

## Anonymous Referee #1

Received and published: 21 December 2020

The authors aim to explore the drivers of IASI-observed NH<sub>3</sub> change from 2008 to 2017 over Africa through spatiotemporal analysis together with IASI CO VCDs, precipitation observations, surface temperature reanalysis data, MODIS burned area data and MODIS cropped area data. The topic is interesting and important. However, major re- vision is recommended before being suitable for publication due to some unaddressed issues below.

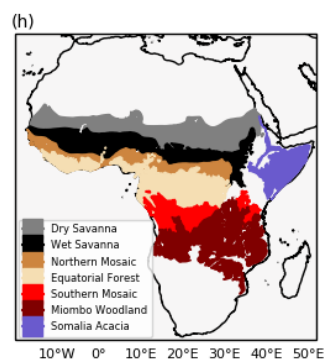
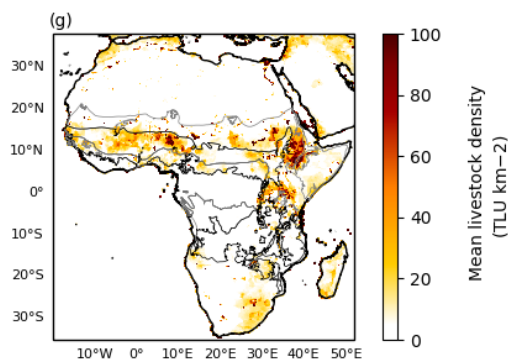
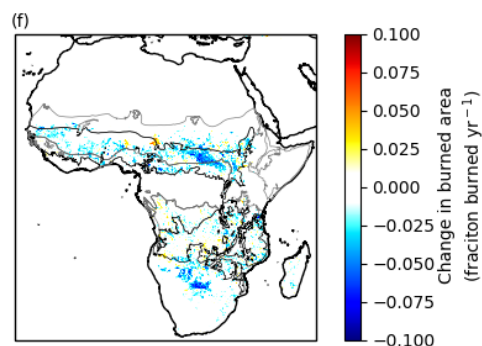
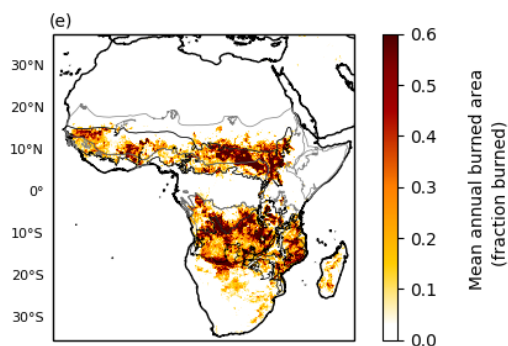
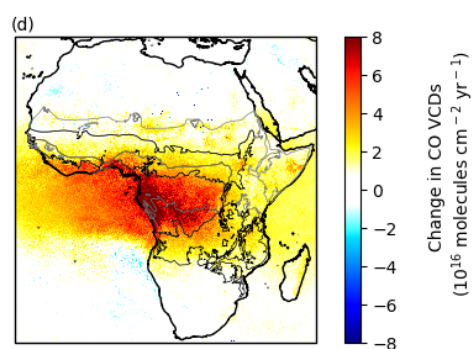
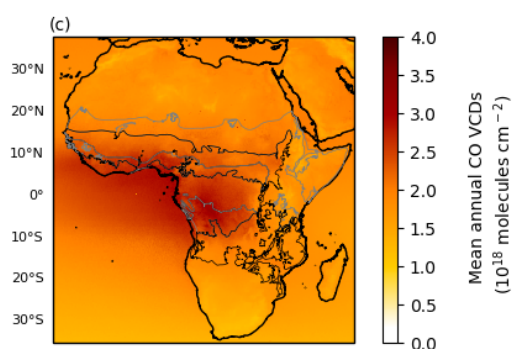
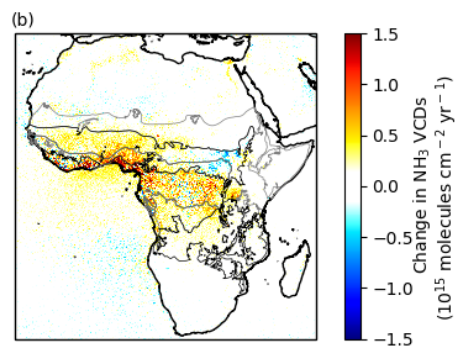
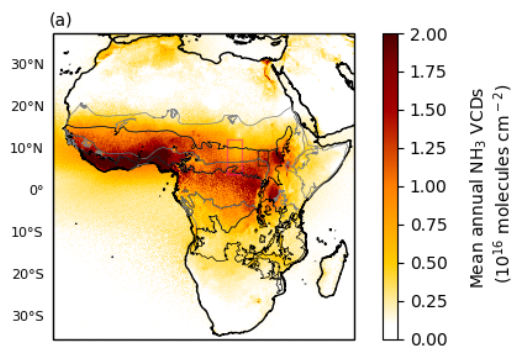
Thank you for the thoughtful review. We have revised our analyses substantially; the effort required has resulted in a delay in submission of our revision, and we thank you for your patience. Most notable in our revision, we now use a new IASI NH<sub>3</sub> product, v3R, which was released after the original submission of the manuscript, and which provides a time series through the end of 2018. We also extend all other data to 2018, and have redone all of our analyses.

The version 3 of the IASI NH<sub>3</sub> dataset has been developed for trends analyses and is extensively described in Van Damme et al., 2021. There is very relatively little difference between the v2.2R and v3R products in mean NH<sub>3</sub> VCDs (see also Van Damme et al., 2021). The main difference for our study is that trends are more broadly characterized by increasing rather than decreasing NH<sub>3</sub> VCDs, putting our results more in line generally with those of Warner et al. 2017 and new trend analyses by Van Damme et al. 2021. However, these changes do not qualitatively change our regional analyses of West Africa, the Sudd, or the Lake Victoria Region—the differences between v2.2R and v3R are most noticeable in central Africa.

Major comments:

1. While Warner et al. (2017) shows increasing trends of AIRS NH<sub>3</sub> across most of Africa from 2002 to 2016, this study shows decreasing trends of IASI NH<sub>3</sub> across most of Africa (except parts of west Africa and Lake Victoria Basin) from 2008 to 2017. In addition, section 3.5 (Figure 8) of this study shows that atmospheric CO, tropical livestock units, crop production and fertilizer N at national scale increased from 2008 to 2017, which seems to support Warner et al. (2017) rather than this study. Those information make me doubt the reliability of the trend analysis (or data processing) in this study. Can you please explain the difference between AIRS-observed NH<sub>3</sub> trends and IASI-observed NH<sub>3</sub> trends across most of Africa?

This is an excellent question, and one we should have spent more time discussing. Much of the difference between Warner et al. and our analyses can be attributed to two factors: 1) differences in the satellite product used (AIRS vs IASI), and 2) differences in the time period examined. In the revised manuscript, we now use v3R of the IASI-NH<sub>3</sub> product; the original version of the manuscript used v2.2R. The updated product results in trends that are more broadly similar to those observed in Warner et al., as can be seen in our revised figure 1b:



**Figure R1 (In manuscript as Figure 1).** Livestock density and annual averages and trends in burned area and atmospheric NH<sub>3</sub> concentrations across seven sub-Saharan African ecoregions. Mean annual (a) and trend (b) in atmospheric NH<sub>3</sub> VCDs from IASI for the period 2008 through 2018. Mean annual (c) and trend (d) in annual atmospheric CO VCDs from IASI for the same period. Mean annual (e) and trend (f) in annual burned area from MODIS for 2008-2018. Livestock densities for 2007 from the FAO (f), and mean cropped area from MODIS for 2008-2018 (g). The border of South Sudan is highlighted in black, and several regions boxed: the Nile region at 30°N, the Sudd wetland in South Sudan, and the Lake Victoria region at the equator.

We do still observe a decrease in NH<sub>3</sub> VCDs over the Sudd and South Sudan, which is not clearly observed in the Warner et al analysis. However, this overall pattern is also observed in the recently published Van Damme et al. 2021 (<https://doi.org/10.1088/1748-9326/abd5e0>), which provides a global evaluation of NH<sub>3</sub> trends using the v3R IASI product. In addition, we believe that our regional analysis provides convincing evidence that the observed decrease is related to interannual variability in Sudd extent, which will be sensitive to the time period observed.

