Response to Review 2:

We thank the reviewer for providing the insightful comments and constructive suggestions, which greatly improve the quality of the manuscript. Please see our responses and related changes (highlighted in red) as below.

I appreciate that the editor has offered this albeit late opportunity to comment. The dataset of historical nitrogen fertilizer use in agricultural ecosystem across the Continental United States itself is already quite important, not to mention its importance to assess the key biogeochemical processes and water quality. I do understand that there is no perfect way to generate such dataset. So I think this dataset and paper is worth to publish in this journal. But there are some information needs to be clarified before its acceptance and publication.

Line 87-90. For different datasets at national level, is there any gap among those datasets or do they have smooth connection among different datasets. For Mehring et al., 1957, do they provide annual data for each year?

Reply: In this study, we developed three national data, including national N consumption, national average crop-specific N use rate, and national consumption of 11 N fertilizer types for different purpose. They are all same source data but cover different periods. Therefore, they are smoothly connected and have no gaps between different datasets.

Mehring et al. (1957), USDA (1971), and USDA-ERS (2013) were used to develop the national N consumption. All three datasets provide annual data.

Five datasets were used to develop the national average crop-specific N use rate. Each dataset provides the average fertilizer use rate of each crop for a certain year or period (Supplementary table S3). They are all sourced from USDA survey but with slightly different features or spanning period. For the period of 1965-2015, the nitrogen fertilizer use survey was periodically conducted by each state based on their own schedules. In addition, the survey schedules also varied among crop types within the state. Collectively, there are many gaps with irregular length in the century-long N fertilizer use data, and most of these gaps are less than three years. In addition, Mehring et al. (1957) reported the sum of multiple fertilizer uses (N, P, K, trace nutrients) for each crop, instead of N fertilizer use, in the year 1927, 1938, 1842, and 1946. They provided data of both N fertilizer and total fertilizer use in 1950. Therefore, the crop-specific N fertilizer use in other years have been reconstructed based on the ratio of N input to the sum of multiple fertilizer uses in 1950.

Four datasets were used to reconstruct the national consumption of 11 N fertilizer types, they provide annual value for different period and are smoothly connected (Supplementary table S6).

Supplementary table S3. Data sources for national average crop-specific N use rate

Data sources	Period	Crop types	Data form
Mehring et al. (1957)	1927, 1938, 1942, 1946, 1950	7[a]	Total fertilizer consumption
USDA (1957)	1954	10[b]	N use rate
Ibach et al., (1964)	1959	10[b]	N use rate
Ibach and Adams. (1967)	1964	10[b]	N use rate
USDA-ERS (2013) and -NASS (2017)	1965-2015	9[c]	N use rate

The number of crop types surveyed in different sources varied. [a], crop types included corn, soybeans, wheat (spring wheat, winter wheat, and durum wheat in total), cotton, sorghum, rice, barley, and cropland pasture. [b], crops included all nine major crops and cropland pasture. [c], crops only included nine major crops. Total fertilizer uses of each crop type reported in Mehring et al. (1957) contained all N, P, K, and trace fertilizers.

Supplementary table S6. Data sources used to reconstruct historical consumption record of 11 N fertilizers across nation

Fertilizers	AnA	AqA	AN	AS	NS	SN	Urea	CN	DAP	MAP	APs
1900-1953									\	\	
1954-1959								\	\	\	
1960-2003											
2004-2011											

Red: Mehring et al. (1957), green: USDA (1966), blue: USDA-ERS (2013), yellow: FAO (2017). AnA: Anhydrous Ammonia. AqA: Aqua Ammonia. AN: Ammonium Nitrate. AS: Ammonium Sulfate. NS: Nitrogen solution. SN: Sodium Nitrate. CN: Calcium Nitrate. DAP: Diammonium Phosphate. MAP: Monoammonium Phosphate. APs is the integration of Ammonium Phosphates, before 1960, the consumption of DP and MP were relatively small, so these two fertilizers were incorporated in APs, after 1960, however, the consumption of these two fertilizers increased to very high amount and were reported separately.

