

# Global and diurnal variations in tropospheric ammonia observed from a constellation of hyperspectral infrared sounders in three different LEO orbits

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Abstract. As a reactive nitrogen compound, atmospheric ammonia (NH<sub>3</sub>) plays a key role in the global nitrogen cycle. Tracking the spatiotemporal dynamics of NH<sub>3</sub> is crucial to quantify its emissions and depositions, as well as offering insights to inform the regulation of anthropogenic emission sources. Currently, the diurnal cycle of NH<sub>3</sub> remains under-constrained, particularly in regions lacking geostationary satellite observations, which poses a challenge to accurate emission quantification. To address this gap, we construct an integrated constellation to achieve quasi-geostationary-like global monitoring coverage, comprising China's FengYun-3 (FY-3) series satellites and the Cross-track Infrared Sounder (CrIS). FY-3E operates in a dawn-dusk orbit with equatorial overpassing time at 05:30 am/pm, while FY-3F operates in a mid-morning orbit with overpassing time at 10:00 am/pm. Both are equipped with the second-generation High Spectral Infrared Atmospheric Sounder (HIRAS-II). CrIS, operating with overpassing time at 01:30 am/pm, provides supplementary observations in an afternoon orbit. In this study, hyperspectral infrared observations from the constellation are utilized to retrieve global NH<sub>3</sub> columns based on the optimal estimation method. Six maps of global NH<sub>3</sub> for every 4-hour in each day are retrieved. The retrieval results in four weeks of different seasons in 2024, as a demonstration, show elevated columns in global major source regions, including the North China Plain, the Indo-Gangetic Plain, North America and Western Europe. In addition, the diurnal and seasonal cycles of NH<sub>3</sub> over these regions using all observations in 2024 are also investigated. The constellation reasonably captures the diurnal (every 4-hour) and seasonal cycles of NH<sub>3</sub> columns, effectively mitigating the constraints in regions without geostationary observations. Further cross-comparisons with the Geostationary Interferometric Infrared Sounder (GIIRS) retrievals and ground-based Fourier transform infrared spectroscopy (FTIR) measurements demonstrate that the polar-orbiting constellation retrievals are internally consistent and broadly consistent with independent observations, supporting their capability to capture major NH<sub>3</sub> spatial patterns and temporal variability. The sensitivity of NH<sub>3</sub> detection in the lower atmosphere as quantified by the column averaging kernel (AVK) from the retrieval shows diurnal variations that dependent on thermal contrast, defined as the temperature difference between the surface and the lower atmospheric layer. This study demonstrates the capability of the integrated constellation, comprising FY-3E/HIRAS-II (dawn-dusk), FY-3F/HIRAS-II (mid-morning), and CrIS (afternoon), to monitor global and diurnal NH<sub>3</sub> variations at unprecedentedly six distinct times of a day, and has the potential to enhance the global climate-monitoring capacity of polar-orbiting meteorological satellites.

## 1 Introduction

As a critical reactive nitrogen compound, ammonia ( $\text{NH}_3$ ) holds considerable importance in the atmosphere and global nitrogen cycle, yet it also acts as a hazardous pollutant affecting human health and ecosystems. Specifically, it contributes to smog via its neutralization of sulfuric and nitric acids, resulting in the generation of secondary aerosols including ammonium sulfate and ammonium nitrate. (Galloway et al., 2004; Behera et al., 2013). These particles can travel long distances, degrading air quality and disrupting ecological balance (Erisman et al., 2013). These ammonium-based particles, along with  $\text{NH}_3$  itself, can be converted into various reactive nitrogen forms that trigger acid precipitation, eutrophication, and reduced biodiversity (Sutton et al., 2011). Under specific conditions, ammonium nitrate particles may volatilize to reform gaseous  $\text{NH}_3$ , creating a dynamic cycle that complicates its environmental impacts (Weber et al., 2016; Guo et al., 2018). Agriculture serves as the dominant source, stemming from fertilizer storage, livestock manure, and the application of mineral nitrogen fertilizers to crops (Fowler et al., 2013). Secondary contributors to  $\text{NH}_3$  emissions encompass biomass combustion, industrial operations, and automobiles fitted with three-way catalytic converters (Galloway et al., 2003; Erisman et al., 2008). After being emitted into the atmosphere,  $\text{NH}_3$  exhibits a brief residence time ranging from several hours to a handful of days, and its primary removal pathways are dry and wet deposition processes (Liu et al., 2013; Dammers et al., 2017; Zhang et al., 2018). These deposition processes accelerate soil acidification, further exacerbating the ecological damage. Beyond air quality and ecosystem impacts, the aerosols derived from  $\text{NH}_3$  also influence climate by scattering solar radiation, modifying albedo, and altering the properties of clouds (Adams et al., 2001; Abbatt et al., 2006; Isaksen et al., 2009; Myhre et al., 2013).

Different technologies have been used to monitor atmospheric  $\text{NH}_3$ . A number of in situ monitoring sites have been established (Jiménez et al., 2015; De Mazière et al., 2018). Moreover, satellites furnished with high-resolution infrared spectrometers have provided unprecedented spatiotemporal sampling of global  $\text{NH}_3$  distributions, and this has driven major progress in understanding  $\text{NH}_3$  emission and deposition fluxes covering spatial scopes ranging from local-level to global-scale, together with their temporal fluctuation characteristics. Polar-orbiting satellites are capable of delivering global observational coverage up to two times per day, including the Infrared Atmospheric Sounding Interferometers (IASI, e.g., Clarisse et al., 2009) at 09:30/21:30 (Local Solar Time, LST), Tropospheric Emission Spectrometer (TES, e.g., Beer et al., 2008; Shephard et al., 2011), the Atmospheric Infrared Sounder (AIRS, e.g., Warner et al., 2016) and the Cross-track Infrared Sounder (CrIS, e.g., Shephard and Cady-Pereira, 2015) at 01:30/13:30 LST, the Thermal and Near-infrared Spectrometer for Observation-Fourier Transform Spectrometer (TANSO-FTS; e.g., Someya et al., 2020) at 01:00/13:00 LST and FengYun-3D (FY-3D, e.g. Zhou et al., 2024) at 02:00/14:00 LST. Recently, diurnal variations of atmospheric  $\text{NH}_3$  over the Asian region have been observed using the Geostationary Interferometric Infrared Sounder (GIIRS) aboard the FengYun-4A (FY-4A/GIIRS, Clarisse et al., 2021) and FengYun-4B (FY-4B/GIIRS, Zeng et al., 2023; Sheng et al., 2025; Guendouz et al., 2026) satellites. **The Infrared Sounder (IRS) aboard the Meteosat Third Generation (MTG), which was launched in 2025, is expected to provide geostationary observations over Europe and Africa (Holmlund et al., 2021; Guendouz et al., 2026).** Such geostationary orbit-based monitoring offers significant advantages and potential for investigating  $\text{NH}_3$  diurnal variations, which in turn facilitates

65 a deeper understanding of its emission, deposition, transport and other related processes. However, there is currently a lack of geostationary orbit observations across most global regions especially in most of the Southern Hemisphere and North America. Atmospheric NH<sub>3</sub> shows notable spatiotemporal variability, largely attributed to its primary emission characteristics and short atmospheric lifetime. This inherent variability underscores the urgent need for a high-resolution and globally comprehensive monitoring system.

70 To address this gap, we construct an integrated constellation to achieve quasi-geostationary-like global monitoring coverage, comprising China's FengYun-3 (FY-3) series satellites and CrIS. Similar constellation has been developed for monitoring volcanic sulfur dioxide (Zeng et al., 2025). NH<sub>3</sub> retrievals employing the optimal estimation method, will be conducted under this framework to enable quasi-geostationary observations up to six times a day globally. This work is expected to provide crucial observational data for clarifying global and diurnal NH<sub>3</sub> variations.

75 The subsequent sections of this paper are organized as follows. Section 2 elaborates on the methodological approaches adopted in the present research. Section 3 presents and discusses the retrieval results from the constellation. Finally, the conclusions are drawn in Section 4.

