

## Response to Referee #1:

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*.

The authors clearly demonstrate the significance and necessity of improved NH<sub>3</sub> flux estimates, and the results present interesting spatial and seasonal patterns that are valuable to the community. However, the methodological description is currently insufficient and relies too heavily on briefly reproducing elements from Ayazpour et al. (2024) without providing enough self-contained derivation or detailed explanation. To ensure clarity and reproducibility, I strongly recommend expanding Section 2 with a more thorough presentation of the directional derivative framework, explicit definitions of all key variables and terms, and clearer justification for the assumptions made. Addressing these issues will greatly strengthen the scientific rigor and standalone value of the manuscript. Furthermore, emissions are presented in quite some detail, but the deposition fluxes seem to draw the short straw. The lack of dry-deposition measurements of course does not help in being able to make a nice comparison, but some discussion or comparison with past modelled results would increase the value of the deposition results. Subsequently linking those results to for example critical load limits could greatly improve the overall value of the manuscript and enhance the impact of all the work that is already performed.

We thank the referee for the constructive suggestions. In the revised manuscript, we have expanded Section 2 to provide a more detailed description of the directional derivative analysis (DDA) framework. We have also strengthened the discussion of deposition fluxes in Section 4.2 (Line 466 in the original manuscript) by comparing with past modelled deposition results: *“Similar hotspots of deposition downwind of intensive agricultural regions have been reported in model-based studies (Ellis et al., 2013; Hu et al., 2021; Zhang et al., 2012). These hotspots often coincide with high NH<sub>3</sub> loadings, large leaf area indices, and micrometeorological conditions favorable for stomatal and cuticular uptake (Sutton et al., 2009).”*

### Main comments:

1. Section 2.2 — Please define “DD” clearly before using it throughout the paper. Also clarify the meanings of “∇” and “Ω” (column density) upon first mention.

We thank the referee for pointing this out. We do agree that the terms “DD”, “ $\nabla$ ”, and “ $\Omega$ ” should be more clearly stated. We have revised the description of Eq.1 in Section 2.2.1 (Lines 154-168) as follows:

*“The estimation of emissions ( $E$ ) from satellite-observed VCDs ( $\Omega$ ) 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. DDA accounts for horizontal transport, topographic effects, and chemical transformation influencing gas distribution. The DD estimator ( $\vec{u} \cdot (\nabla \Omega)$ ) captures the horizontal advection of  $\text{NH}_3$ , representing the directional derivatives of the VCDs with respect to horizontal wind vectors in the planetary boundary layer ( $\vec{u}$ , 100 m winds).  $\nabla = (\partial/\partial x, \partial/\partial y)$  is the horizontal vector differential operator. Ayazpour et al. (2025) evaluated DD estimators calculated using column amounts and winds at a range of altitudes in an atmospheric model with the model-ingested emission and found that winds from 100-800 m give similar and consistent results. We choose 100 m wind because it has been widely used in previous studies (Goldberg et al., 2022; Lonsdale and Sun, 2023) and is readily available from the ERA5 single-level product. The DD\_topo estimator accounts for the topography term ( $X\Omega \vec{u}_0 \cdot (\nabla z_0)$ ), which is driven by directional derivatives of the surface altitudes ( $z_0$ , obtained from Level 2 satellite data) relative to near-surface wind vectors ( $\vec{u}_0$ , 10 m winds). This component captures the influence of terrain on  $\text{NH}_3$  movement. For example, variations in elevation can create localized gradients that resemble  $\text{NH}_3$  fluxes. The DD\_chem estimator considers the chemistry term ( $k\Omega$ ), representing chemical interactions between  $\text{NH}_3$  and atmospheric acids which result in the formation of particulate matter.”*

2. Line 136 vs. Line 223 — There appears to be an inconsistency regarding the emission inventory year: Line 136 mentions using HEMCO for 2016, while Line 223 states the focus is on September 2019 to April 2021. Please clarify which year(s) of the emission inventory were used and how they relate to the period analyzed.

We thank the referee for pointing out this potential inconsistency. We clarify that the gridded monthly HEMCO emission inventory is only available to us for the year 2016, and this is consistently used throughout the study for two purposes: (1) To identify

low-emission regions (where  $E < 1 \times 10^{-9} \text{ mol m}^{-2} \text{ s}^{-1}$ ) that are suitable for fitting the topography and chemistry terms under the assumption of negligible emissions. (2) For comparison with the satellite-derived flux estimates from IASI and CrIS during the analysis period of September 2019 to April 2021. We have updated the manuscript to clearly clarify the use of HEMCO data in Section 2.1 and the role of the emission inventory in the fitting process in Section 2.2.1 as follows:

In Section 2.1 (Lines 140-142): *“We use this bottom-up inventory from HEMCO to compare with satellite-derived fluxes, providing insights into their consistency and helping to assess the utility of satellite-based estimates.”*

In Section 2.2.1 (Lines 187-189): *“In regions where emissions are negligible ( $E < 1 \times 10^{-9} \text{ mol m}^{-2} \text{ s}^{-1}$ , based on bottom-up inventory from HEMCO; see Fig. S1 for spatial distribution), Eq. (1) can be reformulated into a multilinear regression form”.*

3. Line 143-144: Why are the wind data at 100-10m used when the observed air masses are clearly well mixed within the mixing layer, would a wind speed more representative of the mixing layer (500m-1000m) not make more sense? Wind speeds closer to the surface can also be expected to be much smaller, which seems essential to for the resulting fluxes. Either add a reference for this value or better explain the expected uncertainties.

We thank the referee for this thoughtful comment. We acknowledge the importance of wind height selection in estimating surface fluxes. In the DDA framework (Eq. 1), we use 100m winds for the horizontal wind  $\vec{u}$  in DD estimator ( $\vec{u} \cdot (\nabla \Omega)$ ). The 100 m wind reflects horizontal advection near the surface but still within the well-mixed lower boundary layer. Fig. 3c of Ayazpour et al. (2025) below shows that using winds from layer 4 (~100 m) yields strong agreement between DD estimates and true emissions, and that the DD is robust to wind height over layers 4–7 (~100–800 m).

