Land-use Change from Cropland to Orchard Leads to High Nitrate Accumulation in the Soils of a Small Catchment

Jingbo Gao[1, 2], Yongli Lu[1, 2], Zhujun Chen[1, 2*], Lei Wang[3], Jianbin Zhou[1, 2*]

[1]College of Natural Resources and Environment, Northwest A&F University, Yangling, Shaanxi 712100, China

[2]Key Laboratory of Plant Nutrition and the Agri-environment in Northwest China, MOA, Yangling, Shaanxi 712100, China

[3]British Geological Survey, Keyworth, Nottingham NG12 5GG, UK

*corresponding author:

Jianbin Zhou

Email: jbzhou@nwafu.edu.cn

Zhujun Chen

Email: zjchen@nwafu.edu.cn

Tel: +86-29-87082793

Fax: +86-29-87080055

Abstract

Land-use change from cereals to fruit orchards usually results in a high nutrient surplus in soil. The excessive accumulation of nitrogen (N) in soil, mainly as nitrate, leached from the root zone may serve as a long-term source of surface or groundwater pollution. The N balances and nitrate accumulation in the soil profiles of cereal fields and kiwifruit orchards in the Yujiahe catchment, Shaanxi, China, were compared. Excessive N fertilisation resulted in an excessive N surplus (1133 kg N ha^{-1} yr^{-1}) in orchards (8 times higher than that in cereal fields). More than 77.5% of nitrate in the soil profile (0–400 cm) of the orchards was below the 100 cm soil depth. The average accumulated nitrate within the 0–400 cm profile of orchards was 3288 kg N ha^{-1}, which was 16-fold higher than that of cereal fields. The accumulated nitrate in soil profiles on the downslope (5959 kg N ha^{-1}) was approximately 2 times higher than that of the upslope in the same sloping orchards. The accumulated nitrate in soil profiles at the lowland zone of the catchment was higher than that of the upland zone. Excessive nitrate moves not only vertically downwards to deeper soil depth but also laterally into lower elevations at both field and catchment scales. The total stored nitrate in the upper 400 cm soil profile in the catchment was 464.8 Mg N, while 94.8% (440.8 Mg N) was in orchards. Thus, changing land use from cereal crops to orchards leads to a high nitrate accumulation in the catchment.

KEYWORDS

Kiwifruit orchards, Nitrogen surplus, Nitrate accumulation, Soil profile, Catchment

1 INTRODUCTION

Nitrogen (N) fertiliser is essential for feeding the increasing population of the world. N fertiliser plays a more important role in countries with large populations, such as China and India. The grain yields in China have doubled since the 1980s (National Bureau of Statistics of China, 2018); however, China has also consumed approximately one-third of the global N fertiliser (FAOSTAT, 2018). The overuse of N fertiliser has become common in China in recent years, resulting in an N surplus and a low N use efficiency (Ju *et al.*, 2006). The excessive N that has accumulated in soil can be lost to the environment in different ways, such as leaching and volatilization, resulting in negative effects on the water quality, atmospheric environment, human ecosystems and human health (Hakeem *et al.*, 2016; Ahmed *et al.*, 2017; Burow *et al.*, 2010; Savci, 2012; Li *et al.*, 2018).

Residual N may be fixed by clay minerals or immobilised by soil microorganisms or organic matter; it also exists in the form of mineral N, such as ammonium and nitrate, in the soil (Sebilo *et al.*, 2013). The excessive nitrate accumulation in soil profiles is a major environmental risk for the aquatic environment. Numerous studies have shown that excessive nitrate accumulates in field soils, and there has been research on the nitrate accumulation and losses at catchment scales and even global scales (Fang *et al.*, 2006; Van Meter *et al.*, 2016; Akhavan *et al.*, 2010; Ascott *et al.*, 2017). For example, nitrate loading from the Mississippi River basin resulted in more eutrophication in the Gulf of Mexico from 1988 to 2017 (Van Meter *et al.*, 2018; Rabalais *et al.*, 2002; Obenour *et al.*, 2013).

The elevated nitrate concentration of groundwater bodies in agricultural regions is relatively common in many countries, such as the USA, the UK, Spain, Argentina and China (Burow *et al.*, 2010; Costa *et al.*, 2002; Li *et al.*, 2018; Menció *et al.*, 2016; Stuart *et al.*, 2007). A high nitrate concentration in drinking water causes blue-baby syndrome (methaemoglobinaemia) in infants and may even pose a potential cancer risk (Yang *et al.*, 2007; Burow *et al.*, 2010; Heisler *et al.*, 2008).

An N surplus and nitrate accumulation in soils is closely linked to land use patterns (Laurent & Ruelland, 2011; Min *et al.*, 2018; Zhou *et al.*, 2016; Wang *et al.*, 2017). Great changes in land use patterns have occurred in China since the 1980s, mainly driven by economic development and increasing living standards. Large areas of cereal cropland have been converted to horticultural crops (e.g., fruit trees and vegetables) due to their high economic value (Ju *et al.*, 2006; Qiu *et al.*, 2010). Compared to cropland, over-fertilisation is more common in horticultural crops (Ju *et al.*, 2006; Lu *et al.*, 2016; Gao *et al.*, 2012). More studies indicate high nitrate accumulations in the soils of horticultural systems (Shi *et al.*, 2009; Zhou *et al.*, 2016; Zhou *et al.*, 2010). However, these studies mainly focus on the field scale or consider only the accumulation or vertical movement of nitrate in the soil, without considering its spatial variation at the catchment scale. The catchment approach has long been used to evaluate whole-ecosystem nutrient cycling or the impacts of different land use on the local environment (Laurent & Ruelland, 2011; Marwick *et al.*, 2014; Bartoli *et al.*, 2012; Ierodiaconou *et al.*, 2005). The catchment scale is also considered as the appropriate scale or evaluating water quality and the most important scale for establishing policy that addresses water contamination (Clenaghan *et al*., 2005). Catchment characteristics, such as land-use patterns, management practices and topography, can result in complex spatial patterns of soil nutrients (Wang *et al*., 2001). Therefore, it is very important to study nitrate accumulation in different land use patterns and the nitrate spatial distribution at the catchment scale to develop effective management strategies to reduce nitrate losses to freshwater.

Dramatic land use changes have taken place in China since the 1980s. Kiwifruit (*Actinidia deliciosa*) production in the northern sloping region of the Qinling Mountains in Shaanxi is a typical example. Large areas of cereal lands have been converted into kiwifruit orchards since the 1990s due to the high economic value of kiwifruit and the suitable climate (Chen *et al*., 2019; Lu *et al*., 2016). Now, this region is the largest kiwifruit production area in China, contributing to 30% of the global cultivation area (Shaanxi Provincial Bureau of Statistics, 2018; FAOSTAT, 2018). Similar to other horticultural crops, the overuse of N fertiliser and flood irrigation are common practices for kiwifruit production in this region due to the low education levels and less efficient rational fertiliser application recommendation systems. Our previous studies found that substantial N has accumulated in the soils (Gao *et al.*, 2016; Lu *et al.*, 2016). Due to excessive flood irrigation and hilly topography, nitrate accumulation and loss are expected in the region. Therefore, it is urgent and necessary to understand the nitrate accumulation in the soils, the nitrate spatial distribution, and the nitrate potential environmental risk, thereby providing evidence for sound decision making that promotes both environmental management and kiwifruit production in the region.

Therefore, we selected a typical hilly catchment, i.e., Yujiahe catchment, which has intensive kiwifruit orchards. The main focuses of this paper are (1) comparing the nitrate accumulation in soil profiles of cereal fields and kiwifruit orchards to evaluate the effect of land use change on nitrate accumulation in soils; (2) studying the spatial variation of nitrate accumulation in the soils at the orchard and catchment scales; and (3) investigating the status of nitrate storage in the soils at the catchment scale and its potential environmental risk.

2 MATERIALS AND METHODS

2.1 Study site

The study site is located in the Yujiahe catchment (4.12 km^2, 33°42′–34°14′N, 107°39′–108°37′E) Zhouzhi County, Shaanxi, China (Fig. 1). The site is a typical intensive agricultural catchment in the northern sloping region of the Qinling Mountains. There is a stream originating from the mountains and a reservoir at the end of this catchment. Approximately 85% of the arable lands in this catchment are located on 2–15° slopes. The main soil types include Typic Usti-Alluvic Primosols, Loessi-Orthic Primosols and Typic Hapli-Ustic Isohumosols (Chinese Soil Taxonomy Research Group, 2001). The average annual temperature and precipitation (from 1957 to 2016) in this region is 13.2℃ and 708 mm, respectively. A total of 60–80% of the annual precipitation occurs between July and September. The groundwater is pumped for drinking and irrigation in the catchment.

