Response to reviewer comments

We thank the reviewer for the precious and constructive suggestions to improve our manuscript. We have addressed all the comments raised by the reviewer. Please find our point-by-point response below.

Reviewer 1:

This study, entitled ”Historical nitrogen fertilizers use in China from 1952 to 2018”, tried to develop a new detailed map of nitrogen fertilizer inputs in China. It is within the scope of the ESSD. It contains some new perspectives (crop-specific, crop rotation, etc.) and is basically worthy of publication, but it requires for some corrections before the publication. Some of the descriptions regarding the process of making the data set are unclear. Major comments are below on the points that need to be corrected.

Response: We thank the reviewer for valuing our work! The suggestions are very helpful for improving the manuscript. Below we have made the corrections as per his suggestions.

General comments

1. First of all, thank you very much for citing Nishina et al. (2017) (Nishina map from here). However, please note that the Nishina map is slightly misinterpreted in your comparison. As it uses LUHa (Hurtt et al., 2011) as a map of agricultural land, so it does not consider N inputs where LUHa has 0% agricultural land area. This means that there are at least some areas with zero at all in Nishina map. Therefore, Figure 6 is incorrect. In addition, Nishina map consider the double cropping region, which is based on the crop use intensity (CUI) map provided by Siebert et al. (2010), In the double cropping region, it is needed to be twice the annual input. Otherwise, the FAO input cannot be reproduced. Please refer to Figure 1 in Nishina et al. (2017), which illustrates where we treated as two-season crop areas (map for double cropping areas in Nishina map can also be shared). As for the annual nitrogen input per halfdegree grid cell, if you add up the individual fertilizer input data for NH4+ and NO3- by month, and then aggregate that data for each year. After this, the N input per unit area of cropland can be calculated by dividing by the area of farmland in

LUHa. I think you can create a comparable map.

Nevertheless, this issue has nothing to do with the quality of your dataset.

Response: We thank the reviewer for pointing out the issues in Figure 6. We have updated the Figure 6 with total N use annually aggregated from the monthly $NH4^+$ and $NO3^-$ input data. Besides, we also derived the N input per unit area of the cropland using the approach suggested. Specifically, we downloaded LUHa data from https://luh.umd.edu/luh_data/. The data (LUHa.v1) covers the period of 1500 to 2005, and the years 1961, 1980, 1990, and 2005 (the most recent year is 2005 in LUHa.v1 data) were used for calculating the nitrogen fertilizer use rate per unit cropland area (see Figure 6). If the updated version of LUH data is more appropriate (e.g. LUH2), we are also happy to further update the calculations.

Besides, we also used the Nishina et al. (2017) data for the comparison of nitrogen fertilizer use rate at per square meter of cropland in 2010. Please also see the updated comparisons in Figure 7.

The updated Figure 6:

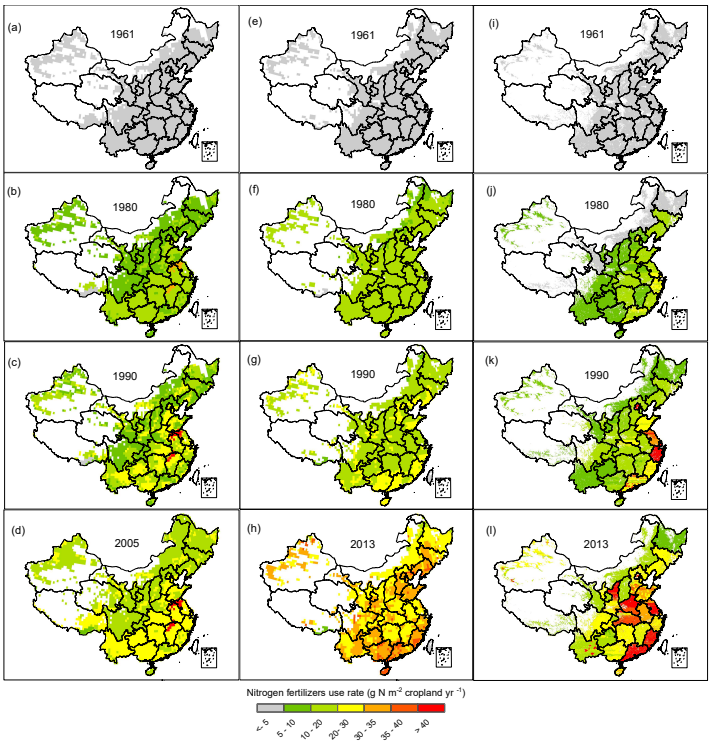

Figure 6. Distribution of nitrogen fertilizers use (a-c) in 1961, (d-f) in 1980, (g-i) in 1990, and (j-l) in 2005 or 2013 (left column: Nishina et al. (2017)'s data; central column: Lu et

al. (2017)'s data; right column: this study; the values indicate N fertilizers use rates per square meter cropland of each grid-cell).

The updated Figure 7:

