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Effect of Long-Term Nitrogen Addition on Wheat Yield, Nitrogen Use Efficiency, and Residual Soil Nitrate in a Semiarid Area of the Loess Plateau of China

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Abstract: Nitrogen (N) fertilizer plays an important role in wheat yield, but N application rates vary greatly, and there is a lack of data to quantify the residual effects of N fertilization on soil N availability. A 17-yr experiment was conducted in a semiarid area of the Loess Plateau of China to assess the effects of N fertilization on spring wheat (*Triticum aestivum* L.) grain yield, N uptake, N utilization efficiency, and residual soil nitrate. Treatments included a non-N-fertilized control and annual application of 52.5, 105.0, 157.5, and 210.0 kg N ha^{-1} in the first two years (2003 and 2004). In the third year (2005), the four main plots with N fertilizer application were split. In one subplot, N fertilization was continued as mentioned previously, while in the other subplot, N fertilization was stopped. The concentration of NO_3-N in the 0–110 cm depth soil layers was significantly affected by N application, with higher N rates associated with greater soil NO_3-N concentration. With the annual application of N over 17 years, residual soil NO_3-N concentration in the 100–200 cm soil layer in the last study year was significantly greater than that in the non-N-fertilized control and was increased with rate of N application. There was a significant positive relationship of soil NO_3-N in the 0–50 cm and 50–110 cm soil layers at wheat sowing with wheat grain N content and yield. Wheat grain yield in the third year (2005) was significantly, i.e., 22.57–59.53%, greater than the unfertilized treatment after the N application was stopped. Nitrogen use efficiency decreased in response to each increment of added N fertilizer, and was directly related to N harvest index and grain yield. Therefore, greater utilization of residual soil N through appropriate N fertilizer rates could enhance nitrogen use efficiency while reducing the cost of crop production and risk of N losses to the environment. For these concerns, optimum N fertilizer application rate for spring wheat in semiarid Loess Plateau is about 105 kg N ha^{-1}, which is below the threshold value of 170 kg N ha^{-1} per year as defined by most EU countries.

Keywords: grain yield; nitrogen use efficiency; nitrogen harvest index; nitrate accumulation

1. Introduction

Wheat (*Triticum aestivum* L.) is one of the three major cereal crops in the world [1]. Wheat, rice (*Oryza sativa* L.), and corn (*Zea mays* L.) provide more than 30% of food calories to humans in 94 developing countries [2], and represent 99% of the total cereal production in China [3]. Nitrogen (N) is one of the major nutritional elements of wheat and other crops, and is widely used to increase yield and improve end-use quality [4]. The application of N fertilizer can improve soil fertility and maintain sustainable soil productivity [5,6]. In the past, the grain yield of wheat was often increased through high rates of N fertilizer application [7]. Therefore, it has been common for farmers in much of the world to overapply N fertilizer to reduce the likelihood of crop N deficiency and ensure high crop yield and quality [8]. The use of N fertilizer has increased dramatically worldwide, from 112.5 million tons in 2015 to about 118.2 million tons in 2019 [9]. In China, more than 31 million tons of N fertilizers were used in 2014, accounting for about 29% of the total global consumption [10]. Based on a national survey of farms, Li et al. [11] found that the average rate of N fertilization for wheat in China was 229 kg N ha^{-1}. However, only about 30–50% of the applied fertilizer N is absorbed by the growing crop in the year of application, depending on the crop species and cultivar [12–15]. Therefore, a key challenge is to satisfy crop N requirements while minimizing N losses to maintain a sustainable environment and economic benefits to farmers [9].

Nitrogen fertilizer is the largest source of N input and loss in cereal cropping systems [16]. With a high rate of N fertilizer application, some fertilizer N may remain in the soil at the end of the growing season, which can be absorbed by the following crops [17] or lead to the negative environmental impacts such as nitrate leaching, greenhouse gas emission, and eutrophication of water bodies [18–21]. Nitrate leaching and water pollution are major environmental problems in Europe, the USA, China, and elsewhere in rainfed areas due to the excessive application of N fertilizer [22–24]. Therefore, most countries in the European Union restrict the rate of N application to 170 kg N ha^{-1} per year [25,26].

A primary goal of N management is to optimize crop yield by applying sufficient N to the crop [20]. Nitrogen use efficiency (NUE), i.e., the amount of N fertilizer absorbed and retained by crops at harvest, is widely used as an index to measure the efficiency of N uptake based on the quantity of N applied [27]. The excessive use of N fertilizer leads to low NUE and economic loss [28–30]. Therefore, the optimization of N nutrition for crops as well as the rational use of N fertilizers and soil N are some of the most important tasks of agronomy [31]. Crop recovery of fertilizer N is relatively low, accounting for only 25–50% of the applied N [32,33], largely due to poor synchronization between N application and crop demand [34,35]. Lower utilization efficiency of N occurs when the amount of applied N exceeds the crop requirements, which thereby increases the risk of N loss [36]. High N input coupled with low NUE threaten the sustainability of agroecosystems [3]. Therefore, exploring approaches to increase NUE in crop production is essential for agricultural sustainability [37]. Numerous studies have been published on the limiting factors of yield and NUE [9,38,39]. Extensive research on the effect of residual fertilizer N on wheat yield and soil nitrate have been conducted in many areas of Germany [40], France [41], Netherlands [42], Canada [14], and Ethiopia [40]. However, few long-term experiments on the relationship between residual effect of N fertilizer and NUE have been conducted, especially in the semiarid Loess Plateau of China.

Crops have been cultivated for thousands of years in the Loess Plateau of China, and wheat is one of the main crops [43]. Long-term annual application of N fertilizer is a common practice in this region [43]. However, former studies have mainly focused on the effect of N fertilizer application rate on wheat grain yield [44], and paid little attention to residual soil nitrate and NUE [18]. Avoiding the buildup of excessive residual soil nitrate and effective utilization of residual soil nitrate are key to enhancing NUE [14]. The objectives of this study were to: (i) determine the effects of long-term N fertilizer application on spring wheat yield (GY), crop N accumulation, soil NO_3-N concentration, and soil total N (TN); (ii) analyze the relationship between soil NO_3-N and wheat N accumulation and GY; and (iii) assess the effects of long-term N fertilizer application rate on NUE, nitrogen uptake efficiency (NupE), nitrogen fertilizer productivity (NfP), and nitrogen harvest index (NHI) to provide a theoretical basis for the rational and efficient application of N fertilizer.