For completeness, we also answer the reviewer's question as to why the earlier v2.2R analysis differed from the Warner et al. analysis:

First, we can note that there are several differences between AIRS and IASI. IASI is approximately 4 times more sensitive to NH<sub>3</sub> than AIRS. The lower sensitivity in AIRS is a particular problem in the boundary layer, where thermal contrast is low, and where we may expect much of the NH<sub>3</sub> column to be located.

In addition to the differences between the instruments and retrievals, there are several key differences in the Warner et al. 2017 AIRS analysis and our analysis of IASI data:

1) Warner et al. evaluate the 2003-2016 period whereas we evaluate the 2008-2017 period;

2) Warner et al calculate mean NH<sub>3</sub> changes using daily observations subjected to a smoothing function. We conduct trend analysis based on annual means calculated based on monthly means. This approach neglects seasonal cycles, but it effectively weights each month of observations equally. This approach ameliorates some issues introduced by the fact that cloud cover causes predictable seasonal variation in valid retrievals (see Figure S2), which creates the potential for introducing biases in analyses.

3) the Warner et al. analysis uses 1 x 1 degree resolution vs. 0.25 x 0.25 degree in our analysis;

4) Warner et al. exclude pixels where >90% of pixels were less than 2ppb and had records shorter than 10 years.

An examination of publicly available L3 AIRS  $\text{NH}_3$  observations suggests that the sensitivity of the trend to the time period examined explains some of these broad differences. We conducted analyses of AIRS data using the same approach we used for IASI, examining trends over two periods: 2003-2016 and 2008-2016. Note that our analysis for 2003-2016 is broadly similar to the Warner et al. 2017 results, though we used a different approach; we believe this supports the reliability of our approach. Also note that the 2008-2016 analysis exhibits patterns more similar to our original IASI v2.2R analysis, including the negative trend over the Sudd.

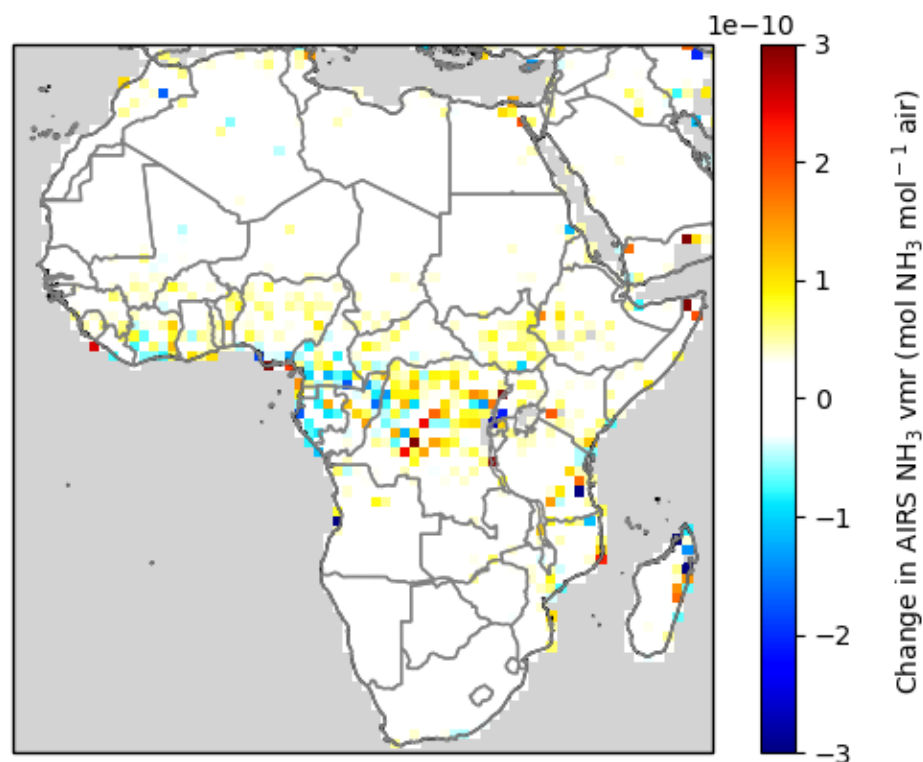


Figure R2: Change in  $\text{NH}_3$  VCDs as observed by AIRS, 2003-2016.

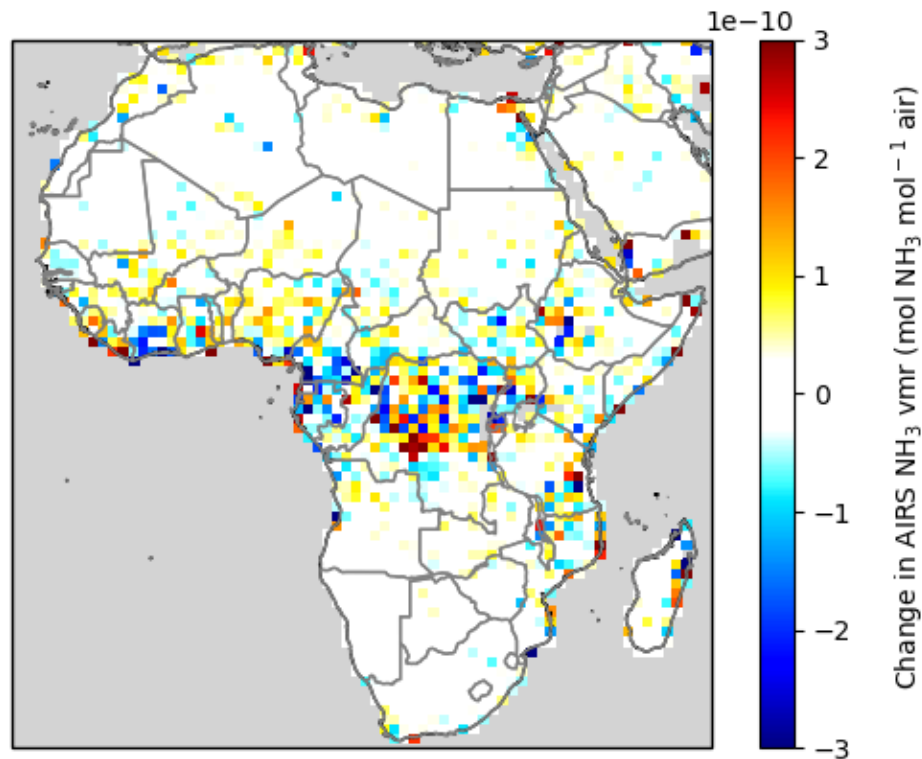


Figure R3: Change in  $\text{NH}_3$  VCDs as observed by AIRS, 2008-2016.

We want to emphasize that the FAO data, which are the source of the national statistics on changes in crop production, fertilizer use, and tropic livestock units, are known to be unreliable for sub-Saharan African countries. In one example, a remote sensing analysis argues that crop production figures reported to the FAO had been grossly inflated in Malawi, and that the reported year-on-year increases in production did not actually occur (Messina et al., 2017).

To demonstrate that our analytical approach of using mean annual  $\text{NH}_3$  VCDs (as derived from monthly means) does not contribute to the differences, we also conducted an analysis that includes explicit analysis of seasonal variability in which we fit a polynomial equation to weekly means of  $\text{NH}_3$  data for the 2008 – 2017 period. The result of this analysis reveals an overall pattern that matches our original analysis. However, using weekly means results in more missing values at some point in the 10-year record.