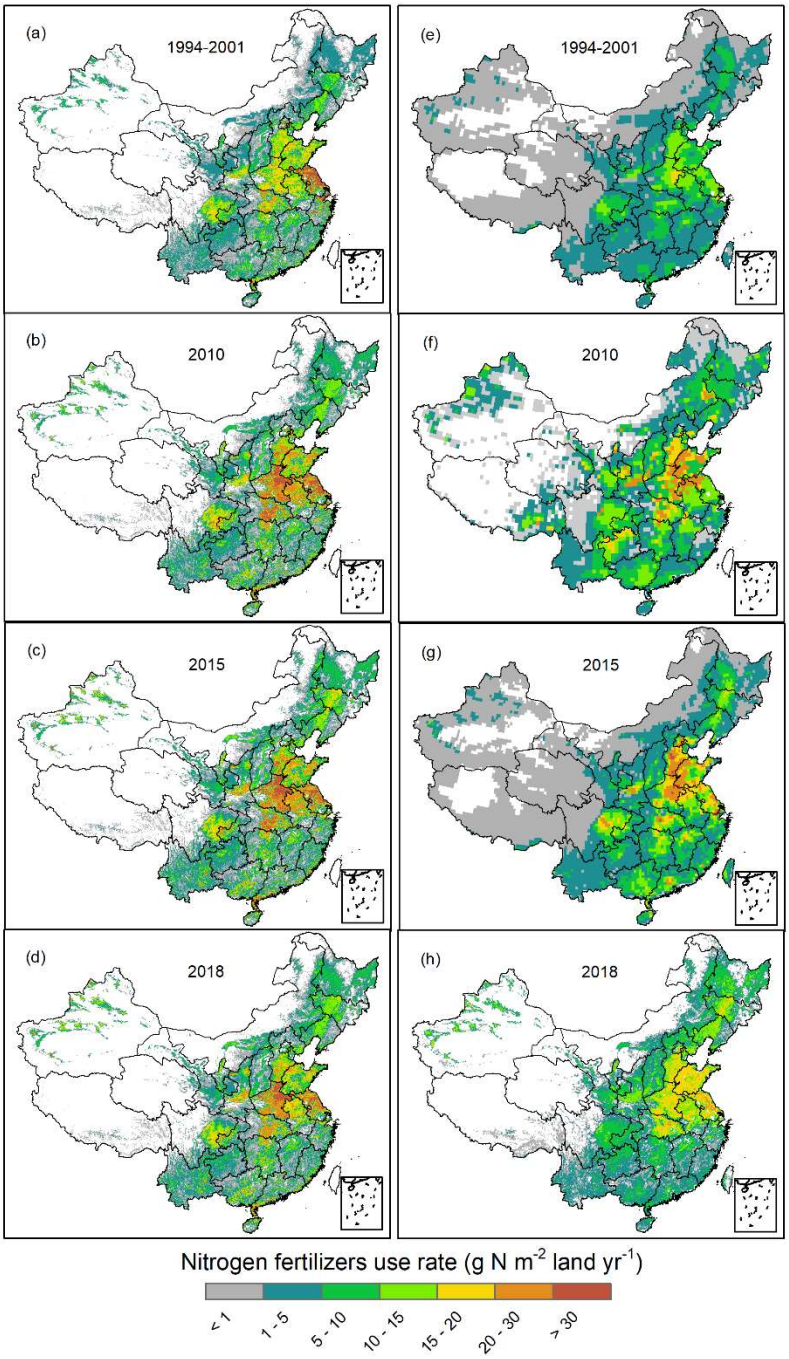

Figure 7. Comparisons of the nitrogen fertilizers use in different studies (the most recent year with both data from this study and other study available was used in comparing; panels a-d: this study: panels e-h: data from Potter et al. (2010), Nishina et al. (2017), Houlton et al. (2019), and Tian et al (2022); the value indicates N fertilizers use rates per square meter of land).

2. As a matter of fact, even if you use Global maps for comparison, there are better maps available for China. One is under review, but I can refer you to two papers by Wang et al. (2020) or Tian et al (under review in ESSD). Please consider comparing with this one.

Response: We thank the reviewer for the suggestion. We agree and have added the map comparisons with Tian et al (2022)'s data as suggested (please see changes made in Figure 7). We also contacted Wang et al. (2020) asking for the data. The comparisons will then be made by adding their global maps data once it is available.

3. The method of Gap-filling is not clear. Especially when Gap-filling is applied to space, the total amount of nitrogen fertilization may be larger than the statistics used, depending on the method.

Response: We thank the reviewer for pointing out this. We realize that our descriptions about gap-filling (i.e., temporal gap-filling and spatial gap-filling) were misleading. In this study, temporal gap-filling and spatial gap-filling were both performed at rebuilding the provincial, crop-specific N fertilizer use table. Specifically, the table describes the N fertilizer use rate for each of the 10 major crop types in each province from 1952 to 2018. We first allocated the N fertilizer use rates obtained from the Cost-benefit Report of the National Agricultural Products (CBR) reports. There were many missing data for certain years and crop types. If a crop type in a province had N fertilizer use rate reported in the CBR, gap-filling of the missing years were treated as "temporal gap-filling". If a crop type in a province had N fertilizer use rate never reported, then we treated the gap-filling as "spatial gap-filling" (because the N use rate was derived from national average or nearby provinces). Specifically, the "spatial gap-filling" was a process to allocate N use rates for crops planted in a province (areas are usually very small) but their N fertilizer use were never reported in the province (usually minor crops for the province). Therefore, both gap-fillings were done in the table, and the gap-filled N use rates were further adjusted to maintain the total N input close to national statistics.