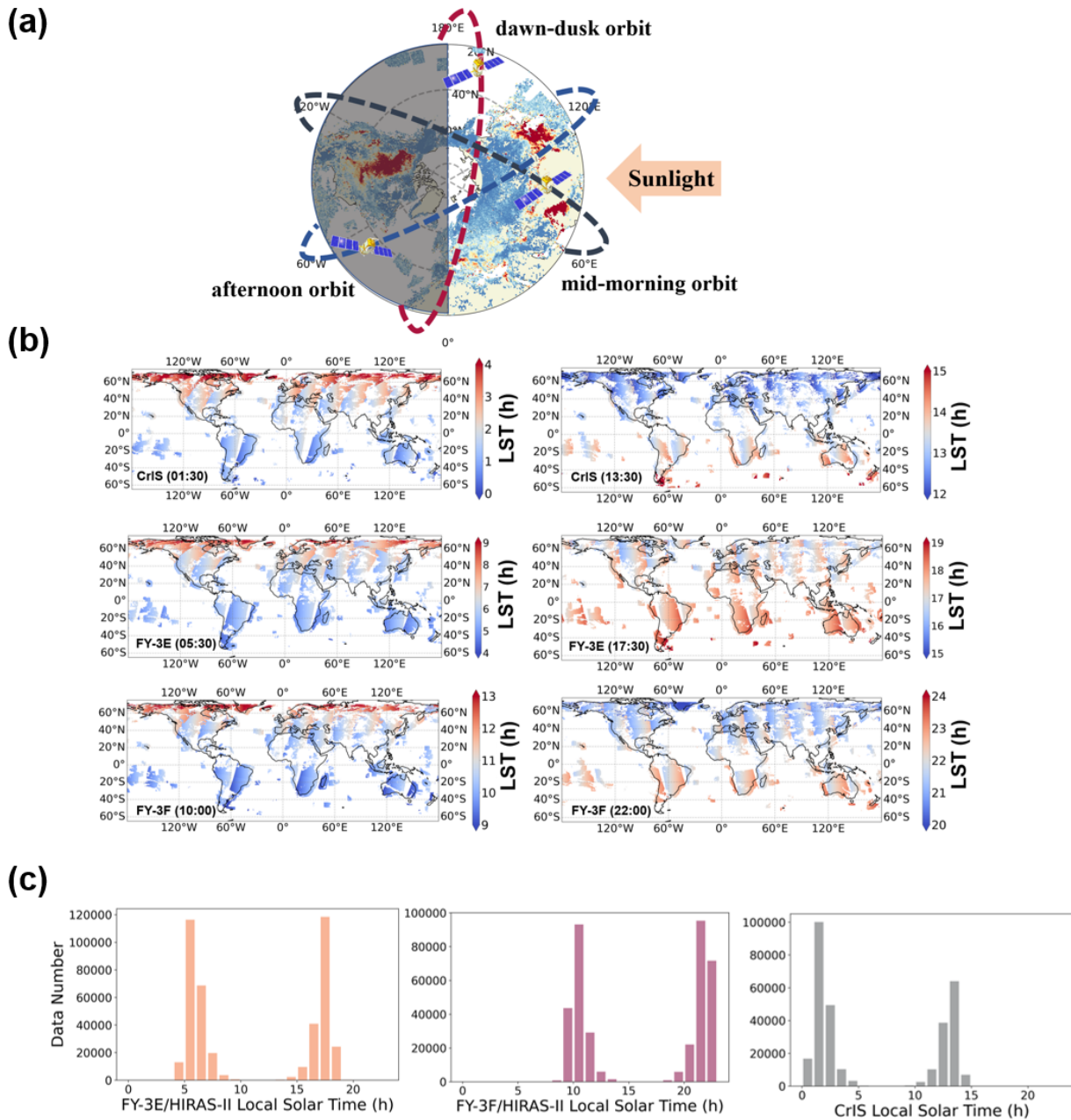
## 2 Methodologies

### 2.1 A constellation of hyperspectral infrared sounders

80 The High Spectral Infrared Atmospheric Sounder (HIRAS) aboard the FY-3 satellite series is a Fourier Transform Michelson interferometer. It measures upwelling infrared radiative signals across the short-wave, mid-wave, and long-wave infrared bands. HIRAS-II is the second-generation of the HIRAS sensor and is carried onboard both FY-3E and FY-3F, which were successfully launched in 2021 and 2023, respectively (Zhang et al., 2022a, b). FY-3E/HIRAS-II is deployed in a dawn-dusk orbit, with an equatorial overpass time of 05:30 am/pm (Local Solar Time, LST), while FY-3F/HIRAS-II operates in a mid-morning orbit featuring an overpass time of 10:00 am/pm LST. Combined with CrIS aboard the Joint Polar Satellite System-1 (JPSS-1) platform, which has an overpass time of 01:30 am/pm LST, these three sensors form a global constellation observation system (Fig. 1a). Note that IASI's overpass times, approximately 09:30 and 21:30 LST, are close to those of FY-3F/HIRAS-II at 10:00 and 22:00 LST and thus can provide additional mid-morning orbit observations. For this concept study, however, only FY-3F/HIRAS-II is used. Their overpass times are shown in Fig. 1b, and the corresponding histograms are presented in Fig. 1c.

90 Both HIRAS-II instruments aboard the FY-3E and FY-3F satellites, as well as the CrIS share key technical specifications with a nadir spatial resolution of 14 km and an unapodised spectral resolution of 0.625 cm<sup>-1</sup> (Zhang et al., 2024). Specifically, HIRAS-II provides continuous spectral coverage across 650–2550 cm<sup>-1</sup> via 3041 channels, while CrIS matches this spectral resolution, ensuring all three instruments fully cover the characteristic rotational-vibrational ν<sub>2</sub> band of NH<sub>3</sub> centered near 10.5 μm. For NH<sub>3</sub> retrieval, we use the 955–975 cm<sup>-1</sup> band extracted from observations of all three instruments. No empirical sensor-specific radiometric bias correction or additional spectral harmonization is applied before the retrievals. We randomly

select one day of observations from the three satellites over the North China Plain and calculate their spectral noise. The noise equivalent differential temperature (NedT) of HIRAS-II aboard FY-3E and FY-3F is approximately 0.08 K, while that of CrIS is around 0.05 K. These noise levels are of a comparable magnitude and sufficiently sensitive to detect variations in NH<sub>3</sub>,  
100 ensuring uniform observational quality for cross-instrument analysis (Zavyalov et al., 2013; Shephard and Cady-Pereira, 2015; Zhang et al., 2024). While CrIS already has well-established and mature NH<sub>3</sub> products from the CrIS Fast Physical Retrieval (CFPR) dataset developed by Environment and Climate Change Canada (Shephard et al., 2020), we adopt a unified retrieval algorithm for all three instruments. This choice is driven by the need to ensure consistency across the satellite constellation, enabling direct inter-comparability between FY-3E/HIRAS-II, FY-3F/HIRAS-II, and CrIS observations. For CrIS retrievals,  
105 we use JPSS-1 CrIS Level 1B Full Spectral Resolution V3 data as input, maintaining methodological coherence with our processing of HIRAS-II observations.



110 Figure 1. (a) Schematic diagram of three polar-orbiting satellites with different orbits. FY-3E/HIRAS-II operates in a dawn-dusk  
 orbit, with an equatorial overpass time of 05:30 am/pm LST, FY-3F/HIRAS-II is in a mid-morning orbit with an equatorial overpass  
 time of 10:00 am/pm LST, and the CrIS has an equatorial overpass time of 01:30 am/pm LST. The color gradient in the background  
 represents the retrieved global distribution of NH<sub>3</sub> columns on 19 July 2024, as will be discussed in subsequent analyses. (b) Local  
 Solar Time (LST) of observations acquired by FY-3E/HIRAS-II, FY-3F/HIRAS-II and CrIS over terrestrial and offshore regions.  
 115 Observations over polar areas and remote oceans are excluded. These maps are constructed from cloud-screened observations  
 collected on 19 July, 2024. (c) Histograms of the overpass Local Solar Time (LST) for FY-3E/HIRAS-II, FY-3F/HIRAS-II, and CrIS.  
 Examples are derived from observations taken on 19 July, 2024.

## 2.2 Optimal estimation methods

For the retrieval, only cloud-screened spectra from FY-3E/HIRAS-II, FY-3F/HIRAS-II, and CrIS are used. To remove cloud-contaminated pixels and ensure consistency across sensors, we apply a unified cloud-screening procedure following the brightness-temperature-difference method of Wells et al. (2020, 2022). Specifically, for each observation, the observed brightness temperature near  $900\text{ cm}^{-1}$  is compared with the nearest-hourly ERA5 surface skin temperature. The cloud-screening threshold is adjusted according to the ERA5 water vapor column, and no fixed cloud-fraction threshold is used. Observations for which the  $900\text{ cm}^{-1}$  brightness temperature is lower than this water-vapor-dependent threshold relative to the surface skin temperature are flagged as cloud-contaminated and excluded. The same cloud-screening procedure is applied to FY-3E/HIRAS-II, FY-3F/HIRAS-II, and CrIS to maintain consistency across different satellites.

The  $\text{NH}_3$  retrieval is based on the FengYun-Low Earth Orbit Atmospheric Infrared Retrieval algorithm (FY-LeoAIR), which adapts the optimal-estimation framework and forward-model structure of the FY-GeoAIR algorithm originally developed for FY-4B/GIIRS  $\text{NH}_3$  retrievals by Zeng et al. (2023). FY-LeoAIR has previously been applied to trace gas retrievals from FY-3E/HIRAS-II observations, including CO and VOCs (e.g., Zeng, 2025; Hua et al., 2025). In this study, FY-LeoAIR is extended to global  $\text{NH}_3$  retrievals from FY-3E/HIRAS-II, FY-3F/HIRAS-II, and CrIS.

The retrieval is performed in the  $955\text{--}975\text{ cm}^{-1}$  band, which contains strong  $\text{NH}_3$  absorption features and is fully covered by all three sensors. Sensor-specific spectral noise estimates are used to construct the diagonal measurement-error covariance matrix. The same forward model, retrieval method, and quality-control procedure are used for all three sensors to maintain internal consistency.

FY-LeoAIR combines a clear-sky infrared radiative transfer forward model with an optimal-estimation-based inverse model. The forward model uses atmospheric temperature,  $\text{H}_2\text{O}$ ,  $\text{O}_3$ , and surface pressure from ERA5,  $\text{CO}_2$  from ECMWF CAMS greenhouse-gas products, and other weakly interfering trace-gas profiles from standard atmospheric profiles, following the configuration of Zeng et al. (2023). Surface skin temperature is initialized from ERA5 and retrieved simultaneously with the trace gases. The a priori land surface emissivity used in the forward model is taken from the University of Wisconsin-Madison global infrared land surface emissivity database (UOW-M), as in Zeng et al. (2023). The a priori surface emissivity spectrum is adjusted within the retrieval window using a fourth-order Legendre polynomial expansion, with the first- to fourth-order coefficients retrieved simultaneously. The parameters in the retrieval state vector are summarized in Table S1.

The main modification relative to Zeng et al. (2023) is that  $\text{NH}_3$  is retrieved using a single multiplicative profile-scaling factor applied to a fixed a priori  $\text{NH}_3$  profile, rather than as independent layer-by-layer  $\text{NH}_3$  elements. The same profile-scaling strategy is also applied to  $\text{H}_2\text{O}$  in this implementation. This approach is adopted because the  $\text{NH}_3$  information content in individual thermal infrared observations is generally limited, with the degree of freedom for signal mostly below 1, making independent layer-resolved retrievals insufficiently constrained for global multi-sensor processing. The profile-scaling

approach also improves computational efficiency for the global constellation retrieval. Similar retrieval strategy has also been adopted by FY-4B/GIIRS NH<sub>3</sub> retrievals (Sheng et al., 2025).

150 The fixed NH<sub>3</sub> a priori profile is constructed following Zeng et al. (2023), based on GEOS-CF NH<sub>3</sub> simulations over representative polluted land regions in East Asia and South Asia. The same normalized profile shape is used for all retrievals, while the retrieved scaling factor determines the retrieved NH<sub>3</sub> abundance. This choice helps avoid introducing artificial spatial or temporal variability from a time-varying prior into the retrieved columns. Retrieval uncertainty and the column averaging kernel are used to characterize the information content of each retrieval. We also conduct an a priori profile sensitivity test  
155 (see Text S1 and Figs. S1-S6 in the supplementary material).

Because NH<sub>3</sub> is mainly concentrated in the lower troposphere, the retrieval state vector only adjusts NH<sub>3</sub> in 11 atmospheric layers from the surface to 200 hPa. The reported NH<sub>3</sub> total column is calculated as the sum of the retrieved surface-to-200 hPa NH<sub>3</sub> column and the fixed a priori contribution above 200 hPa. Since the NH<sub>3</sub> abundance above 200 hPa is generally small and is not independently constrained by the spectra, the spatial and temporal variability discussed in this study is  
160 dominated by the retrieved surface-to-200 hPa component.

An optimal state vector for minimizing discrepancies between forward model-simulated spectra and sensor-measured spectra is derived by the optimal estimation framework. In practice, the retrieval algorithm outputs the state vector that minimizes the cost function defined as:

$$J(\mathbf{x}) = \chi^2 = [\mathbf{y} - \mathbf{F}(\mathbf{x}, \mathbf{b})]^T \mathbf{S}_\epsilon^{-1} [\mathbf{y} - \mathbf{F}(\mathbf{x}, \mathbf{b})] + (\mathbf{x} - \mathbf{x}_a)^T \mathbf{S}_a^{-1} (\mathbf{x} - \mathbf{x}_a), \quad (1)$$

165 Here,  $\mathbf{y}$  denotes the satellites measured radiance within the NH<sub>3</sub> retrieval band.  $\mathbf{F}$  is the forward model that produces simulated radiance for the retrieval process.  $\mathbf{x}$  is the retrieval state vector, including the NH<sub>3</sub> profile-scaling factor, the H<sub>2</sub>O profile-scaling factor, scaling factors for interfering trace gases, surface skin temperature, atmospheric temperature scaling, and four surface-emissivity Legendre polynomial coefficients.  $\mathbf{x}_a$  denotes the a priori vector.  $\mathbf{b}$  stands for a set of fixed non-retrieved auxiliary parameters.  $\mathbf{S}_a$  is the a priori covariance matrix corresponding to the state vector.  $\mathbf{S}_\epsilon$  represents the  
170 measurement error covariance matrix, treated as a diagonal matrix derived from spectral noise estimates. Retrieval outputs include the NH<sub>3</sub> column, posteriori column retrieval uncertainty estimation, and column averaging kernel (AVK). The column AVK quantitatively represents the response of total NH<sub>3</sub> column to variations in the partial column at each layer, which is important for cross-comparison and model assimilation of satellite retrievals. A more detailed description of column AVK is provided in (Sheng et al., 2025).