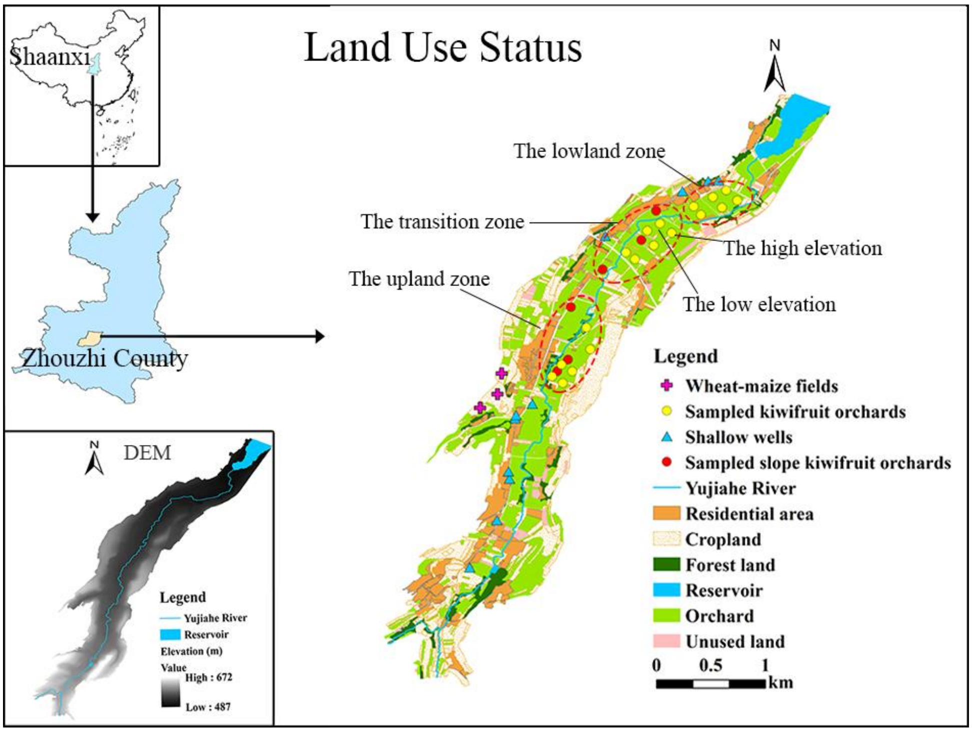

Fig. 1. The location, digital elevation model (DEM), land-use status and sampling sites of the study catchment.

The wheat-maize field was the main land use pattern in the catchment before the 1990s. The kiwifruit orchards were mainly established after the early 1990s (Lu *et al*., 2016). Cereals occupy 28.5% of the entire catchment, and cereals are mainly located in the high-elevation areas (6–15° slope) in the catchment. There is no irrigation for the cereals. The kiwifruit orchards, which cover 32.5% of the catchment, are located at the lower-elevation area (2–6° slopes) of the catchment, where irrigation is available. The average root distributions of the kiwifruit vines in the 0–20, 20–40, 40–60, 60–80, and 80–100 cm layers are 39.8%, 34.0%, 18.0%, 6.1%, and 2.1%, respectively (Fan & Yang, 2003; Wang *et al.*, 2010). The annual irrigation rate in kiwifruit orchards is mainly determined by the local rainfall frequency and rate. Usually, approximately 3–4 irrigations (~100–150 mm each event) are normally required during summer each year. The precipitation in 2016 in this catchment is shown in Fig. S1. The groundwater pumped from the wells (80–200 m depths) represents the main irrigation resource in this catchment. The averages of pH, electrical conductivity (EC), NO_3^--N and NH_4^+-N of well water were 7.83±0.13, 545±125 μs cm^{-1}, 7.32±6.11 mg L^{-1}, and 0.08±0.03 mg L^{-1}, respectively.

2.2 Study Methods

We studied the spatial variation of nitrate accumulation in soil profiles at the field scale and catchment scale in October 2016. To evaluate the N surplus in orchards and croplands, a field survey was conducted to collect soil samples.

To understand the spatial variation of nitrate accumulation at the field scale, we chose 6 kiwifruit orchards with 3°–5° gradients and 57 m slope lengths, on average, along the sides of the Yujiahe River. Soil samples were collected from both upslope and downslope sites of each sloping orchard. Three soil cores (0–400 cm depth and 20 cm depth intervals) were collected at the same elevation using a soil auger for both upslope and downslope sites within each orchard. The triplicate samples from each soil depth were mixed to form a composite sample for an upslope or downslope site, and the sample was packaged into a labelled sample bag and immediately stored at 4℃.

To study the nitrate accumulation in soil profiles at the catchment scale, we collected soil profile samples from the kiwifruit orchards of the upland, transition, and lowland zones in the catchment (Fig. 1). In each zone, three kiwifruit orchards located at high elevation and another three orchards located at low elevation were selected.

Therefore, 18 orchards that covered both sides of the river were chosen in this study. The soil profiles from three wheat-maize fields were also collected for comparison with those from kiwifruit orchards (Fig. 1). The soil sample depth and methods were similar to those used in the sloping orchards. The soil texture was clay loam. The average contents of sand, silt and clay in the soil profiles (0–100 cm) were 32.4–37.2%, 33.1–35.7%, and 29.7–30.2%, respectively.

The survey information for each field included the area, the application rates of synthetic fertiliser and manure, the irrigation rate and time, the kiwifruit yield, the biomass of vine pruning, and the grain and straw of wheat and maize. To calculate the total N removed by pruning and fruit harvesting from kiwifruit orchards, we selected 10 of the 24 orchards to determine the N concentration in fruits and branches during harvesting in early October 2016 and winter pruning in November 2016. Approximately 10–15 kiwifruit samples were collected from 10 kiwifruit orchards, from which all pruned branches were collected and weighed. The N concentrations of wheat and maize were cited from Lu *et al.* (2016), which was one of our other studies conducted in this catchment.

2.3 Sample analysis

Soil samples were extracted with 1 M KCl (soil: solution, 1:10) and shaken for 1 h followed by filtration. The mineral N concentrations of the KCl extract and groundwater samples were determined by a continuous-flow N analyser (Branand Luebbe AA3, Norderstedt, Germany). The water content of the soil samples was determined with the method of oven-drying at 105℃. The collected plant samples were placed in the oven at 70°C and dried to a constant weight. The dried samples were ground and passed through a sieve (0.25 mm). H_2SO_4 and 30% H_2O_2 were used to digest the crushed plant samples before determining the N concentration using the Kjeldahl method (Bao, 2000).

2.4 Calculation method

The apparent N surplus per unit area of soil in this catchment was calculated using the following equation (Oenema *et al.*, 2003; Lu *et al.*, 2016):

Apparent N surplus (kg N ha^{-1}) = inputs (inorganic fertiliser + manure + seed + deposition + irrigation) – outputs (nitrogen removed by straw and grain or by fruits and pruning).

The inputs of N from synthetic inorganic fertilisers and manure were calculated based on the N contents and the application rates of each fertiliser. The annual input from atmospheric N deposition was 28.9 kg ha^{-1}, as determined by Liang *et al*. (2014). There was no N input from irrigation in the wheat-maize rotations as a result of there being no irrigation. The N input to the kiwifruit orchards from irrigation in this catchment was 33.6 kg N ha^{-1}, which was calculated based on the N concentration of the groundwater and the annual irrigation rates in the orchards. The annual N input to the cereal fields from the seeds was 4.4 kg N ha^{-1}, which was calculated by multiplying the N content of the seeds by the sowing rate (Table 1). The N input from the other surface vegetation in the orchards was not considered because farmers usually keep the surface clean to prevent plants from competing for nutrients with kiwifruit vines, and the biomass of the vegetation was very low.

The output of N in kiwifruit orchards mainly consists of N removed by fruit harvest, vine pruning in winter, and N stored in kiwifruit vines. The amount of N removed by kiwifruit harvest was calculated based on the N concentrations of the dried kiwifruits, the water contents and the yields of the kiwifruit. The annual N stored in mature kiwifruit vines was estimated as 37.1 kg N ha^{-1} in this region, and this value was obtained from Wang & Tong (2008). The N removed by pruned vines was calculated using the N concentration (dry weight) of the kiwifruit vines, the total branch weight and the water content of pruned vines. The outputs of N by the wheat and maize harvest were calculated based on the N concentrations of grains and straw and their biomasses. The N losses from the wheat and maize residual straw and fallen leaves of the kiwifruit vines were not considered because the N was returned to the system.

The nitrate accumulation (kg N ha^{-1}) in soil was calculated using the following equation:

$$\text{Nitrate accumulation} = \frac{BD \times d \times Con_i}{10} \qquad (1)$$

where BD is the soil bulk density (g cm^{-3}) of different copping systems, *d* is the soil sampling depth (cm), and *Con* is the nitrate content in soil (mg N kg^{-1}) of the crop *i*. The soil bulk densities (BDs) of the top 60 cm were determined from 3 orchards and fields using samples collected at 20-cm depth intervals, and the BDs of the deep soil layers below 60 cm were considered the same as that of the 40–60 cm depth because of small variations in the deep layers (Yang *et al.*, 2015). The average

BD values in the orchards and fields of the 0–20, 20–40 and 40–60 cm soil depths were 1.28, 1.37, 1.38 g cm^{-3} and 1.37, 1.45, 1.47 g cm^{-3}, respectively.