References:

FAO (Food and Agriculture Organization of the United Nations): FAO online database, available at: http://www.fao.org/faostat/en/#data/RF, last access: 19 October 2017, 2017.

Ibach, D. B. and Adams, J. R.: Fertilizer Use in the United States by Crops and Areas, 1964 Estimates, USDA-Economic Research Service and Statistical Reporting Service, Statistical Bulletin No. 408, Washington, D. C., 1967.

Ibach, D. B., Adams, J. R., and Fox, E. I.: Commercial Fertilizer Used on Crops and Pasture in the United States, 1959 Estimates, USDA-Economic Research Service and Agricultural Research Service, Statistical Bulletin No. 348, Washington, D. C., 1964.

Mehring, A. L., Adams, J. R., and Jacob, K. D.: Statistics on Fertilizers and Liming Materials in the United States, USDA-Agricultural Research Service, Statistical Bulletin No. 191, Washington, DC, 1957.

USDA-ERS (U.S. Department of Agriculture-Economic Research Service): Fertilizer Use and Price, available at: https://www.ers.usda.gov/data-products/arms-farm-financial-and-crop-production-practices/, last access: 19 November 2017, 2013.

USDA (U.S. Department of Agriculture): Fertilizer Used on Crops and Pasture in the United States, 1954 Estimates, USDA-Agricultural Research Service, Statistical Bulletin No. 216, Washington, D. C., 1957.

For the crop pasture N consumption, did the author assume the N were evenly distributed?

In addition, I was a little suspicious about using the ratio for the year 1964 to cover entire period during 1945-2015. At least, the authors should discuss the related uncertainties raised.

Reply: Thanks to the reviewer for raising this insightful question and we made improvements in our data. In our study before improvements, we inferred the N fertilizer use for cropland pasture of 1945-2015 based on state-level percentage of cropland pasture N consumption in state total. We used the percentage of the year 1954 for 1945-1959 and the year 1964 for 1960-2015. So the N use in cropland pasture were not evenly distributed among states.

We made improvements of replacing the fixed state-level ratio of cropland pasture in 1954 and 1964 with dynamic annual state-level ratio from 1945 to 2015.

We made the modifications in the **2.1 Historical N fertilizer use rate reconstruction, Estimation crop-specific N use rates at state-level** of the manuscript**,** and **10 pencentage of N fertilizer use in nonfarm, permanent pasture, and cropland pasture to national N fertilizer use** in the supplementary file.