We realized these descriptions might be confusing. To avoid misunderstanding, we have rephrased this part by abandoning the use of the terms "temporal gap-filling" and "spatial gap-filling". Here is the revised text:

"The CBR data provides officially released fertilizers use information summarized from thousands of samples collected in each province in China. First, we created an empty table to record the N fertilizer use rate for each province with all the 10 types included. Second, the N use rate of the table was allocated using data obtained from CBR when available. Third, if a crop type was never planted in the province, the N use rate was set to 0. Fourth, we checked and gap-filled the missing N fertilizers use rates in the province. For crop type with N use rate intermittently reported, we linearly interpolated the rate using the two nearest data reported before and after the year (see equation 2). While for crop type been planted in the province but its N fertilizer use were never reported, two fertilizers use scenarios were considered."

4. On a related point, each and every procedure should be formulated in mathematical formulas (even with respect to simple tabulations). Everything is described by text, making it difficult to see the validity of the method. The same applies to the description of Gap-filling. As other example, I could not see how to calculate and define uncertainty (shown in Fig 5) in the current manuscript.

Response: We thank the reviewer for the suggestion. We agree and have added formulations for describing the method. Specifically, for the uncertainty analysis, the it was derived from the eight scenarios considered. To make it clearer, we have added a table to clarify further. **Since we have been advised not to submit the revised manuscript as supplement, we only attached the pages we made the corresponding revisions here. Please check our revisions and the table we added in the end of this letter.**

5. Please add units to all drawings. Indeed, different units are mixed (e.g., "g N per unit land per year", "N fertilizers use rate per square meter of cropland ") in the text. So, it is not clear at first glance what you are referring to in the figure.