The total N inputs of this catchment via the cropping system were calculated using the following equation:

$$\text{Total N inputs} = \sum_{i=1}^{n} N_{ratei} \times S_{croppingi} \quad (2)$$

where N_{ratei} is the input rate of N (kg N ha^{-1}) in cropping system i, $S_{croppingi}$ is the cultivated area (ha) of cropping system i, and n is the total number of cropping systems.

The total soil nitrate storage within the 0–400 cm depth of all cropping systems in this catchment was calculated as follows:

$$\text{Total nitrate storage} = \sum_{i=1}^{n} N_{accumulationi} \times S_{croppingi} \quad (3)$$

where $N_{accumulationi}$ is the average value of accumulated soil nitrate within the 0–400 cm depth (kg N ha^{-1}) of cropping system i, $S_{croppingi}$ is the cultivated area (ha) of cropping system i, and n is the total number of cropping systems.

2.5 Statistical analysis

The significance of fertiliser inputs, mineral-N contents and accumulations within the soil between two cropping systems and the mineral-N contents and accumulations within the soil at different locations were evaluated by analysis of variance (ANOVA) with SAS 9.0, followed by the least significant different (*LSD*) test for comparing the mean values at the 1% and 5% levels.

3 RESULTS

3.1 N balance and mineral N accumulation in orchards and cereal fields

Compared with wheat-maize fields, kiwifruit orchards had a much higher average inorganic N fertiliser application rate. Only 9.6% of the total N input of kiwifruit orchards was used for growing fruits and vines, thus leading to a high N surplus (1133 kg N ha^{-1}) (Table 1).

Table 1. Annual nitrogen balance in kiwifruit orchards and wheat-maize fields (kg N ha^{-1}).

Cropping system	Wheat-maize (n=3)	Kiwi-orchards (n=24)
Inputs		
Chemical N fertiliser	356±55a	978±435b
Manure N	-	212±155
Irrigation water N	-	33.6±12.6
Wet and dry deposition N	28.9±4.3	28.9±4.3
Seed N	4.4±0.4	-
Total input	385±55A	1253±440B
Outputs		
Plant remove	242±31A	120±22B
N balance (input-output)		
N surplus	147±47A	1133±440B

Note. N input consists of synthetic inorganic fertilisers, manure, atmosphere deposition, irrigation and seeds; N output in kiwifruit orchards consists of N removed by fruit harvest (54.8±21.9) and pruned branches (28.1±7.2) and N stored in kiwifruit wines. N output in wheat-maize fields is removed by grain and straw. Values are the means ± standard error. Different lowercase letters in a row indicate the significant differences by ANOVA least significant different (*LSD*) test at $p<0.05$, and uppercase letters in a row indicate the significant differences by ANOVA least significant different (*LSD*) test at $p<0.01$. There is no irrigation and manure application in wheat-maize fields. Perennial kiwifruit vines have no seed N input.

The NH_4^+-N contents and accumulation in the soil profiles (0–400 cm) were very low, and the differences between the orchards and fields were not significant (Fig. 2a and 2c). The soil nitrate contents in kiwifruit orchards were significantly higher than those in the fields, especially below the 40 cm soil depth. The average nitrate content was 50.35 mg N kg^{-1} at the 400 cm depth (Fig. 2b), indicating that nitrate leached into deeper soils. The nitrate accumulation in the soil profiles of kiwifruit orchards in the

0–100, 100–200, 200–300, 300–400 and 0–400 cm profiles were 739, 990, 809, 750 and 3288 kg N ha^{-1}, respectively. These values were significantly higher than the values for wheat-maize fields (58, 62, 37, 47 and 204 kg N ha^{-1}, respectively) (p<0.01). Approximately 77.5% of nitrate was distributed in soil depths deeper than 100 cm (Fig. 2d).

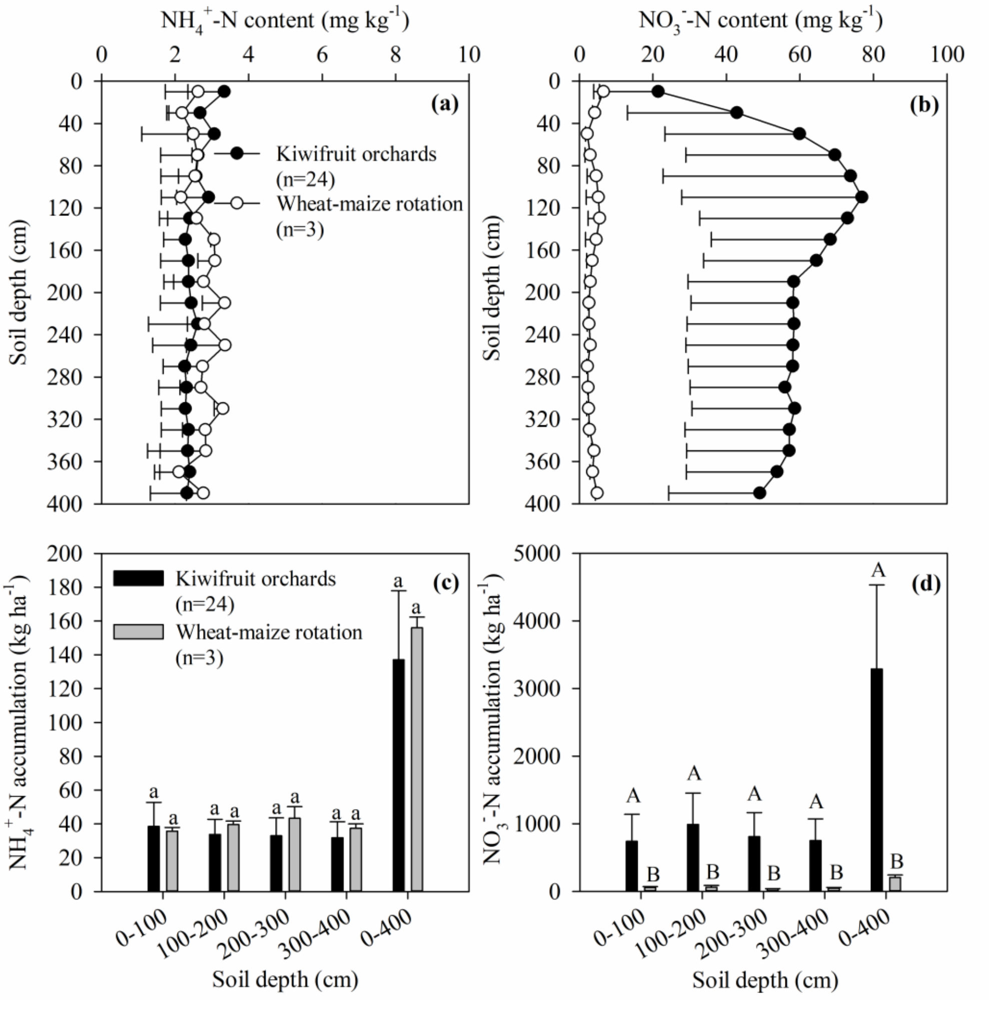

Fig. 2. The distribution (a-b) and accumulation (c-d) of mineral-N within the 0–400 cm soil depths in the kiwifruit orchards (n=24) and the wheat-maize fields (n=3). Note: the same lowercase letters in (c) indicate no significant differences between the two systems in the same soil depth by ANOVA least significant different (*LSD*) test at p<0.05; the different uppercase letters in (d) indicate that the significant differences between the two systems in the same soil depth by ANOVA least significant different (*LSD*) test at p<0.01. Error bars indicate the standard errors of the mineral-N concentration and accumulation.

3.2 Spatial variation in nitrate accumulation in sloping orchards

The soil nitrate contents at each depth within the 0–400 cm profile in the downslope of the sloping orchards were more than 2 times higher than those of the upslope ($p<0.05$) (Fig. 3a). The total nitrate accumulation in the soil profiles at the downslope was 5959 kg N ha^{-1}, which was higher than that at the upslope (3044 kg N ha^{-1}) ($p<0.05$) (Fig. 3b).

