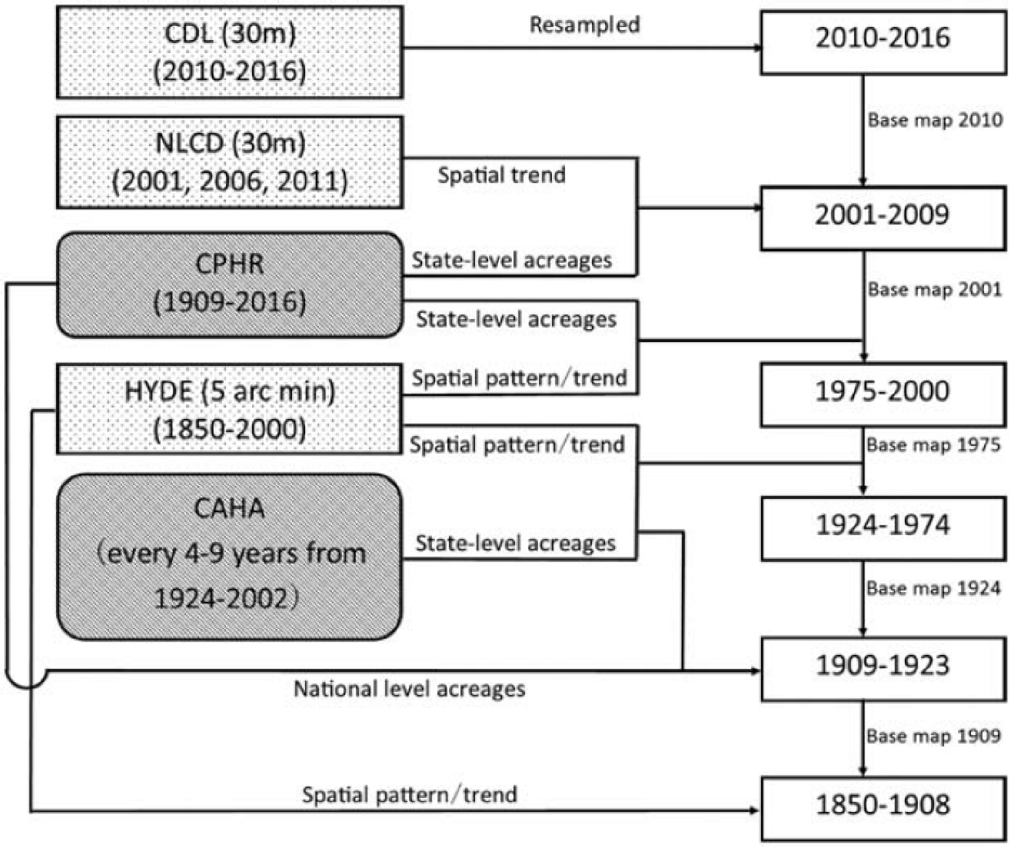
Response to Review 2:

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

The manuscript by Cao et al. estimates NH3 emissions from fertilizer in the US over the past century. By tracking different types of fertilizer and crops, they identify variability in the spatial distribution of fertilizer emissions and emissions factors. Their results are consistent with previously noted studies of shifting spatial distribution in NH4 deposition, for example, but provide additional valuable levels of detail. My main suggestion would be to provide some quantitative assessments of uncertainty, which I think may constitute minor revisions, as they have at least qualitatively identified the key sources of uncertainty. This and a few other minor comments are included below.

Comments: 57-58: It's not clear to me what land and fertilizer use is being referred to here as distinct from the studies cited in the preceding lines.
107-119: I realize this lies somewhat outside the present paper and is likely within the work of Yu 2018, but could the authors briefly comment on how such spatial resolution was known for these distributions prior to the satellite era? Here they mention how satellites were used to determine spatial reconstructions but do not comment on any other method, which presumably would be necessary for the first half of the century, nor how such different methods have been harmonized into a single consistent dataset.

Reply: To reconstruct the spatially explicit cropland distribution maps that go back to 1900, we harmonized multiple state- and national-level inventory data and remote sensing products in different periods. USDA-CDL and NLCD provide the detailed spatial distribution of crop information and are directly resampled for the reconstruction of cropland maps during the recent decade. Another satellite-based database HYDE cropland maps, which were developed by assimilating both inventory and satellite data, was used to reconstruct the spatial maps before 2000 by depicting the potential distribution of agricultural land. Meanwhile, adjusted state- and national-level crop-specific land acreage from USDA survey data was used to limit the acreage of each crop for each state on maps. We think incorporating a detailed description of the methodology of cropland distribution map reconstruction is irrelevant to this study and therefore gave a brief summary and referred to the article that elaborates the cropland maps reconstruction process.



Yu, Z, Lu, C. Historical cropland expansion and abandonment in the continental U.S. during 1850 to 2016. Global Ecol Biogeogr. 2018; 27: 322–333. https://doi.org/10.1111/geb.12697

Fig 1 (a): I think it would be clearer to refer to this as loss "from" N fertilizer, not loss "to" fertilizer, since the process being described here is NH3 from fertilizer to the atmosphere, correct?

Reply: We agree with the reviewer and we have corrected it.

