Authors' Response to Reviews of

South Asia ammonia emission inversion through assimilating IASI observations

Ji Xia*, Yi Zhou*, Li Fang, Yingfei Qi, Dehao Li, Hong Liao, and Jianbing Jin
Atmospheric Chemistry and Physics Discussions,

RC: Reviewers' Comment, AR: Authors' Response, □ Manuscript Text

General Comments:

RC: ***The paper describes the application of an ammonia emission inversion system over South Asia. As emissions of ammonia are rather difficult to estimate by emission models, this approach is very useful to obtain insight in the actual emission strength. Especially the time period of the seasonal emission peak(s) is difficult to model, but the described system seems able to provide a better estimate for that. The system uses IASI satellite observations to constrains the emissions; results are validated by comparing posterior simulations with observations from the CrIS satellite instrument and observations from a ground network. The temporal resolution of the emission estimates is monthly, which is rather course compared to the high frequency changes present in ammonia emissions. For the described study that seems a logical choice, as also the prior emission inventory is monthly.***
Although the paper focusses on the application of the inversion system, the setup of the inversion is described sufficiently well. For some parts a more detailed description could be useful, as described below in the Specific Comments. Overall, the paper is easy to read, and could be published after some minor modifications.

Response to Referee #2: We would like to thank the referee for the careful review throughout the paper and the in-depth comments that help to improve our paper.

Specific Comments:

RC:1) could the authors discuss the potential of their system for higher temporal resolution estimates? What are the current limitations for application on weekly or even daily scale? Is the availability and/or quality of the satellite observations a limitation, or simply the computing resources?

AR: Thanks for comment. The system already has the potential for higher temporal resolution estimates. With data available from the FY-4 satellite, we can access daily or even two-hourly observations. These datasets allow us to explore higher temporal resolution (e.g., daily or weekly

scale) spatiotemporal variation characteristics. Our next steps will focus on further refining the spatiotemporal patterns at the daily or weekly scale, building on the current posterior results.
As for the limitations, the primary constraint is not the availability or quality of satellite observations, but rather the computational resources required. While satellite data is sufficient to support higher temporal resolution estimates, processing these datasets at high spatial and temporal resolutions demands significant computational power and storage. Moving forward, we will optimize the computational methods and leverage more powerful computational platforms to achieve higher resolution temporal estimates.

Text in manuscript:

4 Summary and conclusion

…

The top-down NH_3 emission inversion system driven by IASI observations has demonstrated superior performance in enhancing the NH_3 emission estimates. Nevertheless, several challenges persist, such as the requirement for simulations at finer resolutions to precisely capture very local emission dynamics. Furthermore, observations from stationary satellites, such as FY-4B, also deserve attention for exploring the diurnal variations of the NH_3 emission. Our next steps will focus on further refining the spatiotemporal patterns at the daily or weekly scale, building on the current inversion system.

RC: 2) The inversion now uses IASI observations to constrain emissions, and CrIS observations for validation. Would it be possible to use instead the CrIS observations to constrain the emissions? The results show quite some differences between IASI and CrIS NH_3 columns; would inversion of CrIS data give very different results?

AR: Appreciate your comment. The results of emission inversions are highly dependent on the type of observations used. Different sensors provide different sensitivities and spatial coverage, which can lead to variations in the inversion outcomes. We chose to use IASI observations for the inversion because IASI provides a larger volume of data, which increases the robustness of the emission estimates. Since IASI observations are collected from three satellites, this results in an even larger dataset. While CrIS observations are valuable, we did not use them in this study because we have indicated underestimation issues in South Asia region with CrIS NH_3 columns. At the surface, where CrIS typically has lower sensitivity, it tends to overestimate in low-concentration conditions and underestimate in higher atmospheric concentration conditions (Dammers E et al., 2017). As a result, we opted for IASI data, which provides more reliable constraints in this case. Additionally, the posterior emission estimates, which are based on CrIS, have now been included as supplementary material. Furthermore, we have also added this point in the manuscript to ensure clarity.

Text in manuscript:

3.2 Spatial and Seasonal variation of NH3 emission

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The convergence of prior and posterior emission intensities in June is attributed to the overall offsetting of negative and positive increments in the region, as shown in Figure S7 (f). As depicted in panel (c) of Fig. 3, the negative increments observed in January and April primarily originate from the Indian region, while the positive increments in July and September are predominantly observed in the same area. Additionally, the posterior emission estimates, which are based on CrIS, have now been included as supplementary material.

RC: 3) p 3, line 32: When the monthly averages over grid cells are calculated, is there any spatial or temporal weighting applied? For example, a spatial weight based on the overlap between a pixel footprint and the target grid cell, or a temporal weight based on the instrument error?

