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ORIGINAL ARTICLE

Soil nitrate accumulation and leaching to groundwater during the entire vegetable phase following conversion from paddy rice

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Abstract Large amounts of farmland have been converted from traditional cereal cropping to intensive vegetable cropping in China, creating environmental risks. Previous studies utilized several methods of estimating nitrate accumulation in soil and leaching to groundwater under intensive vegetable cultivation, such as comparing intensive cropping to non-intensive cropping, or investigating the seasonal dynamics of soil nitrates in the short term, such as 1 or 2 years. This study tried to utilize a different method, considering the effects of total nitrogen (N) input during the whole vegetable cropping phase (up to 9 years) after conversion from traditional cereal cropping on soil and groundwater. A large-scale investigation was performed in the study area. Vegetable, soil, and groundwater samples were collected from 22 fields. Results showed that soil nitrate was significantly correlated with total N input from chemical fertilizer during the whole vegetable cropping phase, and groundwater nitrate was significantly correlated with soil nitrate. Nitrate concentration in 45 % of wells exceeded the drinking water maximum contaminant level, 10 mg L^{-1}. These results suggest that soil nitrate accumulation in the area resulted from long-term application of large amounts of N chemical fertilizer, and not from manure. Furthermore, high irrigation levels caused the accumulated nitrate in soil to leach into the groundwater, resulting in nitrate contamination of the groundwater.

Keywords Chemical fertilizer · N input · Nitrate accumulation · Nitrate leaching · Intensive vegetable cropping

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Introduction

Compared to cereal cultivation, vegetable cultivation in China can obtain greater annual yields and higher profits. Consequently, since the end of 1970s, large amounts of farmland have been converted from traditional cereal cropping to intensive vegetable cropping (Min et al. 2011). Based on FAO data, the cultivated area of vegetables increased tenfold from 1980 to 2012 (FAO 2013). Vegetable cultivation often requires a greater degree of management and larger input of N than cereal cultivation (Power and Schepers 1989). Over-application of N fertilizer for vegetable cultivation is very common in China (Chen et al. 2004), and entails high environmental risks (Ju et al. 2009). Excessive accumulation of nitrate in soil is unsustainable in the following ways. First, it decreases pH and increases the electrical conductivity (EC) in soil (Guo et al. 2010; Shi et al. 2009); second, greenhouse gas emissions caused by gaseous nitrous oxide losses from N fertilizer, as well as its production, is a major contribution to air pollution, contributes significantly to particulate matter with diameter smaller than 2.5 μm ($PM_{2.5}$) (Gu et al. 2014; Liu et al. 2013); finally, nitrate runoff and leaching is thought to be the principal source of fresh water eutrophication (Ongley et al. 2010; Zhang and Schilling 2006).

To estimate nitrate accumulation in soil caused by intensive cropping, many studies have compared soil nitrate levels under intensive cropping to those under non-intensive cropping. For instance, one study reported large amounts of nitrate accumulated in vegetable soils in comparison to cereal and orchard (Ju et al. 2007). Moritsuka et al. (2013) also compared vegetable cultivation to paddy cultivation to reveal greater accumulation of soluble nutrients caused by intensive cropping. In addition, some studies have investigated the seasonal dynamics of soil nitrate accumulation over the short term (1 or 2 years) under intensive cropping (Fang et al. 2006; Min et al. 2011; Shi et al. 2009). Given the tendency of farms in China to convert completely from traditional cereal cultivation to intensive vegetable cultivation, it would be useful to consider the effect of total input of N fertilizer during the whole vegetable cropping phase after conversion (i.e., several years rather than 1 or 2) on soil nitrate accumulation, but studies using this method are limited.

Due to the poor extension of rational fertilizer recommendations in areas with rapidly expanding production systems in China, farmers have applied fertilizer based on individual habits or inclinations (Sonntag et al. 2005), suggesting that the N application at different plots in the same intensive cropping system would be quite different. For instance, in Shouguang city, the largest vegetable production base in China, the N inputs from mineral fertilizer and organic fertilizer for a crop season ranged from 190 to 504 kg ha^{-1} and from 8 to 310 kg ha^{-1}, respectively (Song et al. 2009). The majority of farmers considered that high yield could be achieved with higher application of N fertilizer, even at levels that exceeded the crop's requirement, resulting in N loss (Li et al. 2015). Consequently, different total N fertilizer application during the whole vegetable cropping phase probably results in different conditions of nitrate accumulation in soil.

