We would like to thank the reviewers and editorial staff for their assessments of our revised manuscript. We appreciate the insightful comments of Reviewer 3. In this work, we explore a new proxy-based approach to estimate cropland N2O emissions using space-based NO2 observations using (limited) airborne measurements of N2O and NOx available at the field scale. Further atmospheric measurements would be fantastic to further evaluate such an approach. We think our results are encouraging and can be used to motivate such further measurements. In the end, we agree with the reviewer that such a proxy-based approach warrants further exploration. We thank the reviewer for their insight into the complexities of resolving cropland N2O from space using the proposed approach. We have incorporated the reviewer's additional suggestions, as detailed below. Our responses to individual comments are provided in indented italics, which the relevant text supplied in "quotes."

Thank you,

Taylor Adams

On behalf of all authors.

Reviewer 3:

This manuscript presents a preliminary attempt to investigate whether soil N2O emissions can be inferred from NOx emission estimates determined from satellite data on NO2 columns and a box model. First, they examine aircraft measurements of N2O and NOx downwind of California croplands to obtain central estimates and distributions of N2O:NOx emission ratios. The then apply these emission ratios estimates of agricultural NO emissions derived from calculations of NOx emissions based on TROPOMI NO2 observations. They compare these N2O flux estimates to independent ground and airborne studies in the US Corn Belt and Mississippi River Valley, which are shown to be "broadly consistent." The authors posit that the method is promising enough to warrant further studies.

I agree that the idea was worth pursuing and could be pursued further. I also feel that this the research was sound and the manuscript well-written, so that this effort can and should be documented in the peer reviewed literature. That said, significant challenges remain to prove that this approach will provide useful estimates of N2O emission, which I describe below. I also have minor suggestions for improving the clarity of the manuscript.

For this approach to be proven useful, it will need to demonstrate confidence in spatial and temporal variation in N2O emissions. For example, the flux estimates in Griffis et al. (2013), based on N2O concentrations measurements at 100m on a very tall tower in Minnesota and accompanying micrometeorological measurements and budgeting analyses, show temporal variation on the order of 0 to nearly 3 nmoles N2O/m2/s for the tower footprint area, with a distinct peak in June and much lower fluxes during the late summer and non-growing seasons. At the cornfield plot scale in Maryland, a micromet study using short towers showed fluxes ranging from near zero to a peak of about 8 nmoles N2O/m2/s following a fertilization and wetting events (Zhu et al. 2023). Wagner-Riddle et al. (2007) showed similar peak fluxes after fertilization and also smaller but important peaks of about 1-3 nmoles N2O/m2/s during spring snowmelt in Ontario. So, the question is whether the proposed method based on space-based estimates of soil NO flux, modified by N2O:NO ratios can reveal variation of similar magnitude. On the positive side, the fluxes shown in Fig. 3 are generally within a reasonable range of about 0-4 nmoles N2O/m2/s. A less positive result is that panels a and b show only means and ranges for a measurement period, and panel c has only a one-point-in-time estimate from this method, so we can't tell if there was any temporal variation. Furthermore, panel b suggest that we might not be able to distinguish between a flux of 1 and 3 nmoles N2O/m2/s, which would be disappointing. The SI shows that the mismatch could be larger, depending upon the assumed NOx lifetime. Might this factor vary temporally and spatially, thus making the challenge of detecting spatial and temporal variation on N2O fluxes even larger? A necessary test would be to make estimates over seasons, including periods when fertilization is likely and when it is not and between regions that have a lot of fertilized croplands and those that don't. This is a challenging task, and I understand why it hasn't been done yet, but it should be identified as a key next step.

I give credit to the authors for revealing all of these uncertainties, and I reiterate that I think this was and still could be a worthwhile endeavor, especially given the lack of alternative approaches for space-based estimates of N2O fluxes. Despite my concerns about whether the method will really pan out, I think that this work should be presented in the literature.

Thank you for these thoughtful insights, which we agree with. We have added further language to highlight further measurements would be invaluable for evaluating this:

"Further measurements of N_2O:NO_x emissions at commensurate spatial scales with satellite measurements across different agricultural regions over time would be invaluable to further evaluate this method's validity to capture emissions through potentially complex confounding spatial and temporal factors. "

Here are a few suggestions for minor improvements of the manuscript:

Fig. 1: The inset graphs seem to suggest that the ratios are all integers, but the text gives non-integer values. This should be clarified.