2. Materials and Methods

2.1. Site Description

The long-term experiment, established in 2003, was conducted at the Rainfed Agricultural Experimental Station of the Gansu Agricultural University (35°28′N, 104°44′E, elevation 1971 m above sea level), Dingxi, Gansu Province, northwest China (Figure 1). The soil type of the experimental site is a Huangmian sandy loam [45] and is classified as a Calcaric Cambisol according to the FAO [46]. The basic chemical and physical properties of the soil are shown in Table 1. Based on long-term (2003 to 2019) data from the experimental site, the average annual minimum and maximum air temperatures are −22 and 38 °C in July and January, respectively, the average precipitation is 390.7 mm yr^{-1}, the average annual evaporation is 1531 mm, the annual cumulative air temperature >10 °C is 2240 °C, the annual radiation is 5930 MJ m^{-2}, and the annual sunshine is 2480 h.

Figure 1. Location of the experimental site, Dingxi City, Gansu, China.

Table 1. Soil chemical and physical properties of the experimental site at the beginning of the experiment (2003) [a].

Soil Depth (cm)	TN ($g\ kg^{-1}$)	NH_4-N ($mg\ kg^{-1}$)	NO_3-N ($mg\ kg^{-1}$)	pH	TP ($g\ kg^{-1}$)	AP ($mg\ kg^{-1}$)	TK ($g\ kg^{-1}$)	AK ($mg\ kg^{-1}$)	Bulk Density ($g\ cm^{-3}$)	Organic Matter ($g\ kg^{-1}$)
0–5	0.8	6.4	27.8	8.3	1.9	13.6	18.6	253.1	1.144	12.5
5–10	0.8	5.0	24.0	8.4	1.9	11.8	18.5	329.3	1.144	12.6
10–30	0.7	4.9	31.1	8.3	1.8	4.7	18.3	231.8	1.295	11.7
30–50	0.8	4.5	28.2	8.3	1.7	1.9	18.3	169.1	1.240	11.3
50–80	0.8	4.4	26.9	8.3	1.7	1.9	18.3	123.8	1.180	12.9
80–110	0.8	4.3	24.7	8.4	1.8	2.0	18.2	101.5	1.210	12.6
110–140	0.7	4.4	23.3	8.4	1.7	1.6	18.5	106.0	1.150	11.6
140–170	0.6	5.0	23.4	8.4	1.7	1.7	18.1	101.9	1.150	10.8
170–200	0.6	5.2	21.7	8.4	1.7	2.2	18.1	105.7	1.110	10.7

[a] TN, total nitrogen; TP: total phosphorus; AP: available phosphorus; TK: total potassium; AK: available K.

2.2. Experimental Design and Field Management

The experiment utilized a split-plot arrangement in randomized complete block design with three replications. Each subplot was 3 m × 10 m. The experiment included five treatments applied to main plots in the first two years (2003 and 2004), including a non-N-fertilized control (N1), 52.5 kg N ha^{-1} (N2), 105.0 kg N ha^{-1} (N3), 157.5 kg N ha^{-1} (N4), and 210.0 kg N ha^{-1} (N5) applied as urea. The most commonly used local fertilization rate was 105 kg N ha^{-1}. In the third year (2005), the four main plots with N fertilizer application were split. In one subplot, N fertilization was continued as mentioned previously, while in the other subplot, N fertilization was stopped after 52.5, 105.0, 157.5, and 210.0 kg N ha^{-1} were applied during the first two study years (N2s, N3s, N4s, and N5s, respectively).

All treatments annually received 105 kg P_2O_5 ha^{-1} as calcium superphosphate. Before sowing in each year, all fertilizers were evenly broadcast over each plot area according to the experimentally set amount, and then incorporated into the 0–20 cm soil layer using rotary tillage. Each year, spring wheat (cv. Dingxi 38) was sown in mid-March at a rate of 187.5 kg ha^{-1} in rows spaced 20 cm apart using a grain drill and was harvested in late July to early August. All plots received rotary tillage within one week after wheat harvest. Weeds were removed manually during the growing season and were controlled with herbicides during the fallow periods if needed.

In this study, data from the third experimental year (2005) were used to assess the effects of different N fertilizer application on N uptake and yield of spring wheat. Data were also collected from the seventeenth experimental year (2019) to determine how the long-term N fertilization treatments influenced N uptake and yield of spring wheat, residual soil nitrate, and the persistence of residual effects of N fertilization.

2.3. Soil Samples and Analysis

Soil samples were collected from the 0–5, 5–10, 10–30, 30–50, 50–80, 80–110, 110–140, 140–170, and 170–200 cm soil layers using an auger (inner diameter 4.0 cm) in each plot immediately prior to wheat sowing and after wheat harvest in each year. Soil from three cores of the same layer in a plot was mixed to obtain one sample that was divided into two subsamples: one that was air-dried and ground (<2 mm) for chemical analysis, and another that was immediately transported on ice to the laboratory and stored at –20 °C until analysis for soil NO_3-N and soil water content (SWC) using the oven-dry method [47].

All wheat samples were collected within a 1 × 1 m area at the seedling, tillering, flowering, and maturity stages. The samples were taken to the laboratory and divided into leaf, stem, sheath, glume, and spike-stalk fractions, and dried at 70 °C until constant weight. Each plant fraction was passed through a 2-mm sieve for chemical analysis.

Soil total nitrogen (TN) was determined by the standard Semimicro–Kjeldahl method [48]. Soil nitrate-N (NO_3-N) and ammonium-N (NH_4-N) were determined by the spectrophotometry method with a Discrete Auto Analyzer (Smartchem 450, Beijing, China) [49]. Soil pH was measured using a glass combination electrode in a suspension of soil and water at a ratio of 1:1 [50]. Soil total phosphorus (TP) and soil available phosphorus (AP) were determined by the standard molybdenum antimony colorimetric method [48]. Soil organic matter was determined using the $K_2Cr_2O_7$ external heating method, soil total potassium was determined using the NaOH melting flame photometry method, soil available potassium was determined using 1.0 M CH_3COONH_4 extraction, and total N in grain was determined by H_2SO_4-H_2O_2 digestion and the Nessler colorimetry method, as described by Bao [48]. Soil bulk density was were determined by the ring-knife method [51].