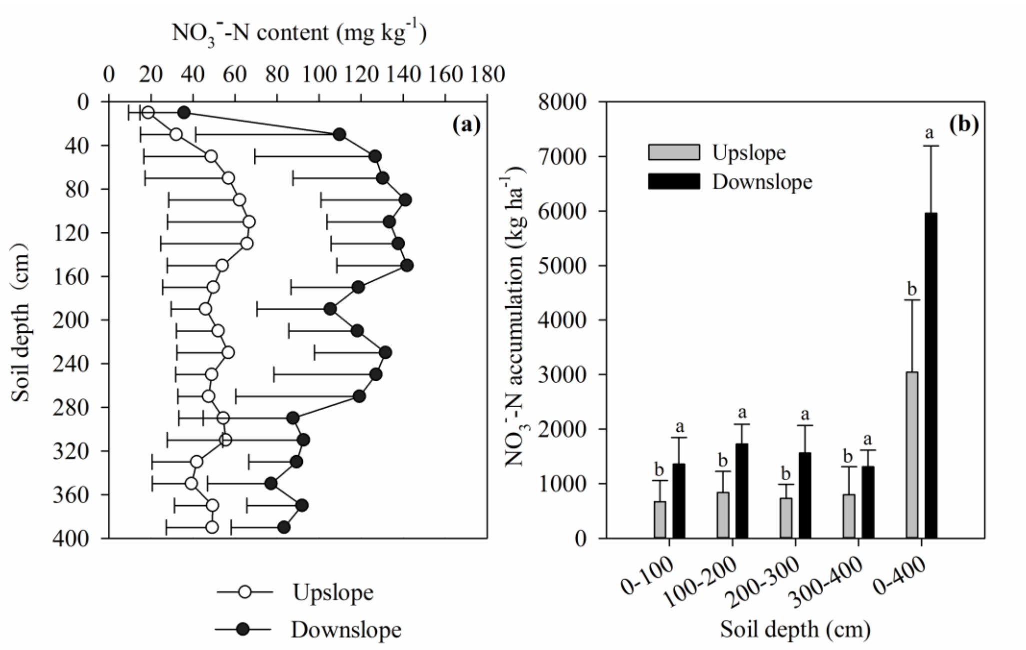

Fig. 3. The nitrate distribution (a) and accumulation (b) in the 0–400 cm soil profiles in kiwifruit orchards with different slopes (n=6). Note: the different lowercase letters in (b) indicate the significant difference between the nitrate accumulation at different slope positions in the same soil depth by ANOVA least significant different (*LSD*) test at $p<0.05$. Error bars indicate the standard errors of the nitrate concentration and accumulation.

3.3 Spatial variation in nitrate accumulation in the catchment

The average nitrate contents in orchard soils at low elevations were higher than those at high elevations in each zone of the catchment (Fig. 4a, 4b and 4c), especially in the upland zone (Fig. 4a). The average soil nitrate accumulation values in the 0–400 cm profiles of the low elevation in the upland, transition and lowland zones were 3946, 4111 and 4214 kg N ha^{-1}, which were 2.0, 1.4 and 1.5 times higher than those of the high elevation site, respectively (Fig. 4d, 4e and 4f).

The average nitrate contents in the upper 200 cm soil profile of the lowland zone were higher than those in the upland zone at the catchment scale (Fig. 5a). A significant difference was found in the soil nitrate accumulation within the 0–100 cm depth between the upland zone and the lowland zone (Fig. 5b). The average soil nitrate accumulation of the lowland zone was 688 kg N ha^{-1} higher than the value of the upland zone at a depth of 0–400 cm (Fig. 5b).

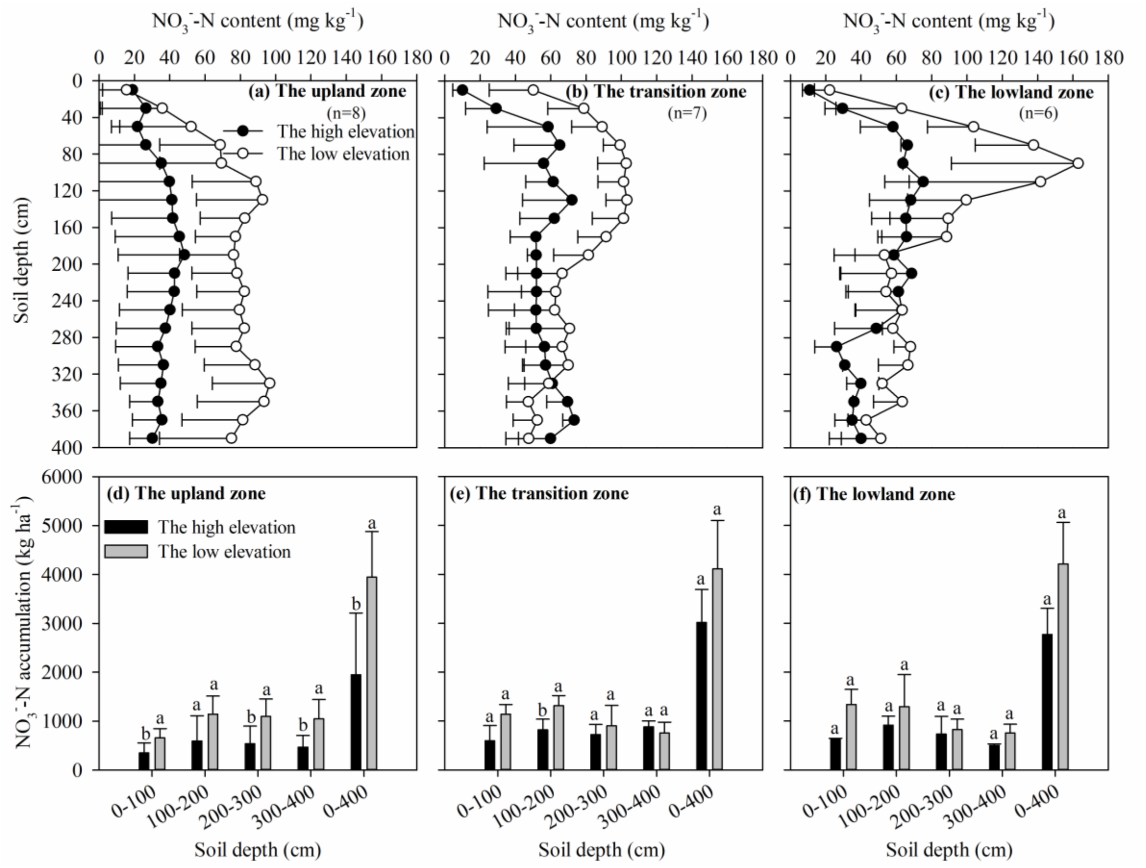

Fig. 4. The nitrate distribution (a, b and c) and accumulation (d, e and f) in the 0–400 cm soil profiles of kiwifruit orchards located in the different terrain areas. Note: the different lowercase letters in (d, e and f) indicate that the significant differences between the average nitrate accumulation at different terrain in the same soil depth by ANOVA least significant different (*LSD*) test at p<0.05. Error bars indicate the standard errors of the nitrate concentration and accumulation.

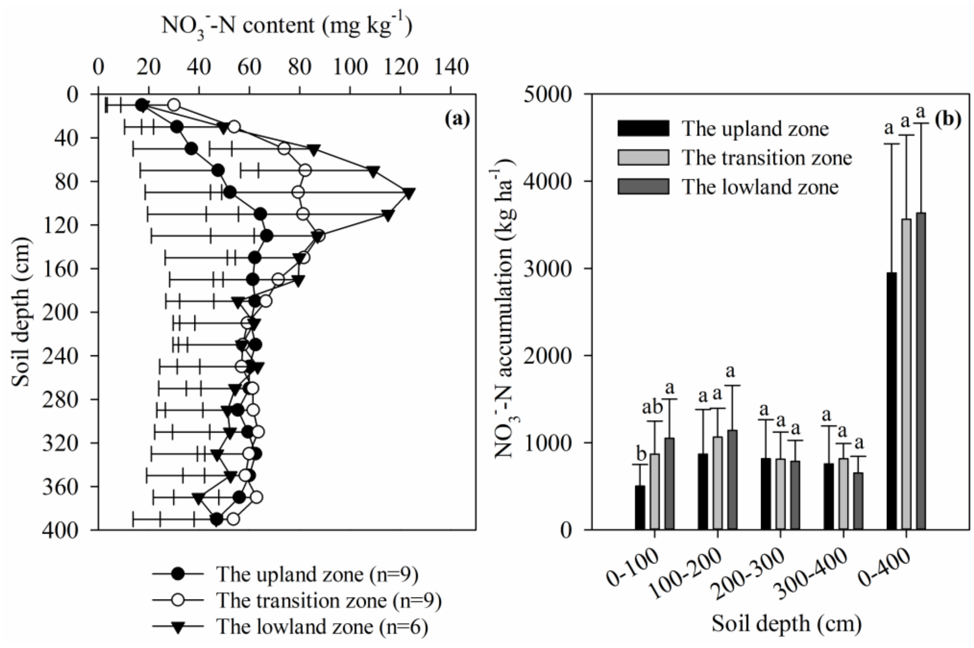

Fig. 5. The nitrate distribution (a) and accumulation (b) in the 0–400 cm soil depths of kiwifruit orchards in different areas of the Yujiahe catchment. Note: the different lowercase letters in (b) indicate that the significant differences between the nitrate accumulation at different areas in the same soil depth by ANOVA least significant different (*LSD*) test at $p<0.05$. Error bars indicate the standard errors of the nitrate concentration and accumulation.

3.4 Total nitrate accumulation in the catchment

The total nitrate accumulation in the catchment was 464.8 Mg N, which was 2.2 times higher than the total annual N inputs (213.2 Mg N). The nitrate accumulation in the kiwifruit orchards was estimated to be 94.8% (440.8 Mg N) of the total accumulation in the catchment (Table 2). The total soil nitrate accumulation within the 0–100 cm profile of the catchment was 105.9 Mg N, which was only 22.8% of that in the 0–400 cm soil profile.

