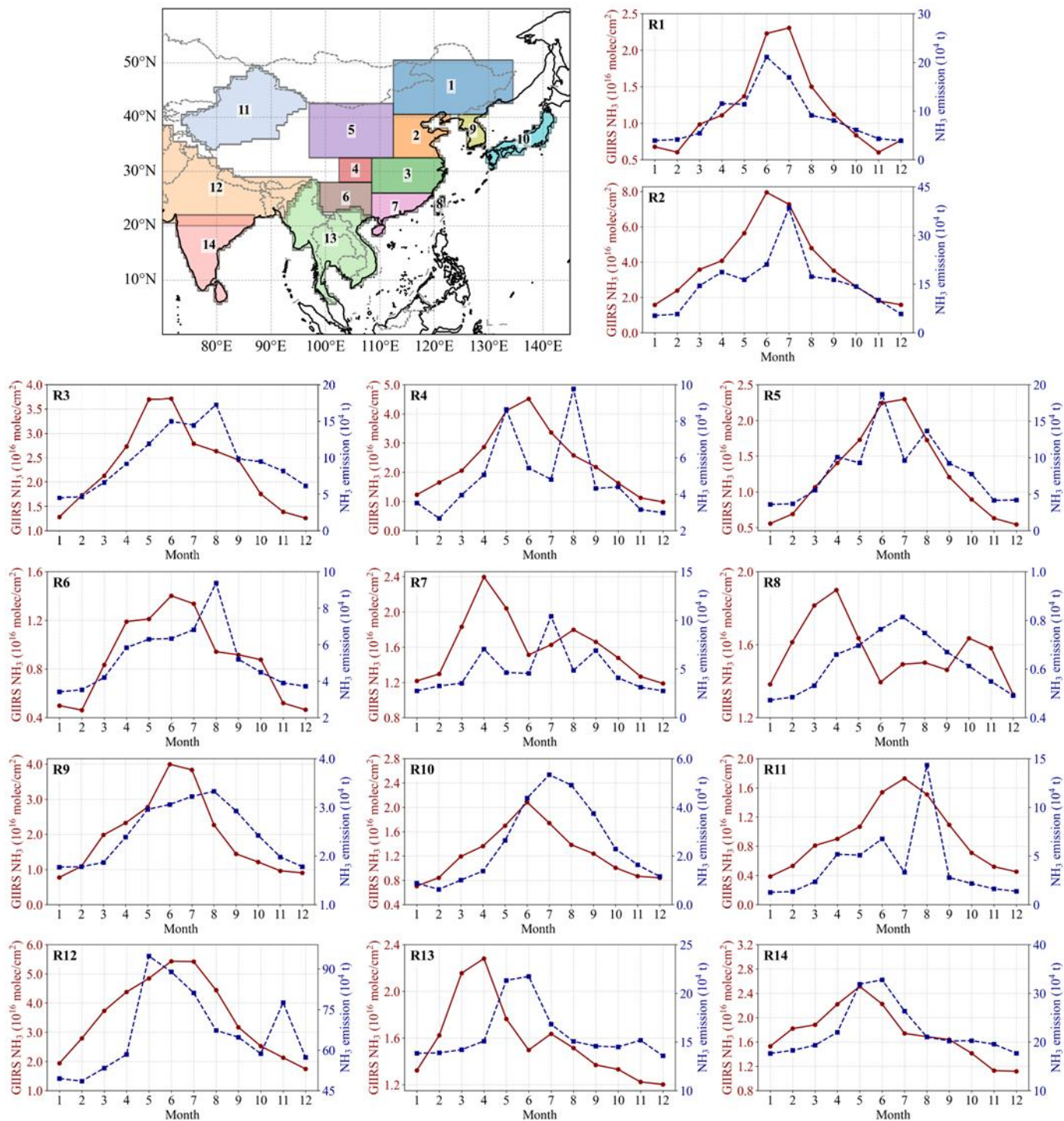
322 Figure 5 illustrates the months corresponding to the maximum NH<sub>3</sub> concentrations derived from GIIRS and IASI at a 0.5°  
 323 grid, while the result from CrIS, which is highly scattered at this scale, is shown on a 1° grid in Fig. S6. The spatial pattern  
 324 shows a clear north-south gradient in the timing of NH<sub>3</sub> peaks. In the northern regions of East Asia, NH<sub>3</sub> columns generally

325 peak in June and July, which could be attributed to enhanced volatilization from agricultural soils, livestock operations, and  
326 other sources (e.g., composting facilities and wastewater treatment plants) under high-temperature conditions (Li et al., 2024).  
327 Irrigated croplands in the North and Northeast China Plains also exhibit a secondary spring maximum (Fig. S7) associated  
328 with extensive fertilizer use and animal feeding operations. In contrast, the regions of southern China, India, and Southeast  
329 Asia, characterized by a tropical monsoon climate, exhibit earlier NH<sub>3</sub> peaks, generally occurring from March to May. The  
330 peak period coincides with their first cropping season, which involves intensive nitrogen fertilizer application. The warm and  
331 humid conditions in these regions further promote NH<sub>3</sub> volatilization from soils and livestock waste. Following the onset of  
332 the rainy season in June-July, atmospheric NH<sub>3</sub> concentrations decline sharply (such as R7, R8, R13, R14 in Fig. 6), primarily  
333 due to enhanced wet scavenging processes. This seasonal pattern aligns with findings from multiple studies reporting a distinct  
334 “late dry season-early wet season” NH<sub>3</sub> maximum in tropical regions (e.g., Chen et al., 2020; Liu et al., 2019). Spring is a  
335 period of intense fire activity in the Indochina Peninsula, which is largely driven by residue burning, land-clearing, and  
336 naturally flammable vegetation during the dry season (Chang et al., 2021; Vadrevu et al., 2019). The NH<sub>3</sub> maxima mainly  
337 occur in March-April, and the consistent spatial distribution of these NH<sub>3</sub> peaks observed by GIIRS and IASI over Southeast  
338 Asia suggests the influence of biomass burning associated with fires on its seasonal variations. In addition, bimodal local  
339 maxima, with peaks of comparable magnitude, are also observed in regions such as central India (from May to September),  
340 Guangdong in southern China (typically in April and August), and the transboundary region between the Mekong Delta in  
341 Vietnam and Yunnan in China (March-April and June-July).

342 In general, the spatial and temporal distributions of NH<sub>3</sub> peaks largely agree with the previous studies of Van Damme et  
343 al. (2015) and Shephard et al. (2020). Minor discrepancies between GIIRS and IASI result from differences in observational  
344 accuracy and retrieval algorithms, as discussed above. Satellite-observed NH<sub>3</sub> seasonal cycles are driven by the interplay of  
345 agricultural activities and climatic conditions, leading to deviations from seasonal patterns of anthropogenic emissions (Fig.  
346 S8). Several studies have reported that NH<sub>3</sub> emissions inferred from satellite observations exhibit distinct seasonal variations  
347 that can differ from those of the NH<sub>3</sub> columns (Li et al., 2026; Kumar et al., 2025). Therefore, we focus on the spatial  
348 distribution of NH<sub>3</sub> enhancements, relative to regional background levels, as captured by different satellites.