**3.1 Global variations of NH<sub>3</sub> columns from the LEO constellation**

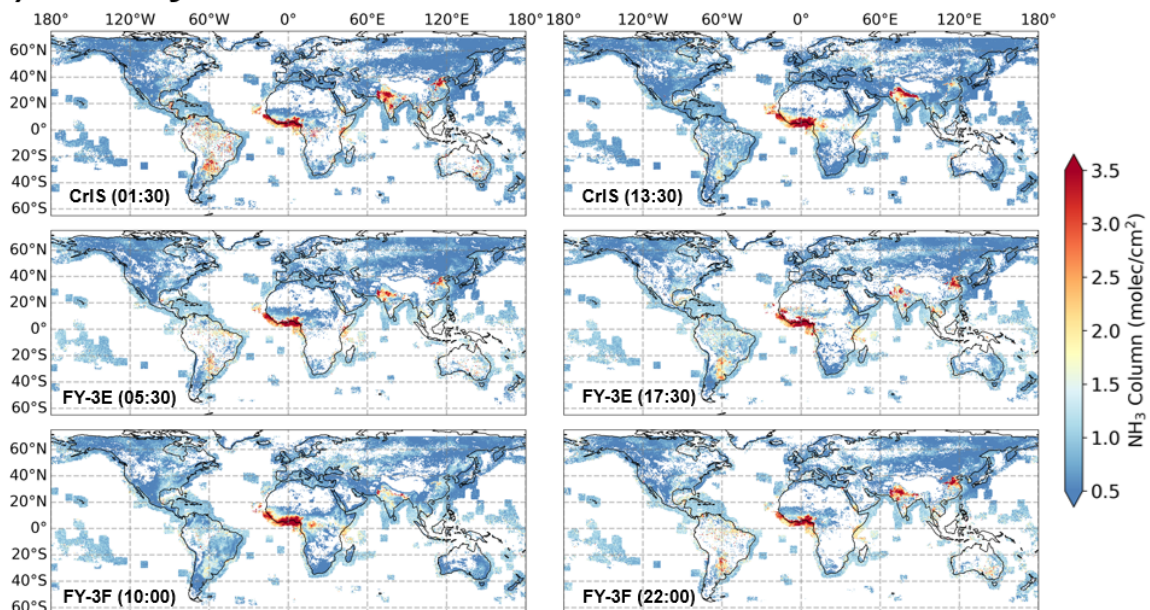
As an illustrative demonstration of the global sampling capability of the LEO constellation, we first examine representative seven-day mean NH<sub>3</sub> column distributions in January, April, July, and October 2024, specifically January 12–18, April 12–18, July 17–23, and October 1–7. These weekly maps are intended to demonstrate the global spatial coverage and broad seasonal contrast captured by the constellation, rather than to provide a climatological seasonal mean. We first retrieve global daily maps of NH<sub>3</sub> columns, re-grid them to a 0.5° × 0.5° spatial resolution, and then calculate seven-day averages for these representative periods. A strict quality control process is applied to ensure the reliability of satellites retrievals. Observations that fail to achieve convergence within 10 iterations are excluded from the dataset. We then implement a series of post-filters, retaining only those retrieval results that satisfy all the following criteria simultaneously. First, the retrieved NH<sub>3</sub> columns are required to be positive. Second, the absolute difference in surface skin temperature between the a priori and retrieved values is less than 10 K. Third, the retrieval error of the column does not exceed 300%. Fourth, the surface AVK is greater than 0.1. Fifth, the absolute thermal contrast (TC), defined as the temperature difference between the surface and the lowest atmospheric layer, is larger than 3 K. The absolute TC > 3 K and surface AVK > 0.1 criteria used here are intended for the global spatial-distribution analysis, where the objective is to illustrate the broad coverage and spatial patterns retrieved by the LEO constellation while retaining sufficient sampling density. These criteria remove retrievals with weak thermal contrast and limited near-surface sensitivity, but they are less restrictive than the criteria used later for the regional hotspot diurnal analysis. The effect of different TC and AVK thresholds on spatial distributions, diurnal cycles, and data retention is further evaluated and presented in the Supplementary Fig. S7-10. Sixth, a threshold for surface emissivity at 8.3 μm is set at 0.9, with only results meeting this threshold retain. This step is designed to avoid the occurrence of abnormally high NH<sub>3</sub> column concentrations over desert regions, consistent with the method of Clarisse et al. (2019). The surface emissivity data used for this filter are obtained from the Combined ASTER and MODIS Emissivity over Land Database Monthly Global 0.05° V003, which provides global monthly emissivity data at a 0.05° spatial resolution.

As shown in Figs. 2 and 3, broad seasonal contrasts in the representative weekly global NH<sub>3</sub> maps are evident across the four selected periods. In January, elevated NH<sub>3</sub> columns are concentrated in the Indo-Gangetic Plain and Central Africa, which is primarily associated with ongoing agricultural activities such as winter fertilizer application and extensive biomass burning for land clearance in these regions. In April, high NH<sub>3</sub> loading persists in the Indo-Gangetic Plain, East China and Central Africa, driven by intensified spring planting and corresponding fertilizer use in agricultural hotspots. Southeast Asia also exhibits increased NH<sub>3</sub> levels during this month, a pattern attributed to widespread slash-and-burn agriculture practices that are common in the regional dry season to prepare farmland. Additionally, scattered high NH<sub>3</sub> columns are observed in parts of North America, likely linked to localized agricultural operations and small-scale biomass burning. In July, high NH<sub>3</sub> columns remain concentrated in the Indo-Gangetic Plain, the North China Plain, Central Africa and Western Europe. The sustained high levels in these areas correlate with summer agricultural activities and elevated temperatures that enhance

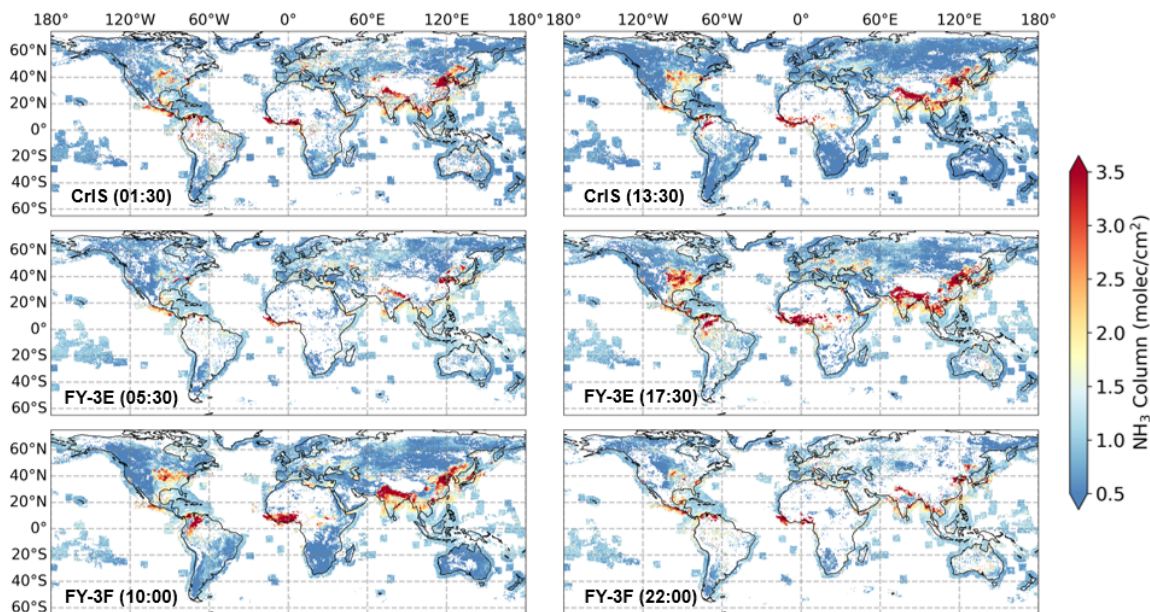
emissions (Ding et al., 2024). In particular, large-scale wildfires during the summer in North America exert a broad-range impact, leading to significantly elevated NH<sub>3</sub> columns across extensive regions as a result of biomass burning releasing substantial amounts of NH<sub>3</sub> (Lutsch et al., 2019). Within the smoke plumes, rapid gas-particle partitioning of NH<sub>3</sub> and favorable ammonium nitrate formation in cold, high-altitude conditions sustain column enhancements even as the plume disperses, reinforcing the widespread NH<sub>3</sub> elevations we observe (Lindaas et al., 2021). October still sees detectable high NH<sub>3</sub> columns over the Iní Plain, where post-harvest agricultural residues burning and late-season fertilizer application maintain elevated emissions. In South America, elevated NH<sub>3</sub> levels are mostly attributed to combined effects of fertilizer usage for autumn crops and biomass burning, as documented by Luo et al. (2022). The seasonal patterns reflect the close linkage between NH<sub>3</sub> distribution and region-specific anthropogenic activities, alongside seasonal natural processes. These spatial patterns exhibit general consistency with results from IASI (e.g., Van Damme et al., 2015) and CrIS (e.g., Shephard et al., 2020).

We note that these weekly global maps may still be influenced by short-term episodic events, such as biomass burning. Therefore, they are used here primarily as representative examples of the constellation-derived global NH<sub>3</sub> spatial distributions. The more quantitative regional diurnal and seasonal analyses in Section 3.2 are based on full-month averages using all available observations in each selected month.