2.4. Methodology

2.4.1. Residual Soil NO_3-N

Residual soil NO_3-N concentration (RSN) was calculated using the following equation [52]:

$$\text{RSN} = \frac{\text{T}i \times \text{D}i \times \text{C}i}{10} \tag{1}$$

where T*i* represents the thickness of the soil layer (20 cm), D*i* represents the soil bulk density (g cm^{-3}), C*i* represents soil NO_3-N concentration (mg N kg^{-1}) of the corresponding layer, and 10 represents the conversion coefficient.

2.4.2. Utilization Efficiency of N Fertilizer

The utilization efficiency of N fertilizer was assessed based on N uptake efficiency (NupE), N fertilizer productivity (NfP), NUE, and nitrogen harvest index (NHI) [53], which were calculated according to the following equations [54,55]:

$$\text{NupE}\left(\text{kg kg}^{-1}\right) = \frac{\text{total N uptake}}{\text{N application rate}} \tag{2}$$

$$\text{NfP}\left(\text{kg kg}^{-1}\right) = \frac{\text{grain yield}}{\text{N application rate}} \quad (3)$$

$$\text{NUE}\left(\text{kg kg}^{-1}\right) = \frac{\text{grain yield}}{\text{total N uptake}} \quad (4)$$

2.5. Statistical Analysis

One-way analysis of variance was conducted using the general linear model function of SPSS 19.0 software (IBM Corp., Chicago, IL, USA) to determine whether soil TN concentration, soil NO_3-N concentration, N accumulation in wheat, GY, NupE, NfP, NUE, and NHI of wheat were affected by N fertilizer treatment. Differences among means were tested with Fisher's protected least significant difference test at $P \leq 0.05$. Pearson's correlation coefficient was used to assess the relationships among soil NO_3-N concentration, N accumulation in wheat, GY, NupE, NfP, NUE, and NHI of wheat across all treatments in 2005 and 2019.

3. Results

3.1. Wheat Yield

Wheat GY increased with increasing N fertilizer application for treatments with and without annual N application in 2005 and 2019 (Figure 2). When N fertilization was applied only during the first two study years, N application rate influenced GY in the subsequent year (2005), but not in the seventeenth year. Wheat GY in the third year (2005) for N3s, N4s, and N5s was significantly 22.57–59.53% greater than the unfertilized treatment (N1), while GY of N2s was not significantly different to that of N1.

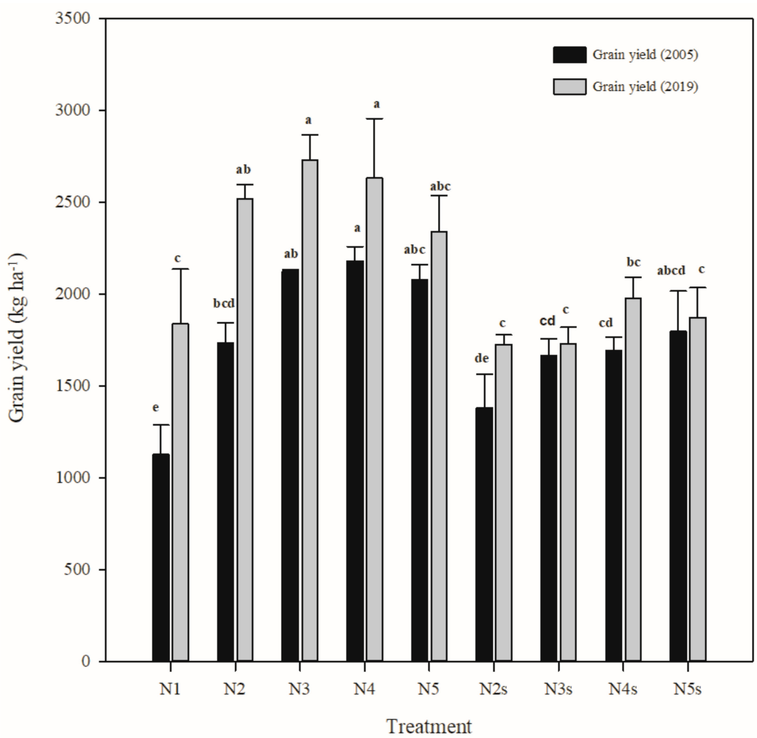

Figure 2. Wheat grain yield (kg ha^{-1} at 13% moisture) in 2005 and 2019 as affected by nitrogen (N) fertilizer treatment. N1, non-N-fertilized control; N2, N3, N4, N5, annual (2003–2019) N fertilizer application at 52.5, 105.0, 157.5, and 210.0 kg N ha^{-1}, respectively; N2s, N3s, N4s, and N5s, N fertilization stopped after two consecutive years (2003–2004) of applying 52.5, 105.5, 157.5, and 210.0 kg N ha^{-1}. Bars with different letters denote means that are significantly different ($P \leq 0.05$).

3.2. Crop Nitrogen Accumulation

Similar to GY, application of N fertilizer increased N accumulation in grain, straw, and total aboveground biomass (grain plus straw) of wheat compared no N fertilization, but the effects were inconsistent (Table 2). Nitrogen accumulation in aboveground biomass with N fertilizer applied annually was significantly greater than that of the non-N-fertilized control in 2005 and 2019. For treatments where N fertilization was stopped, after 157.5 and 210 kg N ha^{-1} was applied in the first two study years (N4s and N5s, respectively), N accumulation in wheat grain, straw, and aboveground biomass in the third year was also significantly greater than that of the non-N-fertilized treatment, but not in the last study year.

Table 2. Nitrogen (N) recovery (kg N ha^{-1}) in the grain, straw, and aboveground biomass (grain plus straw) of wheat grown in 2005 and 2019 as affected by N fertilizer treatment. [b]

Treatment [a]	2005			2019		
	Grain	Straw	Aboveground Biomass	Grain	Straw	Aboveground Biomass
N1	18.34 ± 3.81d	10.22 ± 1.57c	28.56 ± 5.11d	36.37 ± 7.12b	61.82 ± 10.56c	98.19 ± 14.53c
N2	33.04 ± 3.53abc	14.89 ± 4.84bc	47.92 ± 6.06bcd	48.35 ± 1.63ab	143.69 ± 15.47b	192.04 ± 13.95b
N3	40.07 ± 1.18a	22.85 ± 5.3bc	62.93 ± 6.46ab	55.99 ± 3.22a	141.62 ± 12.73b	197.61 ± 15.95ab
N4	39.77 ± 4.69a	26.19 ± 1.51ab	65.96 ± 4.35ab	52.89 ± 10.56a	183.93 ± 29.65ab	236.82 ± 36.77ab
N5	39.18 ± 1.74a	38.61 ± 3.64a	77.79 ± 4.16a	49.96 ± 4.50ab	200.61 ± 19.20a	250.56 ± 21.78a
N2s	22.81 ± 4.28cd	16.83 ± 4.03bc	39.64 ± 8.23cd	33.44 ± 1.18b	68.80 ± 2.30c	102.24 ± 2.54c
N3s	26.15 ± 1.85bcd	21.55 ± 2.25bc	47.7 ± 3.72bcd	33.79 ± 2.68b	56.77 ± 11.34c	90.56 ± 13.74c
N4s	29.38 ± 2.96abc	26.34 ± 8.69ab	55.72 ± 11.13abc	42.83 ± 3.12ab	69.16 ± 12.88c	111.99 ± 14.88c
N5s	34.3 ± 4.54ab	24.49 ± 5.7abc	58.79 ± 9.32abc	35.90 ± 3.05b	73.17 ± 5.85c	109.07 ± 8.34c