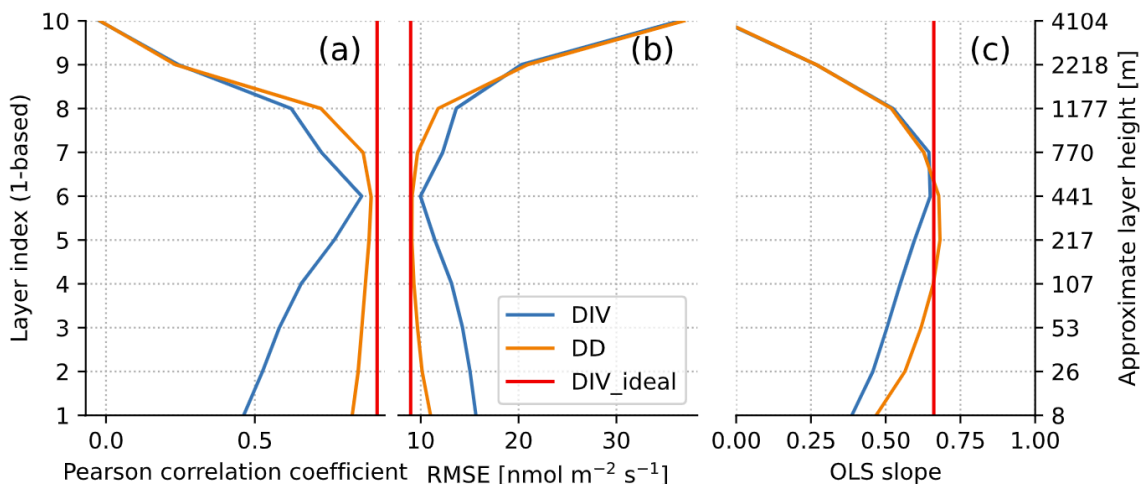


Figure 3 of Ayazpour et al. (2025). (a) Pearson correlation coefficient, (b) root mean square error, and (c) slope of linear regression of DIV\_ideal and DIV estimators (two estimators in the flux divergence approach, not relevant for this work) of the divergence method and the DD estimator of the directional derivative approach framework when compared with model emissions from WRF-CMAQ using winds at different layers. The approximate layer center altitude is shown on the right vertical axis.

We use the 10m winds for the near-surface winds  $\vec{u}_0$  in the topography term ( $X\Omega\vec{u}_0 \cdot (\nabla z_0)$ ). Because this process occurs very close to the ground, using near-surface (10 m) winds is physically appropriate for this component of the model. We have revised the text in Section 2.2.1 (Lines 159-160) and added a reference to Ayazpour et al. (2025) to support the choice of wind layer: *“Ayazpour et al. (2025) evaluated DD estimators calculated using column amounts and winds at a range of altitudes in an atmospheric model with the model-ingested emission and found that winds from 100-800 m give similar and consistent results. We choose 100 m wind because it has been widely used in previous studies (Goldberg et al., 2022; Lonsdale and Sun, 2023) and is readily available from the ERA5 single-level product.”*

4. Line 197–199 (Fitting criterion) — The statement “This fitting was limited to rough terrains...” conflicts with the description “This step was conducted in flat terrains...” and also appears to misrepresent the original criterion (Ayazpour et al., 2024, Sect. 3.2: “which eliminates open water and very rough terrain”). Please clarify whether the fitting excludes both open water and very rough terrain, or whether it is limited to rough or flat terrains, and explain whether the fitted parameters from flat terrain are appropriate for application in mountainous areas or for the entire CONUS domain.

We thank the referee for this helpful comment. Consistent with the approach in Ayazpour et al. (2025), our fitting process excludes both open water bodies and very rough terrain to ensure the reliability of the parameter estimates. Specifically, we performed a two-step fitting: (1) the scale height ( $X$ ) was fitted in moderately rough terrains, where elevation gradients are sufficiently present to constrain the relationship, and (2) the chemical term ( $k$ ) was fitted in flat terrains, where topographic influences are minimal. Moderately rough terrains are broadly distributed across the CONUS and provide representative conditions for fitting  $X$ , while flat terrains are best suited for isolating  $k$ . We have clarified the terrain selection criteria in the revised manuscript below.

In Section 2.2.1 (Lines 195-200), we have added the description of the fitting process as follows: *“We conducted a two-step fitting process to estimate  $X$  and  $k$  following (Lonsdale and Sun, 2023). The two fitting processes exclude open water bodies and*

*very rough terrains. The first fitting step focused on  $\beta_1$  since the fitting results for  $\beta_2$  are usually noisy. The first-round fitting for  $\beta_1$  was limited in moderately rough terrains with  $0.001 \text{ m s}^{-1} < \langle \vec{u}_0 \cdot (\nabla z_0) \rangle < 0.1 \text{ m s}^{-1}$ . Once  $\beta_1$  was determined and then fixed, the second-round fitting for  $\beta_2$  was conducted in flat terrains ( $\langle \vec{u}_0 \cdot (\nabla z_0) \rangle < 0.001 \text{ m s}^{-1}$ ) with moderate  $\text{NH}_3$  VCDs ( $\Omega > 2.5 \times 10^{-5} \text{ mol m}^{-2}$ ) and minimal emissions ( $E < 1 \times 10^{-9} \text{ mol m}^{-2} \text{ s}^{-1}$ ) to isolate chemical transformation.”*

5. Scale Height Assumption — The manuscript assumes a regionally constant scale height. Considering the substantial local variability in boundary layer depth and surface conditions, could the authors discuss how this assumption affects the flux estimation, particularly over complex terrain, and whether a spatially or seasonally varying scale height was tested?

We thank the reviewer for pointing this out. Fundamentally, the inverse scale height and chemical reactivity in DDA are empirical fitting parameters that ensure the emissions center around zero in low-emission regions. They are tied to physically meaningful parameters through the theoretical derivation of the DDA equations, but they should not be constrained or prescribed. This is because the fittings are already “optimal” as they are directly informed by satellite data and auxiliary variables (wind, surface altitude). Prescribing these numbers, potentially in a complicated, spatiotemporally resolved way (i.e., from a model), will deviate the derived emissions over confidently low-emission regions from zero and thus bias the emissions over source regions. This also allows the large number of assumptions and approximations in various model settings to influence the outcomes of DDA and negates our aims to provide lightweight and timely observation-based emission estimates. The downside of always fitting the emission estimator to zero in low-emission regions through linear regression is the constraints from the quality and quantity of available satellite data. In the case of IASI and CrIS, we do expect the quality of emission estimation to degrade over complex terrain because individual IASI and CrIS pixels are separated by gaps (in contrast to tiled pixels like TROPOMI) and therefore undersample the topography. Disaggregating satellite data spatially and/or temporally may better resolve the fitted parameters but at the expense of further thinning the data and losing the signal under the noise. To clarify these points, we rewrite Lines 201-205 of Section 2.2.1 in the original manuscript as separate paragraph:

*“ $X$  and  $k$  represent the inverse scale height and chemical reactivity, in which scale height represents the characteristic height of the species’ vertical distribution, and chemical reactivity represents the inverse of average time before the species being removed by chemical reactions. To improve the performance of the flux estimates, we*

*treat the  $X$  and  $k$  as fundamentally empirical fitting parameters within a data-driven approach to ensure the resultant emission estimator centers around zero where emissions are negligible. Although in theory  $X$  and  $k$  are tied to physically meaningful quantities, their main purpose is to enhance emission estimators in the presence of topography and chemistry using information contained by satellite observations. As such, the quality of fitted  $X$  and  $k$  is subject to the quality and quantity of available satellite data. Because of the gaps in individual IASI and CrIS pixels that lead to undersampling of topography, we expect that  $X$  cannot fully account for topography effects over complex terrains, an inherent limitation for scanning Fourier Transform Spectrometers like IASI and CrIS."*

To identify the optimal spatial granularity, we divided the domain into quantile-based bins according to topographic conditions, represented by  $\langle \Omega \vec{u}_0 \cdot (\nabla z_0) \rangle$ . Within each bin, the scale height  $X$  was fitted independently. This binning approach allows regions with similar topographic forcing to be treated consistently, but it also increases the noise level when the number of bins becomes large.  $X$  remains positive when using a single bin (Fig. S2), whereas using multiple bins can introduce excessive noise, sometimes leading to negative fitted values. Temporal variability was assessed by fitting with different temporal aggregation windows, where individual flux estimates were averaged over fixed periods before fitting (e.g., 1-, 3-, 6-, and 9-month intervals). Longer windows (>6-months) produce stable and positive fitted  $X$  values, while shorter intervals yield larger noise and sometimes negative fitting results (Fig. S3).