AR: Many thanks for your feedback. We did not apply any spatial or temporal weighting. The data is processed by reading daily NH_3 values and adding them to the corresponding grid cells. For each valid data point, if its latitude and longitude fall within the specified range, its NH_3 value is added to the corresponding grid cell.

RC: 4) p 4, line 14-15: What are the units of these variables? A more standard formulation of the kernel application would look like:

Xm = Xa + A (m - B)

Could Eq (1) be rewritten to this actually?

How is the averaging kernel applied to the model data exactly? Is a monthly averaged kernel applied to monthly averaged concentration? If so, how is the monthly averaged model concentration calculated, as an average over all ours, or using time of overpass only? Or are the individual pixels simulated from the model first, and then averaged over grid cells and months?

AR: Thank you for your valuable comment. Apologies for the confusion. There are two methods in (Clarisse et al., 2023) and we used the model vertical profile as a prior to recalculate the ammonia column concentration from IASI. So the formula for this method is still different from Xm = Xa + A (m - B). To clarify, in Eq. 1:

$$m_z = \frac{M_z^m - B_z}{M^m - B}$$

M_z^m represents the modeled concentration of NH_3 at altitude **z**.
B_z is the background concentration of NH_3 at the same altitude.
M^m represents the total modeled concentration of NH_3 in the atmosphere.
B is the total background concentration.

$$A_z^a = \frac{1}{N}\frac{\hat{X}^a - B}{\hat{X}^{|z} - B}$$

$\hat{X}^{|z}$ represents the a priori (or assumed) concentration of NH_3 at altitude **z**.
B_z is again the background concentration at that altitude.
$\hat{X}^a$ is the total a priori concentration.
N is a normalization factor, ensuring the matrix A_z^a sums correctly to account for all altitudes.
The specific description of the relevant part is as follows:

Text in manuscript:

2.1 *IASI satellite measurements*

…

Here, $\hat{X}^m$ represents the IASI column concentration retrieved with model profile. $\hat{X}^a$ denotes the initial IASI column concentration, with the background concentration B. The Aa z values are AVK for each vertical layer, with the model profile m_z. More detailed information and the corresponding equations are provided in the supplementary materials equation S8 and S9.

Supplement

…

$$m_z = \frac{M_z^m - B_z}{M^m - B}$$

here M_z^m represents the modeled concentration of NH_3 at altitude $z. B_z$ is the background concentration of NH_3 at the same altitude. M^m represents the total modeled concentration of NH_3 in the atmosphere. B is the total background concentration.

$$A_z^a = \frac{1}{N}\frac{\hat{X}^a - B}{\hat{X}^{|z} - B_z}$$

here $\hat{X}^{|z}$ represents the a priori (or assumed) concentration of NH_3 at altitude $z. B_z$ is again the background concentration at that altitude. $\hat{X}^a$ is the total a priori concentration. N is a normalization factor, ensuring the matrix A_z^a sums correctly to account for all altitudes.

RC: 5) Would it also be possible to not use monthly averaged observations, but simply all observations individually? The estimated emission state could still be monthly, so what is the reason for using monthly averaged observations?

AR: Thanks for comment. Using all individual observations without averaging would indeed be possible, but there are two main reasons we use monthly and grid averaging. First, averaging aligns the satellite observations with the model simulation's grid resolution, ensuring that we are comparing like with like. Second, averaging dramatically reduces the size of the observational vector used in the assimilation, which in turn lowers the computational cost. For example, without averaging, the observational vector y might have a size of around 1,000,000, whereas with monthly and grid averaging, it is reduced to about 1,000. This reduction makes the assimilation analysis far more computationally efficient while still accurately representing the monthly emission state. We have also added relevant explanations for this section in the article belew:

Text in manuscript:

2.1 IASI satellite measurements

…

The assimilated observations for estimating the NH_3 emissions were the monthly IASI column concentration means over the 0.5 ◦× 0.625 ◦GEOS-Chem grid cell. These values were derived from the latest ANNI-NH_3-v4R-ERA5 product. Despite improvements in NH_3 column retrievals from satellite observations, there remains substantial variability in measurement uncertainty, ranging from 5 % to over 1000 %. (Van Damme et al., 2014; Whitburn et al., 2016; Van Damme et al., 2017). Data selection was performed by excluding nighttime observations, irrational values (<0), and only using data with a cloud fraction< 0.1 (Van Damme et al., 2018) and skin temperature > 263 K (Van Damme et al., 2014) during the calculation of the monthly mean. Additionally, while negative values are not necessarily incorrect, they are considered unrealistic in the context of NH_3 concentrations. To improve the quality of the monthly average, we removed those negative values. It is also important to note that we used daily observations from three satellites, each with a pixel resolution of approximately 12 km × 12 km, which provided us with sufficient observations to calculate the monthly average. We applied a selection criterion, using only grid averages that contain a minimum of 80 observations. This ensures that the grid-averaged values are statistically representative and that the monthly mean is of high quality. Notably, the time coverage of the available version 4 IASI product used was limited: Metop-A provided data for the entire year of 2019, Metop-B provided data from January to July 2019, and Metop-C did not have data for 2019. Therefore, only the data from Metop-A and Metop-B within the 2019 time frame were used in this study. To further improve the data quality and ensure consistency, we performed monthly and grid averaging of the observations. This approach not only allows for a fair comparison between the observed and modeled NH_3 concentrations but also reduces the computational cost of the assimilation process. Using individual observations without averaging would result in an excessively large observational vector, which would significantly increase the computational burden. For example, without averaging, the size of the observational vector could reach 1,000,000, while with monthly and grid averaging, it is reduced to a manageable size of around 1,000. This reduction in size helps to optimize the data assimilation process while maintaining the integrity of the emission estimates.

RC: 6) Negative values are not necessarily wrong. The uncertainty of these values is probably high, so the "true" value is still a very likely outcome. By removing the negative observations, the monthly average will have a positive bias. Could this be discussed?

AR: Thank you for your comment. We acknowledge that negative values are not necessarily wrong in an assimilation context if they have large uncertainties. However, in a physical sense, negative NH_3 concentrations are not realistic. To improve the quality of the monthly averages, we removed these negative values. It is also important to note that our monthly averages are calculated using daily observations from three satellites, each with a pixel resolution of 12km × 12 km. This means we have a large number of observations available for each grid cell. Additionally, we apply a selection criterion: a grid average is only used if it contains a minimum of 80 observations. This ensures that the final grid-averaged value is statistically representative and minimizes potential biases in the monthly mean. Given this approach, we believe that removing negative values does not introduce a significant positive bias but rather enhances the reliability of the data used in the assimilation. We have also added relevant explanations for this section in the article belew:

Text in manuscript：

2.1 IASI satellite measurements

…

The assimilated observations for estimating the NH_3 emissions were the monthly IASI column concentration means over the 0.5 ◦× 0.625 ◦GEOS-Chem grid cell. These values were derived from the latest ANNI-NH_3-v4R-ERA5 product. Despite improvements in NH_3 column retrievals from satellite observations, there remains substantial variability in measurement uncertainty, ranging from 5 % to over 1000 %. (Van Damme et al., 2014; Whitburn et al., 2016; Van Damme et al., 2017). Data selection was performed by excluding nighttime observations, irrational values (<0), and only using data with a cloud fraction< 0.1 (Van Damme et al., 2018) and skin temperature > 263 K (Van Damme et al., 2014) during the calculation of the monthly mean. Additionally, while negative values are not necessarily incorrect, they are considered unrealistic in the context of NH_3 concentrations. To improve the quality of the monthly average, we removed those negative values. It is also important to note that we used daily observations from three satellites, each with a pixel resolution of approximately 12 km × 12 km, which provided us with sufficient observations to calculate the monthly average. We applied a selection criterion, using only grid averages that contain a minimum of 80 observations. This ensures that the grid-averaged values are statistically representative and that the monthly mean is of high quality. Notably, the time coverage of the available version 4 IASI product used was limited: Metop-A provided data for the entire year of 2019, Metop-B provided data from January to July 2019, and Metop-C did not have data for 2019. Therefore, only the data from Metop-A and Metop-B within the 2019 time frame were used in this study. To further improve the data quality and ensure consistency, we performed monthly and grid averaging of the observations. This approach not only allows for a fair comparison between the observed and modeled NH_3 concentrations but also reduces the computational cost of the assimilation process. Using individual observations without averaging would result in an excessively large observational vector, which would significantly increase the computational burden. For example, without averaging, the size of the observational vector could reach 1,000,000, while with monthly and grid averaging, it is reduced to a manageable size of around 1,000. This reduction in size helps to optimize the data assimilation process while maintaining the integrity of the emission estimates.

RC: 7) p6, lines 8-10: Why is this minimum value chosen, how often does it have this value? Are the gray values in Figure 2 "b" this minimum? Maybe better to move this part to Section 2.1 where the observation uncertainty is discussed.