[a] N1, non-N-fertilized control; N2, N3, N4, N5, annual (2003–2019) N fertilizer application at 52.5, 105.0, 157.5, and 210.0 kg N ha^{-1}, respectively; N2s, N3s, N4s, and N5s, N fertilization stopped after two consecutive years (2003–2004) of applying 52.5, 105.5, 157.5, and 210.0 kg N ha^{-1}. [b] Within a column for a given year, means followed by different letters are significantly different ($p \leq 0.05$).

3.3. Soil NO_3-N

With annual N application, the average soil NO_3-N concentration of the N2–N5 treatments in the third study year was 31.56 mg kg^{-1} at wheat sowing and 21.94 mg kg^{-1} at wheat harvest in the 0–30 cm soil layer, 29.98 mg kg^{-1} at wheat sowing and 24.74 mg kg^{-1} at wheat harvest in the 30–110 cm soil layer, and 45.69 mg kg^{-1} at the wheat sowing and 25.84 g kg^{-1} at the wheat harvest in the 110–200 cm soil layer (Figure 3a,b). Under the condition of stopping the application of N fertilizer after two years, the average total NO_3-N concentration with the different N fertilizer treatments (N2s–N5s) in the third study year was 23.54 mg kg^{-1} at wheat sowing and 14.99 mg kg^{-1} at wheat harvest in the 0–30 cm soil layer, 14.68 mg kg^{-1} at the wheat sowing and 12.13 mg kg^{-1} at the wheat harvest in the 30–110 cm soil layer, and 30.03 mg kg^{-1} at wheat sowing and 21.00 mg kg^{-1} at wheat harvest in the 110–200 cm soil layer.

With the annual application of N fertilizer, NO_3-N concentration in the 100–200 cm soil layer at wheat sowing in the last study year (2019) was significantly greater than that of the non-N-fertilized treatment (N1), and was increased with the rate of N fertilizer application (Figure 3c). However, at wheat harvest in 2019, soil NO_3-N concentration was significantly greater compared to the unfertilized treatment (Figure 3d).

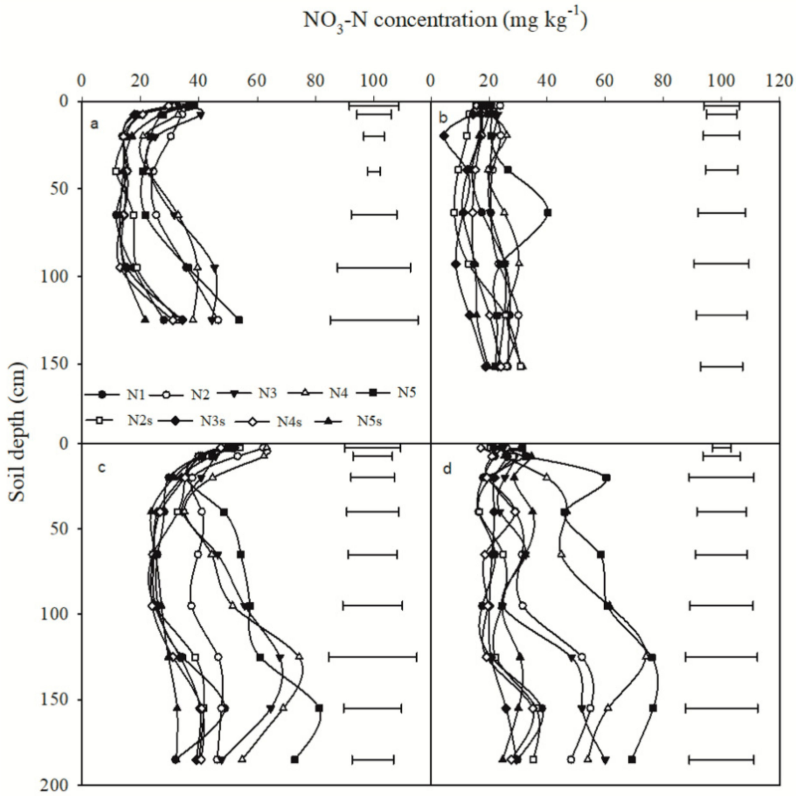

Figure 3. Residual soil nitrate (NO_3)-nitrogen (N) concentration in the 0–200 cm depth in 2005 (**a**,**b**) and 2019 (**c**,**d**) at the wheat sowing (**a**,**c**) and harvest (**b**,**d**) as affected by N fertilizer treatment. N1, non-N-fertilized control; N2, N3, N4, N5, annual (2003–2019) N fertilizer application at 52.5, 105.0, 157.5, and 210.0 kg N ha^{-1}, respectively; N2s, N3s, N4s, and N5s, N fertilization stopped after two consecutive years (2003–2004) of applying 52.5, 105.5, 157.5, and 210.0 kg N ha^{-1}. Horizontal bars are standard errors.

In 2005 and 2019, soil NO_3-N concentration of treatments with annual N application was greater than that of treatments with N applied only during the first two study years, and soil NO_3-N concentration at sowing was higher than that at harvest (Figure 3a–d). The greatest soil NO_3-N concentration with N fertilizer applied annually was obtained within the 110–200 cm soil layer at wheat sowing and at harvest in 2005 and 2019. The lowest soil NO_3-N concentration with N fertilizer applied annually was obtained within the 30–110 cm soil layer at sowing, and in the 0–30 cm soil layer at harvest in 2005 and 2019.