We have added the sensitivity tests of fitted  $X$  in the end of Section 2.2.1 as follows: *"We tested different spatial groupings, temporal averaging windows, and stricter maximum emission thresholds to refine the fitted  $X$  and  $k$ . For  $X$ , fitting the entire domain as a single group produces consistently positive and stable values, whereas subdividing the domain often introduces excessive noise (Fig. S2). Similarly, varying the temporal aggregation affects the stability of the fits (Fig. S3): shorter intervals produce noisier estimates, while longer intervals yield more robust results. We adopted a six-month aggregation interval as it provides reliable estimates across the entire period while retaining seasonal variability."*

The following figures have been added to the Supplementary Materials:

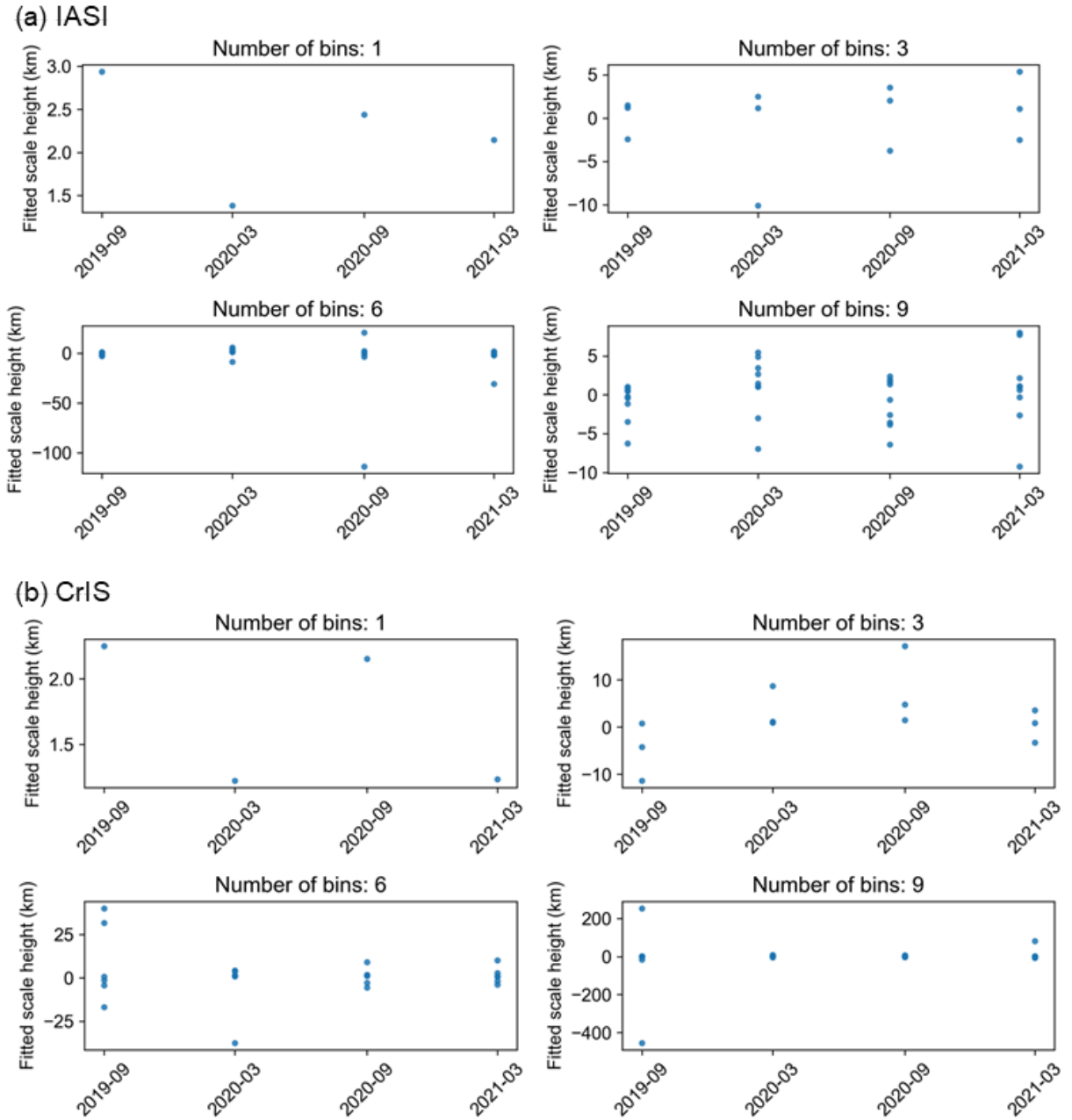


Figure S2. Spatial sensitivity tests for the topography term fitting to derive  $DD\_topo$  from IASI (a) and CrIS (b). The number of bins indicates the wind-topography ( $\Omega \vec{u}_0 \cdot (\nabla z_0)$ ) sectors used in the fitting. The y-axis shows the fitted  $X$ .