AR: Appreciate your comment. The minimum value used for the assimilation process is empirically chosen to avoid overemphasizing extremely low measurements that are likely unreliable due to high uncertainty. This value is rarely used and occurs with very low frequency, approximately 3% of the observations. We also note that the gray values in Figure 2(b) represent the uncertainty in the observations, not the minimum value used in the selection process. However, we have kept this part in the current section because Section 2.1 primarily focuses on the processing of satellite observations and their associated uncertainties, which is distinct from the inversion system discussed here. The details of the observation error covariance matrix (O) and the related formula are directly relevant to the emission inversion process, which occurs in this section. Therefore, we believe that placing this explanation in the current context provides a clearer understanding of how the uncertainties are incorporated into the inversion system.

RC: 8) p 14, section 3.2: Fig 9.a shows prior and posterior model columns, are these after application of averaging kernels? Then the lines should be different for IASI and CrIS. If these are model columns, how well could these be compared to the satellite columns?

AR: Thank you for your comment. As answered above regarding the use of averaging kernels, model column concentrations are not processed with averaging kernels, while satellite column concentrations are corrected using averaging kernels to align them more closely with the vertical distribution of the model. After correction, the satellite column concentrations are adjusted to match the model's three-dimensional structure, ensuring that they have a similar physical basis before comparison. We have also strengthened some of the discussion regarding this figure in the article, as follows:

Text in manuscript

3.1.2 ***Seasonal and annual variation of*** NH_3 ***concentration***

…

The high value in May is attributed to huge amount of biomass burning in South Asia during the spring in Figure S4 (c). We have identified the planting and harvesting times of crops in the South Asia region from USDA(U.S.DEPARTMENT OF ARGRICULTURE, https://ipad.fas.usda.gov/rssiws/al/crop_calendar/sasia.aspx). The heavy use of fertilizers in agricultural activities has resulted in the highest emission throughout the year, as will be illustrated in Fig. 4 (b) in Section 3.2. This has lead to the second NH_3 concentration peak in July. The reasons for higher emissions in July but lower concentration levels compared to May could be attributed to meteorological factors. The monsoon season in South Asia results in increased wet deposition, and notably, 2019 experienced the most intense monsoon since 1994 (NASA, 2020). As shown in the Figure S4 (a) and (b), precipitation and temperature in July are the highest of the year. High temperatures increase ammonia volatilization, and the high precipitation increases the wet deposition of ammonia. These combined factors lead to July having a smaller concentration peak compared to May, despite being another peak month.

…

3.2 ***Spatial and Seasonal variation of*** NH_3 ***emission***

…

The substantial emissions in July, as indicated by the posterior inventory, can be attributed to the increased fertilizer application for rice and corn crops during the summer season (Tanvir et al., 2019). Although biomass burning emissions are generally higher in spring in Figure S4 (c), agricultural activities remain the primary contributors to NH_3 emissions (Huang et al., 2016), resulting in July surpassing May in emission intensity. From July to September, as rice and other crops progress through their growth stages, fertilizer application typically decreases, leading to a gradual reduction in NH_3 emissions. Additionally, temperatures decline from August to September in Figure S4 (b), reducing the volatilization rate of NH_3. This pattern occurs because NH_3 volatilization is strongly influenced by temperature (Fan et al., 2011)