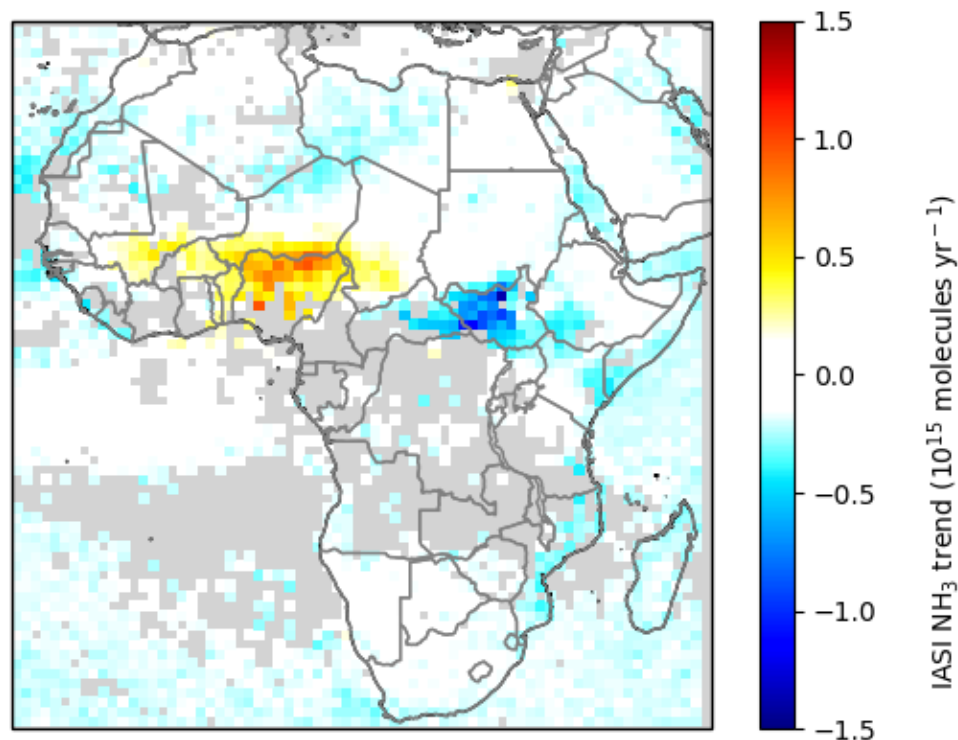


Figure R4: Change in  $\text{NH}_3$  VCDs as observed by IASI v2.2R, 2008-2017, using a deseasonalized trend analysis.

2. Line 128-129: “We regridded the Level-2 IASI  $\text{NH}_3$  and CO products to  $0.25^\circ \times 0.25^\circ$  resolution . . .”. How did you regrid the data? Did you apply any averaging weight? Like column error? How large is the observation error of IASI  $\text{NH}_3$ ? Did you apply any data quality control procedures? Does the number of pixels in each individual grid have large spatial variability? Generally how many pixels are in one  $0.25^\circ \times 0.25^\circ$  grid?

We note that the approach used in this manuscript to gridding the data is a conventional approach, and is the same detailed and used in previous publications, including Van Damme et al., 2021, which conducted a global  $\text{NH}_3$  trend analysis using IASI observations. We now include more detail on the data selection and regridding in the methods section (lines 130-136; please note that all line numbers refer to the version of the revised manuscript \*without\* tracked changes):

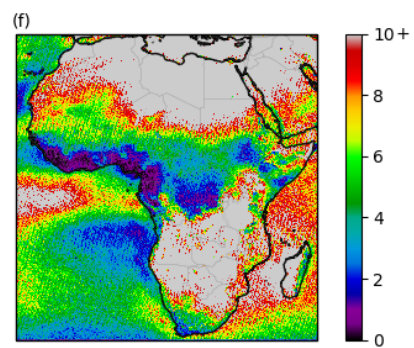
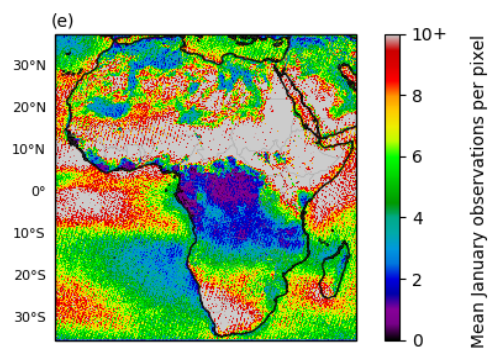
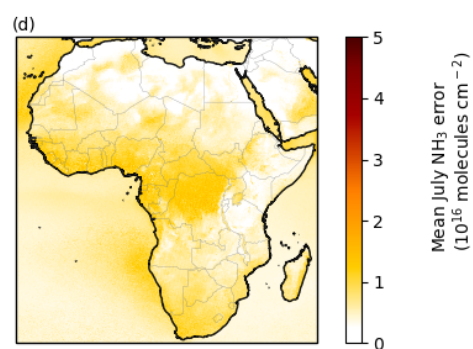
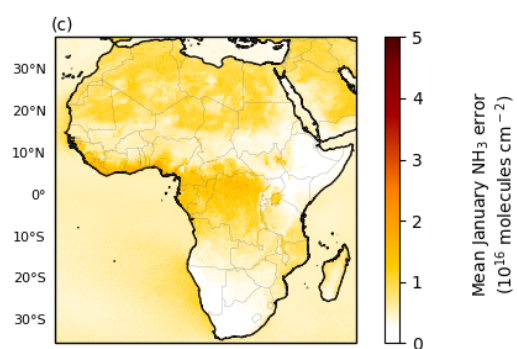
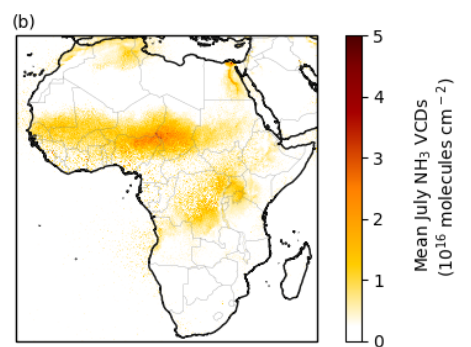
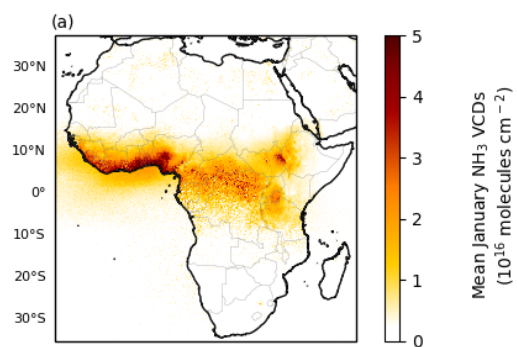
We used a conventional binning approach based on the center of each satellite footprint. We did not apply an averaging weight. Quality control procedures were followed as detailed in van

Damme et al. 2017 and Van Damme et al., 2021. Specifically, the screening of retrievals included filtering of retrievals where cloud cover is over 10%, where the total column density is below zero and the absolute value of the hyperspectral range index (HRI) is above 1.5, and where the ratio of the total column density to HRI is larger than  $1.5 \times 10^{16}$  molecules  $\text{cm}^{-2}$ .

Random errors in observations can be assumed to cancel out in the annual mean, which is what we used in our analysis. With the assumption that random errors cancel out, only systematic errors related to tropospheric vertical column contents remain; these systematic errors do not contribute to uncertainty in trend analyses. In addition, we first take monthly averages based on all daily observations within a given month before calculating seasonal means to minimize any potential effects of temporal variability in cloud cover.

In response to the questions regarding the number of pixels per grid cell and the associated error, please see figure R5, which presents the values for months from two seasons, as well as mean  $\text{NH}_3$  VCDs to provide context:







**Figure R5: Mean NH<sub>3</sub> VCDs, errors, and pixel counts for January and July, 2008-2018.** a) Monthly mean NH<sub>3</sub> VCDs for January. b) Monthly mean NH<sub>3</sub> VCDs for July. c) Monthly mean of the absolute error per pixel for January. d) Monthly mean of the absolute error per pixel for July. e) Monthly mean number of pixels per grid cell for January. f) Monthly mean number of pixels per grid cell for July.

Specific comments:

1. Line 36-41: “. . . with increases of over 6% yr<sup>-1</sup> in Nigeria. . . South Sudan NH<sub>3</sub> VCDs declined by over 2% yr<sup>-1</sup>. . .”. What’s the significance level of these trends?

We have revised these statements after analysis with the v3R product, and update the text accordingly, with significance levels (lines 34-40):

In West Africa NH<sub>3</sub> VCDs are observed to increase during the late dry season, with increases of over 7% yr<sup>-1</sup> in Nigeria during February and March ( $p < 0.01$ ). These positive trends are associated with increasing burned area and CO trends during these months, likely related to agricultural preparation. Increases are also observed in the Lake Victoria Basin, where they are associated with expanding agricultural area. In contrast, NH<sub>3</sub> VCDs declined over the Sudd wetlands in South Sudan by over 2% yr<sup>-1</sup> ( $p = 0.20$ ).