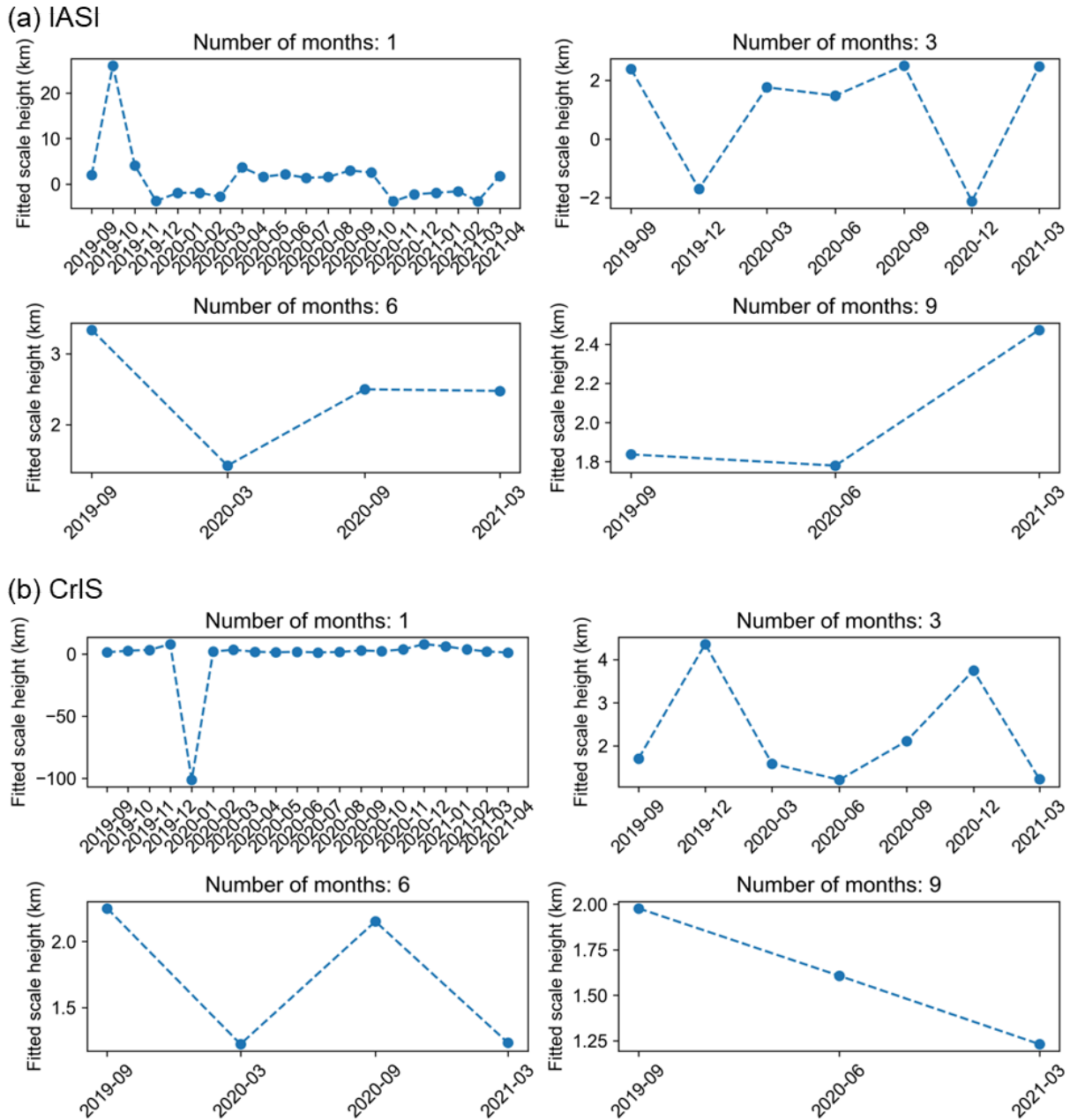


Figure S3. Temporal sensitivity tests for the topography term fitting to derive DD<sub>topo</sub> from IASI (a) and CrIS (b). The number of months indicates the temporal aggregation interval used for the fitting. The y-axis shows the fitted X.

6. Inverse scale height – As stated the scale height has a direct relation to the mixing layer, wouldn't it make more sense to use a boundary layer height product to substitute into these functions instead of deriving them from the satellite observations? This will probably remove potential artifacts from spotty

spatial/temporal measurement records and smooth out the resulting fields. (Based on L253-255 this does not seem to be a bad idea).

We thank the reviewer for this thoughtful comment. The fitted inverse scale height ( $X$ ) in the DDA framework serves a different purpose than boundary layer height. Rather than representing the physical mixing depth alone,  $X$  captures the integrated vertical structure of the observed column, which extends to the top of the atmosphere and also encodes the vertical sensitivity of each instrument. In this sense,  $X$  acts as an empirical correction that improves the fidelity of the  $DD$  estimator—particularly in heterogeneous terrain and under variable atmospheric conditions. The data-driven approach of fitting  $X$  can also absorb some of the errors caused by the simplifications and the choice of near-surface wind. The rationale of these fittings is to explain the residual values of the  $DD$  over locations where the emissions are negligible. We have clarified that the data-driven fitting approach aims to enhance the DDA estimators in Section 2.2.1, which is detailed in our response to main comment #5. Specifically, to improve the performance of the flux estimates, we treat the  $X$  and  $k$  as fundamentally empirical fitting parameters within a data-driven approach to ensure the resultant emission estimator centers around zero where emissions are negligible.

7. Chemical Loss Term — dropping the lifetime term because of a bad fit seems a bit easy and one that potentially has a large impact on the resulting emission and deposition fluxes, especially when moving away from the strongest emission gradients.

We thank the reviewer for this important insight. We agree that omitting the chemical loss term due to a poor fit is a debatable choice, particularly in areas with abundant columns. In response to this concern, we have revised our analysis to retain the chemistry term in the final flux estimator, using the  $DD_{chem}$  estimator as our primary flux estimate throughout the manuscript.

According to Ayazpour et al. (2024), a stricter maximum emission threshold is necessary for fitting  $DD_{chem}$  than for  $DD_{topo}$ . Could the poor fitting performance of the chemical loss term (line 247) be related to an insufficiently strict threshold? Please clarify and discuss whether further refinement of the  $X$  and  $k$  estimates is planned.

We conducted sensitivity tests to evaluate the fitted  $X$  and  $k$ ,  $w$ . For  $X$  fitting, we tested both spatially and temporally varying fittings, as described in our previous response to main comment #5. For  $k$  fitting, we tested spatially and temporally varying  $k$ , as well as stricter maximum emission thresholds, as suggested by the referee. The spatial and temporal variability tests indicate that allowing  $k$  to vary does not improve the fitting performance (Figs. S4-S5). Likewise, applying stricter emission thresholds for  $k$  fitting does not improve the fitting performance (Fig. S6). Nevertheless, changing the

maximum emission threshold from  $1 \times 10^{-9}$  to  $3 \times 10^{-10}$  mol/s alters the area-integrated emission rates by less than  $\sim 7\%$  across regions, indicating that the results are relatively insensitive to this parameter choice. We have added the sensitivity tests of fitted  $k$  in the end of Section 2.2.1 as follows: *“The same settings were applied to  $k$  for consistency, though its performance is largely unaffected by these changes (Figs. S4-S5). Stricter emission thresholds for the chemistry term also have little impact (Fig. S6)”*.

The following figures have been added to the Supplementary Materials:

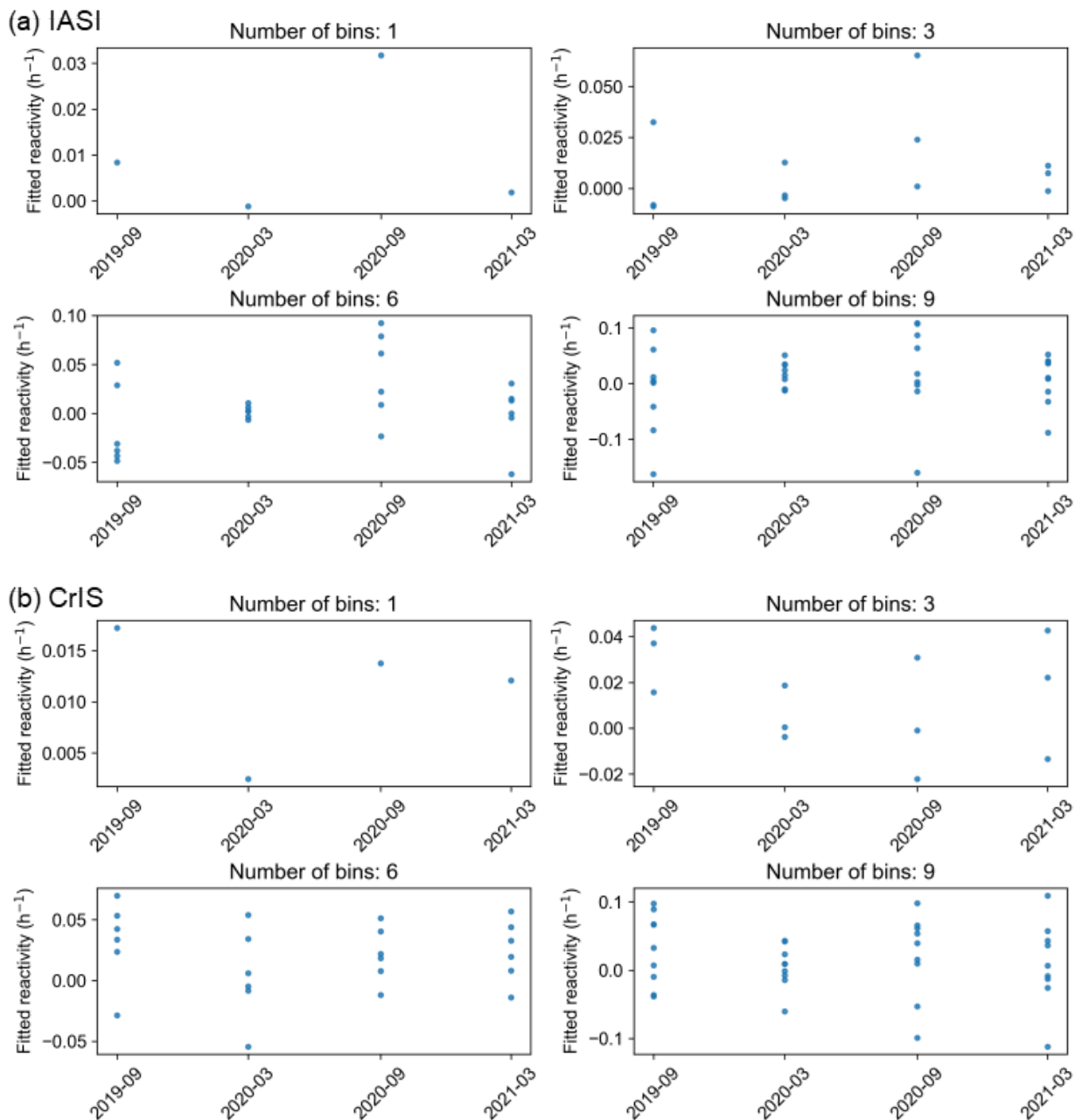


Figure S4. Spatial sensitivity tests for the topography term fitting to derive DD\_chem from IASI (a) and CrIS (b). The number of bins indicates the column ( $\Omega$ ) sectors used in the fitting. The y-axis shows the fitted  $k$ .

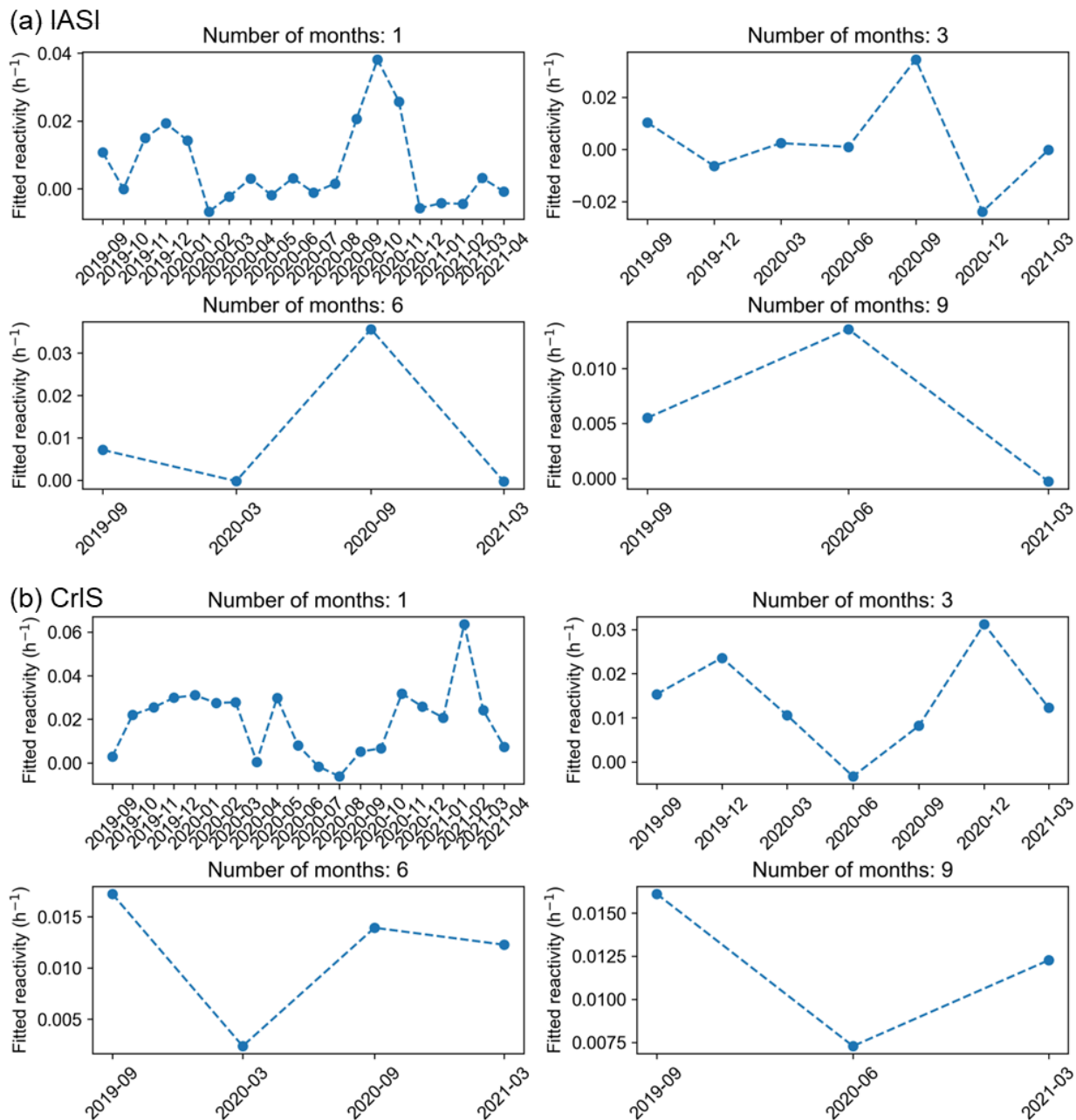


Figure S5. Temporal sensitivity tests for the chemistry term fitting to derive DD\_chem from IASI (a) and CrIS (b). The number of months indicates the temporal aggregation interval used for the fitting. The y-axis shows the fitted  $k$ .