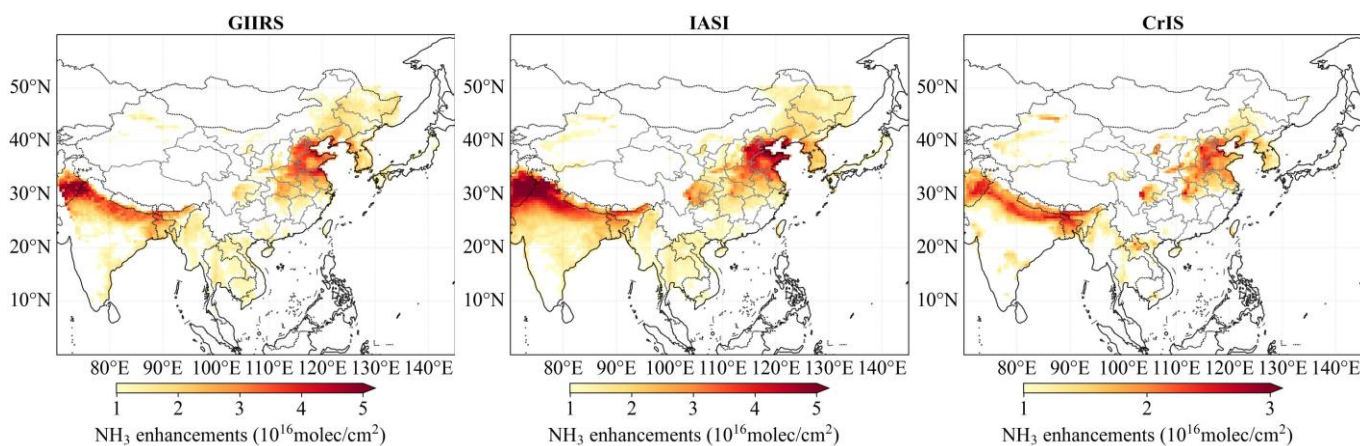
349  
 350 **Figure 5. The months with NH<sub>3</sub> column peaks observed by FY-4B/GIIRS from July 2022 to June 2025 and IASI from January 2022**  
 351 **to June 2025 on a 0.5° × 0.5° grid. GIIRS results for the full daytime period (07:00–19:00 LST) are presented here for clarity, as**  
 352 **morning and afternoon observations show consistent seasonal patterns (not shown).**



353  
354  
355

Figure 6. Monthly variations of GHIRS-derived  $\text{NH}_3$  columns from July 2022 to June 2025 and  $\text{NH}_3$  total emissions from the MIX inventory in 2017. Colored boxes specify spatial domains of 14 subregions (Sect. 2.5.2).

356 After removing monthly backgrounds derived from predefined regions (Fig. S9), NH<sub>3</sub> enhancements (Fig. 7) reveal a  
357 spatial pattern of anthropogenic emissions that is highly consistent with the MIX emission inventory (Fig. 1). Apart from the  
358 major agricultural emission regions mentioned above, some regions (e.g. Hyderabad and Vijayawada in India) also show  
359 elevated NH<sub>3</sub> levels compared to the background during autumn and winter, likely influenced by dense urban and industrial  
360 activities. Due to data discrepancies in local overpass times and sensor sensitivities among the polar-orbiting and geostationary  
361 satellites (Shephard et al., 2025), the enhancement values detected by CrIS are  $0.50 \pm 0.64$  and  $0.74 \pm 0.90 \times 10^{16}$  molec/cm<sup>2</sup>  
362 lower than those detected by GIIRS and IASI, respectively. These inter-satellite differences can lead to inconsistencies in the  
363 observed NH<sub>3</sub> enhancements across various source regions, particularly in the Sichuan Basin, the Ningxia Irrigation Plain, and  
364 the urban and biomass-burning regions of Southeast Asia. Compared with IASI and CrIS observations at different local times,  
365 GIIRS provides consistent and reliable measurements throughout the full daytime, enabling accurate characterization of spatial  
366 and temporal variations in NH<sub>3</sub> concentrations across distinct emission source regions.



367  
368 **Figure 7. Spatial distribution of NH<sub>3</sub> enhancements derived from satellite observations (GIIRS, IASI, and CrIS). Note that the color**  
369 **bar for CrIS differs from those for GIIRS and IASI, and abnormal NH<sub>3</sub> enhancements over the Tibetan Plateau have been excluded.**

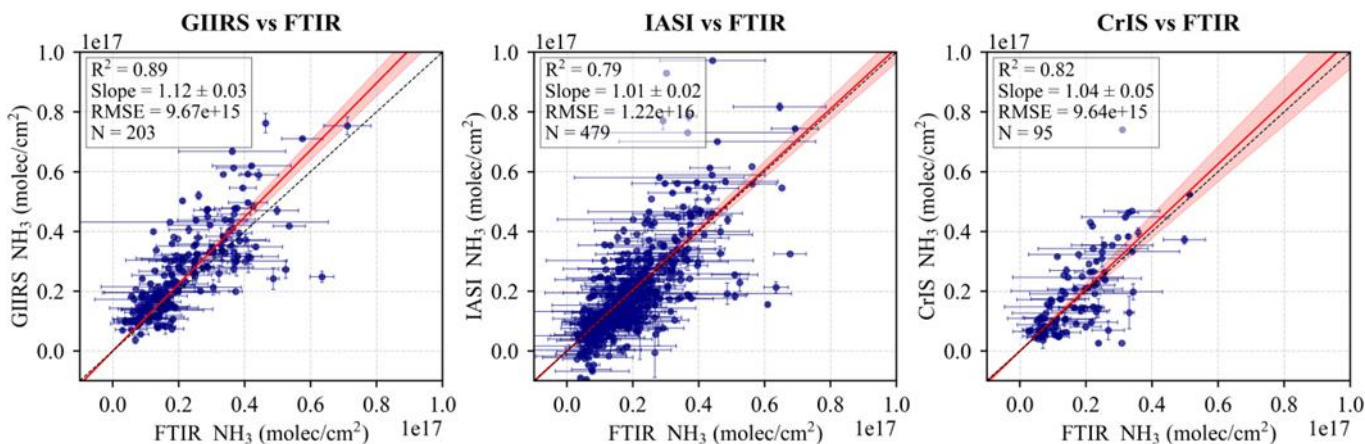
### 370 3.2 Inter-comparison of NH<sub>3</sub> variations at the Hefei station

371 To evaluate the data accuracy of GIIRS-derived NH<sub>3</sub> columns and assess the consistency of temporal variations with  
372 polar-orbiting satellite measurements, we compared the satellite-based observations with ground-based FTIR measurements  
373 at the Hefei station. Satellite data within a 0.25° latitude/longitude radius around the Hefei station (Fig. S10) were selected and  
374 averaged on an hourly scale. A linear regression through the origin was applied to evaluate the proportional agreement between  
375 the datasets.

376 Figure 8 shows scatter plots comparing NH<sub>3</sub> columns retrieved from satellite observations and FTIR measurements during  
377 their overlapping periods. Overall, the satellite-derived NH<sub>3</sub> columns show good agreement with the FTIR data, with Pearson  
378 correlation coefficients (R) exceeding 0.70, although satellite values are generally slightly higher. Among the satellites, GIIRS  
379 agrees best with FTIR in terms of the highest correlation and the low root mean square error (RMSE). However, GIIRS exhibits  
380 a relative systematic bias toward overestimating NH<sub>3</sub> columns, as indicated by its regression slope (1.12) being notably larger

381 than those of IASI and CrIS. When measurement uncertainties were taken into account, regression analysis using the  
382 orthogonal distance regression (ODR) method yielded an increased slope of 1.24 (Fig. S11), indicating that GIIRS tends to  
383 produce systematically higher NH<sub>3</sub> columns than IASI and CrIS.