2. Figure 1: please mark the Lake Victoria basin and Nile delta and river, and South Sudan as well as Sudd in the plots.

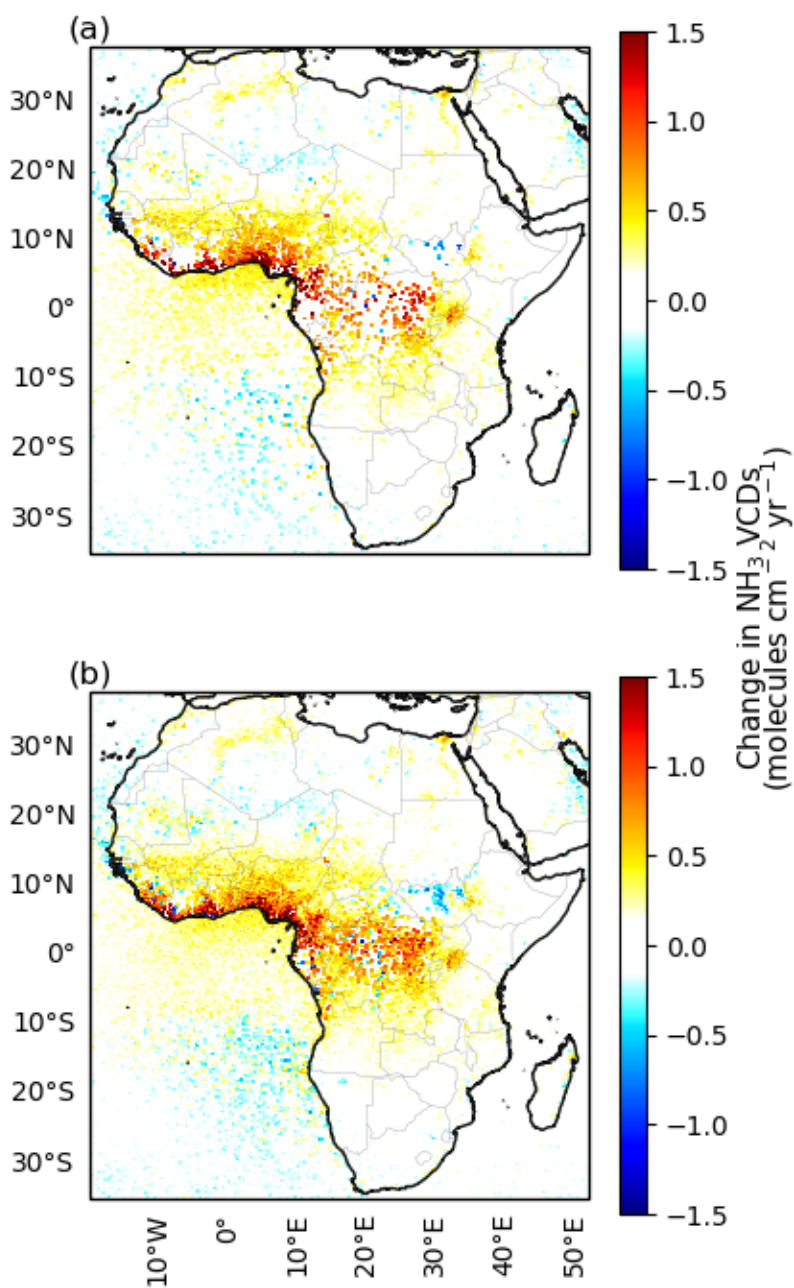
Good suggestion—In Figure 1 we have added boxes around the Nile delta & river region, the Sudd, and the Lake Victoria region as presented in Figure 7. We have also placed the South Sudan boundary in bold. We changed the boundaries from ecoregions to national, since our analysis is more often using countries than ecoregions; this also allowed us to include a map of cropped area in the figure.

3. Line 237: “. . . from 2008 to 2017 (Fig. 1d)”. I think it’s should be Fig. 1b.

Thank you for catching that—we have made the change.

4. Line 236-243: How significant are these trends?

We have elected not to screen the values in Figure 1b for significance to make the figure consistent with the trend results presented in Warner et al. 2017 and Van Damme et al. 2021, which also do not screen for significance. We now include a figure in the SI including only values significant at  $p = 0.1$ , and most of the pixels in Figure 1b are significant at  $P = 0.2$ :



**Figure R6 (In SI as Fig. S1).** Trend in atmospheric  $\text{NH}_3$  VCDs from IASI for the period 2008 through 2018 where trends meet the significance threshold of (a)  $p=0.05$  or (b)  $p=0.20$ .

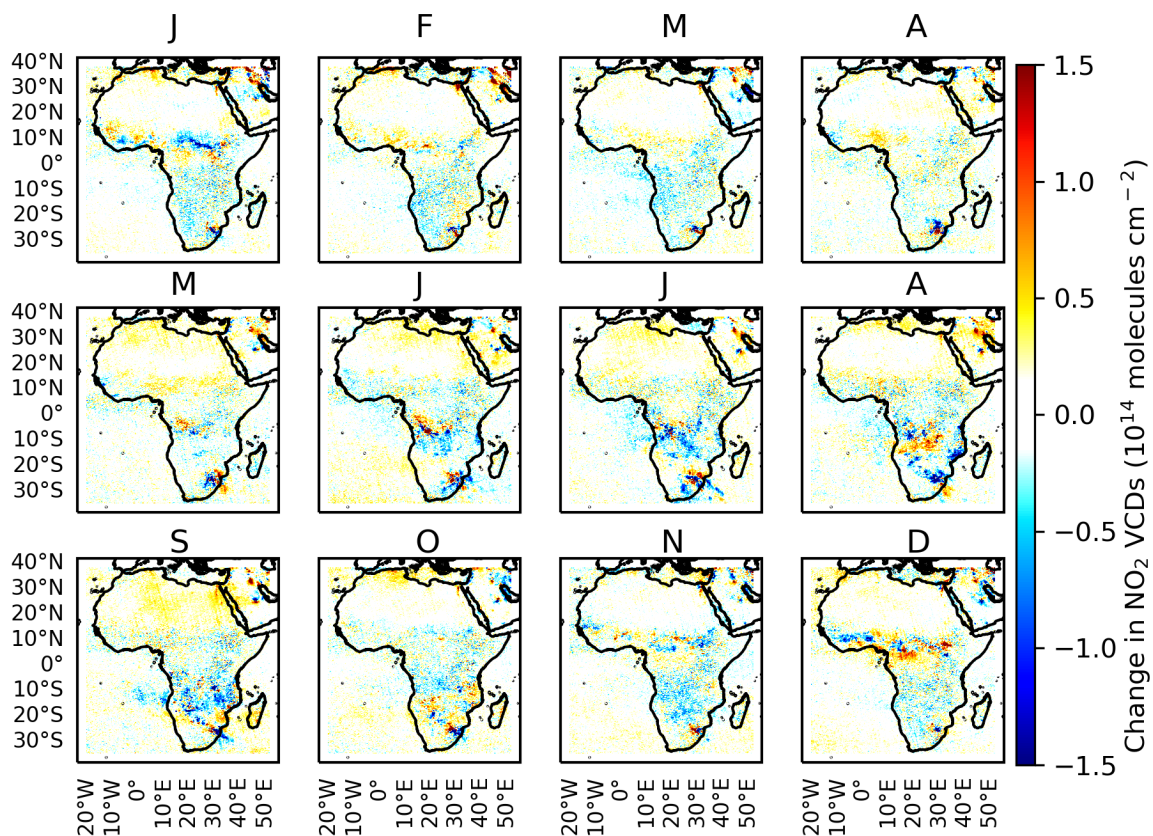
5. Line 279-280: Which months are the transition period, dry season and rainy season, respectively?