### (a) January 12-18, 2024

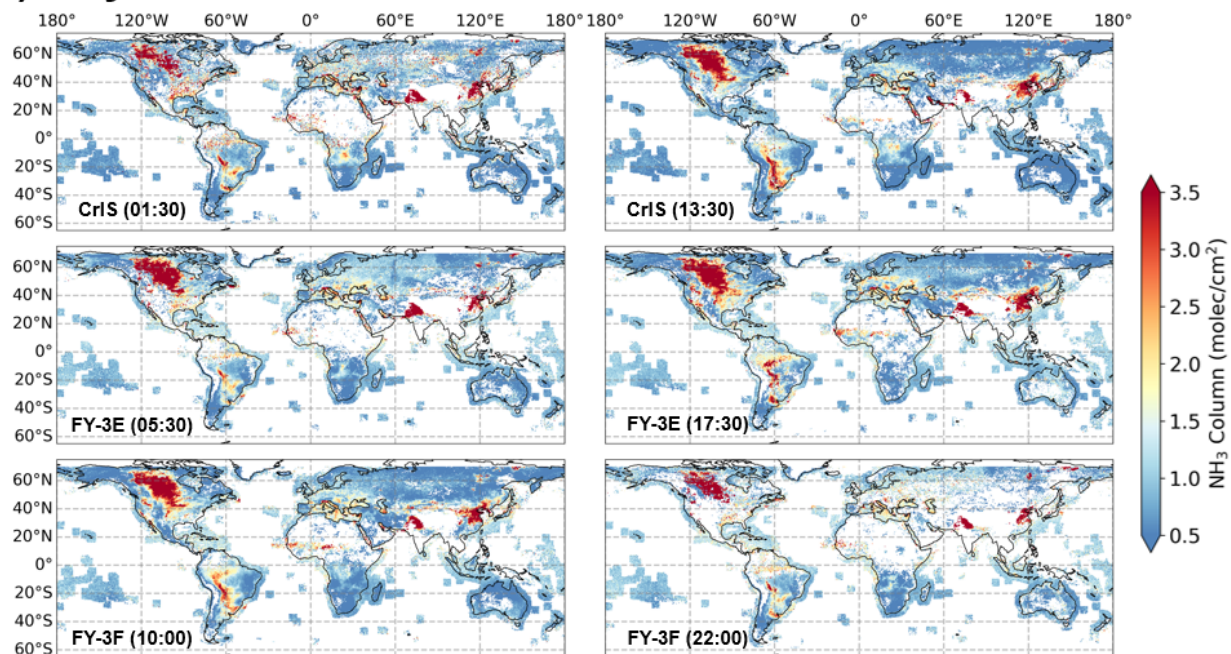


### (b) April 12-18, 2024



225 **Figure 2. Representative seven-day mean NH<sub>3</sub> columns retrieved from FY-3E, FY-3F, and CrIS for six different time slots, gridded to a 0.5° × 0.5° spatial resolution. The data correspond to the period of (a) January 12–18, 2024 and (b) April 12–18, 2024.**

### (a) July 17-23, 2024



### (b) October 1-7, 2024

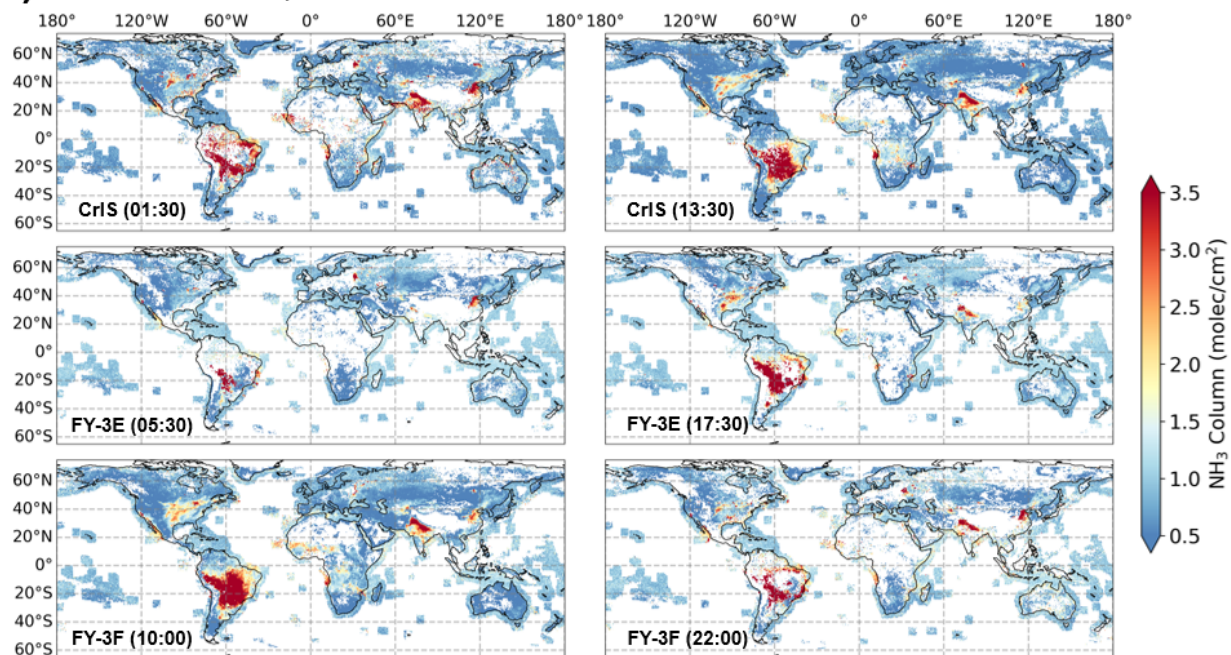


Figure 3. Same as Fig. 2, but for the period of (a) July 17–23, 2024 and (b) October 1-7, 2024.

### 3.2 NH<sub>3</sub> variations in global major source regions from the LEO constellation

230 To conduct in-depth analysis of NH<sub>3</sub> variations, we select three major emission hotspots, locating inside the North China Plain (118°E, 37°N), the Indo-Gangetic Plain (78°E, 28°N), and Central United States (100.5°W, 36.5°N), respectively. These hotspots are characterized by intensive agricultural activities and high livestock densities, consistent with the high-emission areas for their substantial NH<sub>3</sub> column abundances and distinct diurnal and seasonal dynamics. For each hotspot, we extract data within a 2.5° × 2.5° grid centered at the specified coordinates, covering six overpass times per day. A 2.5° × 2.5° box is  
235 selected to balance spatial representativeness and sample size. Because the analysis separates retrievals by satellite overpass time and month and further applies strict quality filters, a smaller box would often lead to insufficient valid retrievals, especially during nighttime or low-sensitivity conditions. The selected box size provides enough samples for stable monthly means while still focusing on the core NH<sub>3</sub> hotspot regions. The monthly mean is calculated only when the number of filtered retrievals within the 2.5° × 2.5° box exceed 10 during that month. TC affects the retrieval accuracy (Bauduin et al., 2017) and that higher  
240 absolute TC contributes to better retrievals (Clarisse et al., 2010). When TC is close to zero, the NH<sub>3</sub> spectral signal becomes weak and the retrieval sensitivity decreases, whereas a larger absolute TC generally enhances the spectral signal and improves the retrieval sensitivity. To ensure high quality NH<sub>3</sub> columns retrievals, we apply stricter sensitivity filters than those used for the global spatial-distribution maps in Section 3.1. Specifically, the same basic quality-control criteria are used, but the TC and AVK thresholds are tightened from absolute TC > 3 K and surface layer AVK value > 0.1 to absolute TC > 5 K and surface  
245 layer AVK value > 0.3. These stricter criteria are adopted to reduce the influence of local-time-dependent retrieval sensitivity on the inferred diurnal cycles, because TC itself has a pronounced diurnal variation.

The diurnal cycles of full-month averaged NH<sub>3</sub> columns for the three hotspots in each selected month are presented in Fig. 4, with IASI NH<sub>3</sub> ANNI V4.0 product overlaid for comparison. Also shown are the monthly mean planetary boundary layer heights (BLH) from ERA5 reanalysis. All three hotspots exhibit consistent seasonal differences in diurnal variability, a  
250 pattern that is consistent with the findings of Clarisse et al. (2021) from GIIRS observations and aligns with broader research on NH<sub>3</sub> dynamics (Tevlin et al., 2017). Warmer months (April–September) show distinct day-night contrasts, while winter months (December–February) display minimal diurnal fluctuations.

For the North China Plain (Fig. 4a), FY-3E, FY-3F, CrIS, and IASI data collectively reveal that warm-season NH<sub>3</sub> columns follow a clear diurnal pattern with a peak between 10:00 and 13:30 local solar time (LST), and nighttime values  
255 approximately 50–60% lower than daytime maxima. The peak corresponds to intensified agricultural activities (e.g., fertilization and livestock management) and temperature-driven volatilization from soils and animal waste, mechanisms identified by Ernst and Massey (1960) and Sutton et al. (2013) as primary drivers of daytime NH<sub>3</sub> emission enhancement. Higher BLH during the day facilitates vertical diffusion of NH<sub>3</sub> and contributes to the observed column dynamics (Saylor et al., 2010). In contrast, winter NH<sub>3</sub> columns over the North China Plain show negligible diurnal variation, which is probably  
260 due to the fact that cold-season emissions are limited by low temperatures and reduced agricultural activity (Kuang et al., 2020; Clarisse et al., 2021).

The Indo-Gangetic Plain (Fig. 4b) exhibits the highest overall  $\text{NH}_3$  columns among the three hotspots, with warm-season daytime columns nearly double those of the North China Plain. This regional difference stems from higher concentrations of  $\text{NO}_x$  and  $\text{SO}_2$  (Clarisse et al., 2021; Ding et al., 2024) over the North China Plain that act as neutralizing agents, shortening the lifetime of atmospheric  $\text{NH}_3$  (Liu et al., 2018; Wang et al., 2020). The most pronounced warm-season diurnal amplitude appears in the Indo-Gangetic Plain, with daytime columns up to three times higher than nighttime values. This pattern reflects the region's intensive irrigated agriculture, high livestock density (Perrone, 2020), and the exponential dependence of  $\text{NH}_3$  emissions on temperature (Hempel et al., 2016). Winter diurnal variations here are similarly weak, though  $\text{NH}_3$  columns remain higher than those in the other two regions due to persistent agricultural activity in milder winter conditions (Saraswati et al., 2018).

For the Central U.S. (Fig. 4c), the diurnal variation pattern is broadly consistent with the Asian regions but with a smaller amplitude. Warm-season  $\text{NH}_3$  columns peak between 11:00 and 14:00 LST, with daytime values approximately 1.5–2 times higher than nighttime levels, while winter columns show little diurnal fluctuation. This consistency supports that temperature, BLH dynamics, and diurnal emission sources (e.g., agricultural activities) are universal drivers of  $\text{NH}_3$  diurnal cycles across high-emission regions globally, which is evidenced by Saylor et al. (2010) from their analysis of  $\text{NH}_3$  measurements in the southeastern U.S.. The smaller amplitude compared to the Indo-Gangetic Plain likely reflects moderate temperature variations and balanced  $\text{NH}_3$  emission and deposition processes, as reported by Warner et al. (2017) in satellite observations of North American agricultural regions. Li et al. (2025) further emphasize that local deposition in vegetation-dense areas offsets large-scale transport, explaining the muted diurnal amplitude relative to Asian regions.

The spatial and seasonal dynamics of  $\text{NH}_3$  columns are further illustrated in Figs. 5–7 for the North China Plain, Indo-Gangetic Plain, Central U.S., respectively, retrieved by FY-3E/HIRAS-II, FY-3F/HIRAS-II and CrIS. Fig. A1 further illustrates monthly mean  $\text{NH}_3$  columns variations for these three regions, confirming consistent time series across the six daily satellite overpasses and coherent seasonal trends. All three regions exhibit a consistent seasonal peak in  $\text{NH}_3$  columns during spring to summer (March–August), with the lowest values in winter (December–February). For the North China Plain, all retrievals at six distinct times show that  $\text{NH}_3$  columns peak in June, with spatial coverage expanding across the entire plain during warm months, reflecting widespread agricultural activities (e.g., wheat fertilization and livestock waste management) and favorable meteorological conditions for emission (Zhang et al., 2010; Zhan et al., 2021). In contrast, winter maps display sparse, localized high- $\text{NH}_3$  patches, consistent with reduced agricultural activity and lower temperature-driven volatilization (Schjoerring et al., 1998).