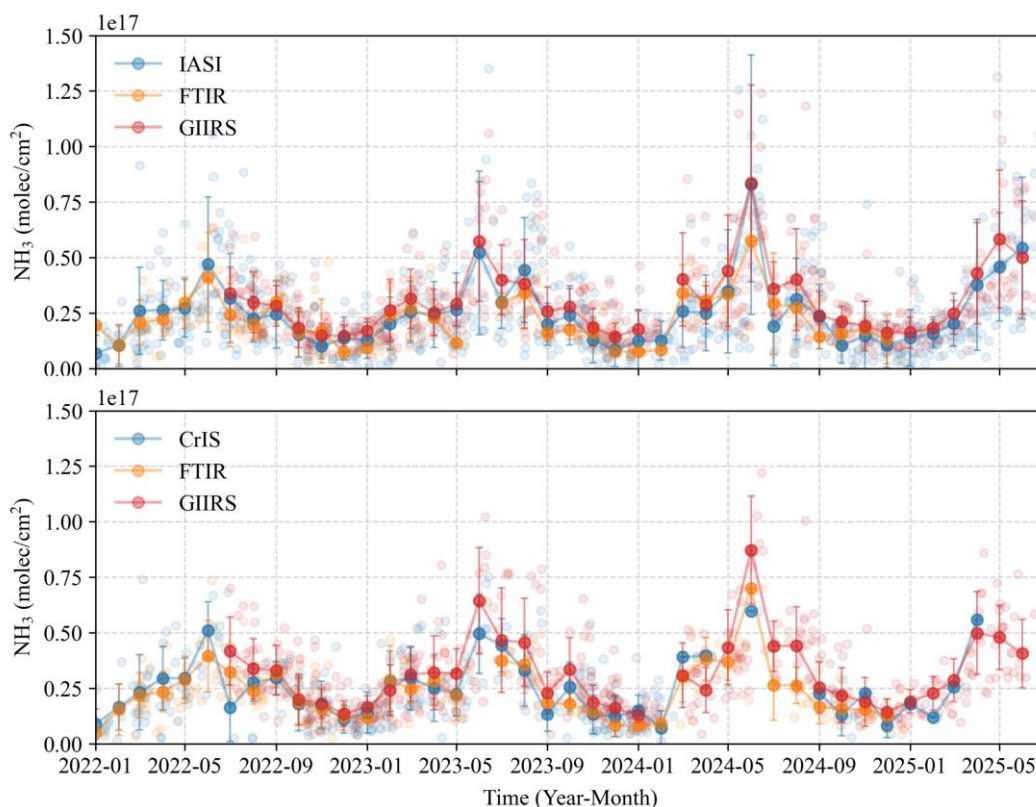
384 Although the overestimation of GIIRS becomes more pronounced in afternoon observations (Fig. S12), the corresponding  
385 data conversely show improved consistency, which is characterized by higher R<sup>2</sup> and lower RMSE values. This is related to  
386 the fact that the inversion accuracy of GIIRS observations is affected by TC and boundary layer conditions. High NH<sub>3</sub> columns  
387 and significant TC represent favorable conditions for retrieving NH<sub>3</sub> from satellite observations (Zeng et al. 2023a; Clarisse  
388 et al., 2010). As illustrated in Fig. S13, large positive TC between 11:00 and 15:00 LST enhances measurement sensitivity,  
389 leading to lower retrieval errors. **GIIRS NH<sub>3</sub> column retrievals are most reliable when TC exceeds 5 K, a condition**  
390 **predominantly observed during summer daytime hours (7:00–19:00 LST).**



391  
392 **Figure 8. Cross-validation of hourly NH<sub>3</sub> columns from GIIRS, IASI, and CrIS with FTIR measurements at the Hefei station. The**  
393 **overlapping periods span January 2017 (IASI), March 2019 (CrIS), and July 2022 (GIIRS) to December 2024. GIIRS data**  
394 **encompass all daytime hours, while IASI and CrIS data correspond specifically to their respective overpass times.**

395 The Hefei station and its surrounding regions predominantly adopt a rice-wheat rotation, with rice and winter wheat  
396 typically sown in spring (around May) and autumn (around October), respectively. According to the MIX emission inventory,  
397 agricultural ammonia emissions in this region exhibit two primary peaks in June and August, which are associated with local  
398 agricultural practices including livestock waste management, nitrogen fertilizer application and irrigation activities during the  
399 rice growing season (Hou et al., 2020). Smaller peaks occur in October-November and April, corresponding to the rice  
400 harvest/wheat sowing period and the wheat regreening fertilization stage, respectively. Driven jointly by agricultural emission  
401 intensity and meteorological conditions such as high temperatures, atmospheric NH<sub>3</sub> concentrations observed by satellites and  
402 ground-based FTIR show consistent seasonal cycles, peaking in summer (June-August) and reaching a minimum in winter  
403 (November-January). Figure 9 shows the monthly NH<sub>3</sub> time series observed by satellites and FTIR at different local overpass  
404 times. GIIRS and IASI show closely matched NH<sub>3</sub> peaks in June. The systematic overestimation of peak values by GIIRS  
405 relative to CrIS and FTIR is consistent with an inherent data bias as shown in Fig. 8.

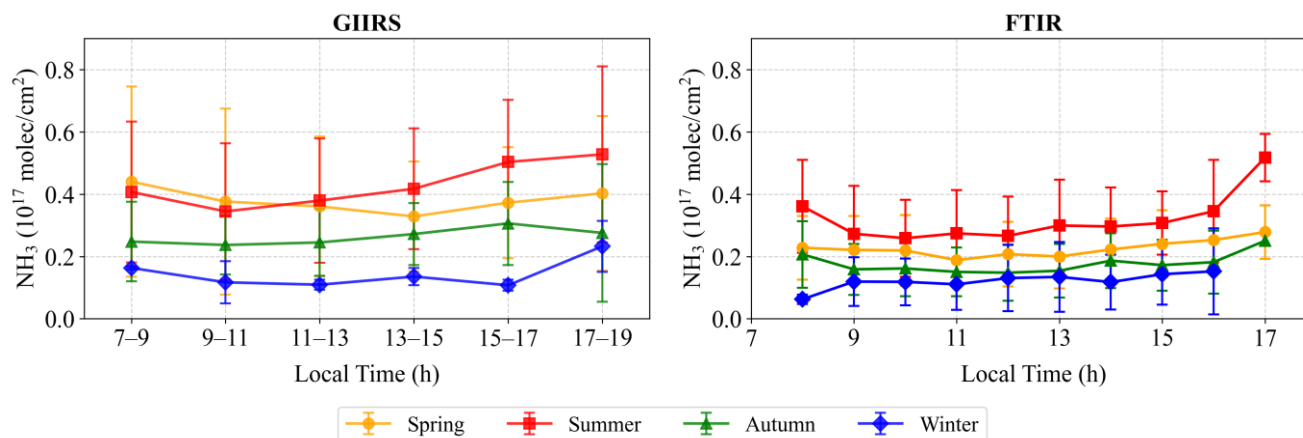
406 Notably, GIIRS-derived  $\text{NH}_3$  columns in June 2024 were significantly higher than those in the same month of 2023 and  
 407 2025, with increases of 1.46- and 1.67-times during morning overpass time ( $\sim 9:30$  LST), and 1.35- and 2.15-times during  
 408 afternoon overpass time ( $\sim 13:30$  LST), respectively. Such pronounced variability is likely linked to extreme weather events  
 409 reported in the region, such as persistent heatwaves and abrupt drought–flood transitions (Zhou et al., 2025; Ding et al., 2024).  
 410 Morning observations from IASI align with GIIRS data, showing that  $\text{NH}_3$  concentrations in June 2024 reached 1.43 to 2.62  
 411 times the levels recorded in the years 2017–2025 (specifically, 1.59 and 1.53 times those of 2023 and 2025, respectively).  
 412 Similar interannual changes were observed by CrIS and FTIR in June 2024, with  $\text{NH}_3$  levels increasing by factors of 1.17–  
 413 1.53 and 1.40–2.28 compared with previous years, respectively, despite a data gap in June 2023 for FTIR.



414  
 415 **Figure 9. Time series of monthly  $\text{NH}_3$  columns at the Hefei station from satellite-based and ground-based observations,**  
 416 **corresponding to different local overpass times of polar-orbiting satellites for morning ( $\sim 9:30$  LST) and afternoon ( $\sim 13:30$  LST).**  
 417 **The light-colored, semi-transparent points represent daily mean values at different local times.**

418 Comparing GIIRS-based  $\text{NH}_3$  columns at different local times, afternoon concentrations are generally higher than those  
 419 in the morning, with the largest differences observed in summer (July–August) and the smallest in autumn and winter. To more  
 420 accurately characterize daytime  $\text{NH}_3$  variations, we further applied strict data filtering criteria by retaining only retrievals with  
 421  $\text{TC} > 5$  K to ensure high sensitivity to near-surface  $\text{NH}_3$ . Over the three-year period, 37 high-quality observation days were  
 422 identified, each providing six consecutive two-hour interval observations from 07:00 to 19:00 LST (Fig. S14). The maximum

423 diurnal amplitude occurs in summer, reaching over  $1.5 \times 10^{16}$  molec/cm<sup>2</sup>. Except for autumn, all three other seasons exhibit  
424 relatively high NH<sub>3</sub> concentrations around 7:00 and 17:00 LST, a pattern that aligns well with the diurnal variations reported  
425 by Wang et al. (2022) based on FTIR measurements (Fig. 10). However, NH<sub>3</sub> retrievals from FTIR lack sufficient valid  
426 observations within a single day, making it difficult to consistently capture the intra-day variability in NH<sub>3</sub> across different  
427 dates.



