Supplementary material

Net anthropogenic nitrogen input and its relationship with riverine nitrogen flux in a typical irrigation area of China based on an improved NANI budgeting model

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1. Land use maps used in this study

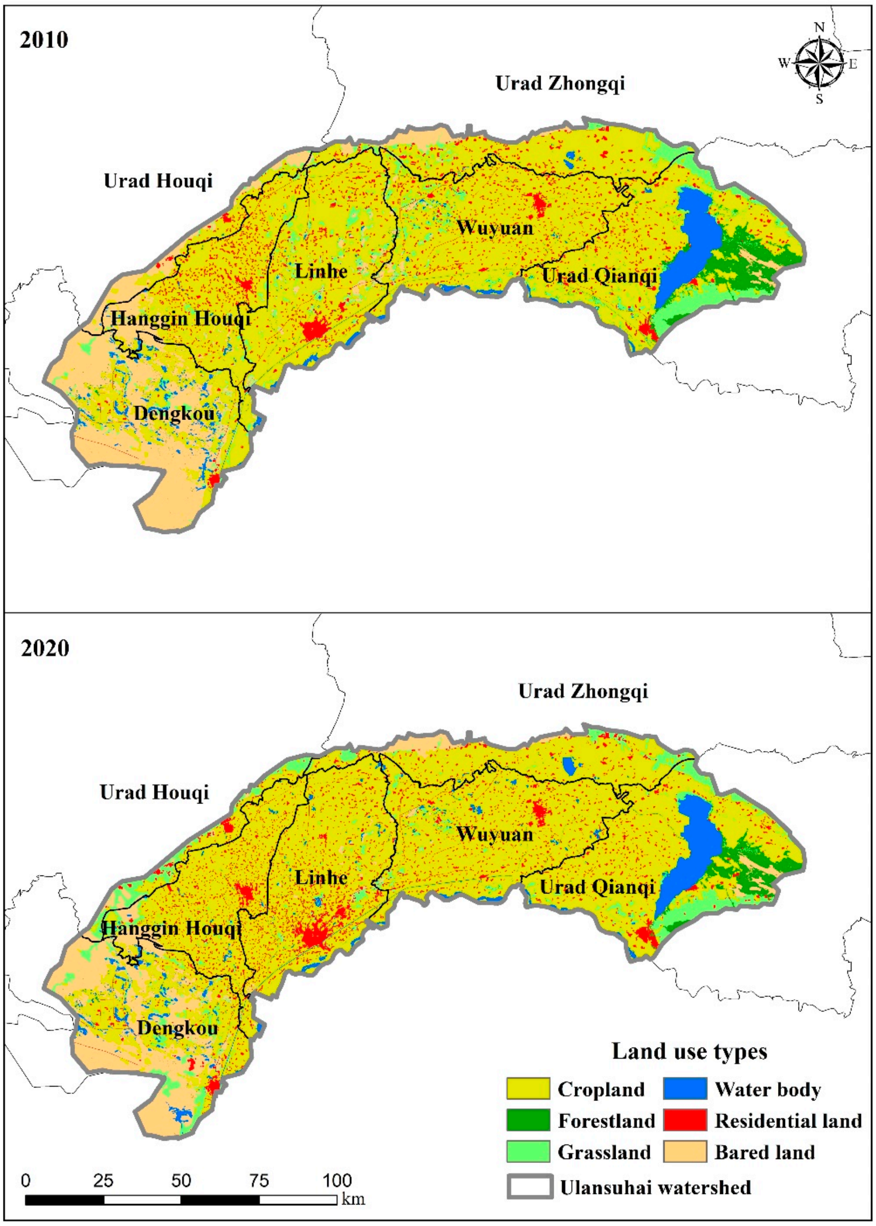

Figure S1. Maps of land use types of 2010 and 2020 in the Ulansuhai Nur watershed (30m resolution).

