Response to Referee #2:

We would like to thank the referee for the useful comments and constructive suggestions. In the following, we address the referee's comments and describe corresponding changes we have made to the manuscript. The referee's comments are listed in blue, followed by our response in black. New/modified text in the manuscript is in *dark orange italics*.

Li et al. presented an analysis using IASI and CrIS ammonia retrievals to study ammonia emissions and deposition distribution in the US and the associated seasonality and trends. This is an interesting study. However, I have several major comments concerning (i) the validity of the presented results, (ii) organization and presentation of the analysis and related discussions. These comments need to be addressed before the paper should be considered for publication in ACP. I would like to encourage the authors carefully address the concerns raised in my major comments #1, #2, and #3, as they may impact the validity of the presented results for a robust flux analysis.

Major comments:

1. Section 2.2.1 (lines 154-171). The description of equation (1) is inconsistent with the labeling of the terms on line 169-171, or at least confusing to me. The sum of all three terms on the right-hand is referred to as DD_chem. But if I understand this correctly, only the third term, k(omega), is the chemical loss. The sum of all three equals on the left hand. Also I am not sure why you want to study DD, and DD_topo (which is the sum of the horizontal wind transport and vertical altitude related derivation), such as that shown in figures 4-9. Why can't you just show the separate impact of 1st and 2nd term, which gives a better sense of the relative importance of each factor? Looking at the figures, DD and DD_topo don't look that different, which implies the 2nd term is possibly not important at all?

We thank the reviewer for this comment. We agree that the description of DDA framework in Section 2 was not clear enough, and we rewrote this part for improved clarity. The three estimators within the DDA framework are: DD, DD_topo, and DD_chem, where DD_topo = DD + topography term, and DD_chem = DD_topo + chemistry term. This formulation follows Ayazpour et al. (2025). Lines 154-168 in the original manuscript have been revised as follows: *"The estimation of emissions (E) from satellite-observed VCDs (Ω) is grounded in the principle of mass conservation as in Eq. 1, which is in the same form as presented in the previous DDA literature (Sun, 2022; Ayazpour et al., 2025). The DDA considers the physical and chemical processes affecting gas distribution, incorporating horizontal transport, topography, and chemical transformation. Three estimators within the DDA framework are labeled in Eq. 1 as DD, DD_topo, and DD_chem, representing the* *directional derivative of column densities, the directional derivative with consideration of topography, and the directional derivative with consideration of both topography and chemistry. The DD estimator ($\vec{u} \cdot (\nabla\Omega)$) captures the horizontal advection of NH_3, representing the directional derivatives of the VCDs with respect to horizontal wind vectors representing the planetary boundary layer ($\vec{u}$, 100 m winds). $\nabla = (\partial/\partial x, \partial/\partial y)$ is the horizontal vector differential operator. The wind height choice is supported by Ayazpour et al. (2025), who reconstructed emissions using model (WRF-CMAQ) column and winds at different model layers and found that winds from ~100 m yield strong agreement between the DD estimator and model-ingested emissions while maintaining robustness across wind heights up to ~800 m. The DD_topo estimator accounts for the topography term ($X\Omega\overrightarrow{u_0} \cdot (\nabla z_0)$), which is driven by the directional derivatives of the surface altitudes (z_0, obtained from Level 2 satellite data) relative to near-surface wind vectors ($\overrightarrow{u_0}$, 10 m winds). This component captures the influence of terrain on NH_3 movement. For example, variations in elevation can create localized gradients that resemble NH_3 fluxes. The DD_chem estimator considers the chemistry term ($k\Omega$), representing chemical interactions between NH_3 and atmospheric acids which result in the formation of particulate matter."*

We have also revised Figs. 4-9, replacing DD and DD_topo with VCDs and fluxes (which is the revised DD_chem estimator). The separate impacts of topography and chemistry terms are shown in the Supplementary materials (Fig. S8). As expected, the topography term has a larger influence in mountainous regions, while the chemistry term has a greater impact in areas with high NH_3 columns. The following figure has been added to the Supplementary Materials:

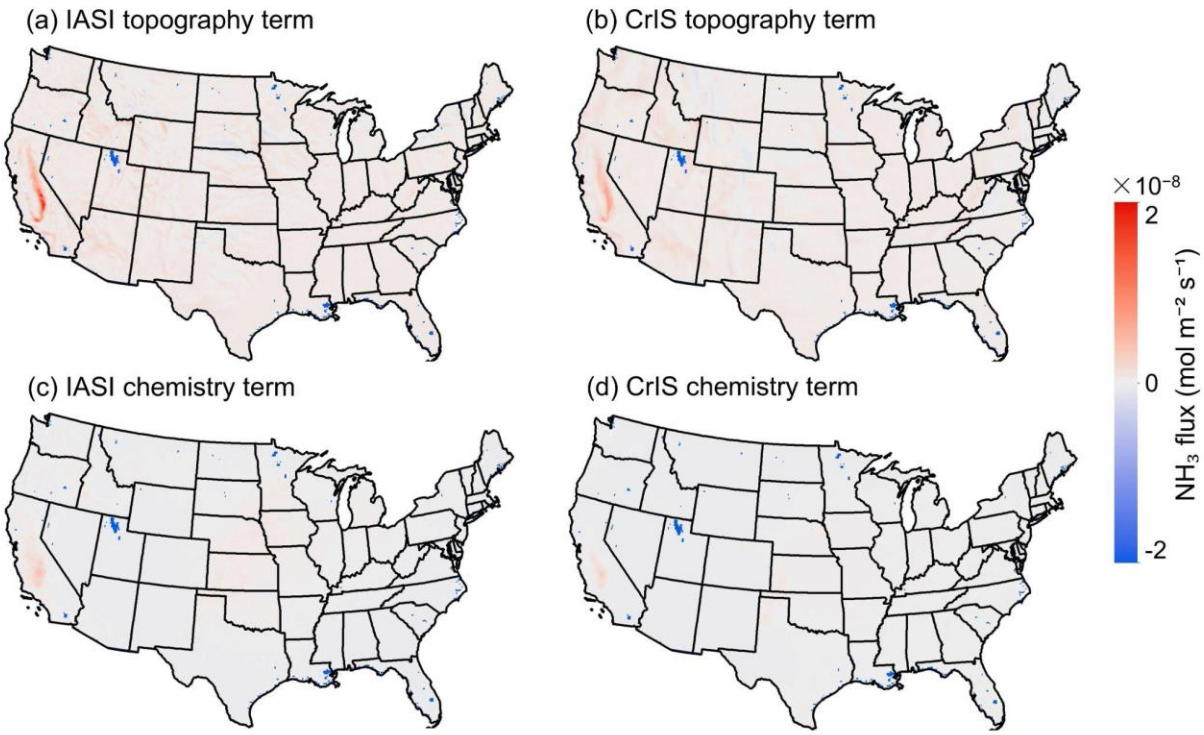

Figure S8. Topography and chemistry terms derived from IASI and CrIS over Sep 2019 to Apr 2021.

2. In addition, on lines 246-248, the authors stated that chemical loss term, the third term, "was excluded from this study due to negligible contribution and poor fitting performance". If the third term is negligible, and 2nd term is very small (see above), that would imply inferred emissions is predominantly controlled by the horizontal transport term? NH3 has an atmospheric chemical lifetime of a few hours to a few days, it is not convincing that atmospheric chemical loss does not play a role in the budget and inferred-emission analysis. More robust analysis is needed to support the validity of these results.

We thank the reviewer for this important insight. We agree that omitting the chemistry term due to a poor fit can introduce significant uncertainty, particularly in areas with abundant columns. In response to this concern, we have revised our results to retain the chemistry term in the final flux estimator, using the DD_chem estimator as our primary flux estimate throughout the manuscript.

3. Figure 11. The relative magnitude of source vs. sink in most regions don't make sense to me. As I mentioned in comment #2, if chemical term is negligible, the sink (which represents surface deposition) is also much smaller that the source term throughout the entire year and can be one magnitude smaller in some regions, shouldn't that imply that the atmospheric budget of NH3 is not in balance and atmospheric NH3 abundance will grow rapidly? The results presented in Figures 10 and 11 does not support the conclusion that chemical loss term being negligible. Some processes, whether it is chemical loss or deposition, must be in play to create the observed the seasonal cycle shown in figure 10. Please clarify and provide adequate analysis result to support your conclusions.

We thank the reviewer for this comment. We agree that, at the global or continental scale, the atmospheric NH_3 budget should be approximately balanced between sources and sinks. However, the regions shown in Figs. 4-9 are relatively small emission-hotspot areas chosen for detailed analysis. In such limited domains, it is expected that local sources will not be balanced by local sinks because a substantial fraction of the emitted NH_3 can be transported downwind and deposited/reacted outside the analysis area. This spatial-scale mismatch means that the "source" term we retrieve may appear larger than the "sink" term within the same region, even if the broader-scale budget is balanced. The imbalance reflects the geographic boundaries of the analysis and transport of NH_3 to surrounding areas, rather than an actual growth in atmospheric NH_3 abundance. Regarding the chemistry term, we have revised our estimation to include the chemistry term and use DD_chem as our flux estimates.