3.4. Soil Total N

Soil total N concentration in the 0–200 cm soil layer at wheat sowing and harvest in 2005 and 2019 decreased with the increase of soil depth, and the decrease within the 0–100 cm soil layer was less than that within the 100–200 cm soil layer (Figure 4a–d). With annual N application, the average soil total N concentration of the N fertilizer treatments (N2–N5) in the third study year was 0.878 g kg^{-1} at sowing and 0.949 g kg^{-1} at harvest in the 0–30 cm soil layer, 0.832 g kg^{-1} at sowing and 0.910 g kg^{-1} at harvest in the 30–110 cm soil layer, and 0.611 g kg^{-1} at sowing and 0.673 g kg^{-1} at harvest in the 110–200 cm soil layer. Under the condition of stopping the application of N fertilizer after two years, the average soil total N concentration of the N fertilizer treatments (N2s–N5s) in the third study year was 0.901 g kg^{-1} at sowing and 0.925 g kg^{-1} at harvest in the 0–30 cm soil layer, 0.891 g kg^{-1} at sowing and 0.943 g kg^{-1} at the harvest in the 30–110 cm soil layer, and 0.649 g kg^{-1} at sowing and 0.699 g kg^{-1} at harvest in the 110–200 cm soil layer.

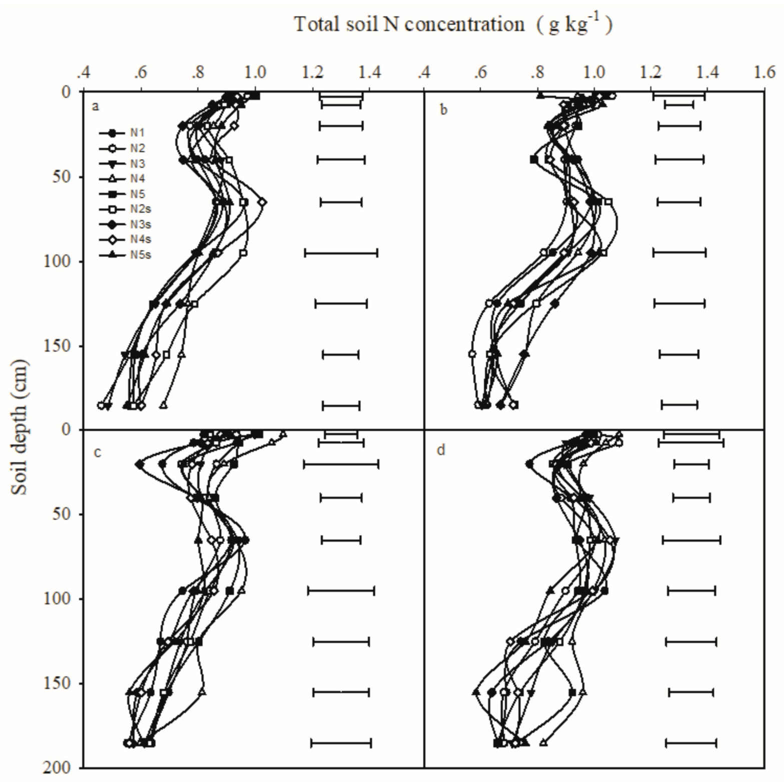

Figure 4. Total soil nitrogen (N) concentration in the 0–200 cm soil layers in 2005 (**a**,**b**) and 2019 (**c**,**d**) at the wheat sowing (**a**,**c**) and harvest (**b**,**d**) as affected by N fertilizer treatment. N1, non-N-fertilized control; N2, N3, N4, N5, annual (2003–2019) N fertilizer application at 52.5, 105.0, 157.5, and 210.0 kg N ha^{-1}, respectively; N2s, N3s, N4s, and N5s, N fertilization stopped after two consecutive years (2003–2004) of applying 52.5, 105.5, 157.5, and 210.0 kg N ha^{-1}. Horizontal bars are standard errors.

In 2005 and 2019, the average soil total N concentration of each soil layer for treatments with N applied annually was significantly greater than that for treatments where N was applied only in the first two study years, and it was significantly greater at wheat harvest than at wheat sowing (Figure 4a–d). The lowest soil total N concentration for all treatments occurred in the 110–200 cm soil layer, while the highest occurred in the 0–30 cm soil layer for treatments that received N fertilizer annually and in the 30–110 cm soil layer for treatments that received N fertilizer in only the first two study years. Across treatments, average soil total N concentration in the 0–30 and 30–110 cm soil layers was not significantly different, and was significantly greater than that in the 110–200 cm soil layer. Across treatments, no significant difference in average soil total N concentration was observed between the 140–170 and 170–200 cm soil layers in 2005 and 2019.

3.5. Relationship between Soil NO_3-N and Wheat N Accumulation and Grain Yield

Wheat GY and grain N accumulation in 2005 were highly correlated with soil NO_3-N concentration in the 0–50 cm soil layer (Table 3). In 2005, there was a significant positive correlation of soil NO_3-N in the 0–50 and 50–110 cm soil layers at wheat sowing and harvest with GY and grain N accumulation, but there was no significant correlation between these variables for the 110–200 cm soil layer. In 2019, there was a significant positive correlation of soil NO_3-N in the 0–50, 50–110, and 110–200 cm soil layers at wheat sowing with GY and grain N accumulation, except for the 0–50 cm soil layer at wheat harvest (Table 4).

Table 3. Pearson's correlation coefficient for correlations of soil nitrate-nitrogen concentration (mg kg^{-1}) at different soil depths and sampling times with wheat nitrogen (N) accumulation in grain, straw, and aboveground biomass (grain plus straw), and grain and straw yield across all treatments in the third year (2005) [a].

Soil Sampling Time	Soil Depth (cm)	N Accumulation (kg N ha^{-1})			Yield (kg ha^{-1})	
		Grain	Straw	Aboveground Biomass	Grain	Straw
At wheat sowing	0 to 30	0.391 *	0.060	0.238	0.379	0.432 *
	0 to 50	0.437 *	0.095	0.283	0.421 *	0.471 *
	0 to 80	0.522 **	0.124	0.345	0.492 **	0.525 **
	0 to 110	0.553 **	0.164	0.385 *	0.506 **	0.526 **
	30 to 110	0.531 **	0.194	0.391 *	0.472 *	0.466 *
	50 to 110	0.504 **	0.179	0.369	0.443 *	0.432 *
	50 to 200	0.460 *	0.199	0.357	0.401 *	0.383 *
	80 to 200	0.425 *	0.211	0.347	0.366	0.348
	110 to 200	0.313	0.198	0.28	0.269	0.242
At wheat harvest	0 to 30	0.454 *	0.198	0.354	0.329	0.351
	0 to 50	0.501 **	0.258	0.414 *	0.365	0.376
	0 to 80	0.536 **	0.348	0.486 *	0.385 *	0.397 *
	0 to 110	0.506 **	0.291	0.436 *	0.379	0.388 *
	30 to 110	0.491 **	0.326	0.450 *	0.376	0.375
	50 to 110	0.459 *	0.301	0.418 *	0.355	0.362
	50 to 200	0.330	0.103	0.233	0.234	0.227
	80 to 200	0.184	−0.081	0.047	0.135	0.121
	110 to 200	0.091	−0.166	−0.052	0.027	0.006

[a] Correlation coefficients followed by * and ** are significant at $P \leq 0.05$ and 0.01, respectively.