2. The description of NANI method

2.1 Fertilizer nitrogen application (N_{fert})

Nitrogen fertilizers used in the Ulansuhai Nur watershed include chemical nitrogen fertilizer and compound fertilizer. N_{fert} was quantified based on the application amounts of nitrogen fertilizers and the nitrogen content in the corresponding fertilizers. The nitrogen content is 46% in urea, 35% in ammonium nitrate, 17% in ammonium bicarbonate, 82.3% in anhydrous ammonia and 16.8% in compound fertilizer (Cao et al., 2020; Han et al., 2014). The application amounts of different nitrogen fertilizers were collected directly from Regional Statistical Yearbooks (Fig.2S).

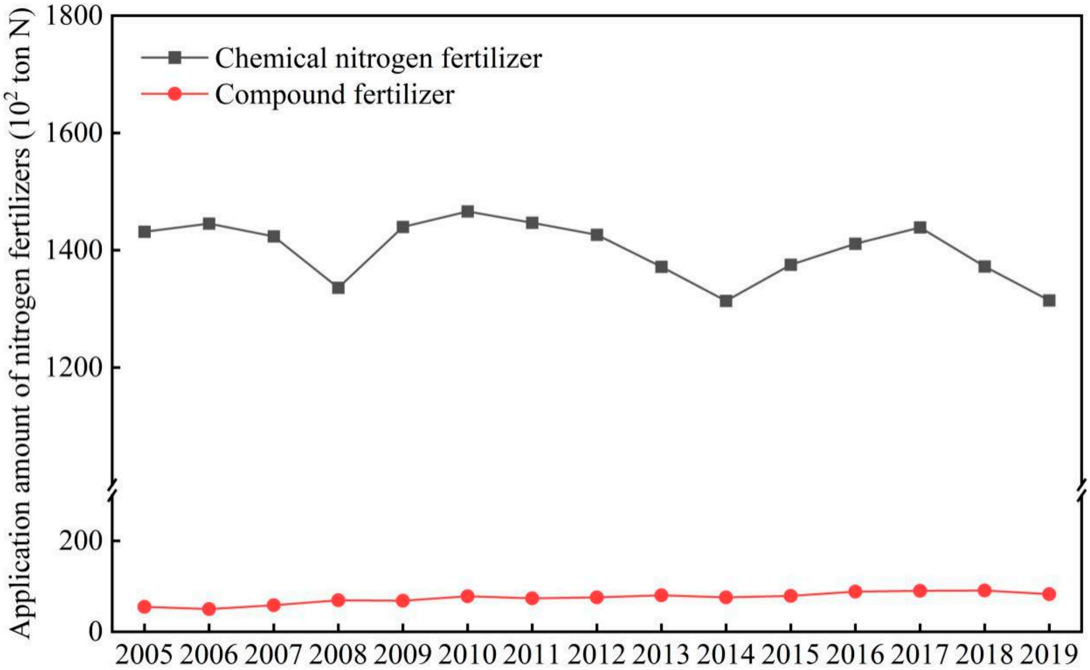

Figure S2. Chemical nitrogen fertilizer application amount in the Ulansuhai Nur watershed from 2005 to 2019.

2.2 Seeding nitrogen input (N_{seed})

Vegetable, fruit and seven major agricultural crops were selected in Ulansuhai watershed to calculate the seeding N input in each research unit. The input amount of seeding N in each research unit was estimated by multiplying the seeding N input per unit area of different crop types and the corresponding sown areas. Seeding nitrogen per unit area for each crop type is listed in Table S1 (Han et al., 2014; Mao et al., 2021). The data on the sown areas of different crop types were derived from Regional Statistical Yearbooks(Fig.3S).

Table S1. Seeding nitrogen input per unit area for each agricultural crop.