4. The side-by-side comparison of IASI and CrIS related results. I strongly support the use of both datasets for this analysis as these datasets provide corroborative as well as complimentary information to study spatial and temporal variability of NH3 abundance and budget analysis. As pointed out by the authors, the different overpass times by the two instruments provide unique information that can be exacted using the right analysis. With that said, I would like to encourage the authors put a bit more thought in when it would be necessary to show panels (or lines) from both instruments and when one dataset would be adequate to convey the scientific message. For example, figures 4-9, I personally don't see the value of showing IASI & CrIS panels side by side. First, they look similar on a broader spatial scale perspective. Second, you don't spend much effort discussing the differences between the two different datasets in the text. Third, repeating IASI and CrIS panels in 6 figures (Figures #4-#9) took up too much space. Therefore, I would recommend showing just one instrument dataset and discuss the relative scientific points. If you prefer, you can include the other instrument result in supplementary material for completeness.

Thanks for this comment. While the general spatial patterns in Figs. 4-9 are broadly similar, we consider it important to show both datasets side-by-side. First, the consistency between two independent instruments with different overpass times, viewing geometries, and retrieval algorithms demonstrates the robustness of the spatial features we report. Second, the differences—though often subtle—are informative, particularly in hotspot regions where diurnal variations, meteorology, or retrieval sensitivity can affect magnitude and spatial extent. We believe retaining both datasets in the main figures improves transparency and illustrates the reliability of our flux estimates.

We have revised the interpretation of Figs. 4-9 in Section 3.3 (Lines 318-331) as follows:

"Figs. 4-9 compare IASI- and CrIS-derived NH_3 fluxes with VCDs and bottom-up inventories across six major high-flux regions. Application of the flux estimator substantially sharpens spatial structures relative to VCDs. For instance, in the Snake River Valley (Fig. 5), enhanced VCDs appear as a broad belt, while the corresponding flux fields resolve into alternating hot and cold spots, indicating localized source–sink variability. Similar sharpening is evident in other regions, demonstrating the added value of the estimator in attributing fluxes to specific land cover types.

The two instruments yield broadly consistent spatial patterns of NH_3 source and sink, although systematic differences are observed. IASI-derived fluxes tend to resolve finer spatial details, consistent with its smaller footprint and denser sampling, whereas CrIS-derived fluxes appear smoother but less noisy. These characteristics are complementary and together provide robust evidence for the spatial distribution of NH_3 fluxes.

Agricultural lands dominate as NH_3 source regions in all cases, with strong fluxes coinciding with intensive cropping and livestock production (San Joaquin Valley, Texas Panhandle, Great Plains). In contrast, natural and semi-natural landscapes function primarily as sinks. Vegetated landscapes—including forests, shrublands, and grasslands (Figs. 4-8), as well as wetlands (Fig. 9)—show consistent negative fluxes, likely reflecting deposition processes in proximity to nearby sources.

Satellite-derived fluxes also align well with bottom-up inventories, with regional correlation coefficients ranging from 0.08 to 0.86 (Fig. S10). Agreement is highest in areas with dense agricultural activity (e.g., San Joaquin Valley), whereas discrepancies in regions such as the Great Plains and Snake River Valley suggest that inventories may not capture the full subregional variability evident in satellite observations. These results highlight both the consistency of satellite-derived fluxes with existing inventories and their capability to provide additional spatial details."

5. Figures 4-9. Here is my recommendation for authors to consider improving these figures for a more informative presentation: a) only show one dataset, either CrIS or IASI, b) show DD, the 2nd term (which is DD_topo – DD) intead of DD_topo, c) add another panel for NH3 VCD. It would be helpful to have the column abundance distribution information on the same figure to better relate with the emission, transport, source and sink information.

We thank the reviewer for these suggestions. Our main objective in this work is to estimate NH_3 fluxes, so we have not placed emphasis on presenting the topography and chemistry terms separately, although their individual effects are shown in the Supplementary materials (Fig. S8). We agree that including column abundance information would improve the interpretability of the figures. In the revised manuscript, we have updated panels (b), (c), (e), and (f) in Figs. 4-9 to display: (b) IASI VCDs, (c) CrIS VCDs, (e) IASI flux, and (f) CrIS flux (now we use the revised DD_chem estimator). These changes are detailed in our previous response to comment #4. Specifically, revised Figs. 4-9 present how the DDA framework transforms satellite-observed VCDs into flux estimates, while retaining both IASI and CrIS datasets in the main figures improves transparency and strengthens the robustness of the flux estimates.

References:

Ayazpour, Z., Sun, K., Zhang, R., and Shen, H.: Evaluation of the directional derivative approach for timely and accurate satellite-based emission estimation using chemical transport model simulation of nitrogen oxides, J. Geophys. Res., 130, e2024JD042817, https://doi.org/10.1029/2024jd042817, 2025.