Table 4. Pearson's correlation coefficient for correlations of soil nitrate-nitrogen concentration (mg kg^{-1}) at different soil depths and sampling times with wheat nitrogen (N) accumulation in grain, straw, and aboveground biomass (grain plus straw), and grain and straw yield across all treatments in the seventeenth year (2019) [a].

Soil Sampling Time	Soil Depth (cm)	N Accumulation (kg N ha^{-1})			Yield (kg ha^{-1})	
		Grain	Straw	Aboveground Biomass	Grain	Straw
At wheat sowing	0 to 30	0.494 **	0.496 **	0.515 **	0.633 **	0.451 *
	0 to 50	0.603 **	0.661 **	0.677 **	0.716 **	0.565 **
	0 to 80	0.683 **	0.783 **	0.797 **	0.775 **	0.640 **
	0 to 110	0.716 **	0.812 **	0.828 **	0.778 **	0.691 **
	30 to 110	0.704 **	0.842 **	0.852 **	0.698 **	0.696 **
	50 to 110	0.671 **	0.786 **	0.798 **	0.662 **	0.665 **
	50 to 200	0.526 **	0.758 **	0.749 **	0.572 **	0.617 **
	80 to 200	0.465 *	0.700 **	0.688 **	0.513 **	0.581 **
	110 to 200	0.409 *	0.693 **	0.673 **	0.483 *	0.550 **
At wheat harvest	0 to 30	0.121	0.555 **	0.505 **	0.165	0.354
	0 to 50	0.241	0.636 **	0.596 **	0.266	0.452 *
	0 to 80	0.334	0.685 **	0.654 **	0.336	0.505 **
	0 to 110	0.419 *	0.740 **	0.716 **	0.399 *	0.555 **
	30 to 110	0.509 **	0.715 **	0.709 **	0.458 *	0.570 **
	50 to 110	0.539 **	0.743 **	0.738 **	0.479 *	0.583 **
	50 to 200	0.637 **	0.819 **	0.821 **	0.628 **	0.731 **
	80 to 200	0.648 **	0.818 **	0.822 **	0.646 **	0.746 **
	110 to 200	0.630 **	0.782 **	0.787 **	0.650 **	0.743 **

[a] Correlation coefficients followed by * and ** are significant at $P \leq 0.05$ and 0.01, respectively.

3.6. Effects of N Fertilizer Application Rate on N Use Efficiency

Nitrogen use efficiency, an index of GY per unit of absorbed N, differed between N fertilization regimes (N applied annually or just in the first two study years) and years (2005 and 2019) (Table 5). The average NUE was significantly less in 2019 than 2005 for both N fertilization regimes (32.92 kg kg^{-1} in 2005 and 11.96 kg kg^{-1} in 2019 when N was applied annually; 33.63 kg kg^{-1} in 2005 and 18.07 kg kg^{-1} in 2019 when N was applied only in the first two study years). Additionally, NUE of the N4 and N5 treatments was significantly less in 2019 when N fertilizer was applied annually compared to when it was applied only in the first two study years, but this difference was not significant in 2005. Under the condition of annual N application or stopping the application of N fertilizer after two years, NupE and NfP decreased with increasing N fertilizer application in 2005 and 2019. Nitrogen uptake efficiency and NfP were lowest in the N5 treatment in 2005 and 2019. Meanwhile, NfP was highest in the N2s treatment in 2005 and 2019. The average NHI was 0.62 in 2005 and 0.24 in 2019 for treatments where N was applied annually, and 0.57 in 2005 and 0.36 in 2019 for treatments where N was applied only in the first two study years. Nitrogen harvest index ranged from 0.55 to 0.70 kg kg^{-1} in 2005 and 0.20 to 0.39 in 2019. With annual N application, NHI with the N2 treatment was significantly greater than that with the N5 treatment in 2005, NHI with the N3 treatment was significantly greater than that with the N5 treatment in 2019, and there were no other significant differences in NHI among N fertilizer application rates in this fertilizer regime in 2005 or 2019. Under the condition of stopping the application of N fertilizer after two years, there was no significant difference in NHI among N rates in 2005 and 2019.

Table 5. Nitrogen uptake efficiency (NupE, kg kg^{-1}), N fertilizer productivity (NfP, kg kg^{-1}), N use efficiency (NUE, kg kg^{-1}) and nitrogen harvest index (NHI, kg kg^{-1}) of wheat at maturity as affected by nitrogen fertilizer treatment in 2005 and 2019 [a].

Treatment [b]	2005				2019			
	NupE	NfP	NUE	NHI	NupE	NfP	NUE	NHI
N2	1.14 ± 0.23bc	33 ± 2.16b	36.79 ± 2.55a	0.70 ± 0.07a	3.66 ± 0.27c	47.99 ± 1.49e	13.22 ± 0.78cd	0.26 ± 0.02bcd
N3	0.8 ± 0.08bcd	20.24 ± 0.1c	34.6 ± 4.05a	0.65 ± 0.05ab	1.88 ± 0.15d	25.99 ± 1.34f	13.91 ± 0.83bcd	0.28 ± 0.01bc
N4	0.56 ± 0.04cd	13.84 ± 0.5cd	33.34 ± 2.52a	0.60 ± 0.03ab	1.5 ± 0.23d	16.72 ± 2.05f	11.28 ± 0.76d	0.23 ± 0.03cd
N5	0.49 ± 0.03d	9.89 ± 0.4d	26.95 ± 2.42a	0.51 ± 0.02b	1.19 ± 0.1d	11.14 ± 0.94f	9.43 ± 0.86d	0.20 ± 0.02d
N2s	2.01 ± 0.42a	52.63 ± 7.03a	36.39 ± 3.76a	0.58 ± 0.02ab	16.55 ± 0.41a	279.66 ± 8.33a	16.89 ± 0.09abc	0.33 ± 0.01ab
N3s	1.21 ± 0.09b	31.7 ± 1.74b	35.05 ± 1.17a	0.55 ± 0.02b	7.33 ± 1.11b	140.22 ± 7.1b	19.99 ± 2.97a	0.38 ± 0.03a
N4s	0.94 ± 0.19bcd	21.5 ± 0.91c	32.13 ± 4.39a	0.55 ± 0.06b	6.04 ± 0.8b	106.66 ± 6.31c	18.15 ± 2ab	0.39 ± 0.03a
N5s	0.75 ± 0.12bcd	17.13 ± 2.08cd	30.93 ± 1.09a	0.59 ± 0.04ab	4.41 ± 0.34c	75.83 ± 6.56d	17.26 ± 1.53abc	0.33 ± 0.01ab