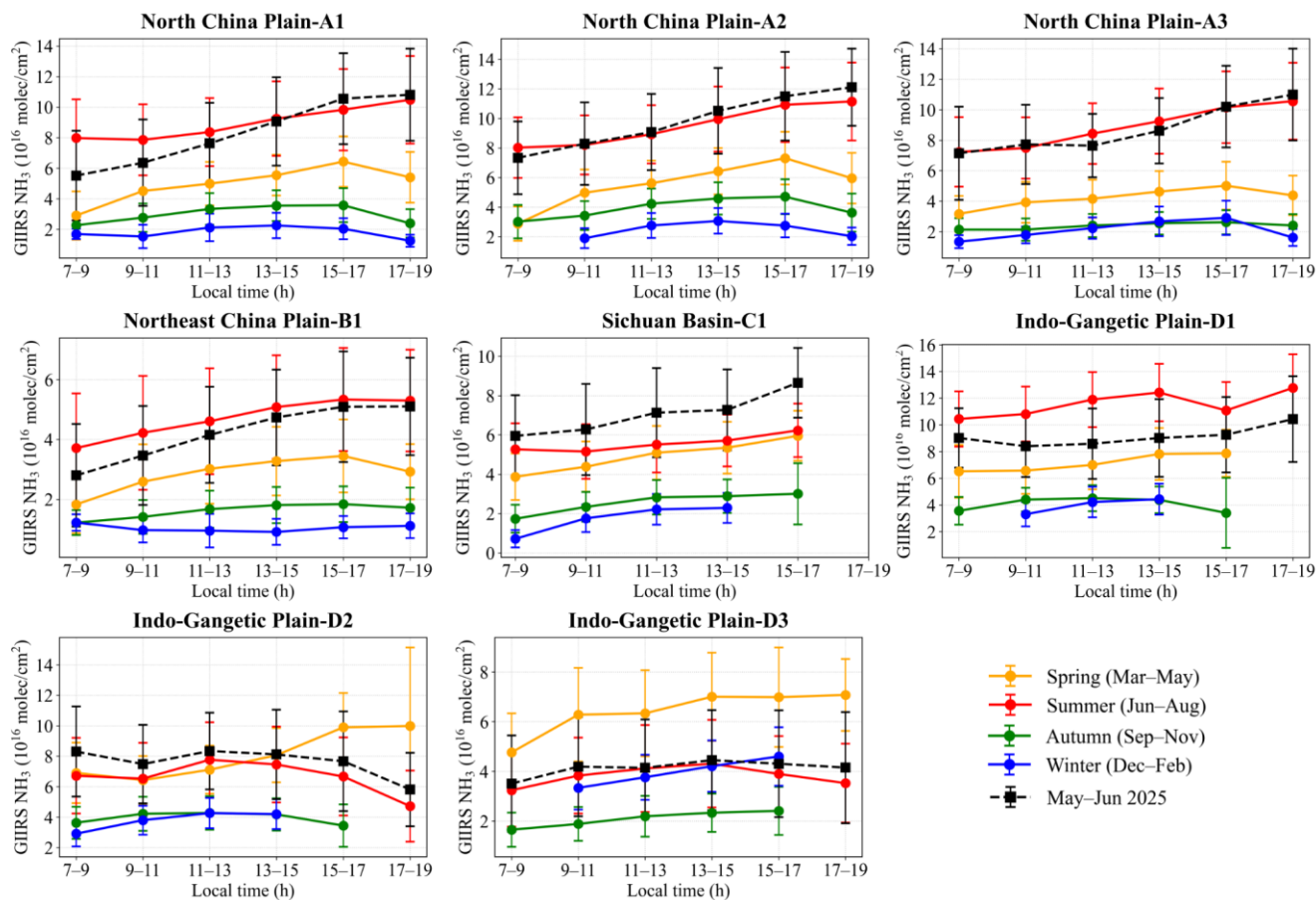
428  
429 **Figure 10. Daytime NH<sub>3</sub> variations observed by GIIRS and FTIR in different seasons. The seasonal mean NH<sub>3</sub> from GIIRS are**  
430 **calculated based on 37 high-quality days in Fig. S14.**

431 Moreover, the FTIR station is situated in a transitional zone between the urban and suburban areas. A spatial domain with  
432 a radius of  $0.25^\circ$  used in this study encompasses GIIRS sampling points from both urban and agricultural regions (Fig. S10).  
433 To assess spatial variability of NH<sub>3</sub>, GIIRS retrievals with TC > 5K within this spatial domain were classified into urban and  
434 non-urban categories, and their daytime variations were statistically analyzed across seasons over three years. As shown in Fig.  
435 S15, NH<sub>3</sub> concentrations in non-urban areas are consistently and significantly higher than those in urban areas during spring  
436 and summer, indicating the strong contribution of agricultural activities to ambient NH<sub>3</sub> levels. Particularly in spring,  
437 statistically significant differences are most pronounced during 7:00–9:00 and 17:00–19:00 LST, corresponding to time  
438 windows of fertilizer application and livestock activity. During winter, higher NH<sub>3</sub> levels in the morning and late afternoon are  
439 mainly observed in urban areas, likely reflecting the impact of traffic emissions. These findings highlight the importance of  
440 high-frequency satellite observations for dynamically monitoring of NH<sub>3</sub> variations, and the contribution of agricultural  
441 activity in shaping the diurnal NH<sub>3</sub> variability.

### 442 3.3 Daytime NH<sub>3</sub> variations in major agricultural regions

443 We focused on eight major agricultural source areas located in the North China Plain and the Northeast China Plain, the  
444 Sichuan Basin, and the Indo-Gangetic Plain, to further investigate daytime variations in NH<sub>3</sub> columns. These areas are  
445 characterized by high NH<sub>3</sub> column concentrations and emissions, generally flat terrain, and a large proportion of irrigated  
446 cropland (as shown in Figs. S16–17). To ensure sufficient measurement sensitivity to near-surface NH<sub>3</sub>, only GIIRS

447 observations with TC greater than 5 K were retained, thereby excluding cases with low information content. During autumn  
 448 and winter, available observations are sparse in the early morning and late afternoon. Here, GIIRS-observed daytime variations  
 449 of  $\text{NH}_3$  columns across different seasons (Fig. 11) and the corresponding spatial distributions for each study region (Figs. 12–  
 450 14, Figs. S18–20) are presented, with comparisons to anthropogenic  $\text{NH}_3$  emissions (Fig. S17) and GEOS-CF model  
 451 simulations (Figs. S21–24). Despite significant differences between model-simulated and satellite-based  $\text{NH}_3$  columns, we  
 452 focused on analyzing the daytime variations and their spatial distributions in representative months of May–June ( $\text{NH}_3$  peak  
 453 months) in 2025.  
 454