> *We believe some confusion likely resulted from unclear axes labels on our previous version of Figure 1. We have updated histogram insets in Figure 1 to clearly identify the emission ratio axis (x-axis) from the frequency/counts axis* (y-axis). *The revised figure, which shows the binned emission ratio data as non-integers, and caption are copied below.*

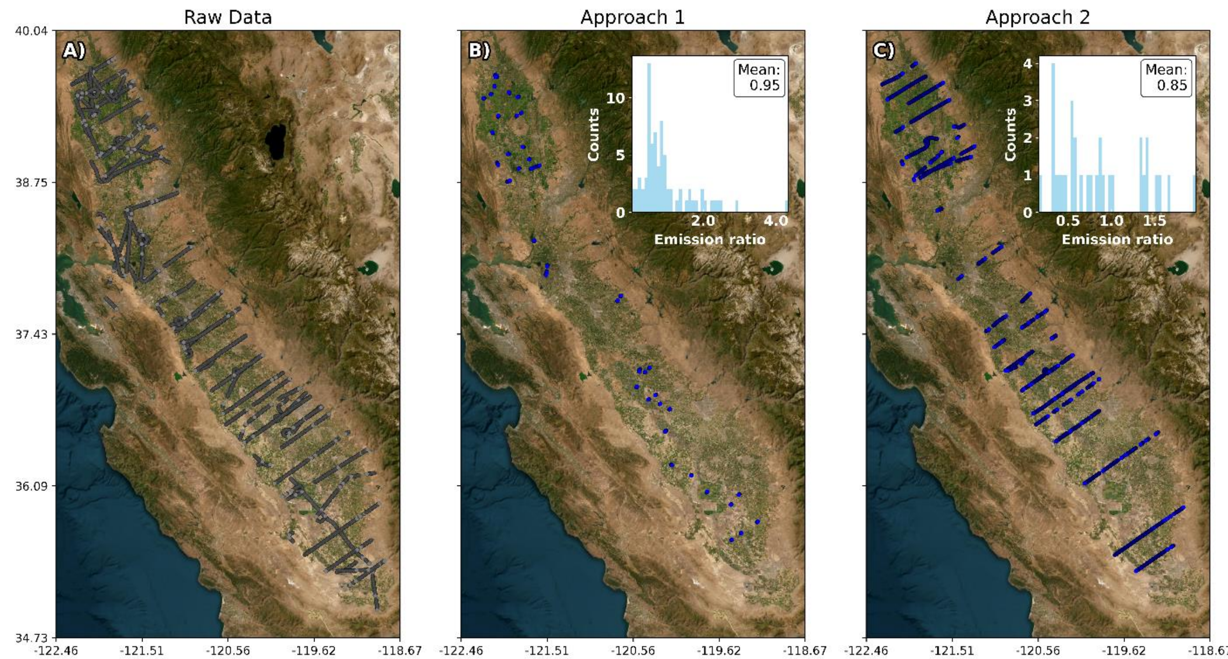

Figure 1: Flight maps corresponding to data remaining after filtering steps in A) the raw CalNex dataset filtered for data within the agricultural fields and away from high NO_2-emission areas, B) in approach 1 and C) in approach 2. A histogram showing the distribution of binned molecular emission ratios determined in each respective approach is overlaid upon the map. Satellite imagery credit: Esri.

Lines 218-220: This is an understatement and it may not be understood by all readers. I had to return to this sentence after reading the results to figure out that the N2O:NO ratios used to estimate fluxes in the Midwestern sites were derived from the study in California. The Central Valley of California is generally drier than the rainfed agricultural areas of the Midwest, so I would expect lower N2O:NO ratios in California. This should be discussed and it should be made clearer later in the manuscript when discussing Fig. 3 results, that the N2O fluxes in the Midwest are based on ratios derived from a study in California. On this point, I disagree with the authors' response to reviewer #1 in the previous round of reviews. Numerous studies in the literature demonstrate that variation in this ratio is large in both space and time.

We agree that it is important to make clear that the N2O fluxes estimated in the Midwest are derived using N2O:NOx emission ratios derived from airborne data in California. As the reviewer suggested, we now include this information in our discussion of our comparison in Figure 3, and we reiterate this at the start of section 5. The relevant section is copied below:

Start of Section 5:

5 Comparison with Independent Estimates of N_2O

We compare our space-based N_2O emissions estimates derived using California-observed $N_2O:NO_x$ emission ratios with N_2O emissions from independent studies. We first compare N_2O emissions derived from TROPOMI-NO_2 observations with those obtained from chamber measurements reported by Lawrence et al., (2021). The chamber measurements, conducted between February 2017 and October 2019 in Iowa crop fields, are compared only for the warm season (May-September) of 2018 and 2019 when TROPOMI was operational, and chamber data was available. The comparison domain spans -94.055 to -93.305 in latitude (0.75°) and 41.605 to 42.355 in longitude (0.75°). The box model domain lies to the north of Des Moines, Iowa, and is centered on the Ames, Iowa field site referenced in Lawrence et al., (2021)(41.98°N, 93.68°W), and is shown in Fig. 2A, with a star indicating the chamber location.