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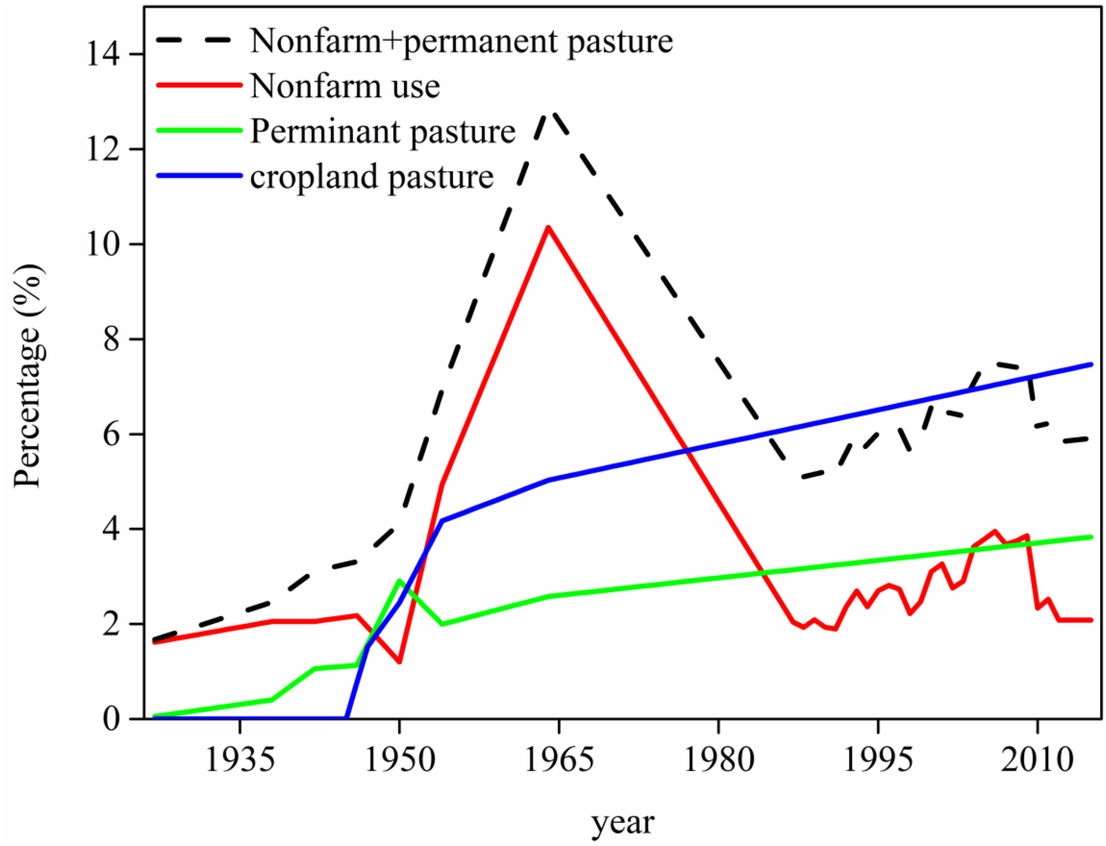
Page 5, line 143. We assumed cropland pasture was not fertilized until 1945 due to a lack of area data and low N use (< 1.5% of national total N use in 1942, Mehring et al., 1957). By timing the annual state-level N consumption with the ratio of cropland pasture N consumption to total N consumption in each state of the same year derived from multiple data sources (see Supplementary table S2 for details), we obtained state-level N consumption of cropland pasture from 1945 to 2015.

Supplementary:

10 pencentage of N fertilizer use in nonfarm, permanent pasture, and cropland pasture to national N fertilizer use

To exclude N fertilizer use in nonfarm and permanent pasture from total N fertilizer use as non-agricultural fertilizer use and introduce the N fertilizer use in cropland pasture (supplementary table S2), we integated the state-level nonfarm, permanent pasture, and cropland pasutre N fertilizer use proportion based on Mehring et al. (1957) for 1927, 1938, 1942, 1946, and 1950, USDA (1957) for 1954, Ibach et al. (1964) for 1959, Ibach and Adams (1967) for 1964, Brakebill and Grinberg (2017) for 1987-2012 (nonfarm use), and IFA (2018) for 2015 (permanent pasture and cropland pasture). According to the newest data set published in IFA (2018), permanent pasture and cropland pasture in the U.S. together accounted for 11.3% of total N fertilzier consumption. We used the individual ratio of these two pastures of 1964 to split 11.3% to 3.8% for permanent pasture and 7.5% for cropland pasture in 2015. Thus, we calculated the increase rates of these two pastures from 1964 to 2015 and increased state-level percentage with the same rate. We assumed the state-level ratio before 1927 kept the same as the ratio of 1927 and adopted linear interpolation to gap-fill the missing years from 1927 to 2015. Supplementary figure S1 shows how the national percentage of N fertilizer use in nonfarm (red line), permanent pasture (green line), cropland pasture (blue line), and nonfarm and permanent pasture together (black dashed line) changed through 1927 to 2015.



Supplementary Figure S1: Percentage of N fertilizer use in nonfarm, permanent pasture, and cropland pasture (classification can be found in Table S2 of Supplementary Information) in the U.S. from 1927 to 2015. Red line represents nonfarm, green line is permanent pasture, blue line is cropland pasture, and black dashed line is sum of nonfarm and permanent pasture.