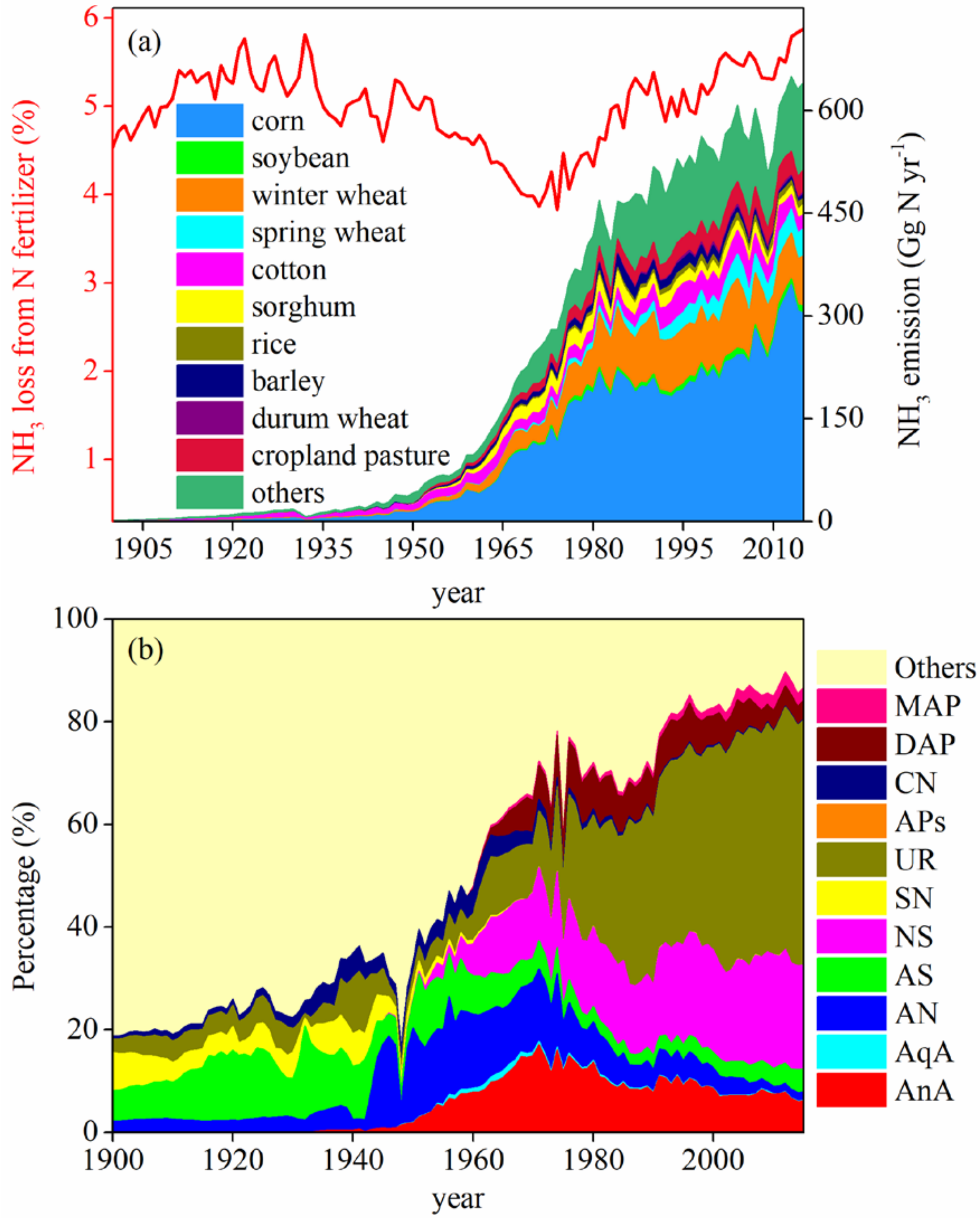


Figure 1. Contributions of major crop types and N fertilizer types to historical NH_3 emissions since 1900. (a) Crop specific NH_3 emissions, (b) Relative contributions of 12 major N fertilizer types to annual total NH_3 emission. Solid line in (a) refers to the NH_3 loss percentage to total N fertilizer input.

173: Here emission growth just refers to fertilizer emission, right? Not emission from livestock, which is the larger component of total emissions.

Reply: We thank the reviewer for pointing out the vague expression here. Our study focused specifically on NH_3 emission from synthetic fertilizer. We added the clarification to address that NH_3 emission in this study refers to N fertilizer-induced NH_3 emission.

Line 62 to 65.

Based on spatially explicit time-series of cropland distribution maps and N fertilizer management database, we adopted empirical modeling of EF to calculate monthly NH_3 emissions from synthetic N fertilizer uses (Hereafter, NH_3 emission refers to the synthetic N fertilizer-induced NH_3 emission unless specified otherwise) in the contiguous U.S. at a resolution of 1 km × 1 km from 1900 to 2015.

Fig 2: What drives the drop in RF from the 40s to 50s in NW, NGP and SW?

Reply: We agree with the reviewer's comment. The widespread popularity of Anhydrous ammonia and Ammonium nitrate occupied the share of Urea in these regions, which largely reduced the ratio of NH_3 emission lost from fertilizer. This phenomenon can be found in five out of seven regions. We revised the sentence in **4.2 Spatiotemporal change in the NH_3 emissions** to include the region into discussion.

Line 317 to 318.

The "V" shape of historical national and regional NH_3 emission factors mainly resulted from the changing preference in using different N fertilizer types (Cao et al., 2018).

197: What do the authors mean by "reportedly" here?

Reply: We have deleted the word.