Further discussion of results from Figure 3:

We find the TROPOMI NO_2-derived N_2O flux compares favorably with various independent measures of N_2O flux and emissions from the corn belt (Dacic et al., 2024; Lawrence et al., 2021) and Mississippi River Valley (Gvakharia et al., 2020). For two of the comparisons, space-based estimates are within ~15% of these independently obtained N_2O estimates, despite this testing capturing multiple regions and time-periods, and the airborne derived emission ratios coming from observations in a completely different agricultural region. The $N_2O:NO_x$ ratios used here were observed with aircraft over California croplands – very different from the rainfed agricultural regions in the Midwest and southern USA. Previous studies have demonstrated strong spatiotemporal variation in emission ratios in response to soil moisture (Anderson and Levine, 1987; Davidson, 1992; Johansson and Sanhueza, 1988; Lipschultz et al., 1981; Tortoso and Hutchinson, 1990) and crop variety (Anderson and Levine, 1987) and suggests in wetter regions $N_2O:NO_x$ ratios would shift upward in general. If that pattern held at the scales observed here, one would expect the satellite method to produce an underestimate of Midwest and southern USA emissions, which is perhaps another explanation for the comparison between the satellite method and Dacic et al., (2024). The general agreement of our proxy-based N_2O estimates with those from independent assessments of cropland N_2O emissions demonstrates the potential of scaling satellite-based NO_2 observations with $N_2O:NO_x$ emission ratios to capture agricultural N_2O emissions, and that such an approach may provide a viable method to estimate N_2O emissions from agricultural regions around the world without necessarily needing location specific $N_2O:NO_x$ emission ratios. Further measurements of $N_2O:NO_x$ emissions at commensurate spatial scales with satellite measurements across different agricultural regions over time would be invaluable to further evaluate this method's validity to capture emissions through potentially complex confounding spatial and temporal factors.

In numerous places in the manuscript the ratio flips back and forth between N2O:NO or NO:N2O (I'm less worried about switching among NO, NOx, and NO2 when using this ratio, for the reasons given by the authors in the responses to the previous round of reviews). It would be less confusing to readers to pick whether N2O appears in the numerator or the denominator and be consistent throughout the manuscript.

> *In the revised manuscript, we now use a consistent form of the emission ratio with N2O in the numerator. We continue to introduce the concept of NO_x:N_2O emission ratios earlier in the manuscript, though. We agree with the reviewer that this improves the readability of the manuscript. As an example of these changes, below is our discussion of the observed N_2O:NO_x emission ratios compared to those in the literature:*
>
> Emission ratios range between 0.038 and 4.34 ppb N_2O / ppb NO_x for *approach 1* and 0.16 and 1.97 ppb N_2O / ppb NO_x for *approach 2*, reflecting expected heterogeneity in N_2O:NO_x ratio over agricultural lands, and demonstrating that increasing the spatial scale aggregated to create these ratios dampens variability. The N_2O:NO_x values we observe are in line with literature from soil-chamber measurements, which report highly heterogeneous emission ratios typically ranging from ~0.1-10, but that in rare cases can be as high as 100. (Johansson and Sanhueza, 1988;Tortoso and Hutchinson, 1990).This variation occurs as a function of factors such as fertilizer application, crop-type, and other management and environmental factors (Anderson and Levine, 1987; Davidson, 1992; Johansson and Sanhueza, 1988; Lipschultz et al., 1981).

In the section on future studies, another approach worth mentioning for estimating N2O:NO ratios would be output from a model like DayCent, which bases estimates of that ratio primarily on soil moisture (specifically on water-filled pore space of the topsoil the last time that I checked, which was a while ago). If soil moisture could be estimated simultaneously from space or from models using ground-based data on soil properties and weather, then a spatially and temporally variable ratio (and its uncertainty) could be estimated.

> *Thank you for this insight into another approach to establish N2O:NO emission ratios for use with the presented N2O proxy method. We have included a discussion of the use of process-based models, such as DayCent, in our closing remarks.*
>
> *Concluding sentence:*
>
> We have demonstrated that space-based NO_2 observations as a proxy for N_2O cropland emissions compare favorably to independent estimates across multiple agricultural areas and years. This suggests that space-based NO_2 retrievals are a viable and robust proxy for N_2O flux at scales of at least 0.75 x 0.75 degrees, and over timescales as short as five days. Further development and refinement of approaches to characterize agricultural NO_2 from satellite observations and link them to N_2O emissions are possible. As presented here, the largest source of uncertainty in the estimated N_2O emissions derives from the large variability in the observed airborne N_2O:NO_x emissions ratio. Improved understanding and definition of this ratio, and what controls variation, could improve the fidelity of this proxy approach. This could be accomplished with airborne observations of NO_x and N_2O. Capturing different crops, agricultural practices and environmental conditions would provide more insight into emissions ratios and best practices on how to apply to independent satellite data in new domains. A complementary approach to the use of direct atmospheric

observations of cropland N_2O and NO_x in our proxy-based method would be to combine satellite-based or ground-based estimates of soil moisture with a biogeochemical model, such as DayCent (Li et al., 1992; Parton et al., 1998), to derive modeled N_2O:NO_x emission ratios. With such an approach one could model emissions, and uncertainties, over many different regions and conditions where measurements may not be possible. The work presented here demonstrates a proxy-based approach that may offer a path towards a more spatially comprehensive constraint on regional and global budgets of agricultural N_2O emissions.