Regarding the temporal change of nitrogen fertilizer consumption, what is the reason for the peak and big drop during 1980s and near 2010?

Reply: Nitrogen fertilizer use can be affected by many factors, such as land use change, agricultural management (crop rotation, fertilizer type change, etc.), technological development (genetic improvement, etc.), economic benefit (labor cost, fertilizer price, crop price, etc.), and local policies. We extended the text in **4.2 Temporal and Spatial change in nitrogen fertilizer use** of manuscript to explain such abrupt changes.

Page 10, line 310. together with the markedly increase in application rate of all crops except cotton, the major agricultural regions received tremendous amount of N during

1970 to 1985, except the conspicuous drop in the year 1983 due to the large cropland area abandon (Yu and Lu, 2017). Driven by the change of grain demand and fertilizer prices, N fertilizer use in the U.S. gradually increased with more fluctuation after 1985, such as the drop of fertilizer consumption in 2008-2009, which may be suppressed by the high price of N fertilizer caused by the 2008 financial crisis (USDA-ERS, 2013).

References:

Yu, Z. and Lu, C. Q.: Historical cropland expansion and abandonment in the continental U.S. during 1850 to 2015, Glob. Ecol. Biogeogr., 27, 322-333, 2017, DOI: 10.1111/geb.12697.

USDA-ERS (U.S. Department of Agriculture-Economic Research Service): Fertilizer Use and Price, available at: https://www.ers.usda.gov/data-products/arms-farm-financial-and-crop-production-practices/, last access: 19 November 2017, 2013.

There are many approximations required to produce the nitrogen fertilizer maps, and then to allocate the Nfer to different crops at different time period. The authors may need to discuss these in greater detail than you do in the "uncertainties" section.

Reply: We agree with the reviewer that there are unavoidable uncertainties in data processing and development. Our N fertilizer data include three final products, i.e., maps of annual N fertilizer use rate in the continental US at 1-km resolution from 1850-2015 (by total rate, at four timings, by application of NH_4^+-N and NO_3^--N at four timings).

We have added the description about the historical land cover dataset developed by Yu and Lu, (2017) to the **2.4 Spatializing state-level crop-specific N fertilizer input to gridded maps**, and relevant uncertainties derived from this into **4.3 Uncertainty and future research needs** of **4 Discussion** in the manuscript.

Page 7, line 197. For spatial analysis, we downscaled the imputed state-level crop-specific N management data to gridded maps based on 1 km × 1 km historical land cover data (including crop density and crop type distribution maps) of the contiguous U.S. from 1850 to 2015 developed by Yu and Lu (2017) and Yu et al. (2018). The cropland density maps, by incorporating various sources of inventory data and high spatial resolution satellite images, were reconstructed to represent the area of cropped land each year while excluding summer idle/fallow. The crop type maps were reconstructed using satellite images and the USDA National Agricultural Statistics Service (NASS) survey data, and state-level land area of each crop type in each year is consistent with USDA survey. More details about cropland maps can be found in Yu and Lu (2017) and Yu et al. (2018).

Page 12, line 362. 6) The historical crop type maps were reconstructed using USDA survey data at state-level. However, spatial distribution of N fertilizer use was uncertain in sub-state level because the lack of finer scale data for crop type map reconstruction.

References:

Yu, Z. and Lu, C. Q.: Historical cropland expansion and abandonment in the continental U.S. during 1850 to 2015, Glob. Ecol. Biogeogr., 27, 322-333, 2017, DOI: 10.1111/geb.12697.

Yu, Z., Lu, C., Cao, P., and Tian, H.: Long-term terrestrial carbon dynamics in the Midwestern United States during 1850-2015: Roles of land use and cover change and agricultural management, Glob. Change Biol., 1-18, 2018, DOI: 10.1111/gcb.14074.