Response: We thank the reviewer for the suggestion. We have added units to all figures as suggested. Please check our revisions in Figures 6&7 before and Figure 5 below:

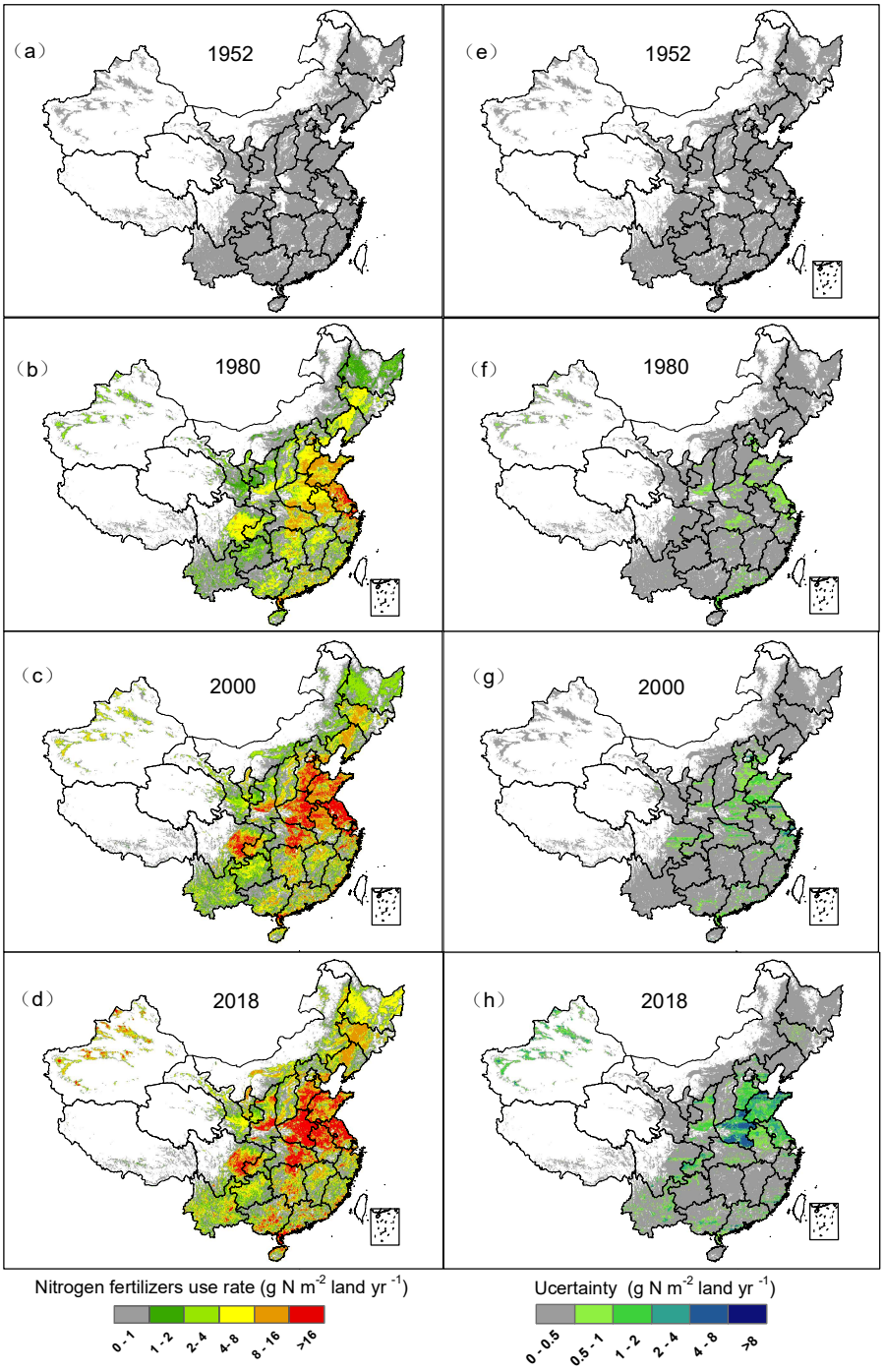

Figure 5. Spatial distribution of the rates (a-d) and the uncertainties (e-h) of nitrogen fertilizers use during different periods in China (the four panels from the top to the bottom indicate the rates (left) and the uncertainties (right) in 1952, 1980, 2000, and 2018, respectively; the value in the scale bar indicates the N fertilizers use rate per square meter of land).

Revisions made in response to suggestion 4:

urgent need for a long-term, spatial explicit N fertilizers dataset to serve the quantification of national and global GHG budgets, and to benefit data analyses and the environment protection including reduced water pollutions and improved land-based ecosystem functions.

In this study, we have reconstructed the annual N fertilizers use in China's cropland using various statistical records, reports, and gridded images at 5 km × 5 km resolution covering the period of 1952 to 2018. We aimed to: 1) develop a continuous dataset depicting the N fertilizers use in cropland in China; and 2) examine the historical distributions and shifts of N fertilizers use cropland in China. Our focus is on the chemical, rather than the organic N fertilizers use, and the term 'N fertilizers' we have adopted here is to refer exclusively to synthetic N fertilizers.

2 Data and methods

2.1 Reconstruction of the national and provincial N fertilizers use

This study focuses on N fertilizers use in the mainland China, while Taiwan, Hong Kong, and Macaw were excluded due to no data availability. The dataset we have adopted here for synthetic N fertilizers use in cropland of China was from N-only fertilizers, and N-mixed fertilizers or 'compound fertilizers' (see equation 1).

$$N_{tot} = N_{only} + N_{mix} \qquad (1)$$

where N_{tot} indicates the total N use in China's cropland, N_{only} indicates the N fertilizer use from N-only fertilizer, and N_{mix} indicates N-mixed fertilizer use. We have adopted two different phrase and units to differentiate the N fertilizers use at national level i.e. 'N fertilizers input' (Tg N per year), and crop field level i.e. 'N fertilizers use rate' (g N per unit land per year).

We first reconstructed the total N fertilizers input in China's cropland at national level. The national fertilizer inputs were provided by both FAO (https://www.fao.org/faostat) and the National Bureau of Statistics of China (Chinese Statistical Yearbook (CSY); also available from https://data.stats.gov.cn). Specifically, the FAO data provides the total N input in China from 1960 to 2018. While in comparison, the CSY data describes total fertilizers use (including N, phosphate, potassium fertilizers such as ammonium phosphate) covering the period of 1952 to 2018, and the N-only fertilizers (e.g. ammonia, ammonium nitrate) from 1987 to 2018. The national total N fertilizer input of the period 1960 to 2018 was directly obtained from the FAO, while the ratio of N fertilizers to the total fertilizers was used to derive the total N fertilizers use from 1952 to 1959.

Second, we compiled the N fertilizers used in each province in China. For the period 1987-2018, N fertilizers use was directly derived from the CSY database. For the period 1952 to 1986 when the provincial data was unavailable, the reconstructed national N fertilizers use was allocated to each province based on the provincial N proportions derived in 1987.