Plant type	Seed N (kg N km^{-2} yr^{-1})
Rice	69.2
Wheat	227.1
Corn	25.9
Sorghum	30.1
Soybean	107.5
Potato	63.2
Sunflower	17.96
Vegetable	2.8
Fruit	2.8

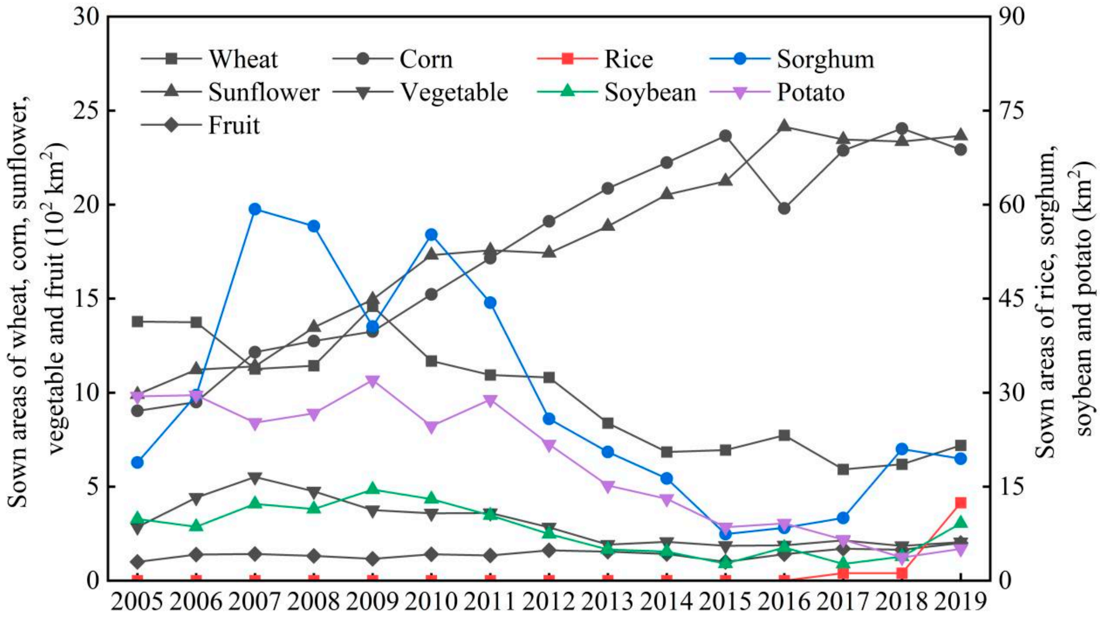

Figure S3. Sown areas for major crops in the Ulansuhai Nur watershed from 2005 to 2019.

2.3 Biological nitrogen fixation (N_{cro})

Biological nitrogen fixation is equal to the sum of symbiotic nitrogen fixation by cultivation of legume crops and non-symbiotic nitrogen fixation by microorganism in natural ecosystem. Biological nitrogen fixation amount in each research unit was derived by multiplying the nitrogen fixation coefficient for each agricultural crops and by its sown area. The biological nitrogen fixation coefficients were 9600, 8000, 3000 and 1500 kg N km^{-2} yr^{-1} for soybean, peanut, rice and other non-symbiotic crops, respectively (Table S2) (Boyer et al., 2002; Chen et al., 2016; Zhang et al., 2015; Zhang et al., 2020). The nitrogen fixation for forest and grass are commonly considered as natural inputs and thus they are not included in the calculation of biological nitrogen fixation in this study (Howarth et al., 2006).

Table S2. Nitrogen fixation coefficients for major agricultural crops in the Ulansuhai Nur watershed.