Table 2. The nitrate accumulation in the 0–400 cm soil profile in this catchment.

Land-use categories	Proportion (%)	Area (ha)	No. of sampling sites	Average of nitrate accumulation (Mg N ha^{-1})	Total annual N input to catchment (Mg N)	Nitrate storage in catchment (Mg N)
Wheat-Maize	28.5	117.3	3	0.20	45.2	23.5
Kiwifruit Vine	32.5	134.1	24	3.29	168.0	440.8
Total	61.0	251.4	27		213.2	464.8

4 DISCUSSION

4.1 Higher nitrate accumulation in kiwifruit orchards than in cereal fields

Our study reveals that nitrate is the major mineral N form in the soil profiles of the catchment. This result might be related to the rapid nitrification in the upland soils (Liu *et al*., 2003). Compared with the cereal fields, a significantly high nitrate accumulation (3288 kg N ha^{-1}) was found within the 0–400 cm depth of kiwifruit orchards in the catchment. Similar results have been reported by other researchers (Lu *et al*., 2016; Qiu *et al*., 2010). A data mining study with more than 7000 samples in uplands of China reported that a large amount of soil nitrate accumulated within the 0–400 cm depth of horticultural systems, such as 1269 kg N ha^{-1} in solar plastic-roofed greenhouse vegetables and 2155 kg N ha^{-1} in orchards (Zhou *et al.*, 2016). The annual fertiliser inputs in intensive systems are almost 1.5 times higher than the recommended rates for the North China Plain (Ju *et al.*, 2006; Zhou *et al.*, 2010). Therefore, the over-application of N fertiliser is the main reason for the high nitrate accumulation in the soil profiles of intensive horticultural systems.

The average soil nitrate accumulation in the 0–400 cm depth of kiwifruit orchards in our study was higher than that in other studies of orchards in China (Ju *et al.*, 2006; Zhou *et al.*, 2016). This excessive accumulation can be explained by the overuse of N fertiliser in kiwifruit orchards in the study region. In an intensive survey at the catchment level, Lu *et al.* (2016) showed that the inorganic N application rate in kiwifruit orchards was approximately 2 times higher than the local recommended N rates (350–500 kg N ha^{-1}). Therefore, it is urgent to optimise the N application rate in kiwifruit orchards to reduce nitrate accumulation in the soil profiles of the study area.

Since the 1980s, the agricultural land use change from cereals to horticultural crops, such as fruit trees and greenhouse vegetables in China, has been substantial due to the high economic profit of the crops (Chen *et al.*, 2019). The planting areas of cereal crops decreased slightly, while the planting areas of fruit trees and vegetable crops increased drastically (Qiu *et al.*, 2010; National Bureau of Statistics of China, 2018). Therefore, higher nitrate accumulation in the soil profiles of horticultural systems can be expected. A larger scale is needed to understand the problem.

4.2 Spatial variation in nitrate accumulation at the catchment scale

There are many studies on nitrate accumulation in the soil profiles (vertical movement) of intensive horticultural systems in China (Ju *et al.*, 2006; Zhou *et al.*, 2010; Zhou *et al.*, 2016). However, few studies have evaluated the spatial variation (both vertical and lateral movement) in nitrate accumulation in the soil profiles of intensive horticultural systems at the catchment scale. Our results reveal the clear spatial variation in nitrate accumulation in the soil profiles of intensive horticultural systems at the field and catchment scales (Fig. 3, Fig. 4 and Fig. 5). For the same orchard, the nitrate accumulation within the 0–400 cm soil profile of the downslope was almost 2 times higher than that of the upslope (Fig. 3b). Similarly, the soil nitrate accumulation within the 0–400 cm profile in the lowland zone was also higher than that in the upland zone (Fig. 5b).

The movement of nitrate in the soils is dependent on the hydrological process (Shrestha *et al.*, 2010). The spatial variation in the soil nitrate accumulation at the catchment could be explained by the precipitation and irrigation patterns, the soil texture and the topography (Costa *et al.*, 2002; Gaines & Gaines, 1994; Maharjan *et al.*, 2014; Gheysari *et al.*, 2009). Approximately 61–84% of the annual precipitation in this region occurs between June and September. Moreover, all irrigation events (average 452 mm) occur between June and August for kiwifruit orchards. Our studies showed that nitrate moved up to 60 cm vertically in the soils after the rainy season (from May to October) (Gao *et al.*, 2016). Topography, which determines the pathways of surface and subsurface flows driven by hydraulic gradients, is another driving factor of lateral nitrate accumulation variability in the soil profile (Wang *et al.*, 2001; Zhu & Shao, 2008; Mei *et al.*, 2018). Elevation has a strong effect on soil moisture (Yang *et al.*, 2017); soil moisture increases from higher elevation to lower elevation (Bi *et al.*, 2009), and higher levels of soil nitrate content were observed at lower elevation sites (Lwiza *et al.*, 2016). In this study, area, the Yujiahe catchment is characterised by hilly topography, in which 85% of the arable lands are located on the slopes (2–15°) (Fig. 1). These special terrain characteristics combined with the intensive rainfall and heavy irrigation led to the lateral movement of nitrate on or near the surface, and hence, the spatial variation in soil nitrate accumulation was observed in this catchment.

4.3 Environmental risk of high nitrate accumulation in soil

The nitrate leaching out of the root-zone depth is regarded as an N loss from the perspective of plant nutrition. The depths of the root zones in the soil profiles depend on the crop, soil type and cultivation method (Fan *et al.*, 2016; Munoz-Romero *et al.*, 2010; Yao *et al.*, 2009). Compared to apple and peach trees in the study region, kiwifruit vines have relatively shallow root systems, with more than 90% of the root systems in the 0–60 cm soil depth (Fan & Yang, 2003; Wang *et al.*, 2010). If the root zone of kiwifruit vines is defined within the 0–100 cm soil depth, our study showed that more than 77.5% of the soil nitrate in the 0–400 cm profile accumulated out of the root zone (Fig. 2d), which could not be easily used by kiwifruit-vine roots. According to Hofman (1999), 90-100 kg N ha^{-1} of the soil nitrate accumulated in the soil depth of 0–100 cm after crop harvest is considered an environmental safety standard in Europe. However, the average total nitrate accumulation in the top 0–100 cm of soils of kiwifruit orchards was as high as 739 kg N ha^{-1}, which was significantly higher than the environmental safety standard in Europe. The average soil nitrate content at the 400 cm depth is 50.35 mg N kg^{-1} (Fig. 2b), thereby indicating there was nitrate leaching loss down into the 400 cm soil profile.

The total soil nitrate accumulation in the 0–400 cm profile of the catchment was 464.8 Mg N, which was 2.2 times higher than the total annual N input (213.2 Mg N). The depths of the groundwater levels of 11 shallow wells in the study catchment ranged from 6.3 to 18.6 m. Substantial amounts of nitrate accumulation in the vadose zone might leach into the groundwater and surface water, thus causing nitrate water pollution (Randall & Mulla, 2001; Burow *et al.*, 2010). Numerical studies have reported that the elevated nitrate in the surface or groundwater is mainly derived from the application of chemical N fertiliser or manure in intensive agricultural regions (Peng *et al.*, 2012; Thorburn *et al.*, 2003; Wang *et al.*, 2017). Therefore, a high potential risk of nitrate loss to fresh water is expected in this catchment.

A large amount of nitrate is stored within the vadose zones at the catchment and global scales (Ascott *et al.*, 2017; Wang *et al.*, 2013). The nitrate time lag in the soils and groundwater system (nitrate legacy) is considered a main reason preventing the achievement of water quality goals in many regions, such as in the Gulf of Mexico (Van Meter *et al.*, 2018). Additionally, in the UK, despite the efforts made under the European Water Framework Directive (Directive 2000/60/EC), the continuous deterioration of fresh water quality is still observed (Stuart *et al.*, 2007). The water quality targets set in 2001 have not been achieved in the Mississippi River basin due to the nitrate legacy (Van Meter *et al.*, 2018). Therefore, the massive accumulation of nitrate in the study area poses a long-term threat to the local fresh water quality. As a result, it is necessary to further investigate the mechanisms of N cycling and the transport pathways in and between the soils, groundwater and surface water at the catchment scale to develop practical measures to realise the sustainable development of both agricultural production and water quality management in this region.