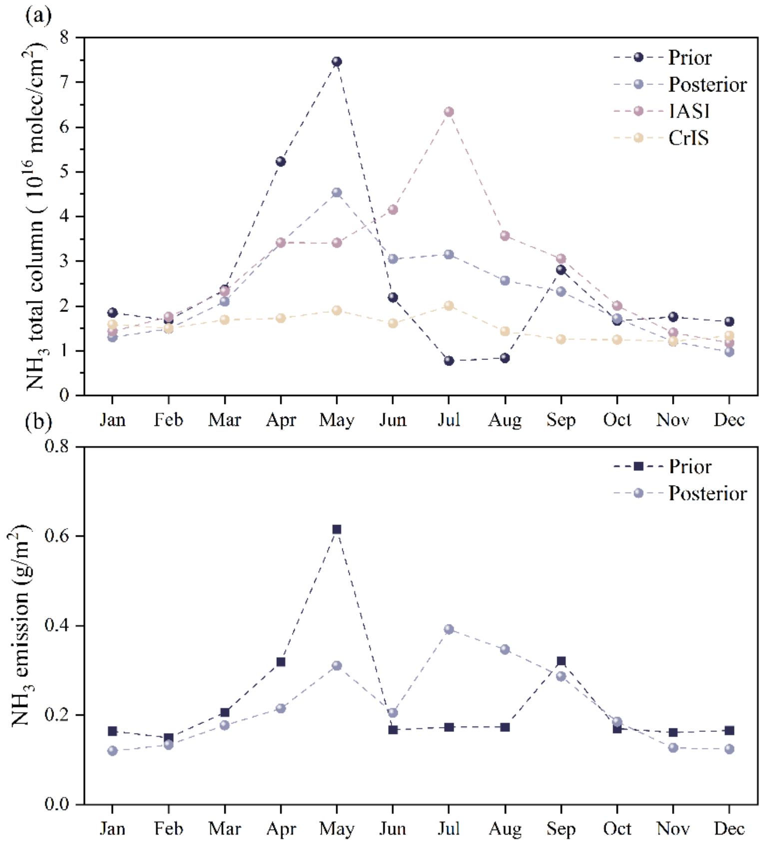

Figure 4. The monthly average total NH_3 column concentrations from the prior and posterior, IASI-observed, and CrIS-observed from January to December (a). The monthly average values of prior and posterior emissions from January to December (b).

Supplement

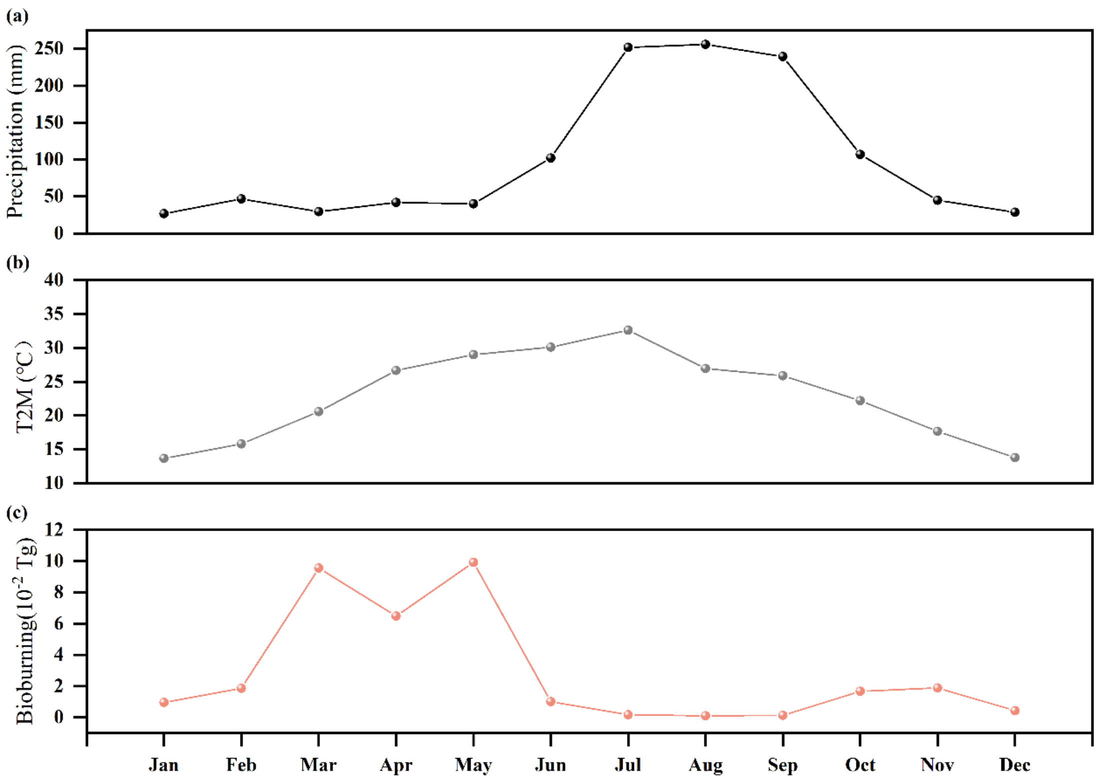

Figure S4. Monthly precipitation (a), temperature (b) from MERRA2 and biomass buring emission (c) from GFED4 in 2019.

RC: 9) p 2, line 13: "compared emissions of other pollutants"

AR: Thanks for comment. I have revised the sentence to clarify the "comparison of emissions of other pollutants". The following is the revised abstract excerpt:

Text in manuscript:

Over the past decade, scientists have predominantly employed the "bottom-up" approach to estimate NH_3 emissions. When combined with chemical transport models, atmospheric NH_3 dynamics can be simulated, enabling the quantification of environmental impacts. Substantial efforts have been made to quantify the spatiotemporal distribution of NH_3 sources and develop global/regional emission inventories, such as the global NH_3 emission inventory (Bouwman et al., 1997), the anthropogenic emission inventory that includes NH_3 estimates (e.g., Community Emissions Data System, CEDS) (Hoesly et al., 2018), as well as regional NH_3 inventories focusing on South Asia (Yan et al., 2003; Yamaji et al., 2004; Liu et al., 2022). However, these bottom-up estimates of NH_3 emissions are generally considered as uncertain (Xu et al., 2019), particularly when compared emissions of other pollutants primarily originating from fossil fuel combustion such as NO2.