Biofixation types	Crop types	Fixation coefficient (kg N km^{-2} yr^{-1})
Symbiotic nitrogen fixation	Soybeans	9600
	Peanuts	8000
Non-symbiotic fixation	Rice	3000
	Other non-symbiotic crops	1500

2.4 Atmospheric nitrogen deposition (N_{dep})

Atmospheric nitrogen deposition in this study includes both wet and dry depositions. In view of the fact that the ammonia and ammonium emission has the short transportation distance and deposits in the same watershed (Prospero et al., 1996; Schlesinger and Hartley, 1992), the deposition of the oxidized form (NOx) was only considered in this study (Howarth et al., 1996). The raster data of atmospheric NOx deposition during 2005-2015 were extracted from Regional Emission inventory in ASia version 3 (REASv3) with a spatial resolution of 0.25° (Kurokawa and Ohara, 2020). The atmospheric deposition data after 2016 was derived from the published literature(Du, 2020). Annual atmospheric nitrogen deposition in each research unit is estimated by multiplying the area of research unit by the nitrogen deposition rate.

2.5 Irrigation Nitrogen input (N_{ir})

Irrigation nitrogen input was estimated by multiplying the irrigation volume per year by annual average total nitrogen concentration at the Sanshenggong Hydrological station as the starting point of Yellow River irrigation in the Ulansuhai Nur watershed. The variations of irrigation volume and total nitrogen concentration during the period of 2005~2019 was shown in Fig. S4. The data on irrigation volume and total nitrogen concentration were derived from the local hydrological department and ecological environment department, respectively.

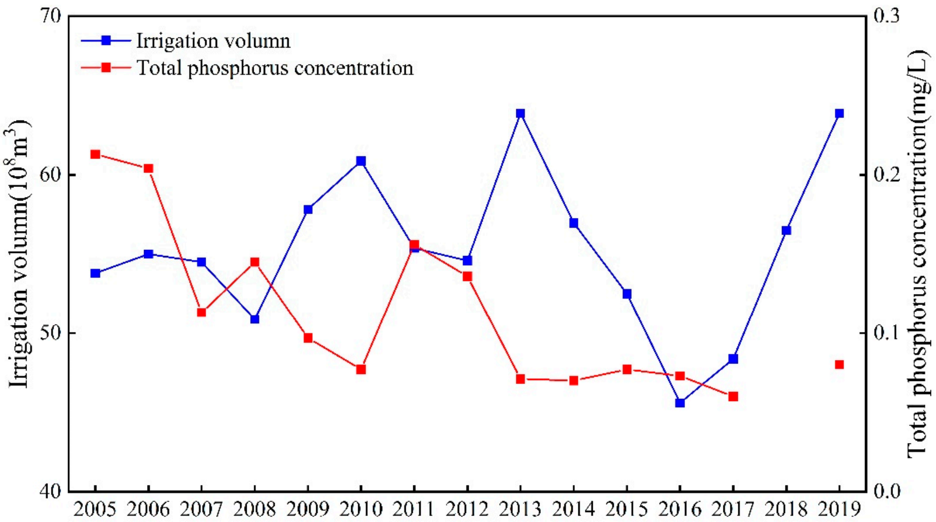

Figure S4. Variations of irrigation water volume from Yellow River and the corresponding total nitrogen concentration in the Ulansuhai Nur watershed from 2005 to 2019.

2.6 Net food and animal feed imports (N_{im})

The net N imports of food and animal feed in the watershed can be calculated by subtracting the N content of crops and livestock products from the N consumed by humans and animals. If the value is positive, it means that the food production in the region is less than the local food demand; if the value is negative, it means that the food production in the region is greater than the local food demand and can be exported. The amount of N_{im} is calculated as follows:

$$N_{im} = N_{hc} + N_{lc} - (N_{lp} + N_{cp})$$

Where N_{hc} and N_{lc} represent the N consumption of Human food and livestock respectively, N_{lp} and N_{cp} represent the N contents of livestock and crop products respectively.