2.2 Reconstruction of the crop-specific N fertilizers use rate in each province

We examined the major crops planted in China and grouped them into 10 types, including early rice, mid-season rice, late rice, wheat, corn, soybean, oil seeds, cotton, vegetable, and other crops. Specifically, other crops include barley, sorghum, sugarcane, tobacco, fruits (e.g. apple, pear, citrus). The N fertilizers use rate for each major crop types (except other crops) was intermittently reported in the Cost-benefit Report of the National Agricultural Products (CBR) covering the period of 2004-2018 (Table 1) (CBR data can be obtained from: https://data.cnki.net/trade/Yearbook/Single/N2021120200?zcode=Z009). The CBR data provides officially released fertilizers use information summarized from thousands of samples collected in each province in China. First, we created an empty table to record the N fertilizer use rate for each province with all the 10 types included. Second, the N use rate of the table was allocated using data obtained from CBR when available. Third, if a crop type was never planted in the province, the N use rate was set to 0. Fourth, we checked and gap-filled t~~Both temporal and spatial gap-filling approaches were implemented for the period of 2004-2018. Temporal gap-filling was applied in province with N fertilizers use rates intermittently reported in few specific years. To do so, t~~he missing N fertilizers use rates in the province. For crop type with N use rate intermittently reported, we ~~were~~ linearly interpolated the rate using the two nearest data reported before and after the year (see equation 2). ~~Spatial gap-filling was applied in province with~~While for crop type been planted in the province but its N fertilizer use were never reported ~~for the crop type(s)~~, ~~in which~~ two fertilizers use scenarios were considered. For the first scenario, we assumed that the N fertilizers use rate of the crop in the province was the same as the average rate at national level (see equation 3). While for the second scenario, we assumed that the N fertilizers use rate of the crop in the province was the average of the rates adopted in nearby provinces (see equation 4).

$$Rate_i = (Rate_j - Rate_k) \times \frac{i-j}{k-i} \quad (2)$$

$$Rate_{p,i} = (\sum_{q=1}^{n} Rate_{q,i})/n \quad (3)$$

$$Rate_{p,i} = (\sum_{q=1}^{m} Rate_{q,i})/m \quad (4)$$

where $Rate_i$ indicate the N fertilizer use rate of a crop type in year i, while j and k indicate the nearest data reported in years j and k representing the years before and after the year i. $Rate_{p,i}$ indicate the N fertilizer use rate of a crop type in province p in year i, q indicates province q with the rate available in CBR, n indicate total number of province with N fertilizer rates available in CBR, and m indicate the total number of nearby province with N fertilizer rates available in CBR.

For the period of 1981 to 2004, the N fertilizers use rates were calculated from the N inputs and the planted areas of each crop type in each province. While for the period of 1952 to 1980 when provincial, crop type-specific N fertilizers use data were unavailable, we proportionally adjusted the N fertilizers use rate of each crop type based on the ratio of N fertilizers use in the year and the amount used in 1981 (see equation 5). This is assuming that the change of N fertilizers use rate of a crop is proportional to the change of the total N fertilizers use in the province.

$$Rate_{p,i} = Rate_{p,1981} \times N_{tot,p,i}/N_{tot,p,1981} \quad (5)$$

where $Rate_{p,1981}$ indicate the N fertilizer use rate of a crop type in province p in 1981, $N_{tot,p,i}$ indicate the total N fertilizer input for province p in in year i, and $N_{tot,p,1981}$ indicate the total N fertilizer input for province p in in year 1981.

As pointed out in the section 2.1, the total N input includes two components, i.e. N from compound fertilizer and N-only fertilizer. Unfortunately, CSY database documented the use of compound fertilizers in major crop types but the N ratio was not specified. Since the N ratio varied between 16%-33.33% in compound fertilizers according to the major fertilizers used in China (http://fgw.kaifeng.gov.cn/info/2107), we assumed two extreme scenarios that the N ratio were either 16% or 33.33% for compound fertilizer being applied to each major crops.

In addition, N fertilizers use in vegetables were also highly uncertain. Specifically, various vegetables were planted in China, while the N fertilizers use rates were missing for the most of the vegetables. However, the vegetables received much higher N fertilizers (71.9 g N m^{-2}) than non-vegetable crops according to the former published study (Huang et al., 2017). In general, the total fertilizers applied in vegetable was about 3.3 times higher than the recommended application rate (Huang et al., 2017). Therefore, two additional scenarios were considered in reconstruction of the N fertilizers use in vegetables. In the first scenario, we assumed that the N fertilizers use rate in vegetable was the same as the average of all other major crop types. In the second scenario, we assumed the N fertilizers use rate in vegetable was 3.3 times of the average rate of other major crop types. Therefore, we considered three uncertainty sources in this study (Table 2).

