**Response to Review 1:**

We thank the reviewer for providing further helpful comments and constructive suggestions, which improve the quality of the manuscript. Please see our responses to the comments as follows.

*The authors have done a great job addressing the comments raised by both reviewers, including revising the methodology. I think the paper has greatly improved. I have a few (mostly) minor comments for the authors to consider before publications.*

*a)*

*Eq. (4)*

*I think there is a mistake. Shouldn't it be (Tmax+Tmin)/2 ?*

**Reply:** We thank the reviewer for pointing out the mistake. We have revised the equation.

*b)*

*line 145. This is an interesting approach, how do the estimated Tb/PHU compared to tabulated values (e.g., Table 1 from Bondeau (2007)).*

**Reply:** We adopted Tb and PHU to estimate the planting, green-up, and harvesting dates for unavailable years. However, the crop progress in our study differs from the crop phenological development defined in the crop model from Williams et al. (1989) and Bondeau et al. (2007). We determine the Tb and PHU based on the days at which crop development were 5%, 15%, 85%, and 95% completed within a state. Whereas the Tb and PHU in Table 1 from Bondeau et al. (2007) are estimated based on the sowing and maturity dates chosen by the farmer under essentially climatic constrains. Therefore, the “planting date” in our study is usually later than the actual sowing date and the “growing season” is shorter than the actual phenological cycle. As a result, the Tb estimated in our study is higher than that in Williams et al. (1989) and Bondeau et al. (2007), while the PHU is smaller. For example, the Tb of corn of Iowa we estimate are 13.7 $^{o}$C, 14.3 $^{o}$C, 16.8 $^{o}$C, and 17.5 $^{o}$C at 5%, 15%, 85%, and 95% completion respectively. The PHU are 1139, 1164, 1201, and 1201 correspondingly. While the Tb and PHU of corn are 8 $^{o}$C and 1670 in Williams et al. (1989), and falls into 5-15 $^{o}$C and 1600 in Bondeau et al. (2007).

*c)*

*line 364: Is the reference for this statement Cao et al. (2018)?*

**Reply:** Yes, it is. We have added the citation for this statement.

*d)*

*This one is probably the most important.*

*Section 2.3: I am still having trouble with this. While maps may not be available for each month, observations certainly are (NADP website). I encourage the authors to look at changes in NH4+ concentration (to limit the impact of precipitation changes) instead.*

*The correlation could be done by aggregating observations within each large regions (as in Fig. 2).*

*I think you mean Fig. 10?*

**Reply:** We appreciate the reviewer’s suggestion. Using $NH_4^+$ concentration significantly improves the association between $NH_3$ emission and $NH_4^+$ deposition and thus strengthens our conclusion.

Based on monthly site monitoring $NH_4^+$ concentration data, we generate the atmospheric $NH_4^+$ concentration maps using Inverse Distance Weighting interpolation method. The relationships between $NH_3$ emission and $NH_4^+$ concentration at each pixel over the past 31 years are examined seasonally and yearly. The correlation coefficient maps from these two time scales show small difference and we put the map using annual data in the discussion section and include the map using spring data as a supplementary figure. We revised the method in section **2.3 Correlation between $NH_3$ emission and deposited $NH_4^+$ concentration** and the discussion in section **4.4 Effects of increasing $NH_3$ emissions on $NH_4^+$ deposition.**

The figure number has been revised to 10 at **line 382**.

**Line 209 to 216**

**2.3 Correlation between NH3 emission and deposited $NH_4^+$ concentration**

We obtained monthly site monitoring data of precipitation $NH_4^+$concentration for the period 1985-2015 in the North America from the National Atmospheric Deposition Program (http://nadp.slh.wisc.edu/data/NTN/ntnAllsites.aspx). After aggregating the monthly data to spring (March-June) and full year at each site respectively, we generated the atmospheric $NH_4^+$ concentration maps using the Inverse Distance Weighting interpolation method and resampled the maps to 1 km resolution to make it comparable to our estimated $NH_3$ emission maps. The associations between fertilizer-induced $NH_3$ emission and $NH_4^+$ concentration in precipitation during 1985-2015 at each grid cell were examined using Pearson correlation coefficients with statistical significance at $p < 0.01$ and $p < 0.001$.