For the Indo-Gangetic Plain, Fig. 6 shows that the region maintains the highest  $\text{NH}_3$  columns year-round, with spatial hotspots concentrated in the central plain during both day and night. However, daytime maps exhibit broader coverage extending to the plain's peripheries, while nighttime maps show more concentrated hotspots near emission sources. For the central U.S., Fig. 7 reveals the lowest overall  $\text{NH}_3$  columns among the three regions, with daytime spatial patterns spreading across the Corn Belt and Great Plains, and nighttime patterns contracting to smaller agricultural clusters.

295 The comparison between daytime and nighttime spatial patterns underscores the critical role of BLH in shaping NH<sub>3</sub>  
distribution. Daytime maps display broader spatial coverage because higher BLH (typically 1–3 km during warm-season days)  
facilitates vertical and horizontal diffusion of NH<sub>3</sub> away from emission sources. In contrast, nighttime maps show more  
localized NH<sub>3</sub> columns, as lower BLH (often <500 m at night) traps emissions near the surface (Walker et al., 2006; Wang et  
al., 2019). This pattern is especially pronounced over the North China Plain, where Figs. 5b-5d show contiguous high-NH<sub>3</sub>  
300 zones across the plain, while Fig. 5a, 5e and 5f limit high columns to central agricultural districts. For the Indo-Gangetic Plain,  
nighttime spatial maps also reveal persistent NH<sub>3</sub> in the residual layer, an effect linked to BLH collapse in the evening, which  
traps NH<sub>3</sub> at higher altitudes for several hours (Lonsdale et al., 2017; Kuang et al., 2020). This residual layer NH<sub>3</sub> explains  
why some nighttime retrievals show broader coverage than expected, even with low BLH, and aligns with Clarisse et al.  
(2021)’s observation that NH<sub>3</sub> vertical profile assumptions can impact nighttime retrieval accuracy.

305 To assess the impact of the filters on the retrieved NH<sub>3</sub> diurnal variation, we perform an additional sensitivity analysis  
using three filtering configurations. A loose filter with surface AVK > 0.1 and absolute TC > 3 K, the current hotspot filter  
with surface layer AVK value > 0.3 and absolute TC > 5 K, and a stricter filter with surface layer AVK value > 0.4 and absolute  
TC > 7 K. The remaining quality-control criteria are kept consistent among the three configurations. The resulting spatial  
distributions over the North China Plain and Indo-Gangetic Plain and the corresponding diurnal cycles are shown in the  
310 Supplementary Figs. S7–S10. The total numbers of quality-controlled retrievals retained for each satellite under the different  
filtering configurations are also reported in the subpanels. The three filtering configurations lead to different data-retention  
rates, as expected, but the major NH<sub>3</sub> hotspot patterns remain spatially similar and the regional diurnal cycles are not  
substantially distorted. This indicates that the main diurnal features discussed here are not strongly affected by the selected  
absolute TC and surface AVK thresholds, although the absolute column values and sampling density remain sensitive to  
315 filtering strength.

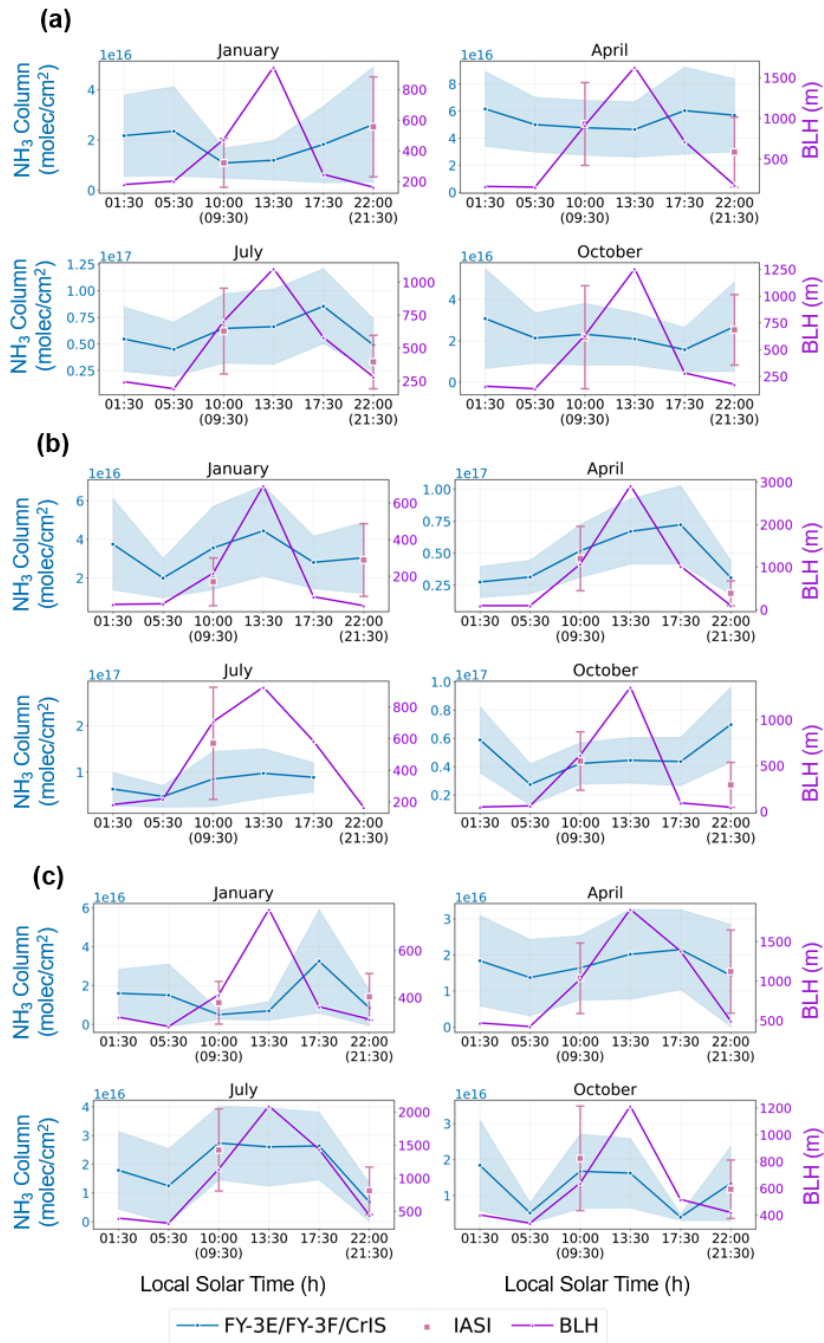
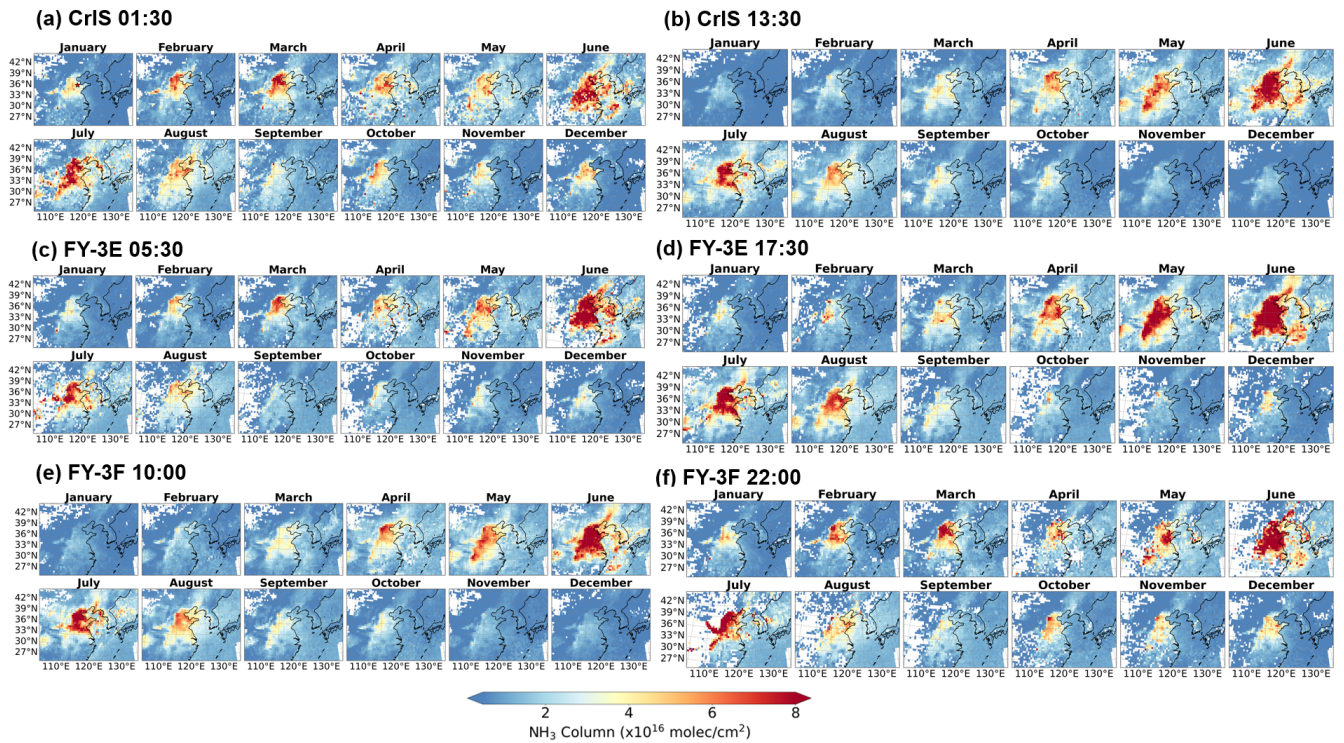
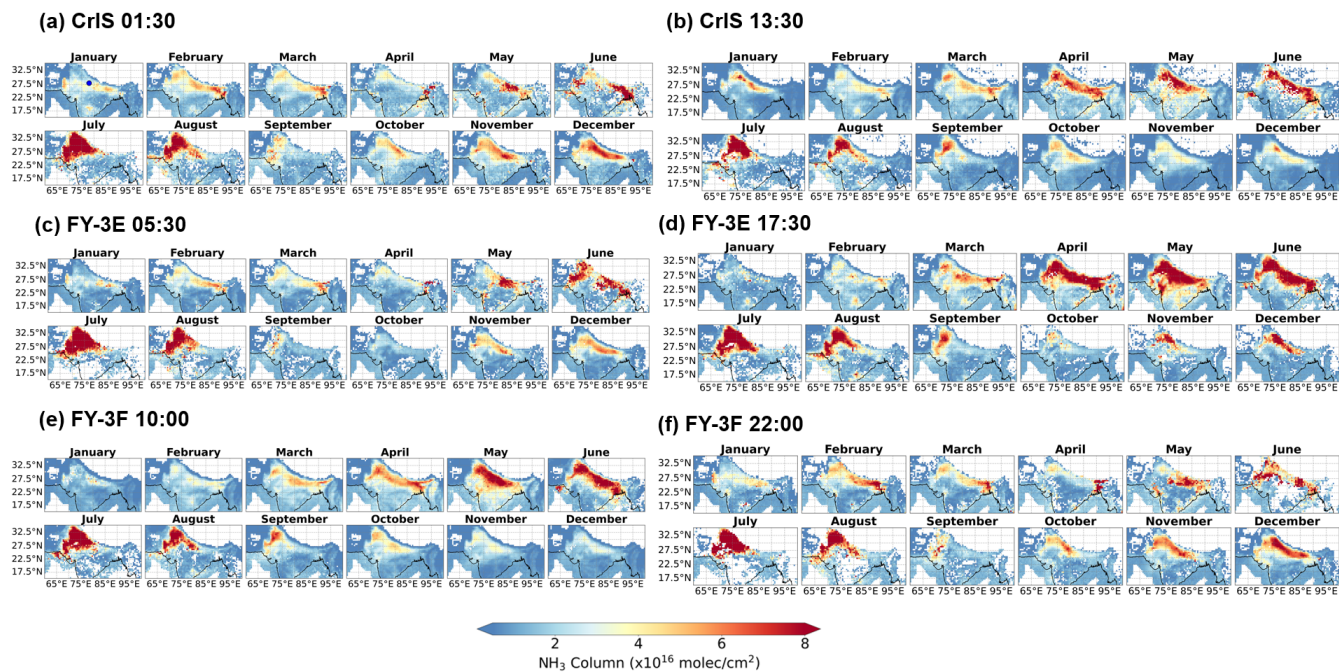