To study the effect of intensive cultivation on nitrate leaching, many studies have compared the nitrate concentration of groundwater in fields under intensive cropping to that in fields under non-intensive cropping (Ju et al. 2006; Morari et al. 2012; Du et al. 2011). In cropping systems of the same intensity, the different nitrate accumulations in soil caused by different N fertilizer application rates probably result in different amounts of nitrate leaching to groundwater, it should be feasible to study the relationship between soil nitrate and groundwater nitrate in greater detail.

A large-scale investigation had been performed in the study area, including vegetable cultivation and paddy rice cultivation (Wang et al. 2015). In this study, we try to (1) indicate that local vegetable cultivation is an intensive cropping system by comparing with paddy rice cultivation; (2) consider total N input from fertilizer during the whole vegetable cropping phase, and evaluate its impacts on nitrate accumulation in soil and nitrate leaching to groundwater.

Materials and methods

Site description

A study area located near the coastal area of southeast Lake Dianchi in Yunnan province, was selected (Fig. 1). The lake is one of the three highly eutrophic freshwater lakes in China (Liu and Qiu 2007). Before 2000, a paddy rice-broad bean system with low input of nitrogen, about 30 g m^{-2} $year^{-1}$, was dominant in this area. Since then, almost all paddy fields have been gradually converted to vegetable fields covered with vinyl houses. The cropping system in the area was multiple cropping of several kinds of leaf vegetables with a short growth duration (30–60 days), such as bok choy *Brassica rapa* L. var.*chinensis*), and stem lettuce (*Lactuca sativa* L.). A large amount of cow manure from exogenous sources with a C/N ratio 27 is typically applied as the base fertilizer in winter or at the beginning of spring. This C/N ratio should be adequate for composting (Bernal et al. 2009). About 0–4050 kg ha^{-1} $year^{-1}$ of urea and

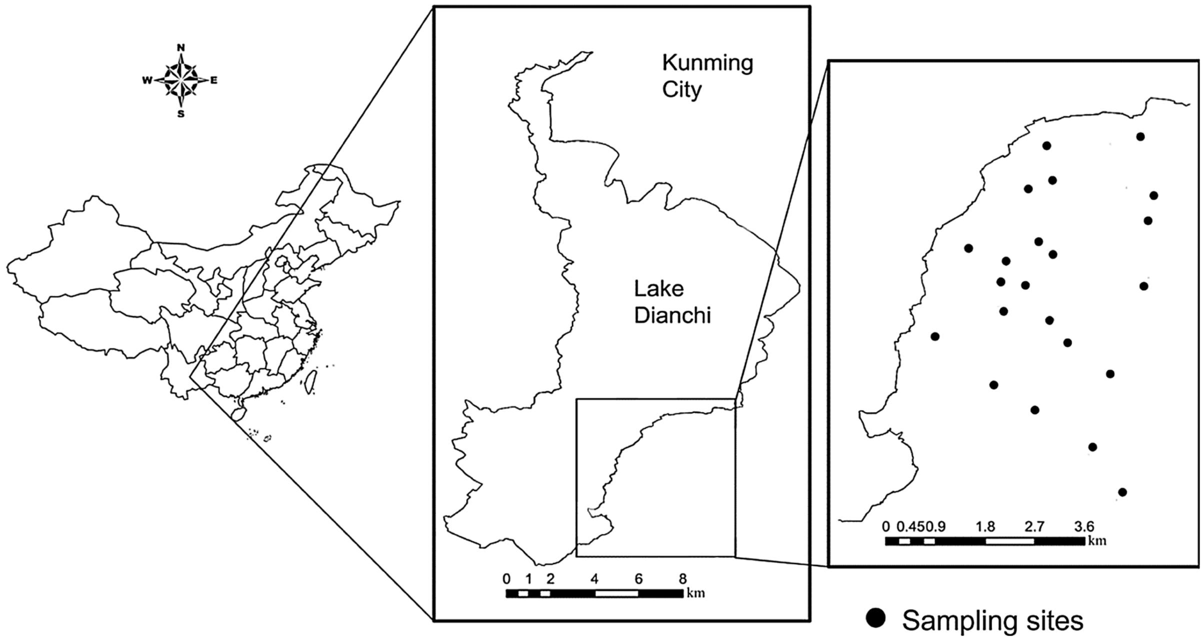

Fig. 1 Location of the study area and sampling sites

900–10,900 kg ha^{-1} year^{-1} of compound fertilizer is applied as topdressing for each crop.