After the gap-filling of the nine of the ten major crop types (except other crops), the total N inputs were calculated by multiplying the rates and the areas of each crop type in each province. The provincial residue N inputs (the difference of the total N inputs calculated, and the total N inputs derived from the FAO and the CSY) were allocated to other crops and the N fertilizer rates were calculated by dividing the residue N inputs to the planting areas obtained from the China Agricultural Yearbook (Table 1).

$$Other\_Rate_{p,i} = [N_{tot,i} - \sum_{t=1}^{9}\sum_{q=1}^{n}(Rate_{q,i,t} \times Area_{q,i,t})] / Area_{p,i} \quad (6)$$

where $Other\_Rate_{p,i}$ indicate the N fertilizer use rate of other crops in province p in year i, $N_{tot,i}$ indicates the total N input in year i, $Rate_{q,i,t}$ indicate the N fertilizer use rate of crop type t in province q in year i, $Area_{q,i,t}$ indicate the planted area of crop type t in province q in year i, and $Area_{p,i}$ indicate the planted area of other crops in province p in in year i.

Table 1. Datasets used for nitrogen (N) fertilizers use reconstruction.

Datasets	Year	Resolution	Variable	Sources
Cropland distribution maps	1900–2016	Annual, 100m-5km	Cropland distribution	(Yu et al., 2021)
China Agricultural Yearbook (CAY)	1980–2018	Annual, provincial	Planted areas of each major crops in each province	National Bureau of Statistics of China

China Statistical Yearbook (CSY)	1952–2018 1987–2018	Annual, provincial	Total fertilizer N fertilizer	National Bureau of Statistics of China
Cost-benefit Report (CBR)	2004-2018	Intermittently, provincial	N fertilizers use by crop types in each province	National Development and Reform Commission of China (Price Department)
FAO N fertilizer	1960-2018	Annual, national	Total nitrogen fertilizers use in China	(FAOSTAT database, 2018)
Rotation maps	1980, 1990, 2000, 2002, 2011	County-level	Crop rotation information	(Liu et al., 2018)

Table 2 The eight scenarios considered in reconstructing nitrogen fertilizer use in China

Uncertainty sources		Scenarios							
		1	2	3	4	5	6	7	8
1	Gap-filling approach	Nation*	Nation	Nation	Nation	Nearby**	Nearby	Nearby	Nearby
2	N ratio in compound fertilizer	16%	16%	33.33%	33.33%	16%	16%	33.33%	33.33%
3	Vegetable N fertilizer use	Ave***	Ave3.3****	Ave	Ave3.3	Ave	Ave3.3	Ave	Ave3.3

*: the N fertilizer rate of missing crop type(s) was derived from national average; **: the N fertilizer rate of missing crop type(s) was derived from nearby province; ***: the vegetable N fertilizer use rate was assumed to be the average of other nine major crops; ****: the vegetable N fertilizer use rate was assumed to be 3.3 time of the average rate of other nine major crops.

2.3 Approach for spatializing N fertilizers use

Before the N fertilizers use rate could be allocated spatially, a crop type map is required. Here, we reconstructed crop rotation maps from 1952 to 2018 using the model we previously developed (Yu and Lu, 2018; Yu et al., 2019) (see Figure S1 for the details). Reconstruction of annual crop rotation map can be divided into two periods, namely the periods before and after 1980. For the period of 1980-2018, county-level crop rotation maps in 1980, 1990, 2000, 2002, and 2011 were used (Table 1) (Liu et al., 2018). More specifically, when allocating a crop type in a province for a year from 1980, the cropland grid-cell located in a county was given priority to be assigned to the crop type identified from the nearest-year rotation map. Due to the lack of data, the crop rotation map in 1980 was used for the period before 1980.