Figure 4. Diurnal cycles of retrieved NH<sub>3</sub> columns from FY-3E, FY-3F and CrIS over (a) the North China Plain (118°E, 37°N), (b) the Indo-Gangetic Plain (78°E, 28°N), and (c) Central U.S. (100.5°W, 36.5°N). The locations of these three sites are indicated in Figs. 5-7, respectively. Blue circles denote the mean values within the 2.5° × 2.5° grid, and the blue shaded area represents the standard deviation for each overpassing time. IASI retrieval results for the same region are overlaid for comparison: redish purple squares indicate IASI mean values, with error bars representing their standard deviation. The time axis is expressed in Local Solar Time. The diurnal variation of monthly mean planetary boundary layer height (BLH) is also plotted in purple.



325 **Figure 5.** Monthly mean  $\text{NH}_3$  columns ( $0.5^\circ \times 0.5^\circ$  grid) over the North China Plain retrieved by (a) CrIS at night overpasses (01:30 LST), (b) CrIS at daytime overpasses (13:30 LST), (c) FY-3E/HIRAS-II at dawn overpasses (05:30 LST), (d) FY-3E/HIRAS-II at dusk overpasses (17:30 LST), (e) FY-3F/HIRAS-II at morning overpasses (10:00 LST), and (f) FY-3F/HIRAS-II at night overpasses (22:00 LST), during 2024. The red star in the January panel of Fig. 5a indicates the location of the site ( $118^\circ\text{E}$ ,  $37^\circ\text{N}$ ) analyzed in Figs. 4 and 8.



**Figure 6.** Same as Fig. 5, but for the Indo-Gangetic Plain. The blue circle in the January panel of Fig. 6a indicates the location of the site (78°E, 38°N) analyzed in Figs. 4 and 8.

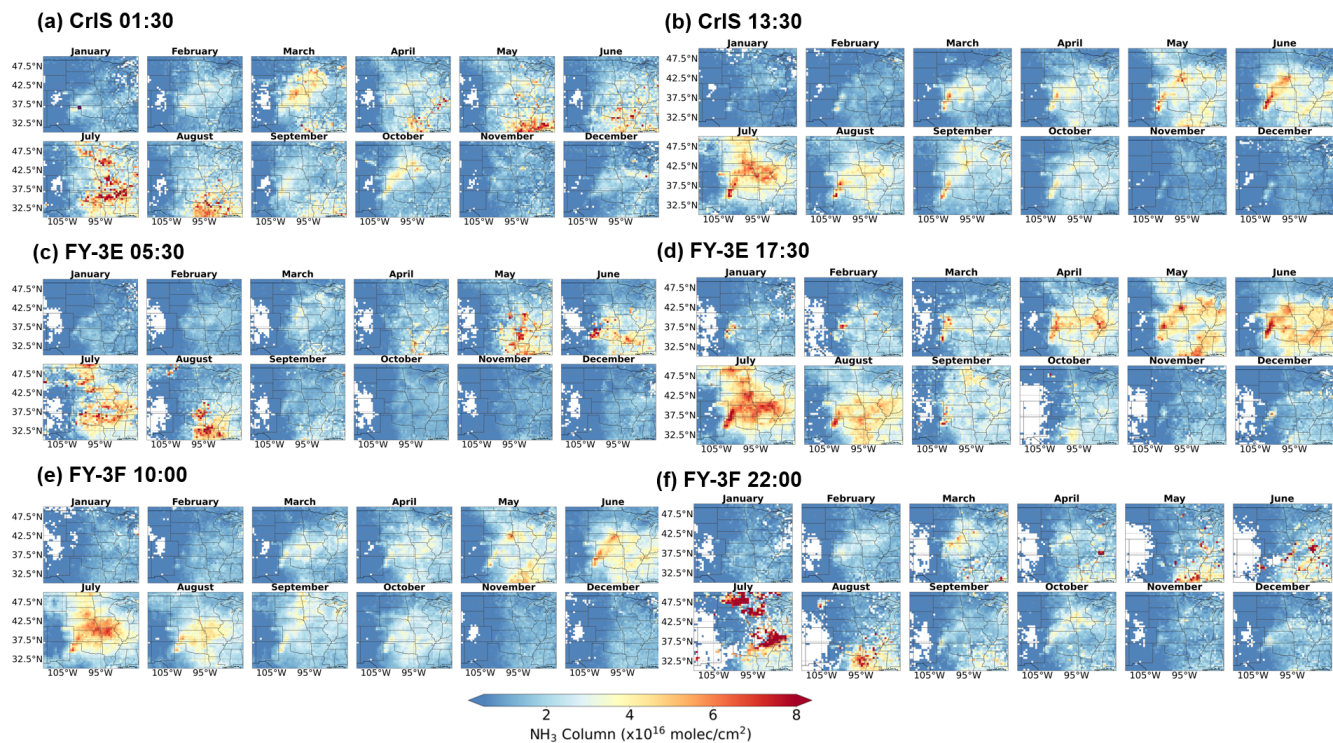


Figure 7. Same as Fig. 5, but for Central U.S.. The purple square in the January panel of Fig. 7a indicates the location of the site (100.5°W, 36.5°N) analyzed in Figs. 4 and 8.

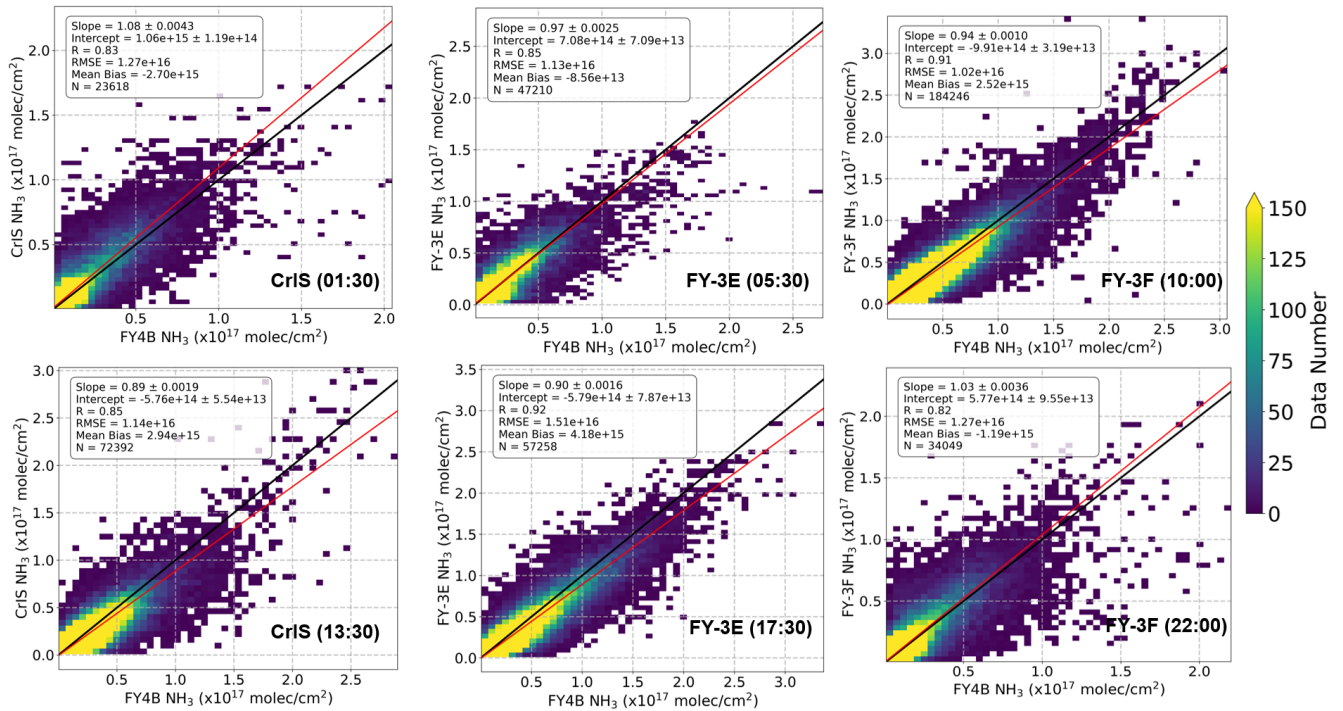
### 3.3 Cross-comparison with geostationary FY-4B/GIIRS and ground-based FTIR NH<sub>3</sub> observations

To evaluate the consistency of NH<sub>3</sub> column retrievals from the polar-orbiting constellation, we perform spatiotemporally collocated cross-comparisons between FY-3E/HIRAS-II, FY-3F/HIRAS-II, CrIS, and geostationary FY-4B/GIIRS retrievals. The comparisons are conducted over the Indo-Gangetic Plain (75°E–95°E, 15°N–35°N) and the North China Plain (110°E–  
345 130°E, 25°N–45°N), where strong NH<sub>3</sub> signals are frequently observed and FY-4B/GIIRS provides geostationary coverage. A polar-orbiting retrieval and a FY-4B/GIIRS retrieval are considered collocated when their longitude and latitude differences are both less than 0.5° and their observation time difference is less than 0.5 h. These thresholds are selected as a compromise between retaining sufficient matched samples and minimizing spatial and temporal representativeness differences.