4.4 Strategies to reduce the N fertilisation and nitrate accumulation

The land use change has led to a high nitrate environmental burden in the study area. Therefore, comprehensive measures are urgently needed to reduce the soil nitrate accumulation and the nitrate losses in this region. First, it is urgent to optimise the N application rate in orchards. Our three-year field experiments in the catchment (Lu *et al.*, 2018) showed that, compared to farmer's conventional N fertilisation, no adverse effects were found on the yield and quality of kiwifruit, with a 25% reduction of the N application rate in 2012–2014 and by 45% in 2014–2015, thereby increasing farmers' economic benefits and reducing nitrate accumulation in soil profiles. Hence, there is a large potential to reduce the N application rate in kiwifruit orchards without compromising crop production in the study region. Flooding irrigation is common in orchards. Fertigation is an effective practice to increase nutrient and water use efficiency (Martínez-Alcántara *et al.*, 2012; Quemada *et al.*, 2013; Siyal & Siyal, 2013). Therefore, adopting fertigation is an efficient way to decrease nitrate leaching in this region. Second, the farmers in the study region, as well as those in other parts of China, pay more attention to crop yield. They usually have less knowledge of how to use fertiliser rationally due to their low education level, and they have a less efficient agricultural extension system. Therefore, educating farmers and establishing more efficient agricultural extension systems are also important. Third, apart from the optimum application of N fertiliser, the measures for reducing nitrate lateral movement from kiwifruit orchards to streams are also needed to mitigate its loss at the catchment scale, such as building the dammed-up beam in the riparian zone. Finally, legislation is another way to limit excessive N inputs in the agricultural system (Appelgren & Burchi, 1993; Louwagie *et al.*, 2011). For example, the Nitrates Directive (91/676/EEC, 1991) has been implemented in Europe for 27 years to control N inputs in agricultural land from livestock effluents and mineral fertilisers (Zavattaro *et al.*, 2016). For small family farms in the study region, educating farmers is a practical way to reduce N inputs in intensive horticultural systems in China with the help of the latest communication techniques, such as the internet and mobile apps.

5 CONCLUSIONS

Our study shows that compared with cereal crops, the annual N surplus in kiwifruit orchards was severe, at 1133 kg N ha^{-1}, thus leading to a large amount of nitrate accumulation in the soils and vadose zones of the catchment. The nitrate stored in the 0–400 cm soil profiles of the catchment was 464.8 Mg N, which was 2.2 times higher than the annual N input (213.6 Mg N). The spatial variation in the nitrate accumulation in the soils was observed in this catchment. The nitrate accumulation in the downslope soils (5959 kg N ha^{-1}) was more than 2 times (significantly) higher than that in the upslope soils. The large amount of nitrate accumulated in the soils and vadose zones will eventually contaminate water bodies in the study area. Therefore, it is urgent to explore measures to reduce nitrate leaching from kiwifruit orchards. Measures of improving N fertiliser efficiency, e.g., straw mulching, building the dammed-up beam, fertigation, and legislation approaches, could be adopted in the study region. Meanwhile, the environmental protection awareness of the residents in the region needs to be improved. Finally, it is important for local policy makers to understand the legacy of the substantial nitrate stored in the catchment before exploring sustainable measures to provide safe drinking water and avoid nitrate-related environmental problems. Therefore, it would also be necessary to regularly monitor water quality and predict nitrate concentration trends in the study area via numerical modelling in the near future.

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REFERENCES

Ahmed, M., Rauf, M., Mukhtar, Z., & Saeed, N. A. (2017). Excessive use of nitrogenous fertilizers: an unawareness causing serious threats to environment and human health. *Environmental Science and Pollution Research*, *24*, 26983-26987. https://doi.org/10.1007/s11356-017-0589-7

Akhavan, S., Abedi-Koupai, J., Mousavi, S. F., Afyuni, M., Eslamian, S. S., & Abbaspour, K. C. (2010). Application of model to investigate nitrate leaching in Hamadan–Bahar Watershed, Iran. *Agriculture, Ecosystems & Environment*, *139*, 675-688. https://doi.org/10.1016/j.agee.2010.10.015

Appelgren, B. G., & Burchi, S. (1993). Technical, policy and legal aspects of chemical time bombs with emphasis on the institutional action required in eastern Europe. *Land Degradation & Development*, *4,* 437-440. https://doi.org/10.1002/ldr.3400040430

Ascott, M. J., Gooddy, D. C., Wang, L., Stuart, M. E., Lewis, M. A., Ward, R. S., & Binley, A. M. (2017). Global patterns of nitrate storage in the vadose zone. *Nature Communications*, *8,* 1416. https://doi.org/10.1038/s41467-017-01321-w

Bao, S. D. (2000). *Chemical Analysis in Soil and Plant*. China Agriculture Press, Beijing (in Chinese).

Bartoli, M., Racchetti, E., Delconte, C. A., Sacchi, E., Soana, E., Laini, A., ... & Viaroli, P. (2012). Nitrogen balance and fate in a heavily impacted watershed (Oglio River, Northern Italy): in quest of the missing sources and sinks. *Biogeosciences, 9*, 361-373. https://doi.org/10.5194/bg-9-361-2012

Bi, H. X., Li, X. Y., Liu, X., Guo, M. X., & Li, J. (2009). A case study of spatial heterogeneity of soil moisture in the Loess Plateau, western China: a geostatistical approach. *International Journal of Sediment Research*, *24*, 63-73. https://doi.org/10.1016/S1001-6279(09)60016-0

Burow, K. R., Nolan, B. T., Rupert, M. G., & Dubrovsky, N. M. (2010). Nitrate in groundwater of the United States, 1991−2003. *Environmental Science & Technology*, *44*, 4988-4997. https://doi.org/10.1021/es100546y

Chen, Z. J., Wang, L., Wei, A. S., Gao, J. B., Lu, Y. L., & Zhou, J. B. (2019). Land-use change from arable lands to orchards reduced soil erosion and increased nutrient loss in a small catchment. *Science of the Total Environment*, *648*, 1097-1104. https://doi.org/10.1016/j.scitotenv.2018.08.141

Chinese Soil Taxonomy Research Group. (2001). *Keys to Chinese soil taxonomy*. Press of University of Science and Technology of China, Hefei, China, 205-206 (in Chinese).

Clenaghan, C., Clinton, F. and Crowe, M., (2005). Phosphorus regulations: national implementation report, 2005. *Ireland: Environmental Protection Agency*.

Costa, J. L., Massone, H., Martınez, D., Suero, E. E., Vidal, C. M., & Bedmar, F. (2002). Nitrate contamination of a rural aquifer and accumulation in the unsaturated zone. *Agricultural Water Management*, *57*, 33-47. https://doi.org/10.1016/S0378-3774(02)00036-7

Fan, C. H., & Yang, X. L. (2003). Study on the roots distribution of Qinmei kiwifruit. *Shaanxi Journal of Agricultural Sciences*, 13-14 (in Chinese). https://doi.org/10.3969/j.issn.0488-5368.2003.05.006

Fan, J. L., McConkey, B., Wang, H., & Janzen, H. (2016). Root distribution by depth for temperate agricultural crops. *Field Crops Research*, *189*, 68-74. https://doi.org/10.1016/j.fcr.2016.02.013

Fang, Q. X., Yu, Q., Wang, E. L., Chen, Y. H., Zhang, G. L., Wang, J., & Li, L. H. (2006). Soil nitrate accumulation, leaching and crop nitrogen use as influenced by fertilization and irrigation in an intensive wheat–maize double cropping system in the North China Plain. *Plant and Soil*, *284*, 335-350. https://doi.org/10.1007/s11104-006-0055-7

FAOSTAT, Food and Agriculture Organization of the United Nations, Statistics division. 2018. http://www.fao.org/faostat/en/#data

Gaines, T. P., & Gaines, S. T. (1994). Soil texture effect on nitrate leaching in soil percolates. *Communications in Soil Science and Plant Analysis*, *25*, 2561-2570. https://doi.org/10.1080/00103629409369207

Gao, J. B., Lu, Y. L., Chen, Z. J., & Zhou, J. B. (2016). Nitrogen inputs and nitrate accumulation and movement in soil of kiwifruit orchards. *Journal of Agro-Environment Science*, *35*, 322-328 (in Chinese with English abstract). https://doi.org/10.11654/jaes.2016.02.016

Gao, J. J., Bai, X. L., Zhou, B., Zhou, J. B., & Chen, Z. J. (2012). Soil nutrient content and nutrient balances in newly-built solar greenhouses in northern China. *Nutrient Cycling in Agroecosystems*, *94*, 63-72. https://doi.org/10.1007/s10705-012-9526-9

Gheysari, M., Mirlatifi, S. M., Homaee, M., Asadi, M. E., & Hoogenboom, G. (2009). Nitrate leaching in a silage maize field under different irrigation and nitrogen fertilizer rates. *Agricultural Water Management*, *96*, 946-954. https://doi.org/10.1016/j.agwat.2009.01.005

Hakeem, K. R., Sabir, M., Ozturk, M., Akhtar, M. S., & Ibrahim, F. H. (2016). Nitrate and nitrogen oxides: sources, health effects and their remediation. *Reviews of Environmental Contamination and Toxicology 242*, 183-217. https://doi.org/10.1007/398_2016_11

Heisler, J., Glibert, P. M., Burkholder, J. M., Anderson, D. M., Cochlan, W., Dennison, W. C., ... & Lewitus, A. (2008). Eutrophication and harmful algal blooms: a scientific consensus. *Harmful Algae*, *8*, 3-13. https://doi.org/10.1016/j.hal.2008.08.006