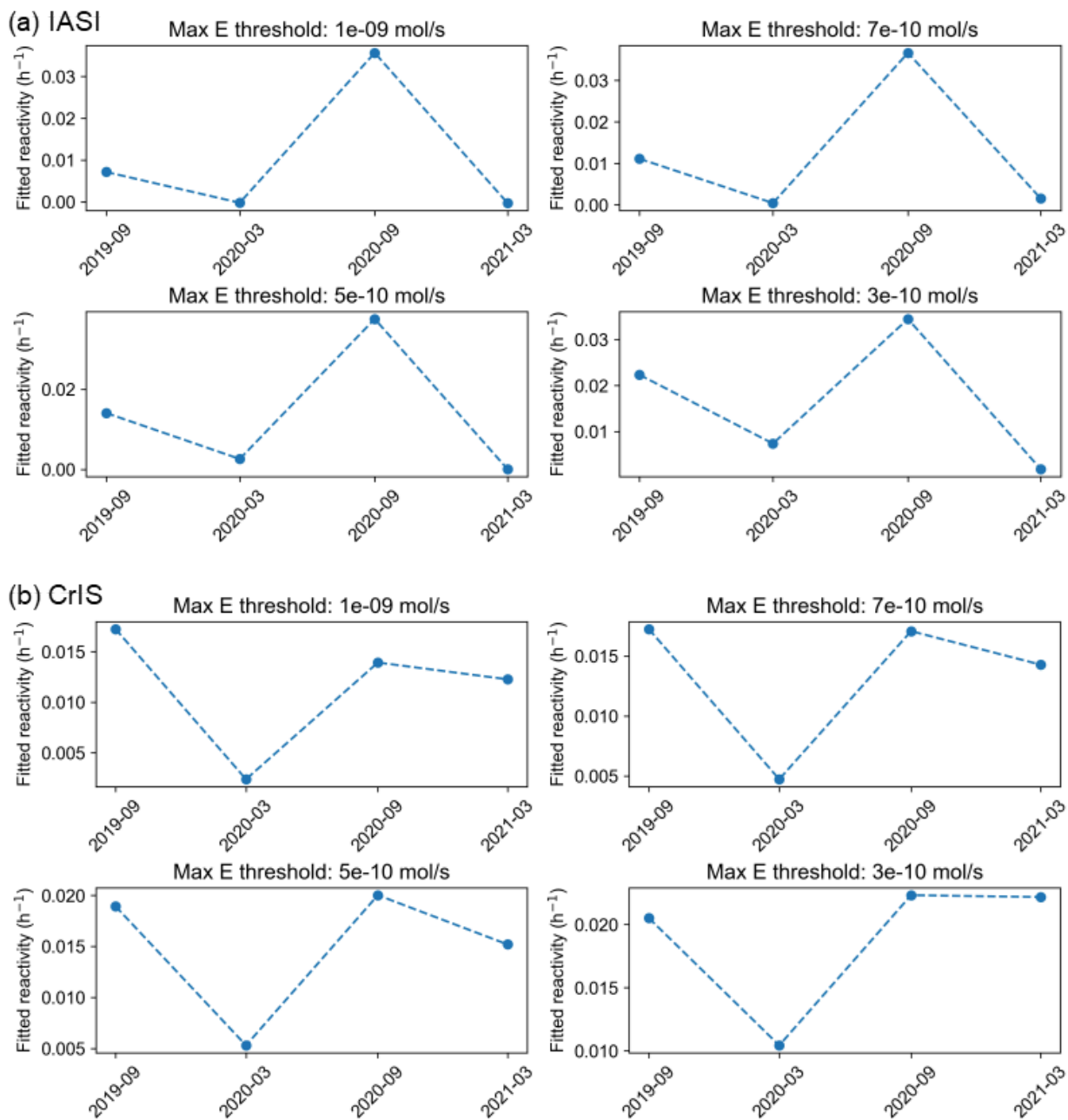


Figure S6. Maximum emission (E) threshold sensitivity tests for the chemistry term fitting to derive DD\_chem from IASI (a) and CrIS (b). The y-axis shows the fitted  $k$ .

Additionally, the fit seems to have been performed on the whole of the CONUS, whereas lifetime will vary strongly depending on the local pollution levels of other species (produced hno3/h2so4). A switch to locally varying fits would make sense from a chemistry point of view.

Thanks for the suggestion. We have addressed this concern in our previous response to main comment #7. Specifically, we have tested spatially and temporally varying  $k$  fittings, and the fitting performance was not improved by these changes (Figs. S4-S5). Although in theory  $k$  is tied to physically meaningful quantities, its main purpose is to enhance emission estimators in the presence of chemistry using information contained by satellite observations. As such, the quality of  $k$  fits is subject to the quality and quantity of available satellite data. Given the data gaps in individual IASI and CrIS pixels, as well as the reduced sample size when applying varying fits, introducing additional spatial or temporal variations led to larger noise in the fitted values without improving performance.

Alternatively, the authors could add an Alinea what the expected lifetime to chemistry is for typical hno3/h2so4 concentrations, and discuss from that point of view if chemistry is important or not. In its current form it's not convincing.

Thanks for the suggestion. As detailed in our response to main comment #7, we conducted sensitivity tests on chemistry term fitting. In the revised manuscript, we have included the chemistry term and use DD\_chem estimator as our flux estimates. We do agree that omitting the chemistry term is debatable, and its spatial pattern indicates that it plays a larger role in column-abundant regions (Fig. S8c, d). The following figure has been added to the Supplementary Materials:

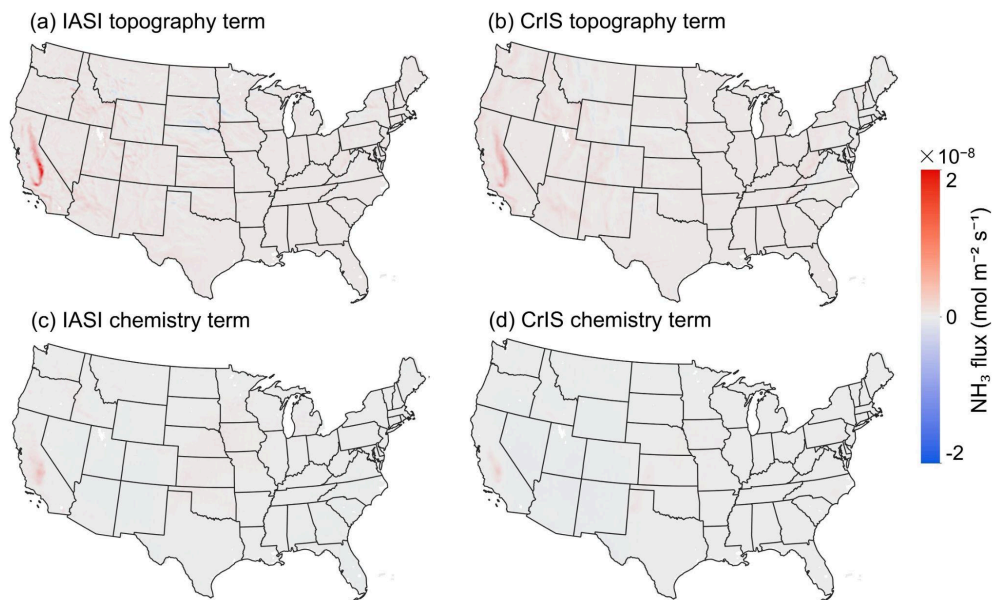


Figure S8. Topography and chemistry terms from IASI and CrIS over Sep 2019 to Apr 2021.