We now clarify the timing of these seasons in the text (new text in italics (lines 308-313):

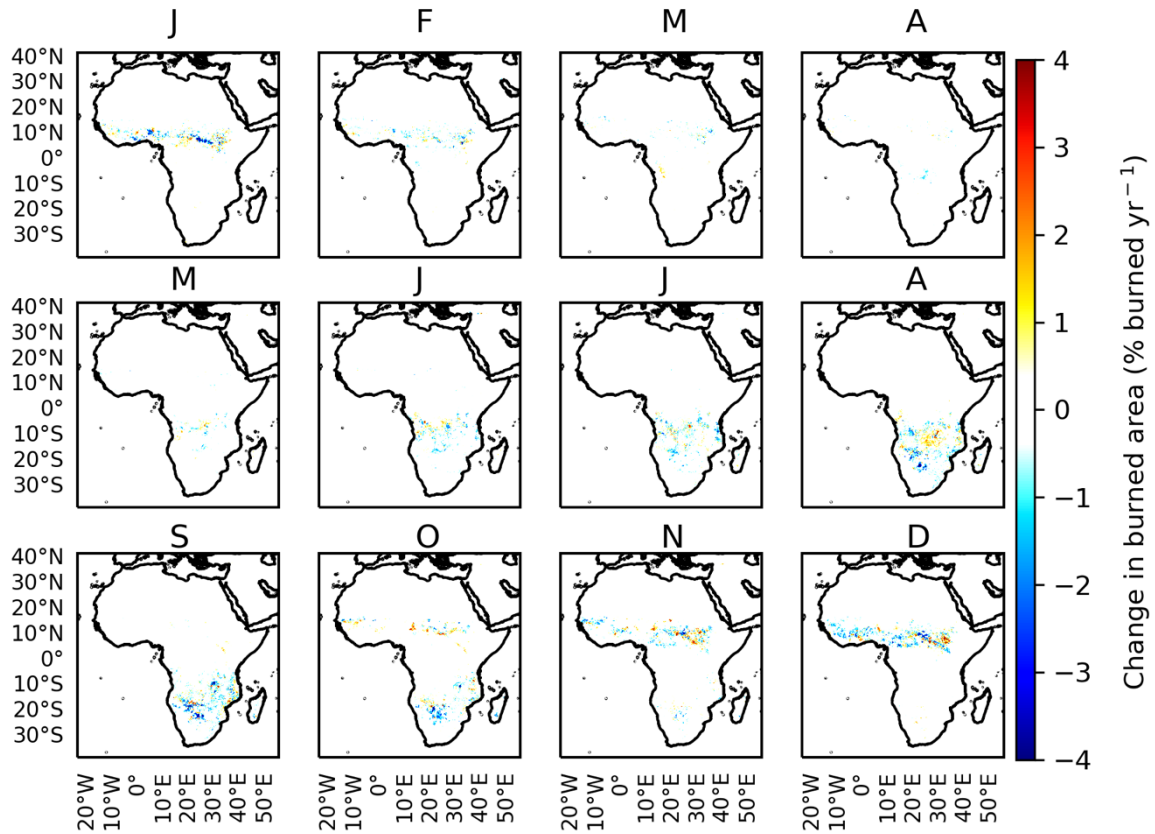
However, the annual decline in burned area and NO<sub>2</sub> VCDs is characterized by heterogeneity when considering individual months. *In West Africa, the dry season is typically November to February or March. During the transition from the dry to rainy season in February and March, both NO<sub>2</sub> VCDs and burned area exhibit increasing rather than decreasing trends in West Africa (Hickman et al., 2021).*

6. Line 278-288: Please show the trends of satellite NO<sub>2</sub> and MODIS burned area during “this transition period” to support your points.

Thank you; we now include two figures in the SI to support these arguments:



**Figure R7 (in SI as Fig. S2).** Change in mean monthly atmospheric OMI NO<sub>2</sub> VCDs for the period 2008 through 2017.



**Figure R8 (in SI as Fig. S3).** Change in mean monthly atmospheric MODIS burned area for the period 2008 through 2018.

7. Line 291-294: Again, show the trends of observed NO<sub>2</sub> VCDs to support your points.

See response to point 6.

8. Line 321-334: Why do you use such old data from the year 1998 while your target period is 2008 to 2017? All the data in this part is before the year 2008. Do you have any recent data to

support your statements? Like sectorial emission estimates from recent bottom-up inventory? What's the percentage change in fertilizer use?

The 1998 year is in error; the data we used and which are cited in the text were for 2005 through 2016, which we now specify in the text, and the citation is updated to 2019, when data were accessed. The percentage change in fertilizer use is presented in Figure 8 and varies by country, but the point that we're trying to make here is that it can be misleading to think about fertilizer use in percentage change terms in this context: a large percentage of a small number is still a small number.

9. Figure 3: I only see significant increases over central Africa and parts of west Africa in Feb and March. For most of Africa during other months, I see more decreases. What's the driver of these decreases?

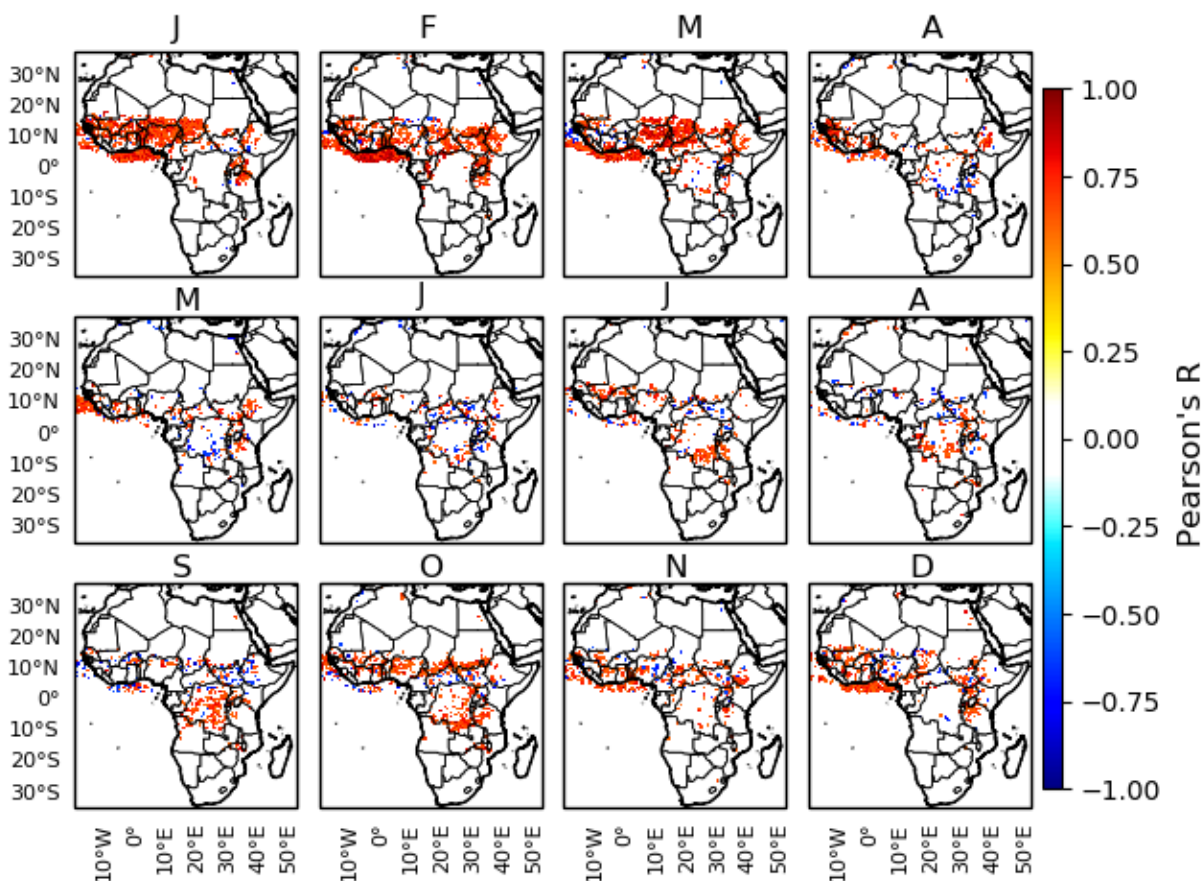
This figure has changed substantially with the shift to the v3R product, in which less area is characterized by decreases in  $\text{NH}_3$ . But it's a great question, whether in the context of the v2.2R or v3R product. The short answer is that in many cases we do not know what is driving changes, and we believe caution should be taken in trying to interpret trends during a given month: these trends are necessarily based on a smaller dataset, and do not account for interannual variation in the timing of seasonal changes in precipitation. We have worked to explain the changes in South Sudan and West Africa in the manuscript, where we were able to marshal other data to provide what we believe are convincing explanations for the trends—changes in wetland extent and in burned area, respectively. The remaining decreases occur largely either in either northern hemisphere sub-Saharan Africa or in the Equatorial Forest ecoregion. Trends in the northern grasslands do not exhibit much spatio-temporal consistency at a regional scale, and it is difficult to speculate what may be leading to them; it's possible that the changes in June, July, and August could be related to precipitation—precipitation is known to cause pulsing of nitric oxide at this time of year (Jaeglé et al., 2004), and interannual variability in precipitation could conceivably contribute to variability in emission pulsing. However, there is less evidence for this pulsing of  $\text{NH}_3$  during this time of year (Hickman et al., 2018). We can also speculate that the changes in the Equatorial forest ecoregion are related to variation in fire emissions ( $\text{NH}_3$  would be expected to be produced at higher rates in the burning of wet, woody vegetation), variation in deposition and emission to the atmosphere related to changes in leaf area index (due either to interannual variation in GPP or in phenological change), variation in soil emissions related to climate. Variation can also simply be an artifact of variation in seasonal climate: a shift of one or two weeks in seasonality could potentially result in apparent trends in a given month, and in some instances we do observe positive trends in one month and negative trends in a neighboring month, which could be indicative of this kind of seasonal shift. It is certainly possible to repeat the types of analyses that we conducted in Figure 2 to explore an anthropogenic or climatic fingerprint, but again we are wary of engaging in what may be a fishing expedition of multiple months, especially given the smaller size of the datasets underlying the trends. Although we are maintaining a focus on long term annual trends in this manuscript, we do believe that a more thorough examination of



