The authors have updated the MS using a recent version of IASI dataset. Although the authors have addressed most of my questions in the first-iteration review, I still have some concerns here.

I think minor revision is needed.

Major comments:
1. The first thing worries me is the number of valid observations in the 0.25o grid cell (as shown by Figure R5 in your response file). Most of your targeted region have less than 7 observations per grid cell per month. Many grid cells have even less than 4 observations per month. Given that NH_3 concentration is quite sensitive to meteorology change, these small numbers of valid observations in each grid cell in each month make me doubt if they can represent the monthly NH_3 concentration levels. In addition, Figure R5 show that the mean July NH_3 error (d) is larger than the mean July NH_3 VCDs (b) over most of your targeted region. Why not try a coarser spatial resolution? Say at 0.5o?

Thank you for the suggestion—we have re-analzyed all the remotely sensed data at 0.5 degree resolution, and have updated all relevant figures using these analyses (i.e., figures including only the GFED4s burned area dataset were kept at the product's native 0.25 resolution, but nearly all figures in the manuscript have been revised). The change in resolution does reduce error and increase the number of observations in many pixels, but fairly modestly (Figures R1 and R2). The central results of the analyses have not changed, with some minor exceptions that we detail below.

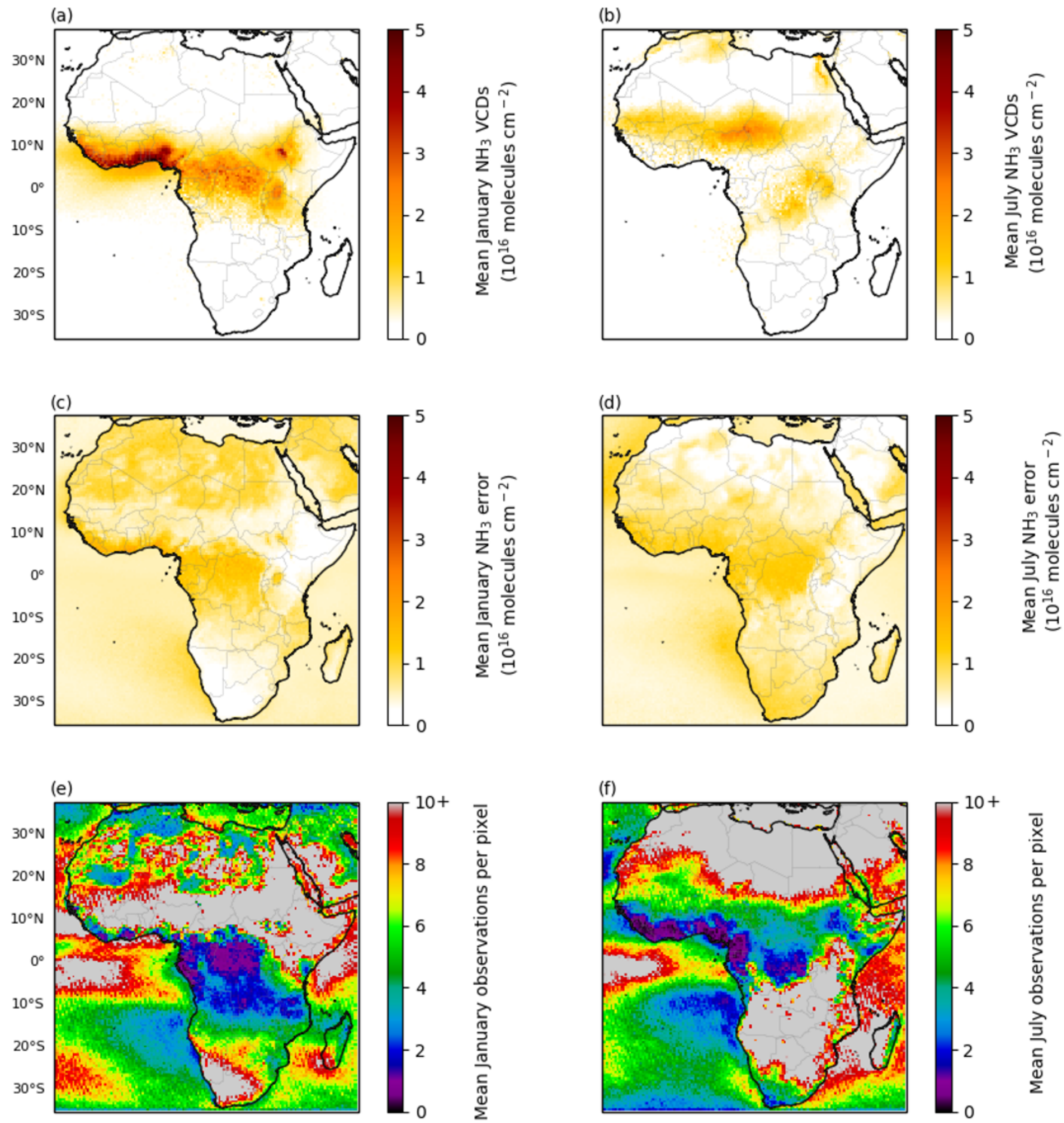

Figure R1: **Mean NH_3 VCDs, errors, and pixel counts for January and July, 2008-2018 using half degree resolution.** a) Monthly mean NH_3 VCDs for January. b) Monthly mean NH_3 VCDs for July. c) Monthly mean of the absolute error per pixel for January. d) Monthly mean of the absolute error per pixel for July. e) Monthly mean number of pixels per grid cell for January. f) Monthly mean number of pixels per grid cell for July.