2.6.1 Human nitrogen consumption (N_{hc})

Human nitrogen consumption in food was calculated by the number of inhabitants including urban and rural population in each research unit and nitrogen consumption per capita per year. The nitrogen consumption (kg N capita^{-1} yr^{-1}) was quantified by human protein consumption (g capita^{-1} day^{-1}) and nitrogen conversion factor of 6.25 (Shaheen et al., 2022). Human protein consumptions for urban and rural population during different periods were listed in Table S3 (Ju et al., 2018; Yu et al., 2016). The population data related to urban and rural population was obtained from Regional Statistical Yearbook.

Table S3. Human protein consumption in food in different periods.

Human protein consumption (g individual^{-1} day^{-1})	2005-2010	2011-2019
Urban population	69.0	65.4
Rural population	64.6	63.6

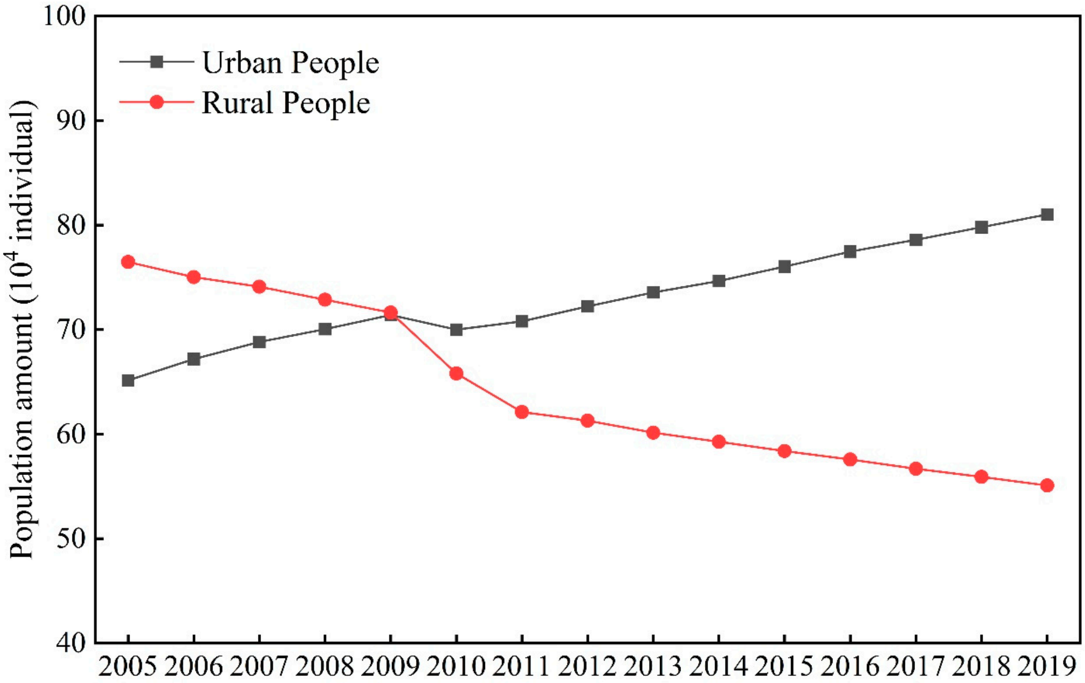

Figure S5. Annual urban and rural population in the Ulansuhai Nur watershed from 2005 to 2019.

2.6.2 Livestock nitrogen consumption (N_{lc})

Livestock included horses and cattle, pig, sheep and poulty in the Ulansuhai Nur watershed. Livestock nitrogen consumption was determined by multiplying the number of each livestock and nitrogen consumption rate per individual. The values of nitrogen consumption rate per livestock listed in Table S4 were acquired from Han et al. (2014). The sales volume and inventory number of livestock were obtained from Regional Statistical Yearbooks.