RC: 10) p 2, line 2: Add a reference here?

AR: Thanks for comment. We have added a reference here and also updated the literature related to climate effects to make it more relevant and up-to-date. The following is the revised abstract excerpt:

Text in manuscript:

Further, ammonia gas, along with its reaction products, plays a pivotal role in soil acidification and the eutrophication of water bodies through both dry and wet deposition (Krupa, 2003), and thereby affecting the balance of ecosystems (Asman et al.,1998) and climate change (Ma et al., 2022; Gong et al., 2024).

RC:11) p 4, line 16: The uncertainty assigned to the IASI measurements is also an essential"

AR: Many thanks for your feedback. We have made the revision accordingly. The following is the revised abstract excerpt:

Text in manuscript:

1.1 IASI satellite measurements

…

The uncertainty assigned to the IASI measurements is also an essential. When calculating the uncertainty of gridded monthly average NH_3 measurements, both instrumental errors $\sigma^{instrumental}$ and representation error $\sigma^{representation}$ are considered. The gridded average uncertainty derived directly from IASI products was designated as instrumental error $\sigma^{instrumental}$, while the standard deviation of the observed samples for the gridded average characterized representation error $\sigma^{representation}$.

RC: 12) p4 line 24: Fig 2 is referenced before Fig 1, change order of figures?

AR: Thanks for comment. We have adjusted the order of the figures accordingly. Below is the revised order of the figures.

Text in manuscript:

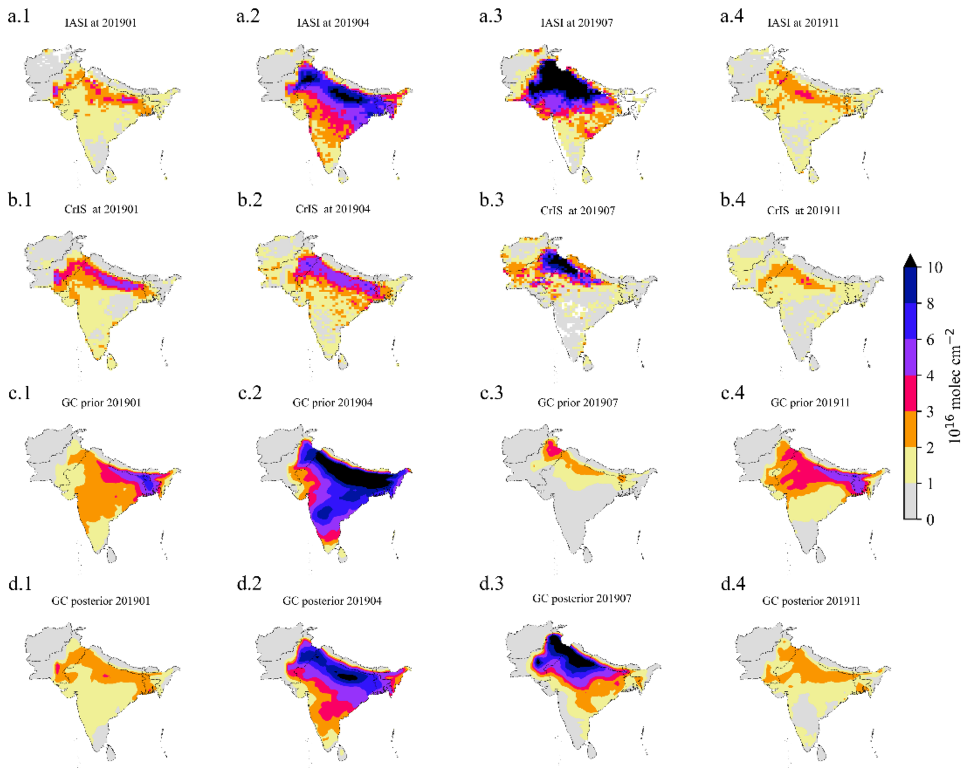

Figure 1. Spatial distribution of the total column NH_3 concentration from IASI (a) or CrIS (b) instruments, and from the GEOS-Chem simulation either using the prior (c) or using the posterior (d) NH_3 emission flex in 2019 January (a.1)–(d.1), April (a.2)–(d.2) , July (a.3)-(d.3) and November (a.4)–(d.4).