Based on the 100-m crop rotation maps developed in the previous step, we link the crop type and the N fertilizers use rate developed for each crop types in each province. Specifically, we spatialized the N fertilizers use rate for each year using the 100-m crop type maps. For grids with multiple crops cultivated in a year, the fertilizers use rate was the total N fertilizers use of all crops (i.e., total N fertilizer applied in a grid-cell in a year). The 100-m resolution N fertilizers use rate maps were then resampled to 5 km × 5 km for a comparison and analyses. In this study, we considered three uncertainty sources and the

uncertainties of the N fertilizer maps were derived from the eight scenarios listed in Table 2. The methodology flowchart is showed in Figure 1.

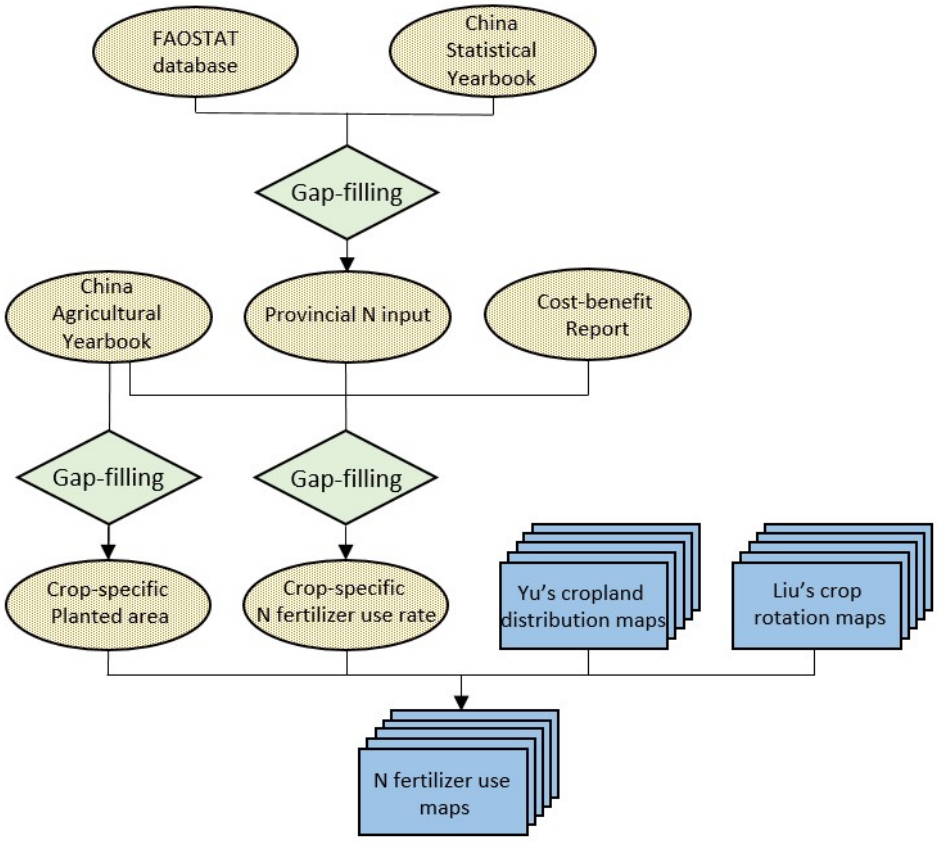

Figure 1. Methodology flowchart of nitrogen (N) fertilizer map reconstruction

3 Results

3.1 National nitrogen fertilizers use for crop production in China

The compiled national total N fertilizers use increased from 0.06 Tg N yr^{-1} in 1952 to the peak of 31.15 Tg N yr^{-1} in 2014 then decreased to 28.31 Tg N yr^{-1} in 2018. The majority of the N input was directly applied as N-only fertilizers (e.g. urea, ammonium carbonate), while the contribution from compound fertilizers (e.g. ammonium phosphate) increased from 16% in 1980 to 24% in the 2010s. Despite the total N input decreased by 9.1% (2.84 Tg N yr^{-1}) from 2014 to 2018, the N input from compound fertilizers increased by 6% (0.43 Tg N yr^{-1}) during the corresponding period (Figure 2).