Considering the difference in feeding period of different livestock, the number of livestock per year during the process of calculation was replaced by average livestock population per year was quantified using data related to sales and inventory of livestock according to equation S1.

$$\mathrm{AL} = Inventory \times \frac{1}{Cylces} + \frac{Sales}{Cycles} \times \frac{Cycles-1}{Cycles} \qquad \text{(S1)}$$

where AL is the annual average number of livestock, Inventory is the number of livestock at the year-end, Sales is the number of livestock slaughtered, Cycles is the duration of the life cycle (the number of days from birth to market) per year, which is determined by the ratio of 365 to life cycle. End-of-year inventory for different livestock types was shown in Fig.S6. The life cycle for horses and cattle, sheep, pigs and poultry in this study are 365, 365, 199 and 55 days, respectively.

Table S4. Annual nitrogen consumption, excretion and production rates for different livestock types.

Livestock species	N consumption (kg N individual^{-1} yr^{-1})	N excretion (kg N individual^{-1} yr^{-1})	N Production (kg N individual^{-1} yr^{-1})
Horses and cattle	54.82	48.79	6.03
Sheep	6.85	5.75	1.1
Pigs	16.68	11.51	5.17
Poultry	0.57	0.37	0.2

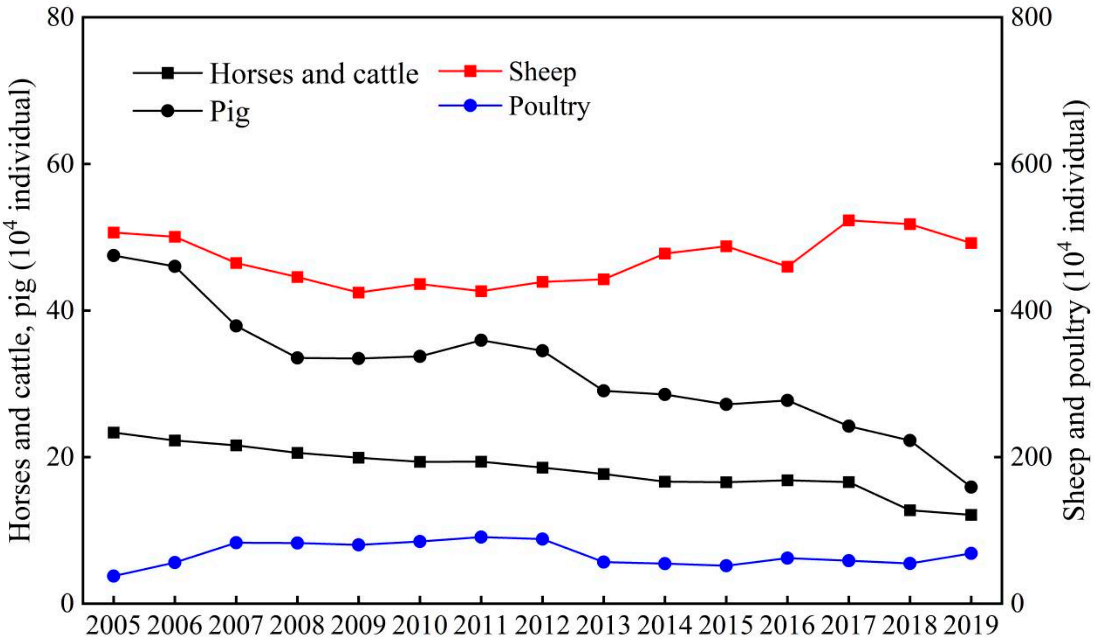

Figure S6. End-of-year number for different livestock types in the Ulansuhai Nur watershed from 2005 to 2019.

2.6.3 Livestock nitrogen production (N_{ln})

Livestock products mainly includes meat, milk, and eggs. Livestock nitrogen production in this study was estimated by the difference between livestock nitrogen consumption and nitrogen excretion (Han et al., 2014). Specifically, livestock nitrogen production was determined by the number of livestock and its nitrogen content. The nitrogen contents (g kg^{-1}) for different livestock were listed in Table S4 and were obtained from Han et al. (2014). In addition, it was assumed that there is 10% loss of livestock products available for consumption due to spoilage and inedible components (Han et al., 2014).