The climate in the area is of the monsoon type; the annual precipitation is 980 mm; and the annual mean temperature is 15 °C. The soils have a clayey texture and a dull brown or dull reddish brown color and are classified as Eutric Cambisol (WRB soil classification) or Dystric Eutrochrept (USDA Soil Taxonomy) (Moritsuka et al. 2013).

Selection of sampling sites

Twenty-two vegetable fields under vinyl houses were selected depending on the annual application amount of N and vegetable cropping phase, and on the existence of a well in the field, with an effort to represent fields in the study area with different N management practices.

Crop management survey

Crop management details after conversion from paddy rice were gathered by interviews with farmers. The questions focused on the varieties of crops, the intensive cultivating duration of vegetable after conversion from paddy rice, the cultivation calendar, and the inputs (amount of chemical fertilizer and manure) in the selected fields.

Vegetable sampling and analyses

Vegetable samples were collected over 1 year from 2011 to 2012. The growth of vegetables at different sites in a vinyl house might be different; for instance, the vegetables near the entrance of the vinyl house might be slightly smaller than those in the center. Thus, we chose an area with average growth status for vegetable sampling. The aboveground biomass of vegetables that were mature enough to be harvested were sampled from three subsample points in each vegetable field. The area of a subsample point was about 0.5 m^2, and the three subsample points were uniformly distributed on the diagonal line of the average growth area. Then, the sampling area was measured accurately. The aboveground biomass of vegetables was sampled from three subsample points in each field at physiological maturity stage. Vegetable samples were oven-dried at 70 °C to a constant weight and ground until they were fine enough to pass through a 2-mm sieve. The plant sample was digested by the Kjeldahl method. Ammonium (NH_4^+) was determined using the cresol red method (cresol red dissolved in sodium hydroxide solution, and then mixed with Hepes, the final solution was used as an indicator) (Schulze et al. 1988) and flow-injection spectrophotometer (WIS-2000, HIRANUMA, Japan). The concentration of nitrate (NO_3^-) was determined using the Cataldo method (Cataldo et al. 1975) and spectrophotometer (U-1500, HITACHI, Japan).

Soil sampling and analyses

Just after plant sampling, surface soil (0–30 cm) samples were collected at biomass sampling points from three sites in each field. Soil nitrate concentrations in this layer (0–30 cm) should reflect the nitrate accumulating degree in the study area (Zhang et al. 2006; Ju et al. 2007, 2006). Soil samples from all three sites in the same field were mixed thoroughly to obtain composite samples. The composite samples were mixed with distilled water (1:5, w/v). The extract was used to determine soil nitrate by the Cataldo method (Cataldo et al. 1975), using a spectrophotometer (U-1500, HITACHI, Japan).

Groundwater sampling and analyses

Each well tested was located inside the corresponding vegetable field, and hence could be presumed to reflect the effects of intensive cropping practices on groundwater. Groundwater was simultaneously sampled with soil sampling, and was then kept in 550-mL polyethylene bottles, sealed and frozen at −35 °C until further use. Samples were filtered through 0.45 µm filter (670-12540-12, SHIMADZU, Japan), and the concentration of nitrate was determined by ion chromatography (PIA-1000, SHIMADZU, Japan).

Data of paddy rice cultivation

Few fields were cultivated with paddy rice for farmers' own consumption. Therefore, the paddy rice cultivation data collected from our previous study were used (Wang et al. 2015).

Manure data

Total N concentrations in manure were cited from *Fertilization Guidance for Main Crops in China* (Zhang et al. 2009).