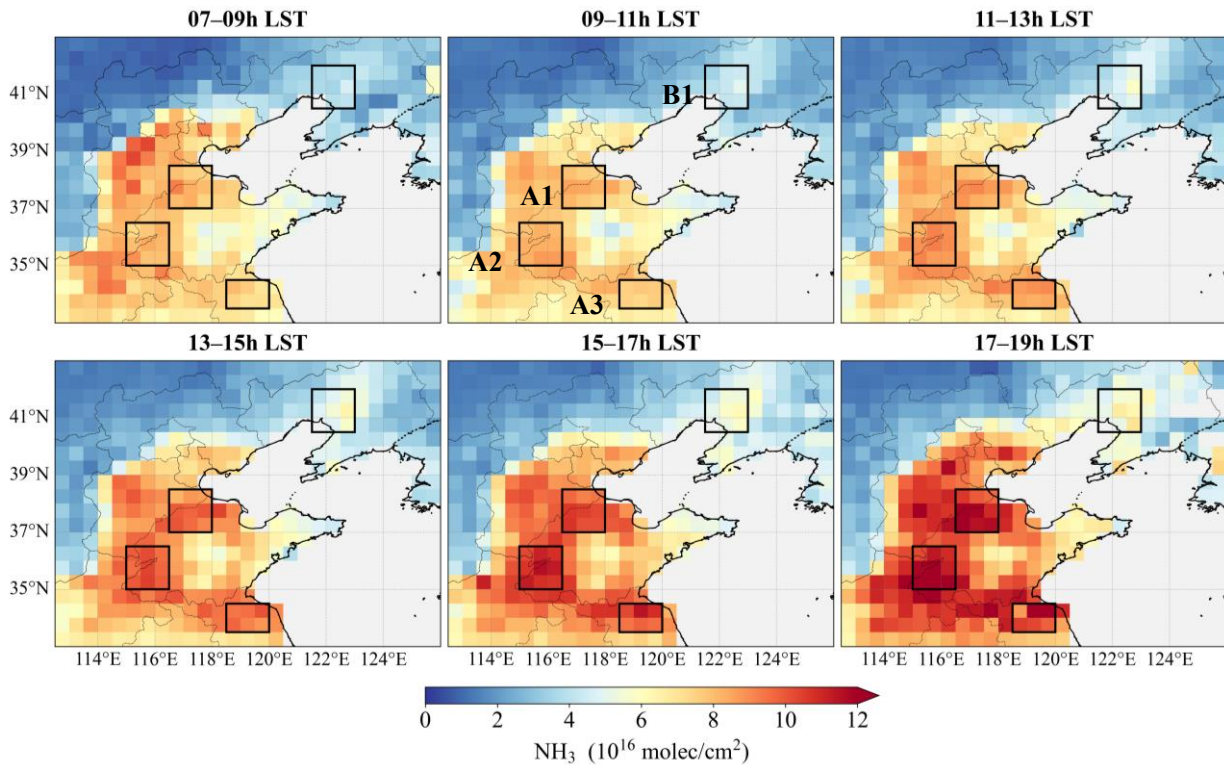
455  
 456  
 457 **Figure 11. Daytime  $\text{NH}_3$  variations for major agricultural emission areas observed by GIIRS in different seasons.**

458  
 459 The North China Plain is characterized by extensive irrigated croplands under winter wheat-maize rotation and  
 460 widespread small-scale livestock farming concentrated in east-central provinces, including Henan, Shandong, and Hebei. Due  
 461 to intensive agricultural and livestock activities, dense industrial emissions and heavy urban traffic, the region ranks among

462 the most polluted regions in China. The spatial distribution of local NH<sub>3</sub> high values across different seasons shows  
463 considerable variations, especially during the late afternoon in the spring and summer, which may indicate a change in emission  
464 hotspots (Fig. 12 and Fig. S18). Across the three selected areas (A1-A3), GIIRS observations reveal a consistent temporal  
465 pattern of NH<sub>3</sub> columns, with values increasing from early morning to late afternoon in summer but generally declining after  
466 15:00 LST in other seasons. In summer, the mean variations in GIIRS NH<sub>3</sub> column amplitudes for the afternoon and morning  
467 are 22%, 27%, and 29%, respectively, while in May and June 2025, these variations increased to 56%, 38%, and 32 %,  
468 respectively.

469 The Northeast China Plain is a key grain production region, particularly for maize, soybeans, and rice. Agriculture is  
470 highly mechanized, and livestock farming is carried out on a large scale. **Because agricultural emission sources are spatially**  
471 **concentrated and geographically stable,** the spatial pattern of NH<sub>3</sub> enhancements shows little variation at different local times  
472 (Fig. 12). The B1 area is located in the estuarine delta, with large cities such as Shenyang and Anshan to the east. Due to low  
473 population density and limited industrial activity, NH<sub>3</sub> emissions in this area are primarily associated with fertilizer application  
474 and animal husbandry, resulting in relatively low levels than those in other areas. GIIRS observations show that NH<sub>3</sub> columns  
475 are at markedly reduced levels, approximately 52% of those in the North China Plain, but follow a similar diurnal cycle, with  
476 the variation amplitude in summer being comparable (25% for 3 years, 43% for May-June 2015). The difference is that in  
477 winter, the diurnal variation also shows higher NH<sub>3</sub> columns in the morning (7:00–9:00 LST), which may be related to traffic  
478 emissions and the effects of meteorological conditions (such as temperature and humidity changes) in the early morning.  
479 Ground-based studies have reported that the peak atmospheric NH<sub>3</sub> concentration typically occurs in the morning, as observed  
480 in the urban area of Beijing (Gu et al., 2022) and in the rural areas of Xianghe (He et al., 2020), Xinxiang (Teng et al., 2017),  
481 Shanghai (Wang S. et al., 2015), and Hefei (Wang et al., 2022).

482

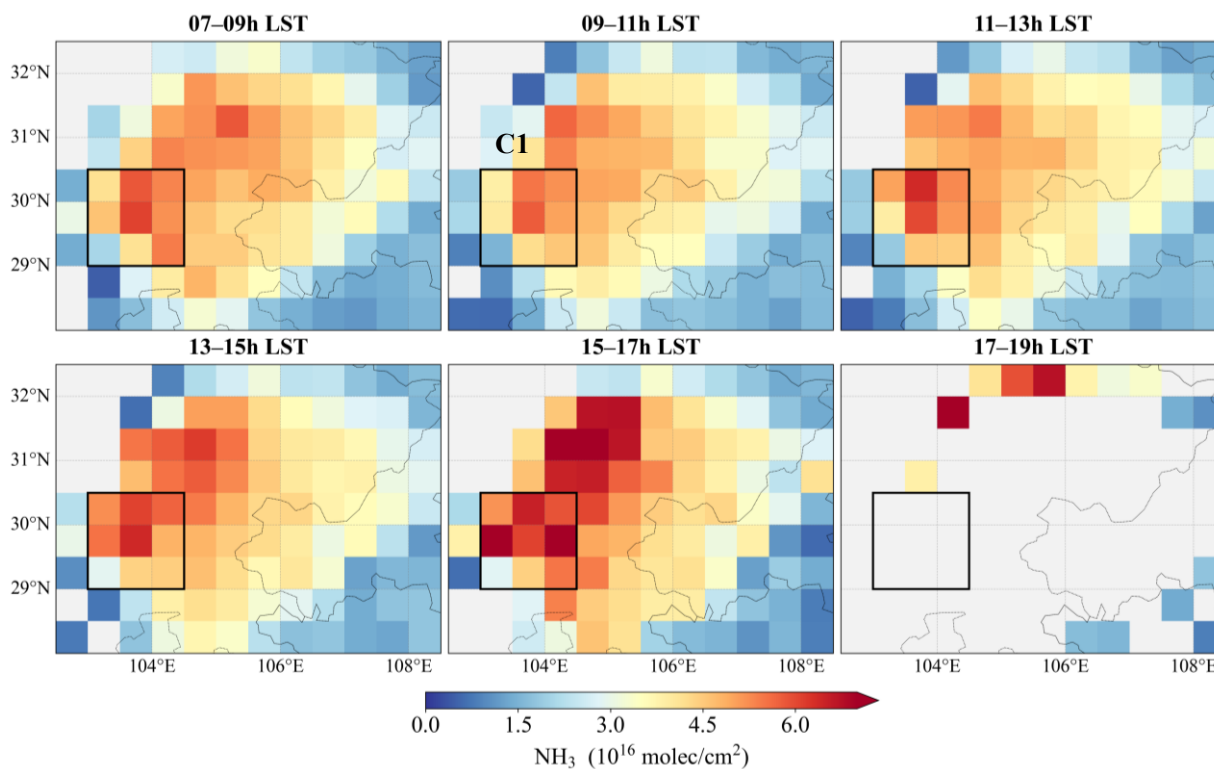


483  
484 **Figure 12. Daytime NH<sub>3</sub> variations observed by GIIRS in summer from July 2022 to June 2025 in the North-Northeast China Plain.**