[a] Within a column for a given year, means followed by different letters are significantly different ($P \leq 0.05$). [b] N2, N3, N4, N5, annual (2003–2019) N fertilizer application at 52.5, 105.0, 157.5, and 210.0 kg N ha^{-1}, respectively; N2s, N3s, N4s, and N5s, N fertilization stopped after two consecutive years (2003–2004) of applying 52.5, 105.5, 157.5, and 210.0 kg N ha^{-1}.

There were significant positive correlations between NupE and NfP, and NUE and NHI in 2005, as well as between NupE and NfP, NupE and NUE, NupE and NHI, NfP and NUE, NfP and NHI, and NUE and NHI in 2019 (Table 6). Nitrogen harvest index was positively correlated with GY, NupE, Nfp, and NUE in 2019, but only positively correlated with NUE in 2005.

Table 6. Pearson's correlation coefficient for correlations between grain yield (GY), nitrogen uptake efficiency (NupE), nitrogen fertilizer productivity (NfP), nitrogen use efficiency (NUE), and nitrogen harvest index (NHI) of wheat across all treatments in 2005 and 2019 [a].

	2005				2019			
	NfP	NUE	NHI	GY	NfP	NUE	NHI	GY
NupE	0.952 **	0.012	−0.100	−0.404	0.982 **	0.424 *	0.430 *	−0.601 **
NfP		0.293	0.097	−0.538 **		0.562 **	0.546 **	−0.640 **
NUE			0.632 **	−0.426 *			0.937 **	−0.534 **
NHI				−0.095				−0.528 **

[a] Correlation coefficients followed by * and ** are significant at $P \leq 0.05$ and 0.01, respectively.

4. Discussion

4.1. Impacts of N Fertilization on Wheat Yield and Crop N Accumulation

The study of fertilizer N fluxes in agroecosystems is important for assessing the effectiveness of its use for wheat yield and in determining possible environmental contamination [56]. Optimal N fertilizer application plays a vital positive role in the growth and productivity of crops [18]. Appropriate N application rates help improve the profitability of crop production and NUE [29]. Some studies have shown that N fertilizer application could increase crop yield, whereas excessive N fertilizer application led to yield reduction [57]. In this study, N fertilizer application could significantly increase the GY of spring wheat in loessial soils of semiarid regions of the Loess Plateau, which is also the main reason for the long-time continuous application of large amounts of N fertilizer in this region [58]. Our findings also suggested that when the amount of N fertilizer application rate increased to a certain extent (N5), the yield of spring wheat would not continue to significantly increase, but there was no significant decline (Figure 2). Therefore, similar to previous research [59], this research also shows that higher N fertilizer application ensured higher crop yield, but was not necessary to achieve optimal crop production.

In China, most farmers often overused and applied N fertilizer in order to pursuit of higher yield [60]. In this study, the optimum N fertilizer application rate for spring wheat in this region was 105 kg N ha^{-1}, which is below the threshold value of 170 kg N ha^{-1} per year as defined by most EU countries [25,26]. Crop N accumulation requires both a N source and a N sink for crop utilization. Similar to the GY, N fertilizer application rate significantly increased crop N accumulation compared with the non-N treatment (N0) (Table 2), which was consistent with the previous studies of Liu and Scholberg et al. [61,62]. However, the amount of N absorbed by crops is affected by genetic and environmental factors that affect crop yield potential, compromising the relationship between N fertilizer application rate and N accumulation in the crop [12–15]. Unfortunately, most farmers usually decide the amount of N fertilizer based on their experience, rather than testing soil nutrients and plant growth [35].

4.2. Soil N Accumulation and N Uptake

When the N fertilizer application rate exceeds crop demand, the proportion of applied fertilizer N used by the crop is reduced, and a larger portion of the applied N remains in the soil or is lost [19]. In contrast, when the N fertilizer rate is less than crop demand, soil N is depleted [63]. Numerous studies have shown that long-term excessive application of N fertilizer can increase soil available N [64,65]. In China, the uninhibited application of N fertilizer over several has caused NO_3-N accumulation in most farmland soils in semiarid regions [23,66]. In this study, with annual application of N fertilizer for three years, the N fertilizer application rate significantly affected soil NO_3-N concentration in the 0–110 cm depth, with higher application rates of N fertilizer associated with greater soil NO_3-N concentration; there was no significant difference in soil NO_3-N concentration below the 140 cm depth both for treatments that received N fertilizer in only the first two study years and for treatments that received N annually (Figure 3a,b), which was consistent with the conclusions of Guo et al. [53]. However, results from this study also indicate that, with annual application of N fertilizer for 17 years, soil NO_3-N concentration was significantly increased in the 100–200 cm soil layer compared to the non-N-fertilized treatment, and was increased with greater rates of N application (Figure 3c,d). This may be due to the continuous and large-scale application of N fertilizer for a long time, which caused the residual N in the soil to move deeper into the soil [61]. Although the accumulation of NO_3-N in the soil profile is a slow process due to the absorption and utilization of crops [67], the accumulated NO_3-N in the soil profile will gradually move downward and leaching to available below the root zone [68] because of the concentrated rainfall in the semi-arid region of the Loess Plateau [43]. Therefore, the accumulation and loss of NO_3-N in soil could be reduced or avoided by applying N fertilizer in an amount less than or equal to that necessary for the optimal crop yield [69,70].