Calculation of annual N balance and total N input during the whole vegetable cropping phase

The annual N balance was calculated as:

$$\begin{aligned}\text{Annual N balance} &= \text{annual N input from chemical fertilizer} \\ &\quad + \text{annual N input from manure} \\ &\quad - \mathit{annualNoutputthroughcropharvesting}\end{aligned}$$

After 2000, there was almost no legume cultivated as main crop or cover crop in local fields. Thus, in the equation of the annual N balance, we didn't take into account N inputs from biological nitrogen fixation. From the result of the above-mentioned interview, local farmers applied fertilizer to vegetables based on individual habits, and the application amount of fertilizer was similar from year to year; thus, we calculated the total N input during the whole vegetable cropping phase as:

$$\begin{aligned}&\text{Total N input during the whole vegetable cropping} \\ &\quad \text{phase} = \text{Annual N balance} \\ &\qquad \times \text{vegetable cropping years after conversion} \\ &\qquad \text{from paddy rice}\end{aligned}$$

Statistical analysis

The significance of annual N input, annual N output, and annual N balance between two cropping systems were analysed by *t* test. The significance among soil nitrate under different vegetable cropping phases were tested by analysis of variance and mean values were compared by least significant difference (LSD) at the 5 % level.

Pearson correlations were calculated to analyse the relationships. All of the analyses were performed with SPSS software (Ver. 16.0 for windows, SPSS Inc., USA).

Results

Intensive cultivation and annual N balance in the study area

The cultivating phases in six vegetable fields were less than or equal to 3 years, those in eight fields ranged from 4 to 6 years, and those in eight fields ranged from

7 to 9 years. Only one crop was cultivated in paddy fields per year in the study area; in contrast, cultivating intensities in 22 vegetable fields ranged from three to seven with an average value of six per year (Table 1). The annual N inputs, N outputs and N balances in vegetable fields were significantly more than those in paddy fields. The annual N inputs were much more than the annual N outputs in vegetable fields, resulting in positive N balances. The annual N inputs from chemical fertilizer, N inputs from manure, and N balances in vegetable fields varied widely, ranging from 89.4 to 2670.8, from 18.5 to 1115.9, and from 89.4 to 2125.7 g m^{-2} $year^{-1}$, respectively. Annual N balance was significantly correlated with annual N input from chemical fertilizer (Fig. 2). The outlier point showed a high annual N balance (161 g/m^2) for a rather low annual chemical N input (82.5 g/m^2), due to extremely large amount of applied cow manure (139.5 g/m^2).

Effect of intensive cultivation on nitrate accumulation in soil

Soil nitrate under 7–9 years of vegetable cropping was significantly higher than that under 1–3 years (Table 2), implying that the soil with a long history of intensive cropping would accumulate more nitrate. The standard deviation of soil nitrate under 4–6 years and 7–9 years was high, reflecting the farmers' wide-ranging fertilizing practices (Table 1).

Soil nitrate was not affected by annual N input from chemical fertilizer and annual N input from manure (Fig. 3a, b). Soil nitrate was significantly correlated with the total N input from chemical fertilizer during the whole vegetable cropping phase; the regression coefficient was 0.23, and the regression constant was 41.94. Under the application of different amounts of N

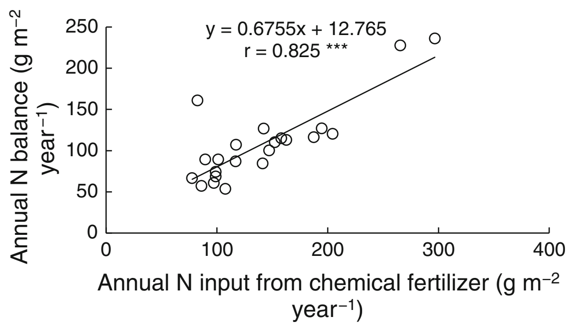

Fig. 2 Relationship between annual N input from chemical fertilizer and annual N balance (n = 22). *** significant at $P < 0.001$

chemical fertilizer, soil nitrate showed a high variance, ranging from 18.6 to 737.4 mg kg^{-1} (Fig. 3c). However, the correlation between total N input from manure during the whole vegetable cropping phase was not significant (Fig. 3d).

Different nitrate concentrations in groundwater caused by different nitrate concentration in soil

Nitrate concentrations in soil and in groundwater showed a strongly significant correlation (Fig. 4). With the increase of soil nitrate, groundwater nitrate increased from 0.3 to 40 mg L^{-1}. Ten of the 22 wells exceeded 10 mg L^{-1}.