seasonal changes in  $\text{NH}_3$  concentrations and their drivers is an interesting question for future research.

10. Figure 4: How significant are these correlation coefficients?

We have added a figure to the SI that includes only those correlation coefficients that are significant at  $p=0.05$ ; these are largely pixels with very high  $r$  values.



**Figure R9 (in SI as Fig. S5).** Correlation coefficient for the relationship between mean annual CO and  $\text{NH}_3$  VCDs over 2008 through 2018 where the relationship is significant at  $P=0.05$ . Regions where mean annual  $\text{NH}_3$  VCDs for the entire period are under  $5 \times 10^{15}$  molecules  $\text{cm}^{-2}$  are screened out.

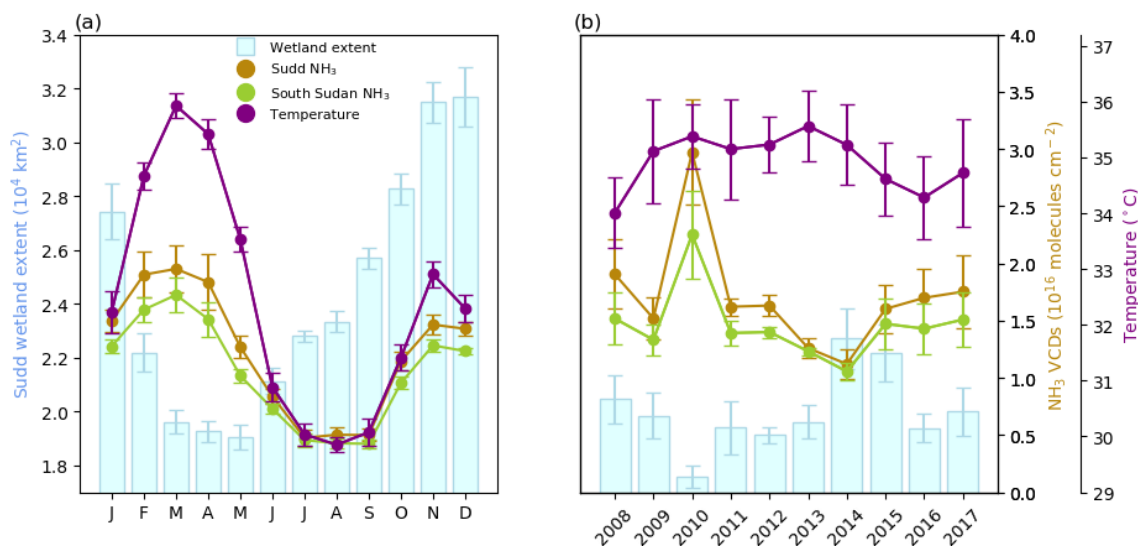
11. Figure S3: mark the Sudd box.

We have added a box marking the Sudd to Figure S3, as well as Figure S6.



12. Figure 5 and Figure 6: It's better to show the standard error of monthly mean NH<sub>3</sub> VCDs for each month or each year.

Thank you for the suggestion. We have added error bars for all variables, and also combined Figures 5 and 6 into a single figure with consistent scales across the two panels:



**Figure R10 (in manuscript as Fig. 5).** Mean (a) monthly and (b) February through May annual mean flooded extent of the Sudd, surface temperatures over South Sudan, and NH<sub>3</sub> VCDs over the Sudd and the entirety of South Sudan for the period 2008 through 2017.

13. Line 670-676: I can't find these two publications online. If they are manuscripts under review, I don't think it's appropriate to cite them here. a. Line 231: "... (Hickman et al., in review). . ." b. Line 276: "... (Hickman et al., in review). . ." c. Line 281: "... (Hickman et al., in review). . ." d. Line 285: "... (Hickman et al., in review). . ."

Thank you; we note that ACP guidelines permit authors to include manuscripts that are in preparation or in review ("Works "submitted to", "in preparation", "in review", or only available as preprint should also be included in the reference list"; <https://www.atmospheric-chemistry-and-physics.net/submission.html#references>). However, the PNAS paper has been published, and we have submitted the revised Global Biogeochemical Cycles manuscript after minor revisions, and hope it may be accepted prior to any decision on this manuscript.

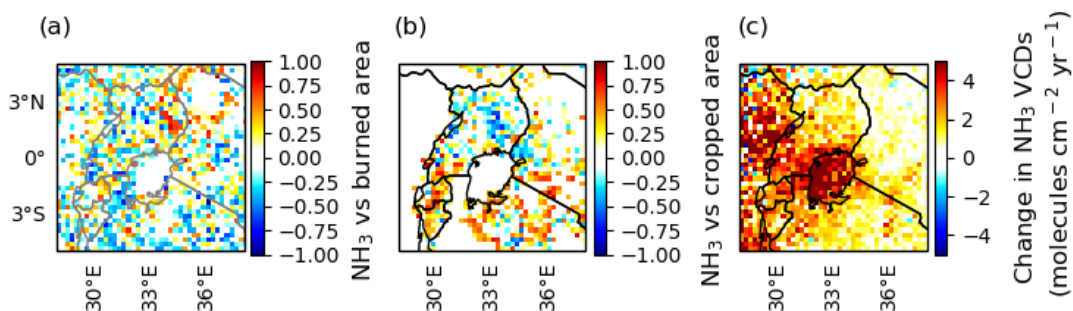
14. Line 386-407: If the civil conflict is not significantly relevant to the  $\text{NH}_3$  change in South Sudan, please concise this part. It's better to move the Sudd flood part to previous paragraph (line 354-373).

Thank you—we felt that it was important to exclude conflict as an important source of the  $\text{NH}_3$  change to strengthen the argument that the change in wetland extent was the likely driver of changes, but agree that the discussion is too extensive; we have moved the full discussion to the SI in revision. We now include three sentences on civil conflict in the main text (lines 424-430):

It is possible that conflict in South Sudan could contribute to the decline in  $\text{NH}_3$  VCDs. In 2013, a civil conflict emerged in South Sudan that was ultimately responsible for the displacement of millions of people (Global Internal Displacement Monitoring Centre, 2020; World Bank, 2019) and the disruption of livestock migration patterns (Idris, 2018). However, these disruptions appeared only after the onset of the long-term change in  $\text{NH}_3$ , and appear unlikely to make an important contribution to the observed interannual variation (SI Text, Fig. S4, S5).

15. Line 426-428: It's really hard to see the increases in the north and south of Lake Victoria Basin in Figure 3. It's better to make similar plots just for this region.

Thank you for the suggestion; we have added additional panels to Figure 6 including results of the analysis relating  $\text{NH}_3$  to changes in burned area and to changes in cropped area for the Lake Victoria Basin:



**Figure R11 (in manuscript as Fig. 6). Changes in  $\text{NH}_3$  VCDs and their relationship with burned area and cropped area over the Lake Victoria region for the 2008 through 2018 period. (a) Correlation coefficients for the relationship between  $\text{NH}_3$  VCDs and burned area. (b) Correlation coefficients for the relationship**

between  $\text{NH}_3$  VCDs and cropped area, including mosaics of crops and natural vegetation cover. (c) Changes in  $\text{NH}_3$  VCDs

16. Section 3.5: So, what's the driver of the  $\text{NH}_3$  declines across those African countries? Did you try the same analysis for temperature,  $\text{NO}_2$  and  $\text{SO}_2$  observations?