Hofman, G. (1999). EU concerted action: nutrient management legislation in European countries. *NUMALEC Report, Fair*. http://www. uni-hohenheim. de/i3v

Ierodiaconou, D., Laurenson, L., Leblanc, M., Stagnitti, F., Duff, G., Salzman, S., & Versace, V. (2005). The consequences of land use change on nutrient exports: a regional scale assessment in south-west Victoria, Australia. *Journal of Environmental Management*, *74*, 305-316. https://doi.org/10.1016/j.jenvman.2004.09.010

Ju, X. T., Kou, C. L., Zhang, F. S., & Christie, P. (2006). Nitrogen balance and groundwater nitrate contamination: comparison among three intensive cropping systems on the North China Plain. *Environmental Pollution*, *143*, 117-125. https://doi.org/10.1016/j.envpol.2005.11.005

Laurent, F., & Ruelland, D. (2011). Assessing impacts of alternative land use and agricultural practices on nitrate pollution at the catchment scale. *Journal of Hydrology*, *409*, 440-450. https://doi.org/10.1016/j.jhydrol.2011.08.041

Li, J., He, Z. B., Du, J., Zhao, L. W., Chen, L. F, Zhu, X., ... & Tian, Q. Y. (2018). Regional variability of agriculturally-derived nitrate-nitrogen in shallow groundwater in China, 2004–2014. *Sustainability*, *10*, 1393. https://doi.org/10.3390/su10051393

Liang, T., Tong, Y. A., Lin, W., Qiao, L., Liu, X., Bai, S. C., & Yang, X. L. (2014). Spatial-temporal variability of dry and wet deposition of atmospheric nitrogen in different ecological regions of Shaanxi. *Acta Ecologica Sinica*, *34*, 738-745 (in Chinese with English abstract). https://doi.org/10.5846/stxb201211011517

Liu, X. J., Ju, X. T., Zhang, F. S., Pan, J. R., & Christie, P. (2003). Nitrogen dynamics and budgets in a winter wheat–maize cropping system in the North China Plain. *Field Crops Research*, *83*, 111-124. https://doi.org/10.1016/S0378-4290(03)00068-6

Louwagie, G., Gay, S. H., Sammeth, F., & Ratinger, T. (2011). The potential of European Union policies to address soil degradation in agriculture. *Land Degradation & Development*, *22*, 5-17. https://doi.org/10.1002/ldr.1028

Lu, Y. L., Chen, Z. J., Kang, T. T., Zhang, X. J., Bellarby, J., & Zhou, J. B. (2016). Land-use changes from arable crop to kiwi-orchard increased nutrient surpluses and accumulation in soils. *Agriculture, Ecosystems &*

Environment, *223*, 270-277. https://doi.org/10.1016/j.agee.2016.03.019

Lu, Y. L., Kang, T. T., Gao, J. B., Chen, Z. J., & Zhou, J. B. (2018). Reducing nitrogen fertilization of intensive kiwifruit orchards decreases nitrate accumulation in soil without compromising crop production. *Journal of Integrative Agriculture*, *17*, 1421-1431. https://doi.org/10.1016/S2095-3119(17)61899-9

Lwiza, M. L. (2016) *Assessment of nitrate levels in water and soils for agriculture and human utilization in Singida district, Tanzania* (Doctoral dissertation, Sokoine University of Agriculture). http://www.suaire.suanet.ac.tz:8080/xmlui/handle/123456789/1574

Maharjan, B., Venterea, R. T., & Rosen, C. (2014). Fertilizer and irrigation management effects on nitrous oxide emissions and nitrate leaching. *Agronomy Journal*, *106*, 703-714. https://doi.org/10.2134/agronj2013.0179

Martínez-Alcántara, B., Quiñones, A., Forner-Giner, M. Á., Iglesias, D. J., Primo-Millo, E., & Legaz, F. (2012). Impact of fertilizer-water management on nitrogen use efficiency and potential nitrate leaching in citrus trees. *Soil Science and Plant Nutrition*, *58*, 659-669. https://doi.org/10.1080/00380768.2012.733678

Marwick, T. R., Tamooh, F., Ogwoka, B., Teodoru, C., Borges, A., Darchambeau, F., & Bouillon, S. (2014). Dynamic seasonal nitrogen cycling in response to anthropogenic N loading in a tropical catchment, Athi–Galana–Sabaki River, Kenya. *Biogeosciences*, *11*, 443-460. https://doi.org/10.5194/bg-11-443-2014

Mei, X. M., Zhu, Q. K., Ma, L., Zhang, D., Wang, Y., & Hao, W. J. (2018). Effect of stand origin and slope position on infiltration pattern and preferential flow on a Loess hillslope. *Land Degradation & Development*, *29*, 1353-1365. https://doi.org/10.1002/ldr.2928.

Menció, A., Mas-Pla, J., Otero, N., Regàs, O., Boy-Roura, M., Puig, R., ... & Folch, A. (2016). Nitrate pollution of groundwater; all right…, but nothing else?. *Science of the Total Environment*, *539*, 241-251. https://doi.org/10.1016/j.scitotenv.2015.08.151

Min, L. L., Shen, Y. J., Pei, H. W., & Wang, P. (2018). Water movement and solute transport in deep vadose zone under four irrigated agricultural land-use types in the North China Plain. *Journal of Hydrology*, *559*, 510-522. https://doi.org/10.1016/j.jhydrol.2018.02.037

Munoz-Romero, V., Benítez-Vega, J., López-Bellido, L., & López-Bellido, R. J. (2010). Monitoring wheat root development in a rainfed vertisol: Tillage effect. *European Journal of Agronomy*, *33*, 182-187. https://doi.org/10.1016/j.eja.2010.05.004

National Bureau of Statistics of China. (2018). *China statistical yearbook 2018*. China Statistical Press, Beijing (in Chinese). http://www.stats.gov.cn/tjsj/ndsj/2018/indexch.htm

Obenour, D. R., Scavia, D., Rabalais, N. N., Turner, R. E., & Michalak, A. M. (2013). Retrospective analysis of midsummer hypoxic area and volume in the northern Gulf of Mexico, 1985–2011. *Environmental Science & Technology*, *47*, 9808-9815. https://doi.org/10.1021/es400983g

Oenema, O., Kros, H., & de Vries, W. (2003). Approaches and uncertainties in nutrient budgets: implications for nutrient management and environmental policies. *European Journal of Agronomy*, *20*, 3-16.

https://doi.org/10.1016/S1161-0301(03)00067-4

Peng, T. R., Lin, H. J., Wang, C. H., Liu, T. S., & Kao, S. J. (2012). Pollution and variation of stream nitrate in a protected high-mountain watershed of Central Taiwan: evidence from nitrate concentration and nitrogen and oxygen isotope compositions. *Environmental Monitoring and Assessment*, *184*, 4985-4998. https://doi.org/10.1007/s10661-011-2314-1

Qiu, S. J., Ju, X. T., Ingwersen, J., Qin, Z. C., Li, L., Streck, T., ... & Zhang, F. S. (2010). Changes in soil carbon and nitrogen pools after shifting from conventional cereal to greenhouse vegetable production. *Soil and Tillage Research*, *107*, 80-87. https://doi.org/10.1016/j.still.2010.02.006

Quemada, M., Baranski, M., Nobel-de Lange, M. N. J., Vallejo, A., & Cooper, J. M. (2013). Meta-analysis of strategies to control nitrate leaching in irrigated agricultural systems and their effects on crop yield. *Agriculture, Ecosystems & Environment*, *174*, 1-10. https://doi.org/10.1016/j.still.2010.02.006

Rabalais, N. N., Turner, R. E., & Scavia, D. (2002). Beyond Science into Policy: Gulf of Mexico Hypoxia and the Mississippi River: Nutrient policy development for the Mississippi River watershed reflects the accumulated scientific evidence that the increase in nitrogen loading is the primary factor in the worsening of hypoxia in the northern Gulf of Mexico. *BioScience*, *52*, 129-142. https://doi.org/10.1641/0006-3568(2002)052[0129:BSIPGO]2.0.CO;2

Randall, G. W., & Mulla, D. J. (2001). Nitrate nitrogen in surface waters as influenced by climatic conditions and agricultural practices. *Journal of Environmental Quality*, *30*, 337-344. https://doi.org/10.2134/jeq2001.302337x

Savci, S. (2012). An agricultural pollutant: chemical fertilizer. *International Journal of Environmental Science and Development*, *3*, 73. https://doi.org/10.7763/IJESD.2012.V3.191

Sebilo, M., Mayer, B., Nicolardot, B., Pinay, G., & Mariotti, A. (2013). Long-term fate of nitrate fertilizer in agricultural soils. *Proceedings of the National Academy of Sciences*, *110*, 18185-18189. https://doi.org/10.1073/pnas.1305372110