485 The Sichuan Basin, located in southwestern China, is an important agricultural region encompassing the Chengdu-  
486 Chongqing urban agglomeration. Ammonia emissions exhibit a clear east-west gradient, reflecting regional differences in  
487 agricultural practices and livestock management (Li et al., 2021; Zhang, L., et al., 2018; Kang et al., 2016). Studies based on  
488 IASI observations indicate that NH<sub>3</sub> hotspots in the Sichuan Basin are primarily concentrated in the Chengdu Plain, and the  
489 southern urban clusters, and northwestern Sichuan, where agricultural sources dominate ammonia emissions (Yang et al., 2024;  
490 Dammers et al., 2019; Van Damme et al., 2018). GIIRS observations show that NH<sub>3</sub> columns are significantly enhanced in the  
491 western regions, particularly along the Mianyang-Deyang-Chengdu-Meishan-Leshan corridor (Fig. 7 and Fig.13), which are  
492 associated with intensive agricultural activity and urban-industrial emissions. The basin's unique topography and  
493 meteorological conditions such as frequent temperature inversions result in very few valid NH<sub>3</sub> observations after 17:00 LST.  
494 Across different seasons, NH<sub>3</sub> columns generally exhibit an increasing trend from morning to afternoon. In spring and summer,  
495 seasonal mean NH<sub>3</sub> concentrations show relatively small differences because agricultural sources dominate and vertical mixing  
496 is enhanced by changes in boundary layer height, with daytime NH<sub>3</sub> variations of 27% and 23%, respectively. The largest  
497 daytime variation occurs in winter, when afternoon values are 1.46 times higher than those in the morning.

498

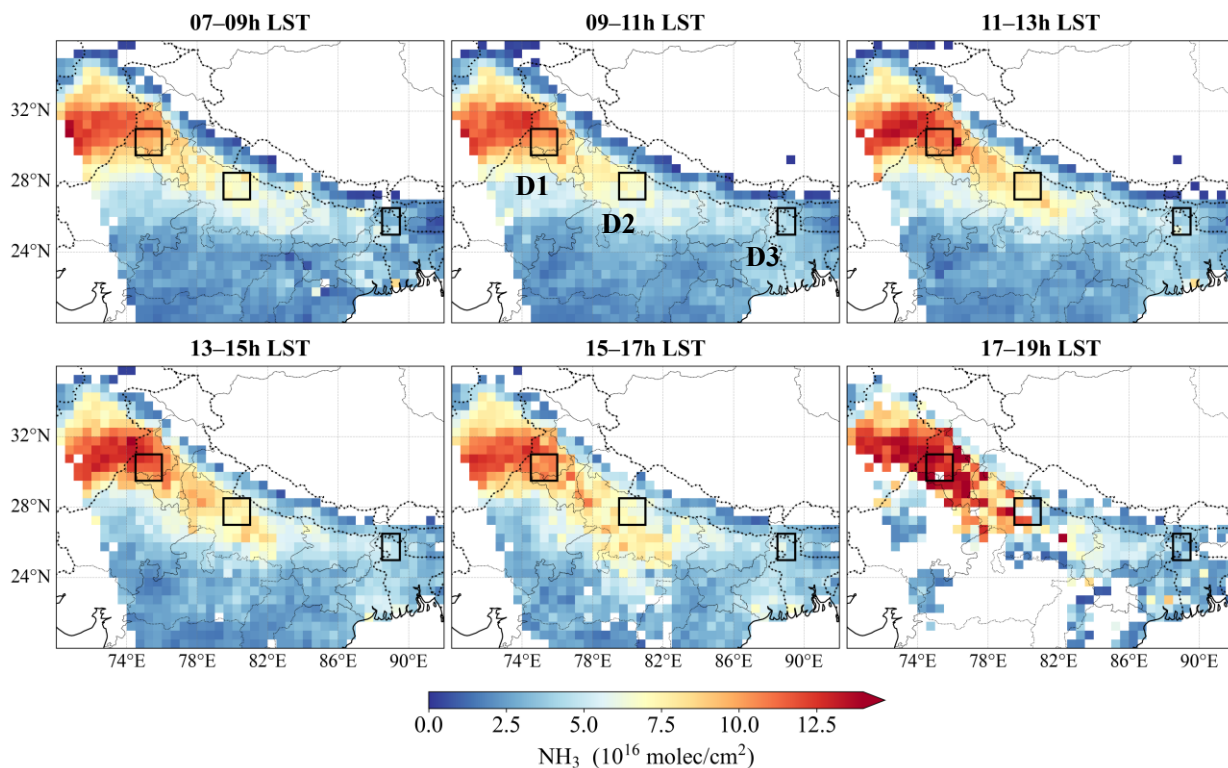
499



500  
501 **Figure 13. Daytime NH<sub>3</sub> variations observed by GIRS in summer from July 2022 to June 2025 in the Sichuan Basin.**

502 The Indo-Gangetic Plain, South Asia's most extensive alluvial plain, spans Pakistan, northern India, and Bangladesh. The  
503 region is densely populated and intensively farmed, producing rice, wheat, sugarcane, and vegetables, often under irrigated  
504 and mechanized conditions. Fertilizer application, livestock excretion, industrial activities, and high population density lead  
505 to significant NH<sub>3</sub> and reactive nitrogen emissions, reaching their maximum over the Punjab region in Pakistan. We selected  
506 three areas that are located in Punjab, Uttar Pradesh, and Bangladesh, respectively (Fig. 14). Differences in NH<sub>3</sub> emission  
507 sources and climatic conditions across these regions lead to pronounced seasonal variations in NH<sub>3</sub> concentrations. The  
508 daytime variation amplitudes of NH<sub>3</sub> columns are highest in spring, which are 17%, 32%, and 21% for D1, D2, and D3,  
509 respectively. However, under the coupled conditions of abundant precipitation and strong solar radiation in summer, NH<sub>3</sub>  
510 columns in the D2 and D3 areas reach their peak around 13:00 LST.

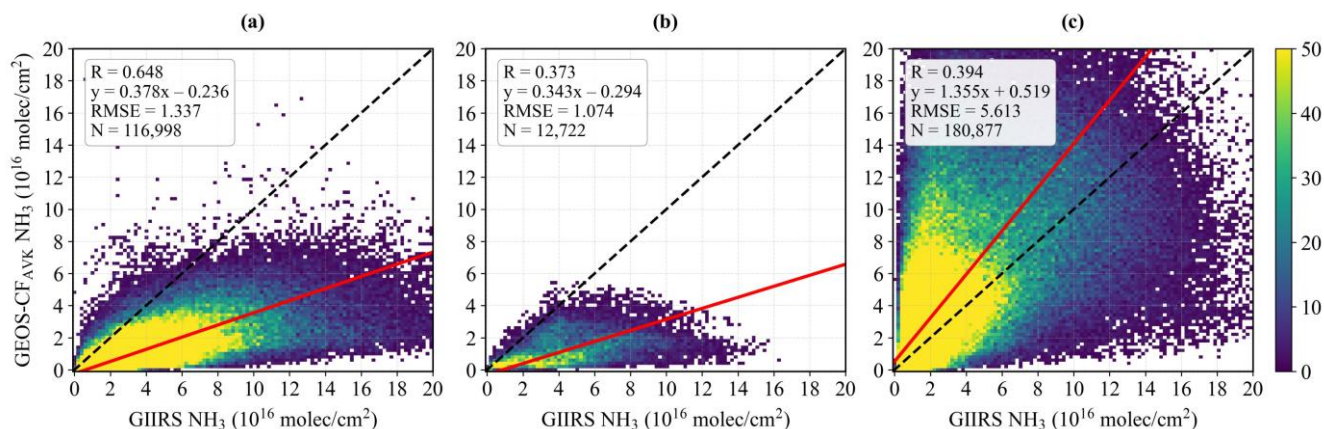
511  
512



513  
514 **Figure 14. Daytime  $\text{NH}_3$  variations observed by GIIRS in summer from July 2022 to June 2025 in the Indo-Gangetic Plain.**