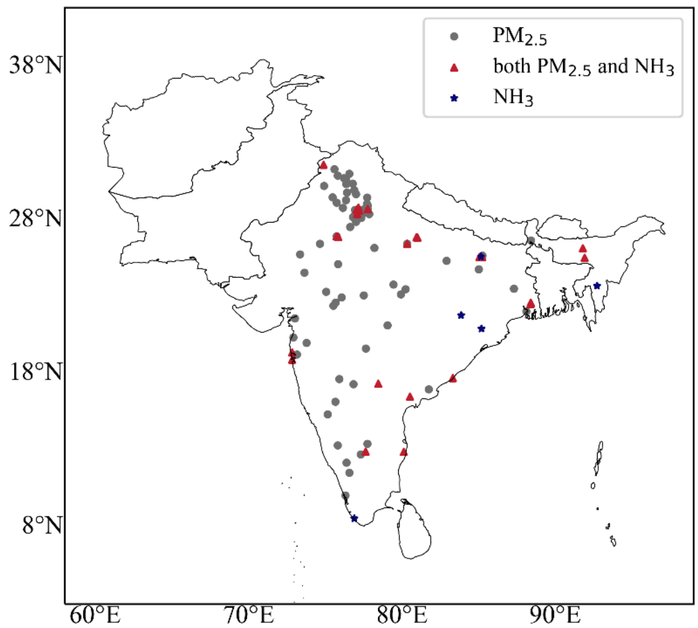

Figure 2. The GEOS-Chem model simulation domain, with dots indicating the locations of ground observation stations from the Central Pollution Control Board (CPCB), India. The three different colored dots represent stations with only PM2.5 observations, stations with both PM2.5 and NH_3 observations, and stations with only NH_3 observations, respectively.

RC: 13) p 5, line 22: Explain that the emission field is "f"; what are the units?

AR: Thank you for your kind comment. We have added a detailed description of this parameter in the manuscript. The relevant information from the article is shown as follows:

Text in manuscript:

2.3Emission inversion system
…
Here, ***f*** denotes the vector of the NH_3 estimated emission field, with its units typically expressed in kg/m2/s.

RC: 14) p 6, line 3: Shouldn't this be fb?

AR: Appreciate your comment. Yes, this should be $\boldsymbol{f_b}$. We have corrected the formula (5) in the manuscript accordingly.

Text in manuscript:

2.3Emission inversion system
…
$$\mathbf{B}(i,j) = \sigma^2 \cdot \boldsymbol{f_b}(i) \cdot \boldsymbol{f_b}(j) \cdot \mathbf{C}(i,j)$$

RC: 15) p 6, line 7: Shouldn't this be "observation representation errors are independent from each other"?

AR: Thanks for comment. I updated this sentence to state "observation representation errors are independent from each other." The following is the revised abstract excerpt:

Text in manuscript:

2.3 Emission inversion system
…
while **O** is the observation error covariance matrix. Here we assume IASI observation representation errors are independent from each other.

RC:16) p6 line 8: "as described in"

AR: Thanks for comment. I added "as described in" for clarity. The following is the revised abstract excerpt:

Text in manuscript:

2.3 Emission inversion system
…
while **O** is the observation error covariance matrix. Here we assume IASI observation representation errors are independent from each other. O therefore is a diagonal matrix filled with the square of the integrated uncertainty as described in Section 2.1.

RC: 17) p8, line 19: "as well as manure from livestock, including cattle, ..."

AR: Appreciate your comment. I revised this line to include the mention of manure from livestock, including cattle. The following is the revised abstract excerpt:

Text in manuscript:

2.4 GEOS-Chem model and emission inventory
The NH_3 emissions inventory employed to drive GEOS-Chem originated from the Community Emissions Data System (CEDS, https://doi.org/10.25584/PNNLDH/1854347) inventory, which was widely used for modeling the South Asia atmo-spheric pollutants, e.g., VOCs (Chaliyakunnel et al., 2019), PM2.5 pollution (Guttikunda and Nishadh, 2022; Xue et al., 2021). CEDS inventory includes various sources encompassing agricultural, energy production, industrial, residential and commercial activities, ships, solvent use, surface transportation, and waste processing (McDuffie et al., 2020), the bulk of NH_3 emissions originate from agricultural practices. Specifically, these emissions stem predominantly from farmlands, including crops such as wheat, maize, and rice, as well as manure from livestock, including cattle, chicken, goats, and pigs (Liu et al., 2022).