Shi, W. M., Yao, J., & Yan, F. (2009). Vegetable cultivation under greenhouse conditions leads to rapid accumulation of nutrients, acidification and salinity of soils and groundwater contamination in South-Eastern China. *Nutrient Cycling in Agroecosystems*, *83*, 73-84. https://doi.org/10.1007/s10705-008-9201-3

Stuart, M. E., Chilton, P. J., Kinniburgh, D. G., & Cooper, D. M. (2007). Screening for long-term trends in groundwater nitrate monitoring data. *Quarterly Journal of Engineering Geology and Hydrogeology*, *40*, 361-376. https://doi.org/10.1144/1470-9236/07-040

Shaanxi Provincial Bureau of Statistics. (2018). *The Shaanxi provincial fruit industry development statistics bulletin 2017 (monitoring)* (in Chinese). http://www.shaanxitj.gov.cn/site/1/html/126/132/141/17458.htm

Shrestha, R. K., Cooperband, L. R., & MacGuidwin, A. E. (2010). Strategies to reduce nitrate leaching into groundwater in potato grown in sandy soils: case study from North Central USA. *American Journal of Potato Research*, *87*, 229-244. https://doi.org/10.1007/s12230-010-9131-x

Siyal, A. A., & Siyal, A. G. (2013). Strategies to reduce nitrate leaching under furrow irrigation. *International Journal of Environmental Science and Development*, *4*, 431. https://doi.org/10.7763/IJESD.2013.V4.387

Thorburn, P. J., Biggs, J. S., Weier, K. L., & Keating, B. A. (2003). Nitrate in groundwaters of intensive agricultural areas in coastal Northeastern Australia. *Agriculture, Ecosystems & Environment*, *94*, 49-58. https://doi.org/10.1016/S0167-8809(02)00018-X

Van Meter, K. J., Basu, N. B., Veenstra, J. J., & Burras, C. L. (2016). The nitrogen legacy: emerging evidence of nitrogen accumulation in anthropogenic landscapes. *Environmental Research Letters*, *11*, 035014. https://doi.org/10.1088/1748-9326/11/3/035014

Van Meter, K. J., Van Cappellen, P., & Basu, N. B. (2018). Legacy nitrogen may prevent achievement of water quality goals in the Gulf of Mexico. *Science*, *360*, 427-430. https://doi.org/10.1126/science.aar4462

Wang, J., Fu, B. J., Qiu, Y., & Chen, L. D. (2001). Soil nutrients in relation to land use and landscape position in the semi-arid small catchment on the loess plateau in China. *Journal of Arid Environments*, *48*, 537-550. https://doi.org/10.1006/jare.2000.0763

Wang, J., & Tong, Y. A. (2008). Study on absorption, utilization and storage of nitrogen of kiwifruit tree. *Plant Nutrition and Fertilizer Science*, *14*, 1170-1177 (in Chinese with English abstract). https://doi.org/10.3321/j.issn:1008-505X.2008.06.023

Wang, J., Tong, Y. A., & Gao, Y. M. (2010). Study on the roots distribution and growth dynamics of kiwifruit in northern area of Qingling. *Journal of Anhui Agricultural Science*, *38*, 8085-8087 (in Chinese with English abstract). https://doi.org/10.13989/j.cnki.0517-6611.2010.15.139

Wang, L., Butcher, A. S., Stuart, M. E., Gooddy, D. C., & Bloomfield, J. P. (2013). The nitrate time bomb: a numerical way to investigate nitrate storage and lag time in the unsaturated zone. *Environmental Geochemistry and Health*, *35*, 667-681. https://doi.org/10.1007/s10653-013-9550-y

Wang, S. Q., Zheng, W. B., Currell, M., Yang, Y. H., Zhao, H., & Lv, M. Y. (2017). Relationship between land-use and sources and fate of nitrate in groundwater in a typical recharge area of the North China Plain. *Science of the Total Environment*, *609*, 607-620. https://doi.org/10.1016/j.scitotenv.2017.07.176

Yang, C. Y., Wu, D. C., & Chang, C. C. (2007). Nitrate in drinking water and risk of death from colon cancer in Taiwan. *Environment International*, *33*, 649-653. https://doi.org/10.1016/j.envint.2007.01.009

Yang, X. L., Lu, Y. L, Tong, Y. A., & Yin, X. F. (2015). A 5-year lysimeter monitoring of nitrate leaching from wheat–maize rotation system: comparison between optimum N fertilization and conventional farmer N fertilization. *Agriculture, Ecosystems & Environment*, *199*, 34-42. https://doi.org/10.1016/j.envint.2007.01.009

Yang, Y., Dou, Y. X., Liu, D., & An, S. S. (2017). Spatial pattern and heterogeneity of soil moisture along a transect in a small catchment on the Loess Plateau. *Journal of Hydrology*, *550*, 466-477. https://doi.org/10.1016/j.jhydrol.2017.05.026

Yao, S. R., Merwin, I. A., & Brown, M. G. (2009). Apple root growth, turnover, and distribution under different orchard groundcover management systems. *HortScience*, *44*, 168-175. https://doi.org/10.21273/HORTSCI.44.1.168

Zavattaro, L., Assandri, D., & Grignani, C. (2016). Achieving legislation requirements with different nitrogen fertilization strategies: results from a long term experiment. *European Journal of Agronomy*, *77*, 199-208. https://doi.org/10.1016/j.eja.2016.02.004

Zhou, J. Y., Gu, B. J., Schlesinger, W. H., & Ju, X. T. (2016). Significant accumulation of nitrate in Chinese semi-humid croplands. *Scientific Reports*, *6*, 25088. https://doi.org/10.1038/srep25088

Zhou, J. B., Chen, Z. J., Liu, X. J., Zhai, B. N., & Powlson, D. S. (2010). Nitrate accumulation in soil profiles under seasonally open ‘sunlight greenhouses’ in northwest China and potential for leaching loss during summer fallow. *Soil Use and Management*, *26*, 332-339. https://doi.org/10.1111/j.1475-2743.2010.00284.x

Zhu, Y. J., & Shao, M. A. (2008). Variability and pattern of surface moisture on a small-scale hillslope in Liudaogou catchment on the northern Loess Plateau of China. *Geoderma*, *147*, 185-191. https://doi.org/10.1016/j.geoderma.2008.08.012

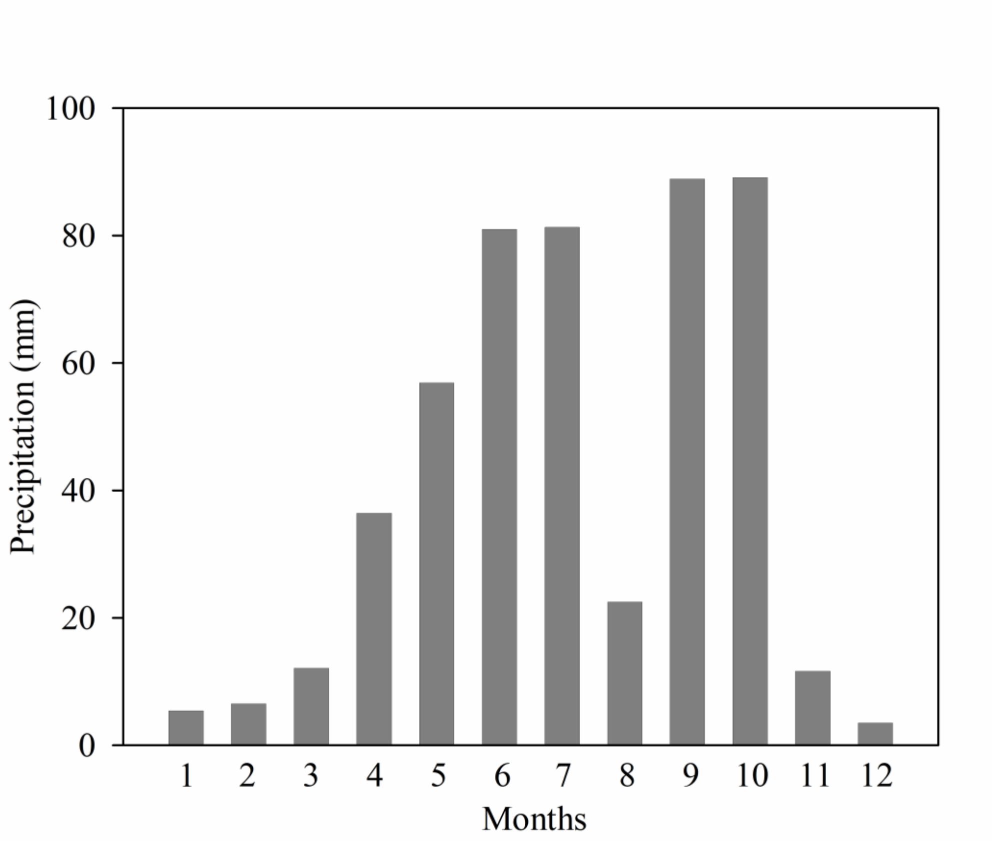

Fig. S1. The mean monthly precipitation for 2016 in this catchment