The comparisons in Fig. 8 also show broad consistency across the matched local overpass periods sampled by the three  
350 polar-orbiting sensors. Here, the observational periods refer to the local-time periods of the polar-orbiting observations collocated with FY-4B/GIIRS, namely CrIS at 01:30/13:30, FY-3E/HIRAS-II at 05:30/17:30, and FY-3F/HIRAS-II at 10:00/22:00. The agreement across these matched local-time periods indicates that the constellation retrievals are generally consistent with FY-4B/GIIRS over both nighttime/dawn and daytime/dusk observations, although the agreement is typically stronger during daytime because of larger thermal contrast and higher retrieval sensitivity.

We further perform seasonal comparisons between each polar-orbiting sensor and FY-4B/GIIRS using the same  
355 collocation and quality-filtering criteria. These results are shown in the Figs. A2. The seasonal regression slopes mostly fall within the range of 0.7–1.1. The slopes slightly below unity in the annual comparison should not be interpreted as clear evidence of a persistent systematic bias in either FY-4B/GIIRS or the polar-orbiting retrievals. Instead, the remaining differences likely due to their differences in retrieval sensitivity, collocation representativeness, sampling footprints, viewing  
360 geometry, and vertical sensitivity. These factors are particularly important for NH<sub>3</sub> because of its short atmospheric lifetime and strong spatial heterogeneity.

We also compare the polar-orbiting satellite retrievals with ground-based FTIR NH<sub>3</sub> column measurements at Hefei, China (Wang et al., 2022). Satellite retrievals within a 1° × 1° spatial window and a 1 h temporal window around the FTIR observations are averaged and compared with the corresponding measurements. The comparison is shown in Fig. A3. The  
365 satellite retrievals show reasonable consistency with the FTIR observations, although the collocated numbers are limited, with slopes of  $0.87 \pm 0.22$ ,  $1.03 \pm 0.08$ , and  $1.13 \pm 0.18$  for FY-3E/HIRAS-II, FY-3F/HIRAS-II, and CrIS, respectively. The corresponding RMSE values range from  $7.80 \times 10^{15}$  to  $1.11 \times 10^{16}$  molec/cm<sup>2</sup>. Overall, the annual and seasonal FY-4B/GIIRS comparisons, together with the Hefei FTIR comparison, support the broad consistency of the LEO constellation retrievals in capturing major NH<sub>3</sub> spatial and temporal variability.



375

**Figure 8.** Comparisons of  $\text{NH}_3$  columns between polar-orbiting satellites (FY-3E/HIRAS-II, FY-3F/HIRAS-II and CrIS) retrievals and geostationary satellite (FY-4B/GIIRS) retrievals over the Indo-Gangetic Plain ( $75^\circ\text{E}$ – $95^\circ\text{E}$ ,  $15^\circ\text{N}$ – $35^\circ\text{N}$ ) and the North China Plain ( $110^\circ\text{E}$ – $130^\circ\text{E}$ ,  $25^\circ\text{N}$ – $45^\circ\text{N}$ ) at different times of the day in 2024. Observations are collocated when the differences in longitude and latitude are both less than  $0.5^\circ$  and observation time is less than 0.5 hours. The black dashed line represents the 1:1 equivalence line, with the red line indicating the orthogonal distance regression (ODR) fit. The fit's slope, intercept, correlation coefficient (R), and root mean square error (RMSE), mean bias (calculated as polar satellite retrievals minus geostationary satellite retrievals), data number are also provided.

### 3.4 Diurnal variations of the surface AVK, the retrieval uncertainty and the TC

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Fig. 9 presents the diurnal and seasonal dynamics of surface AVK, TC, and column retrieval uncertainty over three representative hotspots, locating in the North China Plain ( $118^\circ\text{E}$ ,  $37^\circ\text{N}$ ), the Indo-Gangetic Plain ( $78^\circ\text{E}$ ,  $28^\circ\text{N}$ ), and the Central U.S. ( $100.5^\circ\text{W}$ ,  $36.5^\circ\text{N}$ ). The statistics are derived from FY-3E/HIRAS-II, FY-3F/HIRAS-II, and CrIS retrievals within  $2.5^\circ \times 2.5^\circ$  boxes and are shown as monthly averages for January, April, July, and October across six local overpass times. To clarify the robustness of these monthly averages, the number of valid retrievals used for each region, month, and overpass time is provided in Table S2. Monthly means are shown only when more than 10 quality-controlled retrievals are available.

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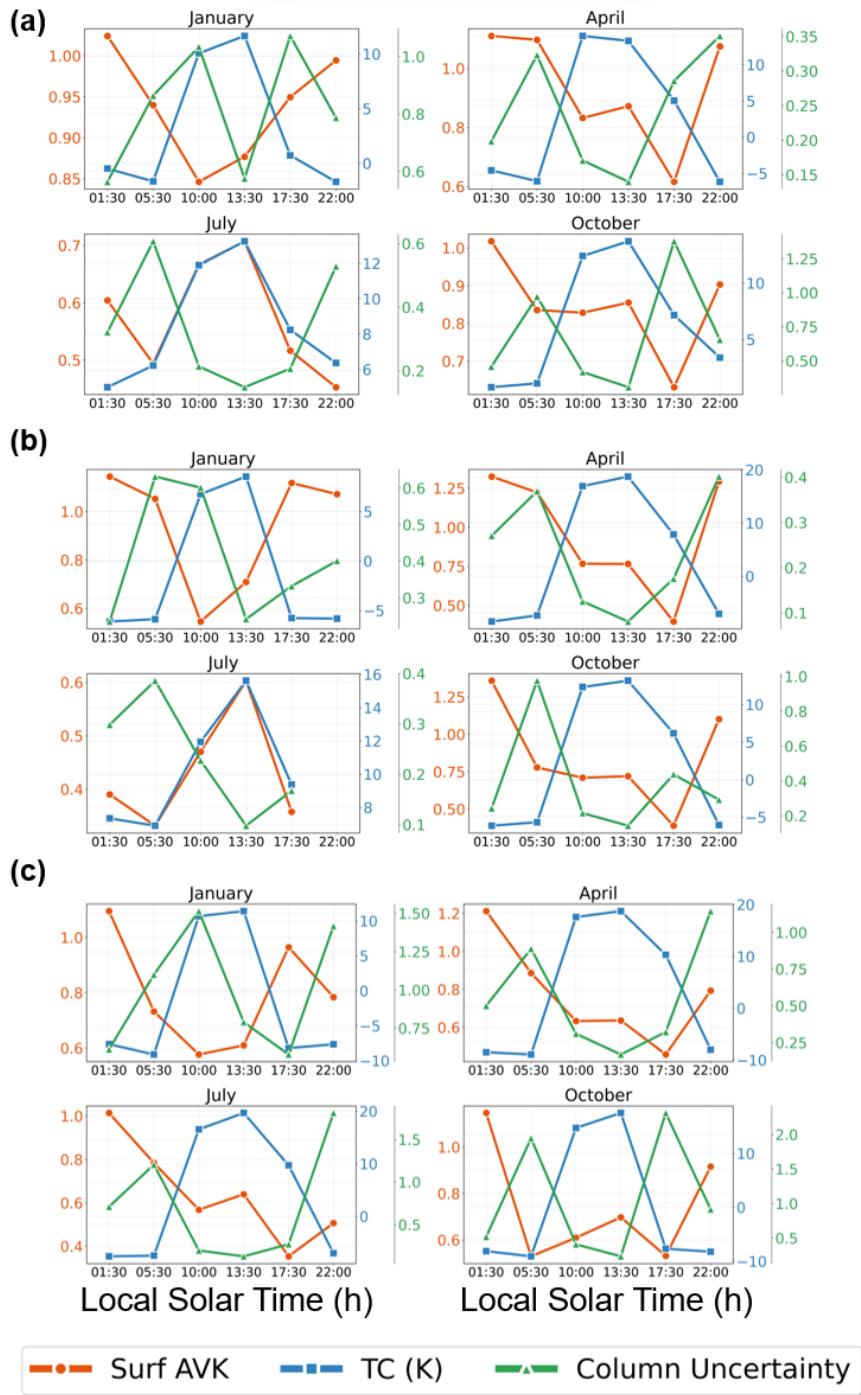
TC exhibits a consistent diurnal pattern across all regions. Midday overpasses (10:00–13:30 local solar time) see TC peak with positive values ranging from 5 to 12 K in warm months (April and July), while nighttime and early morning (01:30–05:30) bring near-zero or negative TC, particularly in January and October. Seasonal contrasts in TC amplitude are striking.

Warm months feature fluctuations of up to 15 K in the Indo-Gangetic Plain, driven by intense solar heating, while cold months (January) show smaller variations, with TC confined to  $\pm 5$  K for most overpasses.

The surface layer value of column AVK, which quantifies the observational information content related to near-surface  $\text{NH}_3$ , tracks TC closely across all regions and seasons. Midday overpasses coincide with peak surface layer column AVK (0.3–0.6), indicating robust sensitivity to near-surface  $\text{NH}_3$  variations when TC is strongest. In contrast, surface layer column AVK at nighttime and dawn drops to 0.1–0.3, especially in January. The Indo-Gangetic Plain exhibits the most pronounced diurnal swings, with July midday surface layer column AVK exceeding 0.6, while the Central U.S. maintains more moderate, stable values (0.2–0.4) across all seasons. Seasonally, surface layer column AVK remains consistently higher in April and July relative to January and October, confirming that warm-season midday conditions offer the most useful observational constraints for  $\text{NH}_3$  retrievals.

Column retrieval uncertainty generally decreases under conditions with larger absolute TC and higher surface AVK, but this relationship is not purely inverse and should not be interpreted as strictly monotonic. In particular, strongly negative TC can also enhance thermal infrared sensitivity because absorption-like spectral features may become emission-like features, as shown for lower-tropospheric pollution retrievals by Boynard et al. (2014). Therefore, TC should be interpreted together with AVK and retrieval uncertainty, rather than as a standalone monotonic sensitivity indicator.

Daytime bins often contain more valid retrievals than nighttime or dusk/evening bins, but the pattern is not uniform across regions and months (Table S2). Some bins have relatively sparse sampling, such as the Indo-Gangetic Plain in July at 05:30 and 22:00 and Central U.S. in October at 17:30, whereas other daytime bins contain more than 1000 valid retrievals. These differences mainly reflect the combined effects of cloud screening and sensitivity-related quality filters.