First off, in the updated analysis using the V3R product, the change in  $\text{NH}_3$  VCDs is now positive in all bins. We did conduct the same analysis for temperature and  $\text{NO}_2$ , but had neglected to evaluate  $\text{SO}_2$  concentrations, which, because  $\text{SO}_4$  can affect  $\text{NH}_3$  lifetime, are worth looking into. We include an updated version of this binned figure in the SI (Fig. S14) that includes  $\text{NO}_2$ ,  $\text{SO}_2$ , and  $\text{CO}_2$  emissions, as well as discussion of  $\text{NO}_2$ ,  $\text{SO}_2$ , and temperature. We now include the following text in the SI:

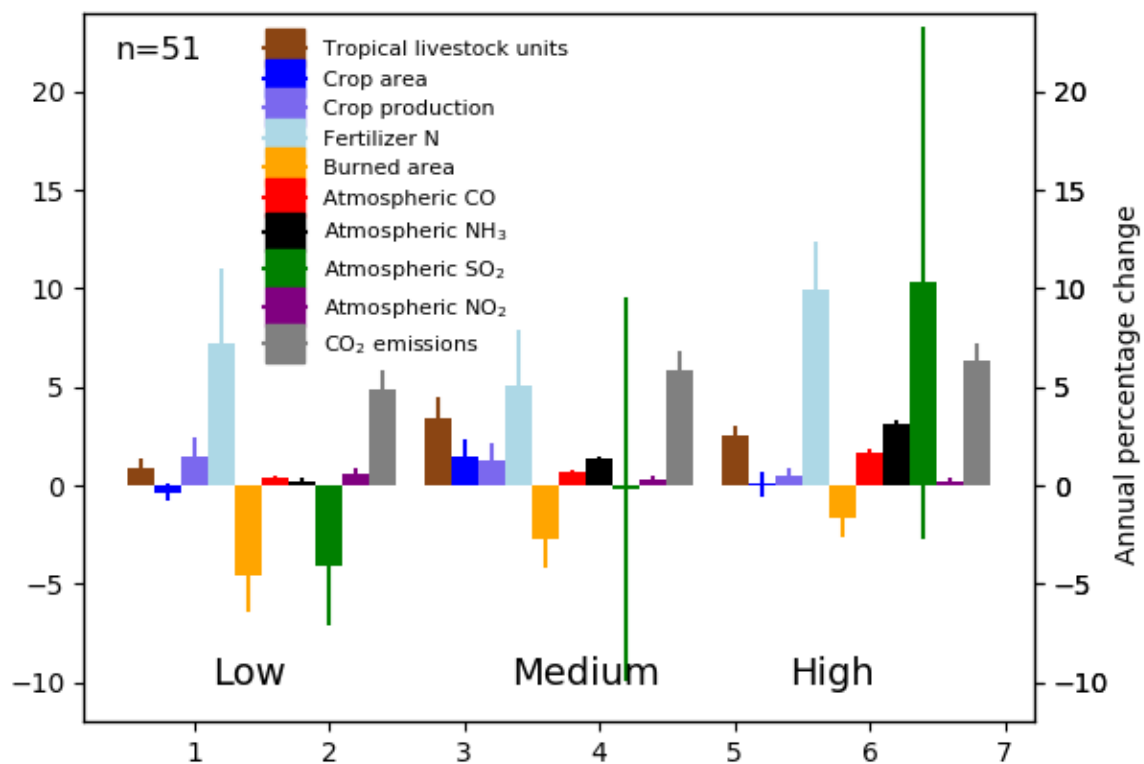
#### **Additional national-scale relationships**

**Temperature:** Mean temperature changes are small—less than  $0.005\% \text{ yr}^{-1}$  with standard errors that bracket zero; the values are too small to appear on Figure S14. They are not significantly different among bins, and do not exhibit a relationship with  $\text{NH}_3$  at the country scale in our binned analysis: the annual rate of warming is lowest in the middle bin ( $0.008 \text{ C yr}^{-1}$ ) and roughly equal in the bottom and top bins ( $0.013 \text{ C yr}^{-1}$  and  $0.017 \text{ C yr}^{-1}$ , respectively).

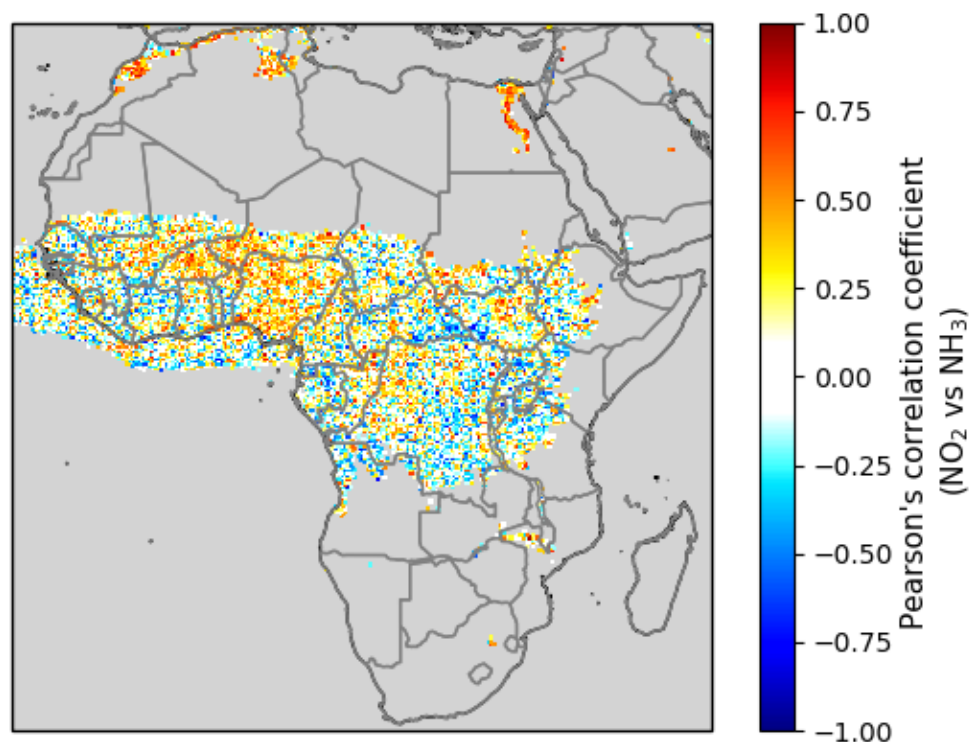
**$\text{NO}_2$ :**  $\text{NO}_2$  exhibits marginal increases in each bin, which appear to decrease across bins, but not significantly so (Fig. S14;  $p=0.63$ ). It remains possible that changes in  $\text{NO}_2$  emissions may influence  $\text{NH}_3$  trends by decreasing the lifetime of  $\text{NH}_3$  in the atmosphere, which would be evident as a negative correlation between the two species. In examining spatial relationships between the two gases there is a lot of noise, but in Nigeria, South Sudan, the Nile Region, and the North African coast, we observe positive correlations, suggestive that changes in  $\text{NH}_3$  VCDs in those regions are not the result of reactions with  $\text{NO}_2$ . Instead, the fact that trends of both gases have the same sign is consistent with changes in emissions being responsible for the trends.

**$\text{SO}_2$ :** As with  $\text{NO}_2$ ,  $\text{SO}_2$  may be expected to decrease the lifetime of  $\text{NH}_3$ . However, in our binned analysis,  $\text{SO}_2$  did not vary among bins (Fig. S14;  $p=0.98$ ). In addition,  $\text{NH}_3$  and  $\text{SO}_2$  may vary increase in parallel, suggesting that chemistry involving  $\text{SO}_2$  does not make an important contribution to the observed  $\text{NH}_3$  trends. Outside of South Africa, variation in  $\text{SO}_2$  emissions in sub-Saharan Africa are largely related to volcanic emissions. During the period of our analyses, emissions from volcanoes in eastern Democratic Republic of Congo and Eritrea play particularly important roles in interannual  $\text{SO}_2$  variability. A 2011 eruption in Eritrea results in an overall decline in  $\text{SO}_2$  concentrations in the region in our analysis. In South Africa,  $\text{SO}_2$  emissions from an array of coal fired power plants in the highveld have declined over the period of our analyses. However, the variation in  $\text{SO}_2$  VCDs is not spatially related to variation in  $\text{NH}_3$  VCDs, which are also small in the continental context.