515 The diurnal variation of atmospheric  $\text{NH}_3$  column concentrations is primarily influenced by surface emissions, boundary  
516 layer dynamics, and gas-particle partitioning associated with meteorological conditions (Lan et al., 2024; Behera et al., 2013).  
517 In these main agricultural source areas,  $\text{NH}_3$  concentrations generally rise from morning to afternoon, driven by diurnal  
518 variations in temperature and humidity that shift the gas-particle equilibrium toward the gaseous phase. The daytime  $\text{NH}_3$   
519 variations simulated by the GEOS-CF model for May-June 2025 range from 7% to 36%, slightly lower than the variations  
520 observed by GIIRS (10% to 56%). However, the GEOS-CF model was unable to accurately reproduce the spatial pattern of  
521  $\text{NH}_3$  enhancements in the Sichuan Basin, which can be largely attributed to the bottom-up emission inventory and the influence  
522 of complex topography and local climatic conditions. After considering about the vertical sensitivities of satellite retrievals,  
523 the AVK-smoothed model data resulted in an overall reduction of  $12 \pm 38\%$  in the model  $\text{NH}_3$  columns over the Sichuan Basin.  
524 However, considerable discrepancies in the spatial distribution remain relative to the satellite observations. Figure 15 presents  
525 a cross-comparison between the satellite-derived  $\text{NH}_3$  columns and the AVK-smoothed model data in May-June 2025. Over  
526 the North-Northeast China Plain, the model underestimates  $\text{NH}_3$  columns by 62% on average, with a standard deviation of  
527 45%, whereas over the Sichuan Basin, the underestimation reaches 65%, with a standard deviation of 30%. In contrast, the  
528 model generally overestimates  $\text{NH}_3$  columns over the Indo-Gangetic Plain. Regarding the spatial and temporal pattern of

529 diurnal variations (Figs. S25–28), the largest discrepancies are observed in the areas of C1 in the Sichuan Basin and D1 in the  
530 Indo-Gangetic Plain, suggesting that the model has limited capability to simulate NH<sub>3</sub> in these areas and may require  
531 improvements through satellite-observed constraints to improve simulation accuracy.



532  
533 **Figure 15. Comparison of NH<sub>3</sub> columns in May and June 2025 from GIIRS observations and GEOS-CF AVK-smoothed model**  
534 **simulations for (a) the North China Plain and the Northeast China Plain, (b) the Sichuan Basin, and (c) the Indo-Gangetic Plain.**  
535 **The model data is calculated by applying the AVKs from satellite retrievals to NH<sub>3</sub> profile (see Text S1).**

## 536 4 Conclusions and Perspectives

537 The geostationary satellite observations from FY-4B/GIIRS provide a unique opportunity to monitor the diurnal variations  
538 of atmospheric NH<sub>3</sub> over East Asia, yet current research on this topic remains limited. In this study, we present a comprehensive  
539 analysis of three years of FY-4B/GIIRS observations from July 2022 to June 2025, revealing the spatiotemporal variations of  
540 NH<sub>3</sub> across East Asia and the distinctive daytime variations in agricultural emission regions.

541 NH<sub>3</sub> column concentrations from GIIRS observations exhibit pronounced local enhancements over major emission  
542 regions, particularly in flat agricultural plains (e.g. the North China Plain, the Northeast China Plain, the Indo-Gangetic Plain,  
543 the Ningxia Irrigation Plain, the Wei River Plain, the Jiangnan Plain, the Mekong Delta in Vietnam, the Chao Phraya River  
544 Plain in Thailand) with intensive crop cultivation and animal husbandry, as well as smaller-scale accumulation in  
545 topographically confined areas such as valleys and basins (e.g. the Sichuan Basin, oasis agriculture in the arid regions of  
546 Xinjiang in China, the Fergana Valley in Uzbekistan). After removing background levels derived from predefined reference  
547 regions, NH<sub>3</sub> enhancements show a consistent spatial pattern with anthropogenic emissions from the bottom-up MIX Asia  
548 emission inventory, and highlight these major emission source regions. The spatial distribution of NH<sub>3</sub> derived from GIIRS  
549 agrees well with polar-orbiting satellite observations from IASI and CrIS.

550 The timing of peak NH<sub>3</sub> in the seasonal cycle primarily reflects agricultural activities related to fertilizer application and  
551 is modulated by climatic factors such as temperature. For example, NH<sub>3</sub> columns generally exhibit a summer maximum (June-  
552 July) and a secondary spring maximum (April) in the North China Plain and Northeast China Plain. In southern regions of  
553 China, India, and Southeast Asia, NH<sub>3</sub> peaks during the late dry season (March-May) and declines sharply in the early wet

554 season (June-July). Overall, regional differences in the timing of peak months follow a north-south gradient. Comparison with  
555 ground-based FTIR measurements and satellite-based IASI and CrIS observations show generally good agreement, yielding a  
556 correlation coefficient of 0.77 and an RMSE of  $9.67 \times 10^{15}$  molec/cm<sup>2</sup> relative to FTIR, while also consistently capturing the  
557 temporal variability of NH<sub>3</sub> columns.

558 This study reports the detailed daytime variations of NH<sub>3</sub> columns observed by FY-4B/GIIRS over the North China Plain,  
559 the Northeast China Plain, the Sichuan Basin, Hefei, and the Indo-Gangetic Plain. The NH<sub>3</sub> columns during the day generally  
560 increase from early morning to late afternoon, showing similar temporal variations to those observed by ground-based FTIR  
561 measurements at Hefei and simulated by GEOS-CF model in the main agricultural source areas. Crucially, GIIRS provides  
562 substantial advantages by accurately characterizing spatial distributions and enabling continuous daily observations. GIIRS in  
563 the infrared is complementary to the Geostationary Environment Monitoring Spectrometer (GEMS; Kim et al., 2020), which  
564 operates in the ultraviolet/visible (UV/VIS) spectral range and provides hourly observations of trace gases such as NO<sub>2</sub> and  
565 SO<sub>2</sub> over East Asia. The synergy between GIIRS and GEMS enables joint investigations of NH<sub>3</sub> and NO<sub>2</sub>, offering new  
566 opportunities to better quantify reactive nitrogen budgets and assess their impacts on air quality and climate. The upgraded  
567 FY-4C/GIIRS was launched in December 2025, further enhancing the FY-4 geostationary observational capabilities. Compared  
568 to its predecessor FY-4B/GIIRS, which provides a spatial resolution of 12 km and a temporal resolution of 2 hours, FY-  
569 4C/GIIRS achieves a significant improvement to 8 km and 1 hour, respectively. This emerging satellite constellation will offer  
570 unprecedented opportunities for seamless hourly coverage to monitor diurnal cycles and emission processes across East and  
571 South Asia, thereby improving our understanding of atmospheric chemistry and supporting the development of more effective  
572 air pollution mitigation strategies.

573

## 574 Appendix: The interpretation and calculation of column AVK

575 In the profile-scaling optimal estimation framework, the column AVK (unitless) represents the change in the retrieved  
576 total  $\text{NH}_3$  column ( $\text{molec}/\text{cm}^2$ ) with respect to a perturbation of the partial  $\text{NH}_3$  column ( $\text{molec}/\text{cm}^2$ ) at a given atmospheric  
577 layer. For example, if the true  $\text{NH}_3$  partial column in the first layer near the surface increases by  $1.0 \times 10^{16}$   $\text{molec}/\text{cm}^2$ , whereas  
578 the retrieval algorithm only detects an increase of  $0.4 \times 10^{16}$   $\text{molec}/\text{cm}^2$  in the final retrieved total column, the resulting column  
579 AVK for the first layer is 0.4. A value of 1.0 would imply perfect sensitivity to that layer. As a hyperspectral sounder has  
580 varying sensitivities to different altitudes and the spectral signatures of  $\text{NH}_3$  molecules vary at different altitudes, the retrieved  
581 total column is not uniformly sensitive to  $\text{NH}_3$  molecules at all heights. The one-dimensional column AVK thus provides a  
582 crucial diagnostic metric to understand where the retrieved column information is originating. This definition of column AVK  
583 is conceptually similar to that from TCCON (Wunch et al., 2011) and IASI (Clarisse et al., 2023). By contrast, the conventional  
584 matrix AVK from optimal estimation quantifies how much information is retrieved from the measurement (Shephard et al.,  
585 2011, 2015), and the sum of each row of AVK provides an estimate of the fraction of retrieval information contributed by the  
586 measurement rather than the a priori at the corresponding altitude. When the information content of the measurement is high  
587 or the retrieval loosely constrained, the AVK values approach the unit matrix. Conversely, when the information content is low  
588 or the retrieval is heavily constrained, the AVK values tend toward zero and the solution remain close to the a priori. A detailed  
589 discussion on the difference between column AVK and matrix AVK can be referred to Clarisse et al. (2023).