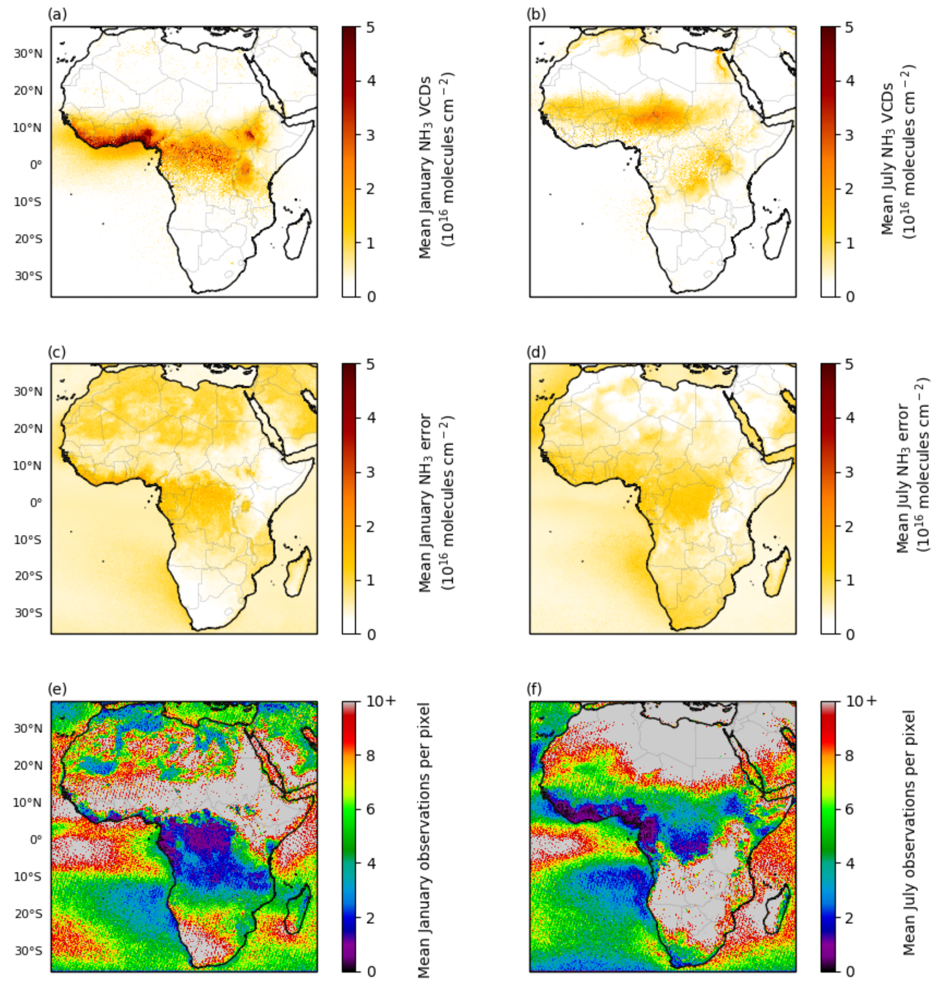

Figure R2: **Mean NH_3 VCDs, errors, and pixel counts for January and July, 2008-2018 using quarter degree resolution.** a) Monthly mean NH_3 VCDs for January. b) Monthly mean NH_3 VCDs for July. c) Monthly mean of the absolute error per pixel for January. d) Monthly mean of the absolute error per pixel for July. e) Monthly mean number of pixels per grid cell for January. f) Monthly mean number of pixels per grid cell for July.

--The main change in results is in Figure 2. The larger pixel size expands these analyses to a larger geographic area: from 1.5 x 1.5 degree areas to 3 x 3 degree areas, contributing to changes the patterns in panels A and B. The main change to the results here is that the positive relationship between population density and NH_3 trends expands to much of West Africa; we also see a little more evidence of a positive relationship between livestock density and NH_3 trends in the Sahel. We have added the following text:

Lines 342-343 (new text in bold):

> "Increases in NH_3 VCDs tend to be higher in grid cells with higher population densities in Nigeria **and other parts of West Africa** (Fig. 2b)."

Lines 423-429:

> "Our analyses do suggest that livestock may contribute to increasing NH_3 VCDs over the Sahel, from roughly 15 to 18N (Fig. 2a). However, many of these pixels are also those where population density appears to be playing a role (Figure 2b) and where correlations between NH_3 and CO VCDs are present during the transition from the dry to rainy season (Fig. S5), which may reflect a contribution from agricultural fires."

Also note that we include pixels over the Sahara and North Africa in panels A and B of Figure 2, which were masked out in the previous version of the figure. This change reveals the positive relationships between population density and livestock density in the Nile region. We have added new text discussing this relationship at lines 285-293:

> "The Nile region exhibits elevated NH_3 concentrations and a modest positive trend over the observation period (Fig. 1a, 1b). This trend appears largely to be related to agriculture and livestock: in a spatial analysis, snapshots of livestock densities and of population densities are both positively related to changes in NH_3 VCDs (Fig. 2). Although there is not a positive relationship between agricultural area and NH_3 VCDs over the Nile region from 2008 to 2018, Egypt's population increased by roughly 25% over that period (World Bank, 2019), and fertilizer N use increased by roughly 8% after a decline in use between 2004 and 2007 (FAO, 2020)."

We also corrected an error in the previous version of the figure—panel D had presented the mean cropped area, not the change in cropped area; that error is fixed in the new figure.

--We also note that there are notable changes in Figure S7 (in the NH_3 trends presented in panel B), but these changes result from the fact that the version of the figure included in the revision relied on the V2.2R IASI NH_3 product rather than the v3R product. We are grateful to have the opportunity to make this correction in this version of the manuscript, and note that the overall result remains consistent with our discussion about the Sudd wetland. These changes did not require substantial revision to the manuscript text, but we did update Figure numbers in the SI text, and made one other change to the SI text:

Original text:

"Maps of annual mean NH_3 VCDs over the IASI lifetime reveal that *a large amount* of interannual variability occurs over the Sudd (Fig. S7, S10), though there is also variability in other parts of the country, though these do not map strongly to interannual variability in precipitation (Fig. S7)."