Discussion

Nitrate accumulation in soil and effective treatment

Only four paddy fields could be found in the study area, thus we collected and calculated the cultivation

Table 1 Cropping systems in paddy and vegetable fields

	Paddy rice cultivation (n = 15)	Vegetable cultivation (n = 22)
Cultivating intensity (crops $year^{-1}$)	1	6 ± 1.2
Annual N input (g m^{-2} $year^{-1}$)		
Chemical fertilizer	30.0 ± 7.6a	142.1 ± 58.6b
Manure	0.8 ± 1.6a	42.8 ± 29.4b
Annual N output through crop harvesting (g m^{-2} $year^{-1}$)	9.6 ± 0.9a	76.1 ± 25.6b
Annual N balance per year (g m^{-2} $year^{-1}$)	21.2 ± 7.1a	108.8 ± 48.0b

Mean ± SD within each row, different letters indicate significant differences ($P < 0.05$)

Table 2 Soil nitrate under different vegetable cropping phases after conversion from paddy rice

	Vegetable cropping phase after conversion from paddy rice (year)		
	1–3	4–6	7–9
Soil nitrate (mg kg^{-1})	118.0 ± 22.7a	191.9 ± 197.0ab	335.9 ± 202.5b

Mean ± SD within each row, different letters indicate significant differences ($P < 0.05$)