590 The column AVK is calculated following the formalism of the full matrix AVK from optimal estimation, which is  
591 expressed as:

$$592 \text{Column AVK} = G K_{GasProf} \quad (\text{A1})$$

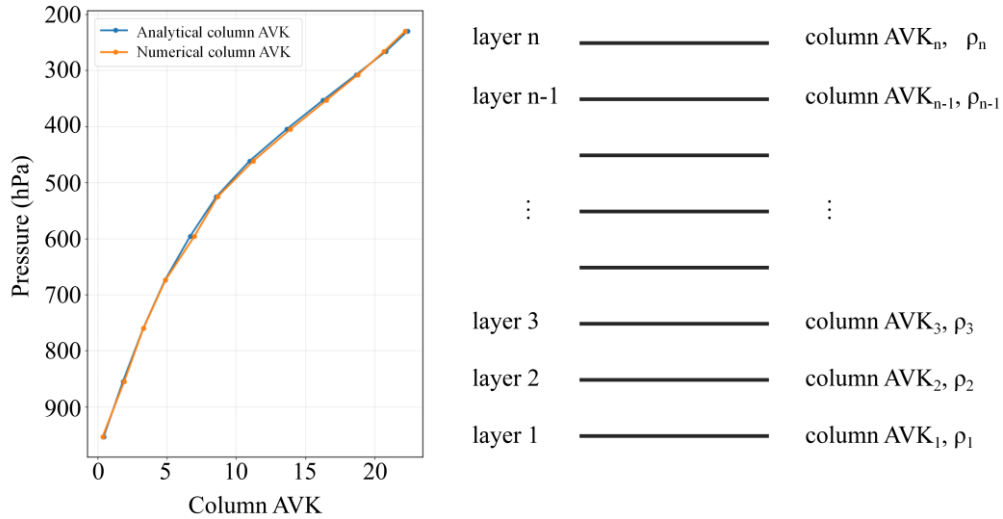
593 where  $K_{GasProf}$  is the layer-resolved  $\text{NH}_3$  Jacobian quantifying the sensitivity of radiance to layer-specific  $\text{NH}_3$  partial column  
594 perturbations.  $K_{GasProf}$  is calculated using the radiative transfer model for each retrieval. The gain matrix  $G$  is given by:

$$595 G = (K_{col}^T S_e^{-1} K_{col} + S_a^{-1})^{-1} K_{col}^T S_e^{-1} \quad (\text{A2})$$

596 where  $K_{col}$  is the total-column  $\text{NH}_3$  Jacobian quantifying the sensitivity of radiance to total  $\text{NH}_3$  column,  $S_e$  and  $S_a$  are the  
597 noise covariance matrix for the measured radiances and the a priori covariance matrix for the retrieval.

598 To verify the analytical column AVK calculated from Eq. (A1), observation simulation experiments were conducted to  
599 calculate the corresponding numerical column AVK for comparison. An observation over the North China Plain (41.97°N,  
600 122.50°E) on 15 July 2025 was selected, with a surface pressure of 1002.65 hPa, surface temperature of 302.49 K, and lowest-  
601 layer air temperature of 297.46 K. The “true”  $\text{NH}_3$  profile was generated by scaling the a priori profile from Zeng et al. (2023a)  
602 by a factor of 10 (approximate total column:  $9.49 \times 10^{16}$   $\text{molec}/\text{cm}^2$ ), which was also used as the a priori profile for all retrievals.  
603 We first simulated the “true” radiance spectrum from the “true” profile, added Gaussian random noise, and ran the retrieval  
604 algorithm 100 times to obtain the mean retrieved total column, which is close to the true total column. The analytical column  
605 AVK solution from this observation was then derived using Eqs. (A1) and (A2). Subsequently, we performed a layer-wise  
606 perturbation test to derive the numerical column AVK. A fixed  $\text{NH}_3$  perturbation of  $1 \times 10^{16}$   $\text{molec}/\text{cm}^2$  was individually added

607 to each of the 12 retrieval layers of the “true” profile, while keeping all other layers unchanged. For each perturbed layer, the  
 608 noise-added retrieval was repeated 100 times to obtain the mean retrieved total column. The numerical column AVK was then  
 609 calculated as the ratio of the retrieved total column change to the imposed “true” column increment ( $1 \times 10^{16}$  molec/cm<sup>2</sup>). **Figure**  
 610 **A1** compares the analytical and numerical AVKs, showing good agreement between the two sets, thereby justifying the  
 611 calculation of column AVK using Eqs. (A1) and (A2).



612  
 613 **Figure A1. Comparison of the analytical and numerical column AVKs for a case over the North China Plain on 15 July 2024 (left**  
 614 **panel). Schematic of the retrieval vertical layers from the surface to 200 hPa (right panel), consisting of n pressure levels (typically**  
 615 **n = 11 or 12), with the layer-dependent column AVK values (column AVK<sub>i</sub>) and the air density (ρ<sub>i</sub>) of NH<sub>3</sub> at each layer.**

616 **Data availability.**

617 The GIIRS NH<sub>3</sub> data (July 2022 to June 2025) used in this study is publicly available on Zenodo (Zeng et al.,  
618 2025; <https://doi.org/10.5281/zenodo.17193848>). Further updates on the data will be provided on the project website  
619 (<https://fengyunair.github.io/>). The IASI L2 ammonia satellite observations are available from the AERIS data infrastructure  
620 (<https://iasi.aeris-data.fr/>, last access: 16 September 2025). The CrIS Fast Physical Retrieval (CFPR) ammonia dataset is  
621 created by the Environment and Climate Change Canada is publicly available  
622 ([https://hpfx.collab.science.gc.ca/~mas001/satellite\\_ext/cris/](https://hpfx.collab.science.gc.ca/~mas001/satellite_ext/cris/)). The Hefei FTIR data are retrieved by Key Laboratory of  
623 Environmental Optics and Technology, Anhui Institute of Optics and Fine Mechanics, Chinese Academy of Sciences (Wang  
624 et al., 2022). The MIX emission inventory can be requested from the MEIC team ([http://meicmodel.org.cn/?page\\_id=2721](http://meicmodel.org.cn/?page_id=2721)).  
625 The GEOS-CF model data are available from High Performance Computing for Science, NASA Center for Climate Simulation  
626 (<https://portal.nccs.nasa.gov/datashare/gmao/geos-cf/v1/das/Y2025/>, last access: 10 August 2025).

627 **Author contributions.**

628 MS and ZZ designed the study. MS carried out the result analysis and prepared the manuscript. ZZ and JH developed the  
629 GIIRS NH<sub>3</sub> retrieval algorithm. MS, RZ, SH, and SL collected and analyzed the emission sets. WW provided the FTIR data.  
630 RD, LZ, HC, ZC, and YG provided guidance on interpreting the satellite data and model simulations. ML provided guidance  
631 on analyzing emission inventory. NG, LC, MVD, and CC provided the IASI data and guided the interpretation. MWS provided  
632 the CrIS data and guidance. LL, CQ, FL, and CH provided guidance on analyzing GIIRS spectra. All authors reviewed and  
633 proofread the paper. LC is a senior research associate supported by the Belgian F.R.S.-FNRS.

634 **Competing interests.** The contact author has declared that none of the authors has any competing interests.

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