2.6.4 Crop nitrogen production (N_{cp})

Vegetable, fruit and major agricultural crops were chosen to calculate crop nitrogen production in each research unit. For a specific crop, crop nitrogen production (Ncp) was estimated by the product of each crop types and its nitrogen content. The nitrogen contents (g kg^{-1}) for different crop products as shown in Table S5 were obtained from China Food Composition Tables (Yang, 2018). Annual outputs of major crops are shown in Fig.S7. In addition, it was assumed that there is 10% loss for all crop products due to pests and spoilage (Han et al., 2014).

Table S5. Nitrogen contents in main agricultural crop products in the Ulansuhai Nur watershed.

Crop types	Nitrogen content (g kg^{-1})

Rice	12.64
Wheat	19.04
Corn	6.4
Broomcorn	16.64
Soybean	56
Potato	4.16
Flaxseed	30.56
Sunflower	38.24
Sugarbeet	1.6
Vegetable	2.56
Watermelon	0.8
Muskmelon	0.64
Back melon seed	48.48
Tomato	1.44
Apple	0.64
Pear	0.48
Grape	0.64

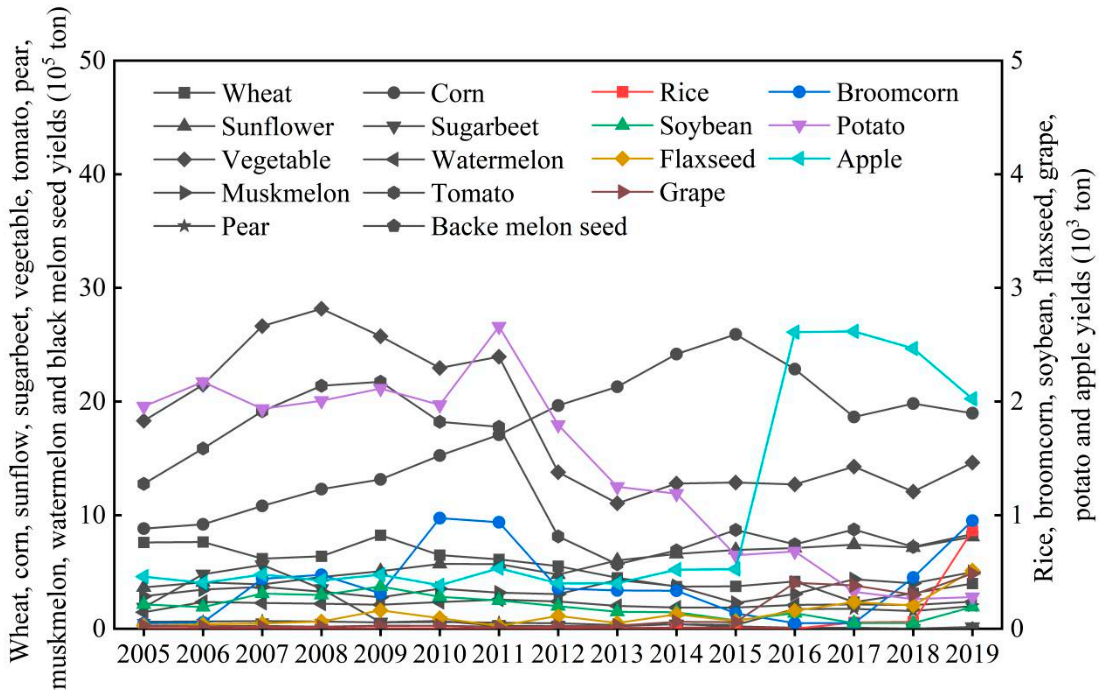

Figure S7. Annual yields of major agricultural crops in the Ulansuhai Nur watershed from 2005 to 2019.

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