RC: 18) p 8, line 25: "posterior result"

AR: I updated "posterior result" as requested. At the same time, we have reorganized the logical structure of this section, and the revised fragment is as follows:

Text in manuscript:

With the assimilation system described above, the monthly NH_3 emission inversion for 2019 over South Asia is conducted. The Spatial of prior and posterior results are in Section 3.1.1. The long-term varying trend of South Asia NH_3 emission is illustrated in Section 3.1.2, followed by an analysis and discussion of its spatial distribution and seasonal profile based on the inversion results in Section 3.2. Then the posterior result is evaluated in Section 3.3.

RC: 19) p 8, lines 26-27: mention 3.2 first, then 3.3

AR: Many thanks for your feedback. I reordered the references to sections 3.2 and 3.3 as per your suggestion.
We have also revised the structure of the Results and Discussion section. The updated structure is as follows:

Text in manuscript:

3 Results and discussion
3.1 Observed NH_3 ***concentrations***
3.1.1 Spatial NH_3 ***total column concentration***
3.1.2 Seasonal and annual variation of NH_3 ***concentration***
3.2 Spatial and Seasonal variation of NH_3 ***emission***
3.3 Validation
3.3.1 NH_3 ***total column concentration validation***
3.3.2 NH_3 ***and PM2.5 ground concentration validation***

RC: 19) p 16, Figure 10 caption: (j) is a time series, not a scatter plot. And the box plots represent yearly averages.

AR: Thanks for comment. I changed the description of panel (j) to indicate that it is a time series, not a scatter plot. I also updated the caption to explain that the box plots represent yearly averages. The following is the revised abstract excerpt:

Text in manuscript:

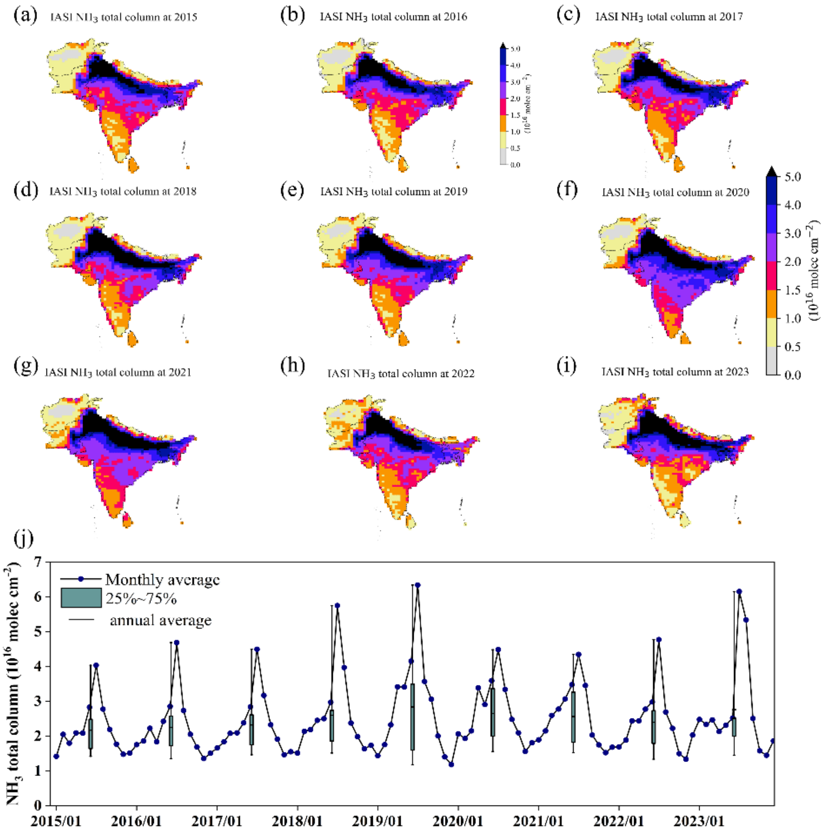

Figure 5. The spatial distribution of the annual averaged IASI column concentrations in South Asia from 2015 to 2023 is shown in panels (a) to (i). Panel (j) presents a time series depicting the monthly variation in IASI-observed NH_3 column concentrations from 2015 to 2023, with the box plots representing the yearly averages showing interannual changes.

Reference

Dammers E, et al. "Validation of the CrIS fast physical NH 3 retrieval with ground-based FTIR。" Atmospheric Measurement Techniques Discussions, 2017(2017): 1-32.

Ma, Ruoya, et al. "Data-driven estimates of fertilizer-induced soil NH_3, NO and N_2O emissions from croplands in China and their climate change impacts." Global Change Biology 28.3 (2022): 1008-1022.

Gong, Cheng, et al. "Global net climate effects of anthropogenic reactive nitrogen." Nature 632.8025 (2024): 557-563.