1. Daily Data and Bias Correction — Using only daytime data may overestimate NH<sub>3</sub> due to satellite sampling biases. Have you considered applying a satellite bias correction, as recommended in Ayazpour et al. (2024, Sect. 2.2.4)?

We thank the referee for the comment. It is true that using only daytime satellite observations may overestimate NH<sub>3</sub> levels due to diurnal variability and the lack of nighttime measurements. However, this limitation is inherent to thermal infrared sounders like IASI and CrIS, which only provide good quality data under clear-sky conditions with adequate thermal contrast between the surface and the lower atmosphere. We acknowledge our DDA-based NH<sub>3</sub> flux estimates are based on daytime data and reflect daytime-effective fluxes, and we added discussion about this limitation in Section 4.1 (Lines 446-448): *“retrievals are limited to daytime clear-sky conditions, potentially biasing results due to NH<sub>3</sub>’s strong diurnal cycle (Blanes-Vidal et al., 2008)”*.

Section 2.2.4 of (Ayazpour et al., 2025) aims to address retrieval biases related to spatial artifacts (e.g., surface reflectance), not temporal sampling biases. They proposed their use as a potential future enhancement, particularly for longer-lived species like CO, CH<sub>4</sub>, and CO<sub>2</sub>. We do not include similar bias-sensitive variables in our NH<sub>3</sub> estimates. This is because (1) there is no clear evidence of systematic spatial biases in our NH<sub>3</sub> retrievals that could be robustly modeled via  $\nabla a$ -type predictor fields, and (2) our regression fitting over low-emission regions inherently absorbs baseline retrieval errors into the fitted parameters for topography and chemistry. Since our primary goal is to estimate NH<sub>3</sub> flux rather than diagnose retrieval errors, we focus on the *DD\_chem* estimator. We acknowledge the potential for retrieval biases and the value of incorporating more advanced correction terms in future studies, as suggested by Ayazpour et al. (2025).

2. Line 301-305: I completely disagree that the directional derivative minimizes the impacts of offsets and scaling differences between the products. As the authors know the bias in the satellite products are not spatially and temporally independent, which means that any offset/scaling difference will vary from point to point, especially around larger shifts in concentration levels. If anything the scaling between the satellite product biases will be enhanced by the derivative. Additionally, the detection limit of both satellites will of course also play an important role in the limitations to detectable gradients. For example the early spring peaks detected by CrIS but not by IASI could be a sign of detection limit (or a strong diurnal variability in emissions of course). Some validation studies are available for both satellite products. These can be used as a basis for an error/uncertainty estimate of the effects. Please show what is the expected uncertainty of the current product bias/scaling on the resulting

fluxes. An update to the limit used later in the manuscript (i.e.  $\pm 2\sigma$ ) might be needed.

We thank the referee for raising these insightful points. We have revised the relevant text in Section 3.2 (Lines 299-305) as follows: *“The VCD comparison reveals systematic differences between IASI and CrIS as different slopes and offsets in different regions (Fig. 3a). These offsets and proportional biases can propagate into flux calculations, where they manifest as amplified variability and reduced correlation (Fig. 3b). The lower correlation in flux estimates compared to VCDs likely reflects compounded noises from derivative-based flux estimates, which relies on external datasets, assumptions, and signal differentiation.”*

We acknowledge the potential for product-specific biases to affect flux estimates, particularly near the detection limits of CrIS and IASI. The detection limit of  $\text{NH}_3$  depends on both thermal contrast and the vertical distribution of  $\text{NH}_3$ . We have added the uncertainties of IASI and CrIS  $\text{NH}_3$  VCDs from previous validation studies in Section 4.1 (Line 448): *“IASI columns differ from ground-based measurements by  $-32 \pm 56\%$  (Dammers et al., 2016), with errors highly dependent on thermal contrast (Van Damme et al., 2014). CrIS retrievals show  $\sim 10\text{--}30\%$  error in total columns and larger uncertainty at low concentrations (Shephard et al., 2020).”*

To mitigate these effects, we focus on hotspot source and sink regions with relatively strong  $\text{NH}_3$  signals, where retrieval uncertainties are lower. We also average over multi-month periods and constrain fitting procedures to low-emission regions where the signal-to-noise ratio is more stable and biases are more consistent. Regarding the  $\pm 2\sigma$  threshold, we have not revised this threshold at this stage, as the regional-scale aggregation and conservative filtering already help reduce the influence of noises. Future work incorporating satellite-specific error propagation into the DDA framework could further refine this threshold.

3. Suggestion for further validation: The comparison with inventories is interesting, but more direct evidence of the value of satellite based emission and deposition estimates would be a comparison with in-situ data, which currently is missing. Feeding back the emissions into a CTM and comparing the resulting concentrations with in-situ data would strengthen the case that satellite based emissions are an improvement over current inventories. Additionally, a comparison of the simulated deposition data (based on the updated emissions) with the satellite derived estimates would further show the value of those deposition estimates. It is quite an effort though so I would understand if the authors state it's beyond the scope of this manuscript.

We thank the referee for the thoughtful suggestion. We agree that using CTM and in-situ measurements for validation would provide valuable, independent evidence of the utility of satellite-based estimates. However, such model integration and evaluation require substantial computational effort, detailed configuration, and rigorous validation protocols, which are beyond the scope of the current study. We encourage future studies to explore this direction.

*Minor edits/comments:*

Abstract L29: “atmospheric” instead of “atmosphere”

We’ve revised as suggested.

L45-46, I’d rephrase this sentence. Spatially the emissions seem to align, but seasonally and in amplitude they do not.

Lines 45-46 changed to: *“Compared with bottom-up inventory, satellite-based estimates capture general spatial and seasonal patterns, while also revealing additional insights into key flux hotspots and peak periods.”*

L74: a few hours is on the low end of the model and measurement estimates, mostly derived from direct fits on satellite data, which are expected to bias low. Estimates of 8-12 or up to 24 hours seems more reasonable based on literature.

Line 74 changed to: *“less than 24 hours”*.

L84: Quite a recent reference, the relation between volatilization and environmental conditions was known much before this point.

Line 84 changed reference to: *“(Sommer et al., 1991)”*.

L103-105: Quite the claim when later analysis mostly focuses on monthly or longer temporal resolutions.

Lines 103-105 changed to: *“We derived top-down NH<sub>3</sub> fluxes at 0.1° resolution using observations from two space-based instruments: the Infrared Atmospheric Sounding Interferometer (IASI) and the Cross-track Infrared Sounder (CrIS)”*.

L124 onward: add the observational periods of each satellite after each satellite name, this will make it easier for the reader to follow what satellite is in orbit when.

Line 123 added: *“from Metop-A (January 2008 to October 2021), Metop-B (March 2013 to December 2022) and Metop-C (September 2019 to December 2022)”*.

Line 132 changed to: *“CrIS-NPP (June 2012 to May 2021) and CrIS-NOAA-20 (March 2019 to December 2022)”*.