Revised text:

"Maps of **trends and** annual mean NH_3 VCDs over the IASI lifetime reveal that **most** interannual variability occurs over the Sudd (Fig. S7, S10)."

--Lastly, we see a large change in the SO_2 values in Figure S14. This change is the result of having Liberia—which had the next-to-lowest increase in NH_3 VCDs in the 0.25 degree analysis--moving from the high bin to the middle bin in the 0.5 degree analysis. Liberia experienced unusually high increases in SO_2 over the observed period, and has a large influence on the mean SO_2 trend for each bin. The changes in the bin composition also changed the p values and r values in our statistical tests, in one case changing a result from not significant to significant at p=0.1 (the difference between the high and bottom bins for change in tropical livestock units).

2. Section 3.4 Lake Victoria Basin region: While you claim that the increases of NH3 VCDs over this region are caused by the increasing cropped area around the lake, both the hot spots of NH3 VCDs (Figure S11) and the largest increase of NH3 VCDs (Figure 7 (c)) were found over the lake, not over the cropped areas to the north or to the south of the lake. In addition, Figure 7 (b) shows that the cropped area to the north of the lake and the NH3 VCDs have negative correlation. Can you explain why?

Great questions. The phenomenon of increasing NH3 VCDs over the lake is also present in the analysis of Van Damme et al. 2020, but the lake has been masked out in studies using satellite NH_3 products from AIRS and CRiS (Dammers et al., 2019; Warner et al., 2017). The conditions over the lake (emissivity, thermal contrast, etc.) are quite different from the surrounding area and we expect this results in retrieval biases which cause some of the mean differences between the lake itself and the surrounding land surface. When it comes to trends over the lake, we expected that this is the consequence of changes in transport from the surrounding land surface. To examine this hypothesis, we analyzed the mean monthly and mean annual NH_3 VCDs over the lake itself and over the surrounding land surface using a pair of masks (Figures S18, reproduced here as Figure R3)

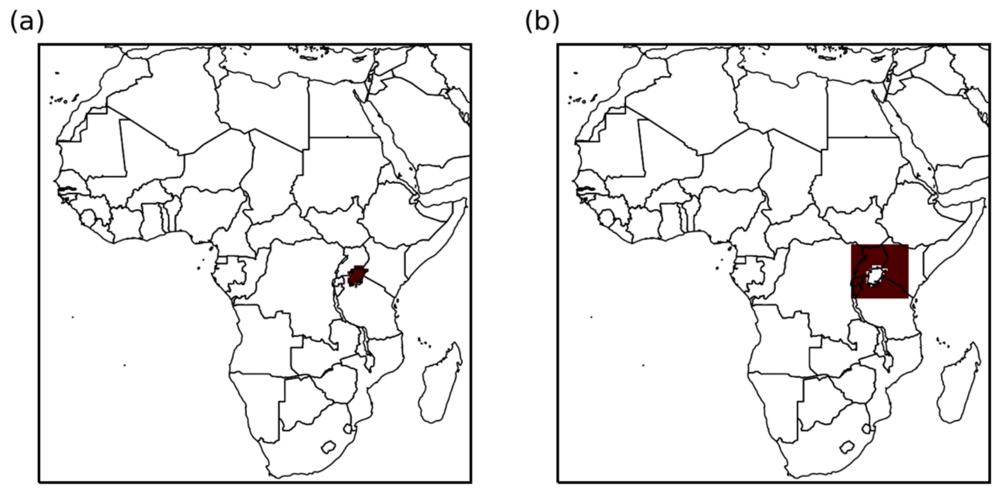

Figure R3 (S18): Masks used in time series analysis of NH3 over Lake Victoria (a) and over the surrounding land surface (b).