Note that we exclude Lesotho from these analyses, which experienced a 1000% rate of increase in SO<sub>2</sub> VCDs from 2005 to 2018; including Lesotho results in a very high mean rate of SO<sub>2</sub> increase in the middle bin.



**Figure R12 (in SI as Fig. S14).** Annual percentage changes in national mean annual Tropical Livestock Units, crop area, crop yield, fertilizer N use, burned area, CO VCDs, NH<sub>3</sub> VCDs, SO<sub>2</sub> VCDs, NO<sub>2</sub> VCDs, and CO<sub>2</sub> emissions for African countries with low, medium, or high rates of NH<sub>3</sub> VCD change. Error bars represent the standard error of the mean. See Table S1 for the list of countries in each bin.



**Figure R13** (In SI as Fig. S15). Correlation between mean annual  $\text{NH}_3$  and  $\text{NO}_2$  VCDs over Africa for the period 2008 through 2018. Pixels where mean annual  $\text{NH}_3$  VCDs are below  $5 \times 10^{15}$  molecules  $\text{cm}^{-2}$  for all years are masked.

#### Anonymous Referee #2

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While long-term surface measurements of ammonia in Africa within the framework of the IDAF have been conducted for years, the spatial and temporal pattern of ammonia in Africa remains unclear due to the limitations of surface measurements. This manuscript investigated ammonia variations in Africa between 2008 and 2017 by using satellite IASI dataset. Compared with AIRS observations, this study show more detailed spatial variations with reasonable explanations, e.g., biomass burning and re- ceded wetland rather than agriculture.

Overall, this manuscript is well written and easy to follow. It will be better if additional field evidences provided, e.g., ammonia isotopic nitrogen tracing for biomass burning and ammonia field flux measurements on wetland. Other suggestions or comments are listed below.

Thank you for the comments. We agree that studies of ammonia isotopes and field flux measurements could help provide important new insights into the sources and emission dynamics of  $\text{NH}_3$ . However, such investigations are unfortunately beyond the scope of the current study.

Line 133, The FTIR observations are limited to few sites. Do we have additional observations to validate the IASI dataset across Africa, e.g., passive samplers available at the surfaces?

Thank you for the suggestion. As the reviewer implies, observations of  $\text{NH}_3$  are very limited in Africa, but there are networks of long-term passive sampling observations at the surface which have been used to evaluate IASI observations. It is important to note that these are comparisons between passive monthly surface observations and total column densities, and so are not comparing like to like, especially as the IASI  $\text{NH}_3$  product does not include an averaging kernel, and so surface column densities cannot be directly inferred from the retrievals. Nevertheless, earlier work has shown fair agreement between IASI total column densities and surface observations of  $\text{NH}_3$  across the INDAAF network in West Africa (Van Damme et al. 2015). And Hickman et al. (2018) found a general correspondence in the seasonal variation of IASI and surface  $\text{NH}_3$  observations across the INDAAF network in West Africa.

We now include these details in the revised text (lines 143-148; please note that all line numbers refer to the version of the revised manuscript \*without\* tracked changes):

Although FTIR observations are absent from Africa, earlier work has shown fair agreement between previous versions of IASI total column densities and surface observations of  $\text{NH}_3$  using passive samplers across the INDAAF network in West Africa (Van Damme et al. 2015), including in observations of seasonal variation (Hickman et al. 2018, Ossouhou et al. 2019).

Line 178, please detail what improvements have been made. Line 179, add reference for the previously published dataset. Line 192, how about other years?

The adjustments were described in the subsequent sentences; we now add the citation to Di Vittorio and Georgakakos 2018 and modify the section slightly to make this clearer (lines 194-198; new text in italics):

A few adjustments have been made to the previously published dataset (*Di Vittorio and Georgakakos, 2018*) for this study. *First*, the classification algorithm has been improved to more accurately capture the inter-annual fluctuations in the permanently flooded areas. *Second*, the dataset was extended through the end of 2017, and the total flooded area was quantified prior to applying the connectivity algorithm.



Regarding the global gridded livestock density dataset from the FAO in line 192: this dataset uses a single reference year to provide a snapshot of the spatial distribution of tropical livestock units. It is not intended to be used to evaluate inter-annual variation in livestock numbers or densities; for those analyses, we have used country-level data (e.g., Figure 8, R11)

Line 230, Some field observations found high ammonia emission at the beginning or during rainfall, likely due to intensive activities of microorganisms. Is this possible in West Africa?

We believe this is indeed possible, but expect the effect to be most pronounced in the drier ecoregions north of the coastal region with high mean annual  $\text{NH}_3$  concentrations. These drier ecoregions experience much less biomass burning, and have conditions—including relatively alkaline soils—that have been shown to favor  $\text{NH}_3$  pulses after rainfall. Indeed, earlier work from some our group found evidence of this phenomenon in the Sahel (Hickman et al. 2018). In that study, we did not observe any evidence for regionally-important pulsed  $\text{NH}_3$  emissions from coastal West Africa, and believe it is safe to conclude that biomass burning emissions are more important.

Line 321, annual average?

Good point—it is indeed the annual average, and we have revised the units to  $\text{kg ha}^{-1} \text{ yr}^{-1}$

Line 349, Can soil/mud itself emit ammonia during the period of drying?

It can: the soil would be considered the likely source of the ammonia emissions as the wetland extent decreases. We indirectly refer to the soil as the source of the  $\text{NH}_3$  emissions in describing the factors that likely contribute to the increase in emissions with soil drying in lines 391-395:

These elevated VCDs are attributed to multiple possible factors, including the effects of drying on concentrations of  $\text{NH}_3$  in solution (which increases the concentration gradient with the atmosphere), reduced biological uptake of  $\text{NH}_3$ , convective transport of dissolved  $\text{NH}_3$  from depth to the soil surface, and increased mineralization of labile organic matter (Clarisse et al., 2019).

Line 393, According to Figure S5, the ammonia concentrations show a declining trend rather than increasing?

That is correct, and apologies for any confusion. In lines 390-391 of the original manuscript, we noted that conflict-related disruptions would be expected to result in a decline in  $\text{NH}_3$ . In line 393 of the original manuscript and Figure S5, we are arguing that although we see a decline in  $\text{NH}_3$  that roughly corresponds to some areas of conflict (as seen in Figure S4), that this declining  $\text{NH}_3$  trend started earlier than the onset of civil conflict in South Sudan, suggesting that another mechanism is responsible. We argue that it is interannual variability in

the Sudd wetland extant rather than civil conflict that is primarily responsible for the observed decline in NH<sub>3</sub> VCDs in the region. Note that much of the discussion of conflict has been moved to the SI at the suggestion of Reviewer 1.

In lines 399-400 of the original manuscript, we do note that during the period of 2013-2017, when the numbers of refugees and internally displaced people increased, NH<sub>3</sub> VCDs also tended to increase—counter to what we would expect if conflict had disrupted agriculture (now in the SI under the heading “South Sudan Conflict”):

The number of refugees and internally displaced people increased substantially from 2013 through 2017, a period during which the dry season flooded extent of the Sudd decreased, and NH<sub>3</sub> VCDs increased (Fig. S5).

Line 429, Is it possible to convert the unit of nutrients to N here?

The data from the World Bank are for units of total nutrients, rather than N, which are not available from that source. We have added additional estimates of N use for Kenya and globally for additional context (lines 451-455):

Fertilizer use in the Lake Victoria region is low: national averages range from about 1 to 3 kg nutrients ha<sup>-1</sup> yr<sup>-1</sup> in Uganda (World Bank, 2019) to about 20 to 50 kg N ha<sup>-1</sup> yr<sup>-1</sup> in Kenya (Elrys et al. 2019); to put these numbers in context, Organization for Economic Cooperation and Development (OECD) countries use about 135-140 kg nutrients ha<sup>-1</sup> yr<sup>-1</sup> (World Bank, 2019).

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