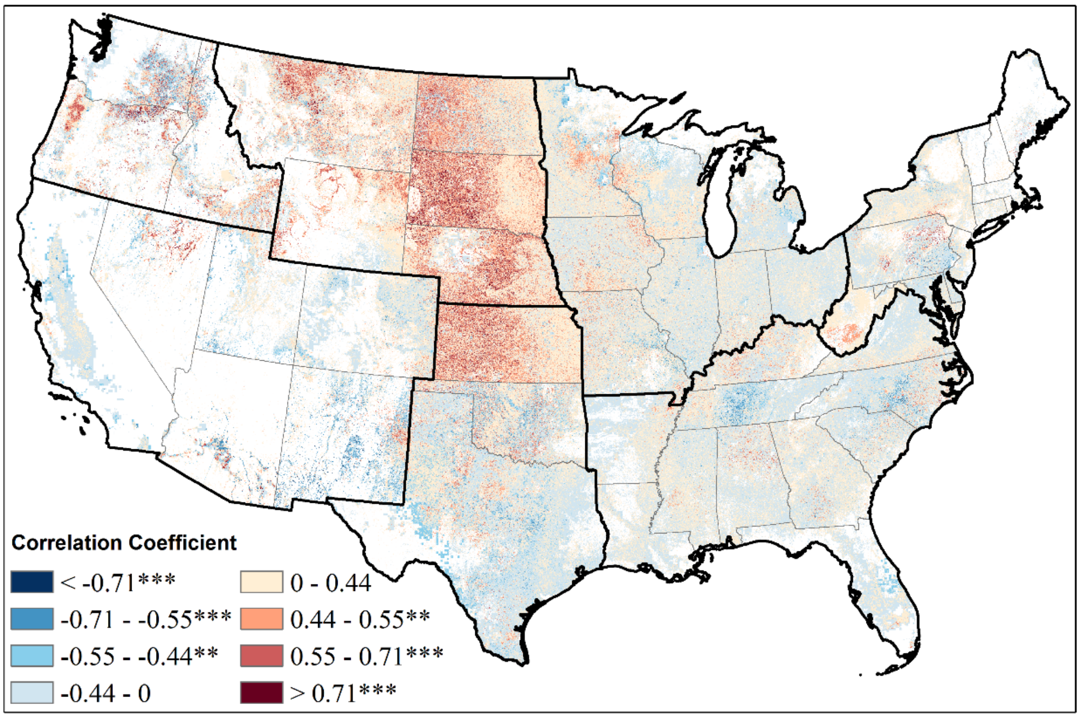
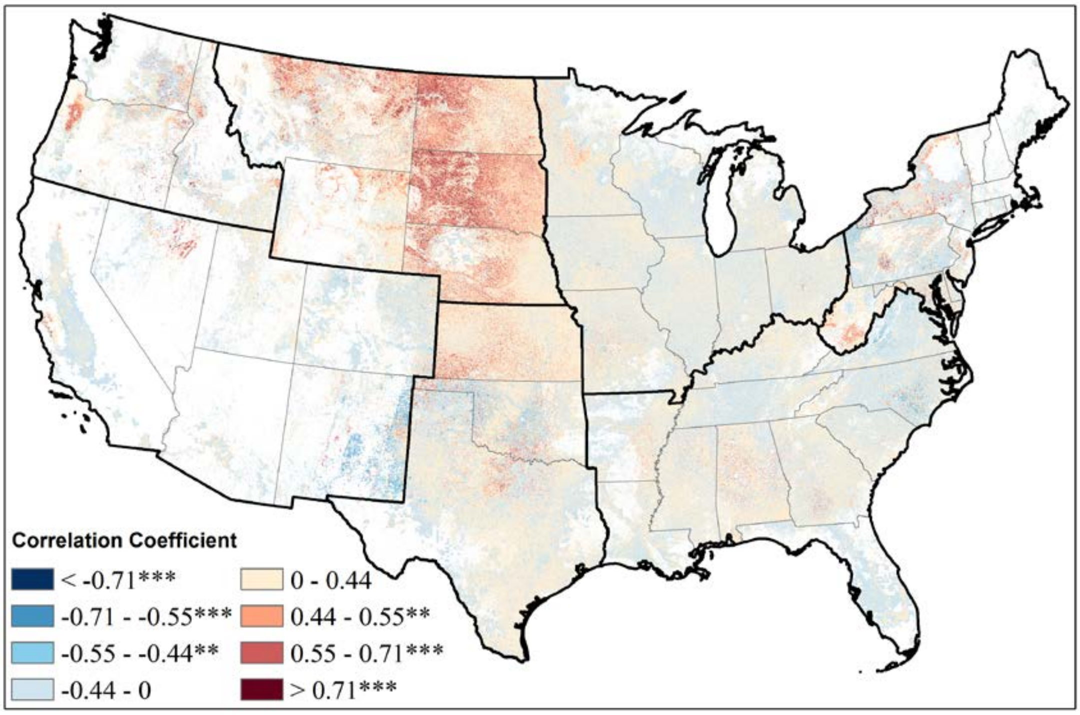



Figure 10. Correlation coefficient between annual $NH_3$ emission from N fertilizer uses and annual $NH_4^+$ concentration in precipitation between 1985 and 2015. The correlation coefficient was calculated between the two time series at each 1km × 1km grid cell. ** refers to P-value < 0.01, *** refers to P-value < 0.001.

**Appendix**

**10 Correlation coefficient between spring $NH_3$ emission and deposited $NH_4^+$ concentration**

The majority of N fertilizer is applied around planting date in spring, resulting in a peak of $NH_3$ emission from March to June in the contiguous U.S. Therefore, we examined the relationship between fertilizer-induced $NH_3$ emission and deposited $NH_4^+$ concentration during this period using Pearson correlation coefficient at each 1 km pixel over 1985-2015 (Fig. S6). The spatial pattern is very similar to the correlation coefficient between the fertilizer-induced $NH_3$ emission and deposited $NH_4^+$ concentration at an annual time scale. It implies the annual-scale relationship is mainly driven by the spring season.




Supplement Figure 6. Correlation coefficient between spring (March-June) $NH_3$ emission from N fertilizer uses and $NH_4^+$ concentration in precipitation between 1985 and 2015. The correlation coefficient was calculated between the two time series at each 1km × 1km grid cell. ** refers to P-value < 0.01, *** refers to P-value < 0.001.

*e)*

*line 375: "Although the intensive NH4+ in wet deposition concentrated in the central U.S." I am not sure what this means.*

**Reply:** We agree with the reviewer that the expression here is not clear. We have rephrased the sentence as follow. **Although the central U.S. is the hotspot of $NH_4^+$ deposition, the largest increase in wet $NH_4^+$ deposition was found in the northern Great Plains and Minnesota from 1985 to 2015 (Du et al., 2014; Li et al., 2016)**