Both the monthly time series and the annual time series show close correspondence between NH_3 over the lake and over the surrounding land surface (Figure S17, reproduced here as figure R4). There are some minor differences in variability between the two time series, but a) that is not surprising given that the mask of the surrounding land area is integrating a much larger area spanning both hemispheres and different seasonal dynamics and b) the overall patterns remain extremely similar.

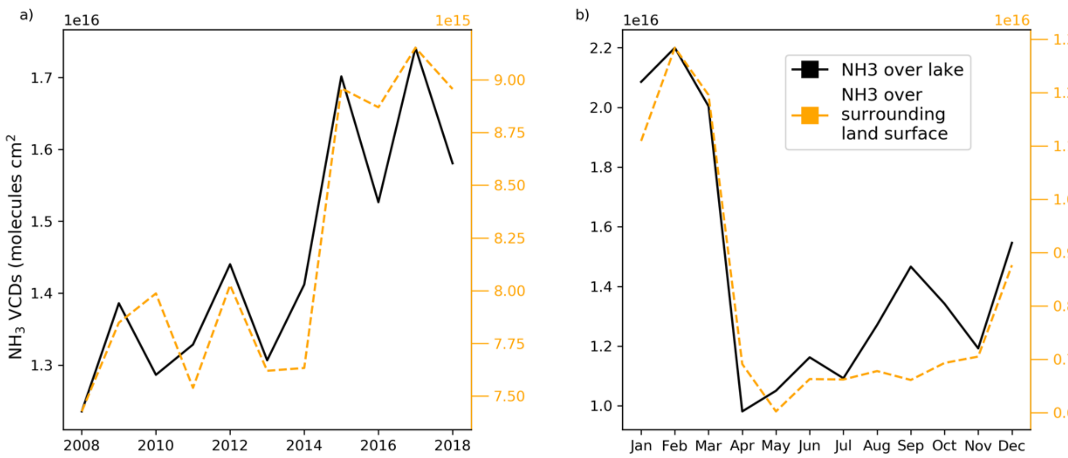

Figure R4 (S17): Mean annual (a) and mean monthly (b) NH_3 VCDs over Lake Victoria and the surrounding land area, 2008-2018.

We have added the following text to acknowledge this point (lines 531-537):

> "We do note that there is an apparent increase in NH_3 VCDs over the lake itself. It is important to note that differences in conditions over the lake and adjacent land cover—e.g., emissivity, thermal contrast, etc.—contribute to substantial differences in mean retrieved NH_3 VCDs over the lake relative to the surrounding land surface. Both monthly and interannual variation in NH_3 VCDs over Lake Victoria correspond closely to variation in NH_3 VCDs over the surrounding land surface (Figure S17, S18), suggesting that the trend over the lake results from transport of NH_3 emitted from the surrounding land surface."

We believe the negative correlation north of the lake is more straightforward. Smallholder farmers in Uganda have notably low fertilizer use rates--about 2-3 kg N ha^{-1} (Masso et al. 2017), which is two orders of magnitude lower than fertilizer use in North America, Europe, or Asia, and an order of magnitude lower than in neighboring countries such as Kenya. As a result, studies suggest that smallholder farmers in Uganda are experiencing negative nutrient balances (Cobo et al. 2010 and references therein)—more N is removed from soils by harvested biomass than added as fertilizer. Under these conditions, expansion of cropland would contribute to reductions in substrate for ammonia volatilization, leading to a negative correlation between cropland and NH_3 VCDs. We have added the following text (lines 527-530):

> "We note that there is a negative correlation between cropland area and NH_3 VCDs in Uganda, north of Lake Victoria (Figure 7b). We expect this is a consequence of the

extremely low fertilizer use in Uganda (Masso et al. 2017), which leads to depletion of soil N—and thus substrate for ammonia volatilization—over time (Cobo et al. 2010)."