Soil N accumulation following excess N application can be substantial in semiarid regions [71], due to persistence of inorganic N and increased mineralizable soil N [65]. In this study, the average soil total N concentration for treatments with N applied annually was significantly greater than that for treatments where N was applied only in the first two study years; soil total N concentration in the 0–200 cm soil layer decreased with the increase of soil depth, and the decrease within the 0–100 cm soil layer was less than that within the 100–200 cm soil layer (Figure 4). There was no significant difference in average soil total N concentration observed between the 140–200 cm soil layers in both 2005 and 2019, which also further proved that the residual soil N that migrated deeper into the soil was reflected in soil NO_3-N, but not in soil TN [14].

According to many studies, the residual NO_3-N in the soil can be absorbed and utilized by following crops. For example, Soperet et al. [72] reported that soil NO_3-N concentration in the 0–60 cm depth can be used as a predictor of soil N available for crop uptake and the need for fertilizer N; Grant et al. [14] also found that soil NO_3-N concentration in the 0–60 cm depth significantly affected N accumulation in wheat. Sieling et al. [73] also reported that the N remained in 90–150 cm soil layer could be taken up by wheat during the subsequent growth period. This study found a significant positive relationship between soil NO_3-N concentration in the 0–50 and 50–110 cm soil layers at wheat sowing with wheat grain N content and GY in 2005, but there were no significant correlations among these variables for the 110–200 cm depth (Table 3). In addition, there was also a significant positive correlation between soil NO_3-N in the 110–200 cm soil layers at wheat sowing with wheat grain N content and GY in 2019 (Table 4). These results not only confirmed the previous research conclusions, but also proved that the use depth of soil NO_3-N by crops might reach to 200 cm. Therefore, it is necessary to find ways to effectively use the accumulated NO_3-N in the soil, while saving the application cost of N fertilizer on future crops, and reduce the leaching of NO_3-N deep in the soil profile [67].

4.3. Residual Effects of N Fertilizer

Some researchers have shown that residual soil N could cause potential N losses, and might also represent considerable fertilizer value [74,75]. This study indicated that for treatments where N fertilizer was applied only in the first two study years (2003–2004), wheat GY in the third year (2005) for N3s, N4s, and N5s was significantly 22.57–59.53% greater than the unfertilized treatment (N1), while N2s was not significantly different than that of N1, which proved that N fertilizer application history in preceding years may affect subsequent soil N supply [14]. For treatments where N fertilization was stopped, after 157.5 and 210 kg N ha^{-1} was applied in the first two study years (N4s and N5s, respectively), soil NO_3-N, N accumulation in wheat grain, straw, and aboveground biomass in the third year was also significantly greater than that of the non-N-fertilized treatment, but not in the 17th year (2019). The results showed that the residual effects of fertilizer N increased with the increase of the previous N fertilizer application rate [67], and decreased over time due to the combination of crop N uptake and N loss pathways [73].

4.4. N Fertilizer Use Efficiency

Nitrogen (N) plays a critical role in crop production, and it may also be an important pollutant that has a significant impact on air and water quality, biodiversity, and human health [76]. From 1987 to the present, China's crop production was at the stage of the highest N input and N loss, higher yields, and the lowest NUE [3]. The large amount of N fertilizer applied to farmland might threaten the environment and the sustainable development of crop production systems [77]. A higher NUE is generally considered to be the adaptation of crops to a habitat with low soil N supply [78]. In this study, NUE varied greatly with the amount of N fertilizer application and the length of N fertilizer application years. The average NUE was significantly less in 2019 than 2005 for both N fertilization regimes (N applied annually or just in the first two study years), and the greatest NUE observed in N2 treatment (applied 52.5 kg N ha^{-1} applied annually) rather than in the higher N fertilizer treatments (N3-N5) or in the treatments N applied only in the first two study years (N2s–N5s) (Table 5). These results also confirmed some previous views that the NUE decreased with increasing N fertilizer application rate and years [79], and didn't increase at very low levels of N fertilizer application (such as N applied just in the first two study years) [80].

In addition, similar to the conclusion of Li et al. [81], the high N rate of 210 kg N ha^{-1} substantially decreased NupE and NfP compared to the lower N rate of 52.5 kg N ha^{-1}. Therefore, farmers' high N rate application should be decreased to improve N fertilizer use efficiency. Generally, it is considered that soil NO_3-N is the major source of N absorbed by crops [82] and the improvement of N recovery depended on the capture of by roots deep in the soil profile during the growing season [83]. Therefore, further research is needed to clarify whether there is scope for increasing N uptake efficiency through increasing utilization of residual soil NO_3-N [84].

In this study, NUE was significantly positively correlated with NfP and NupE in 2019, but not in 2005 (Table 6), and was inconsistent with the conclusions of Guo et al. [53], which might be due to severe water shortage in 2005 that affected crop yields and nitrogen uptake. Previous studies have shown that due to the effect of biomass on nitrogen absorption, the improvement of N uptake for several years may not necessarily improve NHI [80]. This study found that NUE and NHI are significantly positively correlated in both 2005 and 2019 (Table 6). The decrease in NUE and NHI reflects an increase in available N supply that was not utilized by the crop for grain production [80].Therefore, it is essential to increase NUE by reducing the input of N fertilizer and increasing the N absorption of crops, and to achieve the sustainability of crop production.

5. Conclusions

This long-term experiment found that N fertilizer application can significantly increase grain yield and N accumulation of spring wheat. Higher N fertilizer application ensured higher crop yield and N accumulation, but was not necessary to achieve optimal crop production. When N fertilizer application rate exceeds crop demand. Long-time continuous application of large amounts of N fertilizer might cause the residual soil NO_3-N in the soil to move deeper into the soil and seep below the root zone in the semi-arid region of the Loess Plateau, cause potential N losses, and might also be a pollute source. N fertilizer application history in preceding years may affect subsequent soil N supply, and the residual effects of fertilizer N increased with the increase of previous N fertilizer application rate, and decreased over time. Therefore, it is necessary to find ways to effectively use the accumulated NO_3-N in the soil, while saving the application cost of N fertilizer on future crops, and reducing the leaching of NO_3-N deep in the soil profile. The higher rate of N fertilizer application decreased nitrogen uptake efficiency and nitrogen fertilizer productivity. It is essential to increase NUE by reducing the input of N fertilizer and increasing the N absorption of crops, and to improve the sustainability of crop production.

With respect to the above concerns, the optimum N fertilizer application rate for spring wheat in semiarid Loess Plateau is about 105 kg N ha^{-1}, which is below the threshold value of 170 kg N ha^{-1} per year as defined by most EU countries.

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