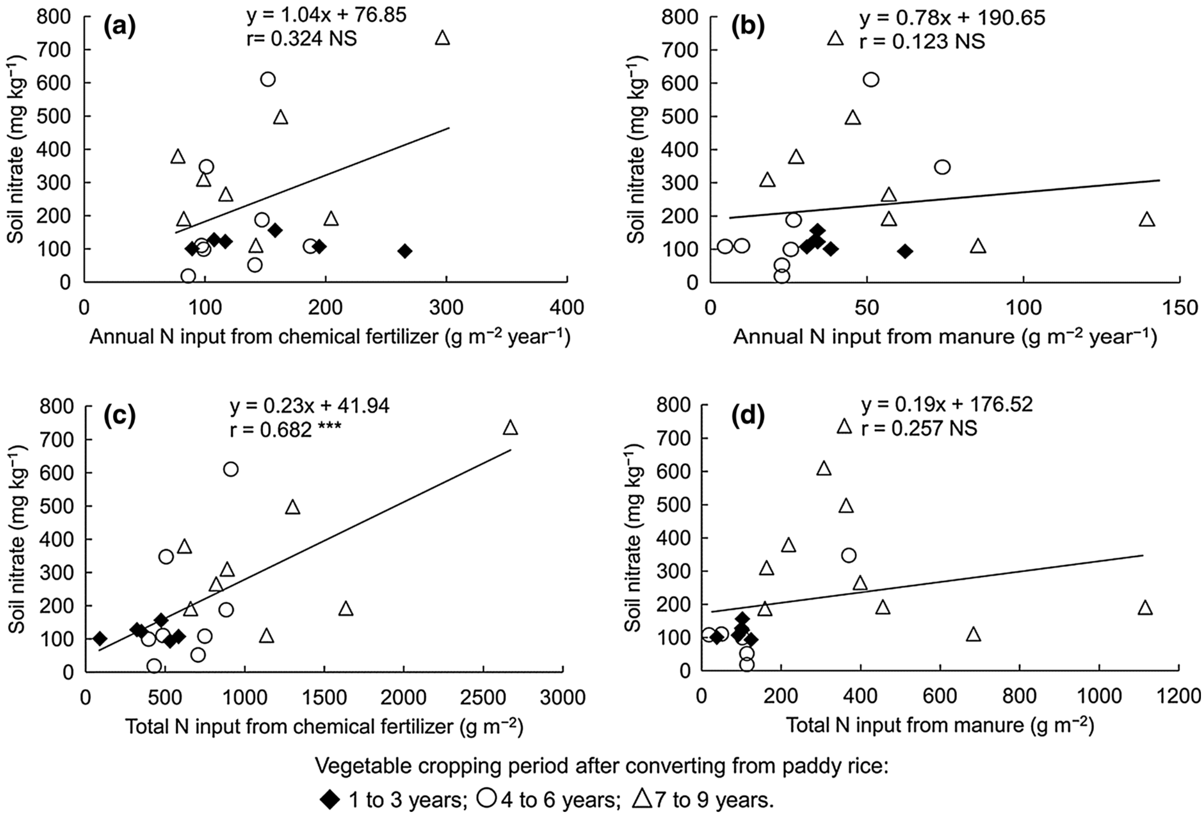

Fig. 3 Relationships between annual N input and nitrate concentration in soil, and between total N input during the whole vegetable cropping phase and nitrate concentration in soil (n = 22). *NS* not significant, *** significant at $P < 0.001$

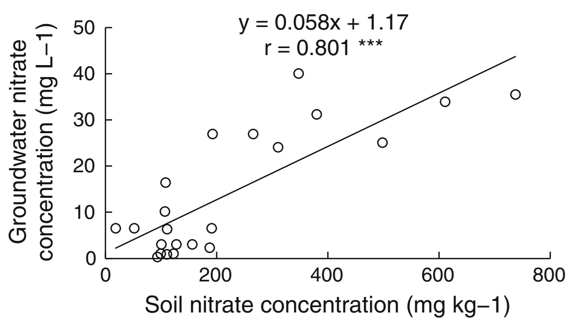

Fig. 4 Relationship between nitrate concentration in soil and in groundwater (n = 22). *** significant at $P < 0.001$

data in ten of the main paddy rice cropping regions in China from published works, and the cropping intensities, N inputs from chemical fertilizer, N inputs from livestock manure, annual N outputs, and annual N balances were 1 or 2, 23.6 ± 6.5, 0, 9.9 ± 5.4, and 13.8 ± 3.0, respectively (Mean ± SD, detailed data not shown) (Zhang et al. 2016; Li et al. 2009a; Li et al. 1998; Roelcke et al. 2002; Zhao et al. 2009; Liu et al. 2015; Tan et al. 2011; Xue et al. 2011; Ju et al. 2009; Zhao and Sha 2014). Compared to paddy rice cultivation in these regions or in the study area (Table 1), vegetable cultivation in the study area was an intensive cropping system, with high cropping intensity and high application of fertilizer. The highly positive annual N balance in vegetable fields indicated that a large amount of N remains in the agroecosystem (Table 1), and the unutilised soil N would continue to rise with the increasing input of N from chemical fertilizer (Fig. 2), thereby increasing the threat to the surrounding environment.

Nitrate accumulation in soil is mainly caused by large application of N chemical fertilizer, not by application of manure (Fig. 3c, d). The nitrate accumulation is a result of long-term successive chemical fertilizer use (Fig. 3a, c). The regression coefficient of the linear regression equation in Fig. 3c reflects nitrate accumulation ratio from chemical fertilizer, suggesting that about 23 % of total N from chemical fertilizer during the whole vegetable cropping phase remains in the soil, while the rest is removed through vegetable harvesting, lost through interflow (Wang and Zhu 2011) or overland flow under heavy irrigation, or lost as gas (Ju et al. 2009). The regression constant reflects the soil nitrate concentration before intensive vegetable cultivation, and it should be similar to the soil nitrate concentration in paddy fields, which are cultivated with a low N input and low N balance. We determined the soil nitrate concentration in paddy fields in the previous study, which ranged from 28 to 60 mg kg^{-1} with an average value of 44 mg kg^{-1} (Wang et al. 2015). The regression constant, 41.94, was close to 44, which indicated that the results obtained from Fig. 3c are close to reality.

Current intensive vegetable cropping systems have a highly positive N balance (Table 1), and our previous studies pointed out that reducing chemical N input was considered to be the best method for reducing the high N balance and the associated accumulation of soil nitrate (Wang et al. 2015, 2016). Moritsuka et al. (2013) reported that the amount of available N accumulated in the greenhouse surface soils (0–10 cm) exceeded 5000 kg ha^{-1} in the study area. Given the high reserves of background soil N, reducing chemical N input was unlikely to affect vegetable yields (Wang et al. 2016). However, at some point in the future when soil available N was excessively consumed, the reduction in chemical N input may result in a negative N balance where yields would be impacted, at which time chemical N input would need to be reassessed.