Line 182-185: Essentially you are gap-filling the record, but I fail to see the basis for just inflating the pixel size without any smart input of additional data. I can imagine this type of gap-filling introducing stronger or weakening gradients in regions with very localized sources and/or very common wind directions. Please add a few words on potential effects on gradients.

Lines 182-185 changed to: *“This is a critical step to enable spatial gradient calculation when data coverage is sparse (e.g., only a single overpass is available). This also limits the spatial resolving power to about twice the pixel size”*.

Figure 1: Whats going on with the few outlier months in the CrIS and IASI records, are these specific periods? And what does excluding these do for your results?

In Fig. 1, a few outlier months with higher-than-expected random errors correspond to periods of low data coverage caused by cloudiness or partial instrument downtime. These months constitute a small fraction of the dataset and have minimal influence on the results, as low-coverage periods are down-weighted in the aggregation.

Line 313-315: what about the instrument detection limit?

We have added *“and differing detection limits”* to Lines 313-315 and moved this sentence to Section 4.1 (Line 428).

Section 4.2: or in discussion: I miss a discussion on the potential effects of instrument/product bias changing over time, and the expected impact compared to the increasing trends you observed here.

We thank the reviewer for this comment. Both IASI and CrIS have operated with multiple instruments over time, but existing studies indicate good temporal consistency. For IASI, the ANNI-NH<sub>3</sub> products include empirical corrections to address small instrument- and processing-related differences (Clarisse et al., 2023) and validation shows good agreement with independent datasets in multiple regions (Van Damme et al., 2015). For CrIS, long-term spectral stability is high (Chen et al., 2014) and validation shows strong correlation and minimal bias (Dammers et al., 2017). We therefore consider any time-dependent bias too subtle for the multi-year increases reported here to be attributed to bias drift. To address this potential limitation, we have added the following text to Section 4.1 (Line 448): *“While both IASI and CrIS have*

*demonstrated good long-term stability through calibration monitoring and ground-based validation (Chen et al., 2014; Clarisse et al., 2023; Dammers et al., 2017; Van Damme et al., 2015), subtle time-dependent biases cannot be fully excluded.”*

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## 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  $DD_{topo} = DD + \text{topography term}$ , and  $DD_{chem} = DD_{topo} + \text{chemistry term}$ . 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 ( $\Omega$ ) 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. DDA accounts for horizontal transport, topographic effects, and chemical transformation influencing gas distribution. 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 in the planetary boundary layer ( $\vec{u}$ , 100 m winds).  $\nabla = (\partial/\partial x, \partial/\partial y)$  is the horizontal vector differential operator. Ayazpour et al. (2025) evaluated  $DD$  estimators calculated using column amounts and winds at a range of altitudes in an atmospheric model with the model-ingested emission and found that winds from 100-800 m give similar and consistent results. We choose 100 m wind because it has been widely used in previous studies (Goldberg et al., 2022; Lonsdale and Sun, 2023) and is readily available from the ERA5 single-level product. The  $DD_{topo}$  estimator accounts for the topography term ( $X\Omega\vec{u}_0 \cdot (\nabla z_0)$ ), which is driven by directional derivatives of the surface altitudes ( $z_0$ , obtained from Level 2 satellite data) relative to near-surface wind vectors ( $\vec{u}_0$ , 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 Figures 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:

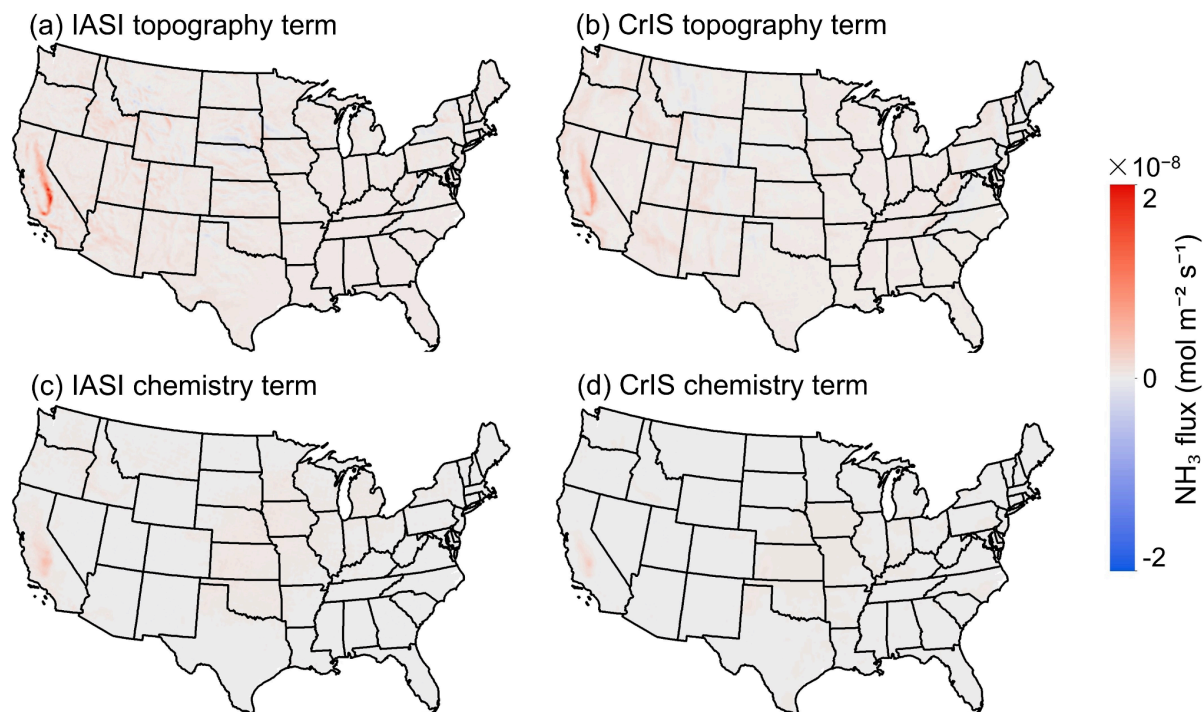


Figure S8. Topography and chemistry terms derived from IASI and CrIS over Sep 2019 to Apr 2021.

- 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?  $\text{NH}_3$  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 chemical loss 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 than 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 NH<sub>3</sub> is not in balance and atmospheric NH<sub>3</sub> 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<sub>3</sub> 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<sub>3</sub> 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<sub>3</sub> to surrounding areas, rather than an actual growth in atmospheric NH<sub>3</sub> 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 NH<sub>3</sub> abundance and budget analysis. As pointed out by the authors, the different overpass times by the two instruments provide unique information that can be extracted 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<sub>3</sub> 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 fluxes 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<sub>3</sub> source and sink, although systematic differences are observed. IASI-derived fluxes tend to resolve finer spatial detail, 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<sub>3</sub> fluxes.*

*Agricultural lands dominate as NH<sub>3</sub> source regions in all cases, with strong fluxes coinciding with intensive cropping and livestock production (e.g., 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 detail.”*

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  $DD_{\text{topo}} - DD$ ) instead of  $DD_{\text{topo}}$ , c) add another panel for  $\text{NH}_3$  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  $\text{NH}_3$  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  $DD_{\text{chem}}$  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.

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