410 **Figure 9. Diurnal cycle of surface AVK, TC and column uncertainty from FY-3E, FY-3F and CrIS over three hotspots in (a) the North China Plain (118°E, 37°N), (b) the Indo-Gangetic Plain (78°E, 28°N), and (c) Central U.S. (100.5°W, 36.5°N). The number of valid retrievals used for each monthly mean is provided in Table S2.**

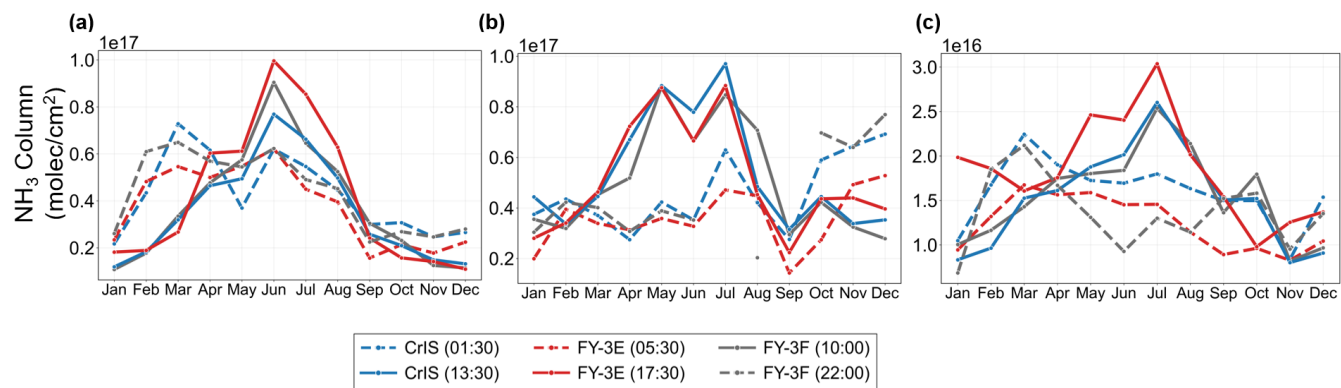
## 4 Conclusions

This study develops a global monitoring framework by integrating three polar-orbiting hyperspectral infrared sounders, including FY-3E/HIRAS-II in a dawn-dusk orbit, FY-3F/HIRAS-II in a mid-morning orbit, and CrIS in an afternoon orbit. This constellation achieves quasi-geostationary-like coverage, enabling six global NH<sub>3</sub> column retrievals based on the optimal estimation method, for every 4-hour per day. This framework successfully captures spatial, diurnal, and seasonal dynamics of tropospheric NH<sub>3</sub> worldwide, with elevated columns consistently identified in major agricultural hotspots. Annual and seasonal cross-comparisons with geostationary FY-4B/GIIRS show broad consistency between the LEO constellation retrievals and GIIRS over the matched local overpass periods, including CrIS at 01:30/13:30, FY-3E/HIRAS-II at 05:30/17:30, and FY-3F/HIRAS-II at 10:00/22:00. An additional comparison with ground-based FTIR observations at Hefei provides independent support for the reliability and consistency of the retrievals.

Key findings indicate that temperature-dependent volatilization, boundary-layer dynamics, and agricultural activity patterns jointly regulate the diurnal and seasonal variability of NH<sub>3</sub>. Differences among regions are further modulated by source strength, fertilizer and livestock management practices, meteorological conditions, and atmospheric chemical processing. The constellation effectively addresses the longstanding gap of high-frequency NH<sub>3</sub> observations in regions lacking geostationary satellite coverage, providing a comprehensive dataset for global NH<sub>3</sub> cycling research.

Looking forward, the upcoming integration of FY-3H/HIRAS-II launched at the end of 2025, equipped with the same HIRAS-II sensor as FY-3E and FY-3F, will homogenize the constellation, further enhancing data consistency and spatial-temporal coverage. This refined system holds great potential to advance NH<sub>3</sub> emission quantification, improve parameterizations in global climate and air quality models, and support evidence-based policies for anthropogenic source management. Future work could focus on optimizing NH<sub>3</sub> vertical profile parametrization, cross-calibrating between sensors onboard different satellite for enhanced retrieval accuracy and consistency, and expanding the constellation's applications to study NH<sub>3</sub>'s links with aerosol formation, acid deposition, and ecosystem impacts. To summarize, this framework demonstrates the value of synergizing polar-orbiting satellites to strengthen global atmospheric composition monitoring, contributing to more effective mitigation of NH<sub>3</sub>-related environmental and climate effects.

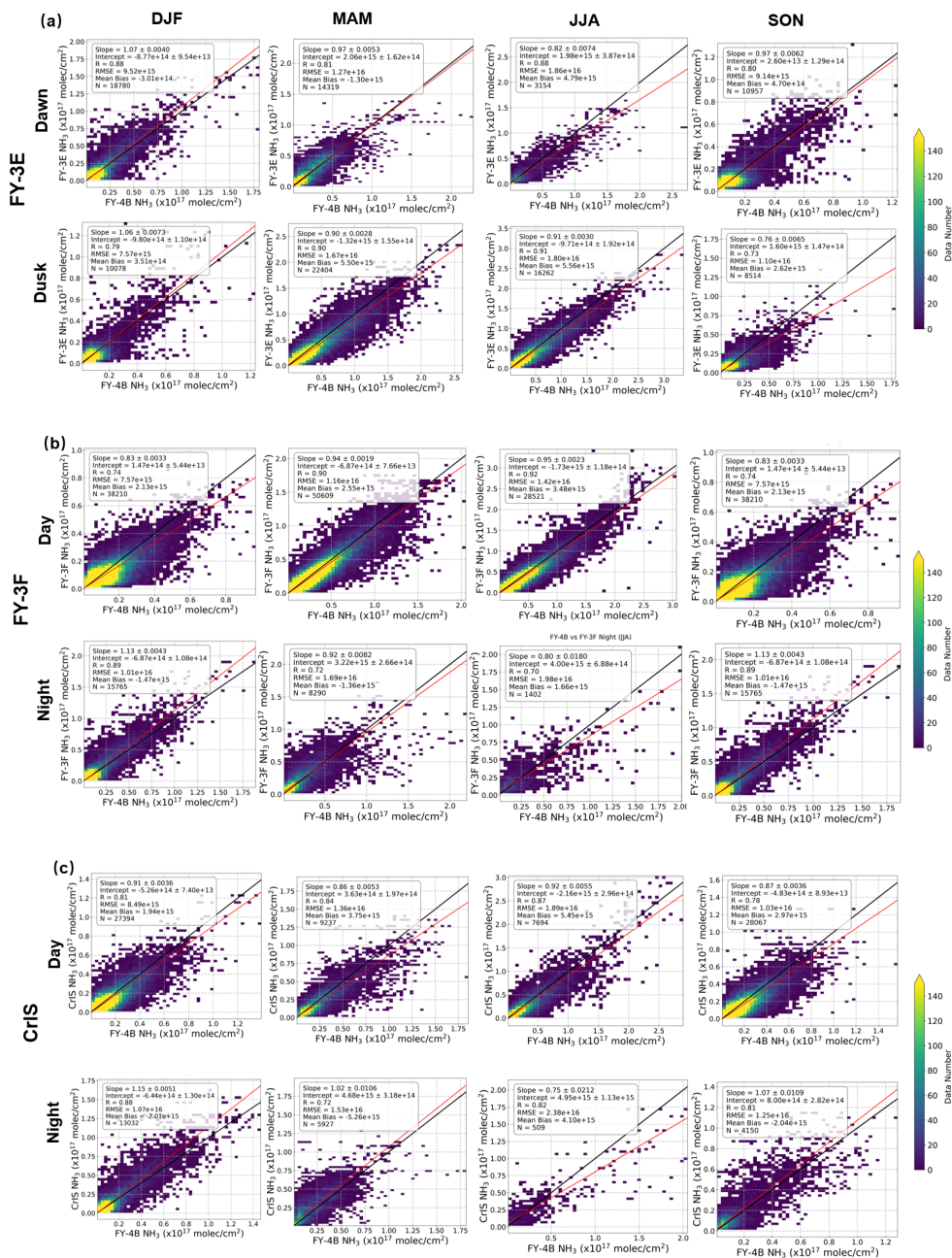
## Appendix A



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**Figure A1. Seasonal variation timeseries of retrieved  $\text{NH}_3$  columns over (a) the North China Plain, (b) the Indo-Gangetic Plain and Central U.S.**

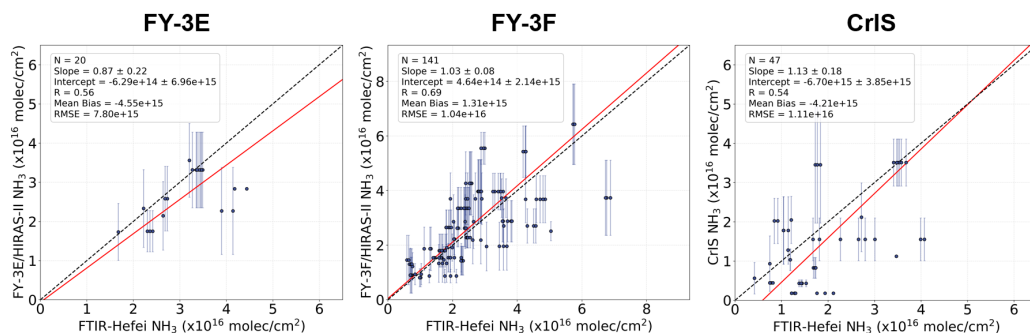
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Figure A2. Same as Fig. 8, but for seasonal comparisons between FY-3E/HIRAS-II (a), FY-3F/HIRA-II (b), CrIS (c) and FY-4B/GIIRS over the Indo-Gangetic Plain and the North China Plain. DJF, MAM, JJA, and SON denote December–January–February, March–April–May, June–July–August, and September–October–November, respectively. The black solid line represents the 1:1 line, and the red solid line indicates the orthogonal distance regression (ODR) fit. The slope, intercept, correlation coefficient (R), root mean square error (RMSE), mean bias, and number of collocated samples are provided in each panel.

455



460 **Figure A3. Validations with ground-based FTIR in Hefei, China. The black dashed line represents the 1:1 equivalence line, with the red line indicating the orthogonal distance regression (ODR) fit. The fit's slope, intercept, correlation coefficient (R), root mean square error (RMSE), mean bias (calculated as polar satellite retrievals minus ground-based measurement), and data number are also provided. The dot represents the mean value of satellite retrieved NH<sub>3</sub> columns and the error bar represents 1 standard deviation. If there is only one match point, the error bars are not displayed.**

#### 465 Data availability

The FY-3E and FY-3F NH<sub>3</sub> retrieval datasets are publicly available at ZENODO (<https://doi.org/10.5281/zenodo.18359451> and <https://doi.org/10.5281/zenodo.18366114>, respectively). Currently, the FY-3E dataset is available from January 2023 to December 2024, and the FY-3F dataset is available from January to December 2024. The GIIRS NH<sub>3</sub> data (from July 2022 to June 2025) used in this study are publicly available on Zenodo (<https://doi.org/10.5281/zenodo.17193848>). Further updates on  
 470 FengYun NH<sub>3</sub> data will be provided on the project website (<https://fengyunair.github.io/>). FengYun satellite Level 1 data are publicly available from the FengYun Satellite Data Center at <http://satellite.nsmc.org.cn/portalsite/default.aspx>. The IASI NH<sub>3</sub> ANNI V4.0 product is available from the AERIS data infrastructure (<https://iasi.aeris-data.fr/>). The CrIS L1 spectra are publicly available from <https://search.earthdata.nasa.gov/>.

#### Author contributions

475 ZZ designed the experiments, and JH carried them out. ZZ and JH developed the model code and JH performed the simulations. MS and RZ assisted the satellite data pre-processing and results analysis. JH prepared the manuscript with contributions from all co-authors.

#### Competing interests

At least one of the (co-)authors is a member of the editorial board of Atmospheric Measurement Techniques.

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