Nitrate leaching to groundwater and potentially effective treatment

Previous research has found that groundwater nitrate in the study area can vary widely, suggesting that the underlying groundwater aquifer systems were localised and not regional (Shi et al. 2005). The localised groundwater systems help to explain the significant correlation between soil nitrate built up in the vegetable fields and the contamination of nearby wells (Fig. 4).

The high accumulation of soil nitrate resulted in about 45 % of wells exceeding the World Health Organization's maximum contaminant level, of 10 mg L^{-1} of nitrate for drinking water (Fig. 4). If current farmers' practices of continuous over fertilising with chemical N remain in place, it is likely that more wells will suffer from nitrate contamination.

The principles for managing nitrate leaching include synchronizing N application with N sinks (Meisinger and Delgado 2002). Some studies have pointed out that soils in vegetable fields in China accumulated very high levels of nutrient under application of high levels of fertilizer (Chen et al. 2014; He et al. 2006; Moritsuka et al. 2013). According to the regression equation in Fig. 4, nitrate in ground water exceeded 10 mg L^{-1} when soil nitrate exceeded 152.24 mg kg^{-1}, suggesting that reducing soil nitrate lower than this level might ensure the safety of groundwater from nitrate contamination. Thus, decreasing the N input–output balance by reducing the input of N from fertilizer not only decreases soil nitrate but also might mitigate nitrate leaching to the groundwater.

Tanaka et al. (2013) reported that nitrate concentration in surface water was observed to have comparatively strong correspondence to the 3-day cumulative precipitation before the sampling date in our study area. But the fact that this kind of relationship was not observed between the cumulative precipitation (1–30 days) before sampling and the concentration of nitrate in groundwater (data not shown) was probably because 91 % of this area is covered by vinyl greenhouses (Anzai et al. 2011). Thus, irrigation should be an important factor in nitrate leaching in this area. Many reports pointed out that high irrigation is an obvious characteristic of vegetable production, which may promote nitrate leaching out of the root zone and into the subsoil or groundwater (Guimera 1998; Kraft and Stites 2003). Although we did not monitor the amount of irrigation at each selected field, we installed a water meter in one field, which should be partly representative of the study area. An average of 848 mm water was applied to each crop in the field. In a previous study in the main vegetable production area of China, on average 887 mm of water was applied to each crop, and 51 % of the nitrate contained in the leached irrigation water had originated from mineral fertilizer sources (Song et al. 2009). Although the present climate and soil environments were different from those in the aforementioned area, a considerable amount of soil nitrate in the study area can be expected to leach to groundwater through irrigation water. It has already been pointed out that improving water use efficiency would be a useful approach to decrease nitrate leaching (Dole et al. 1994). The local irrigation methods are primarily sprinkler irrigation, where water is irrigated from fixed sprinklers mounted at the top of vinyl greenhouses. Utilizing root-zone drip irrigation, which has been adopted elsewhere in China, and found to improved water use efficiency, and may also reduce associated nitrate leaching into groundwater (Deng et al. 2006).

Application of this study

This study should have universal application in other similarly eutrophic agricultural areas. For instance, the continuous conversion from paddy fields to vegetable fields has also taken place in the coastal areas of Lake Taihu and Lake Chaohu, the other two highly eutrophic freshwater lakes in China (Li et al. 2009b, 2014). Consideration of the total nutrient balance during the whole cultivating phase of vegetables, should be useful for evaluating the effect of intensive cropping on the agroecosystem in these areas.

Conclusion

We studied the effects of total N input after conversion from paddy rice to vegetable cropping on nitrate concentration in soil and groundwater. Long-term application of large amounts of N chemical fertilizer, and not manure, resulted in positive N balance and associated nitrate accumulation in soil and subsequent leaching to groundwater. We recommend that current N chemical fertilizer rates applied to intensive vegetable cropping systems in the Yunnan province should be reduced, to mitigate nitrate contamination of underlying groundwater aquifers. In the short term, the reduction in N chemical input is unlikely to negatively impact on vegetable yields, due to the current large reserves of soil nitrate. This study provided a different approach to evaluate nitrate accumulation in soil and leaching to groundwater under intensive cropping, which should have universal application in other similar eutrophic agricultural areas.

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