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Short communication

Wet deposition N and its runoff flow during wheat seasons in the Tai Lake Region, China

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ABSTRACT

Interest in the study of wet deposition N continues to grow for its impact on terrestrial ecosystems. Assessing the contribution of wet deposition N to intensive croplands is significant for recommending N fertilizers and minimizing environmental hazards. NH_4^+–N, NO_3^-–N, and total N concentrations in wet deposition, seasonal deposition N fluxes, and N loads in event-based runoff were determined for four consecutive wheat seasons in the Tai Lake Region. Wet deposition N during wheat seasons ranges from 11 to 15 kg ha^{-1} (average, 13 kg ha^{-1}), 61% of which is in the form of NH_4^+–N. NH_4^+–N concentrations range from 0.3 to 8.5 mg N L^{-1} (average, 1.7 mg N L^{-1}), showing greater temporal variations than do those of NO_3^-–N, which range from 0.2 to 4 mg N L^{-1} (average, 0.8 mg N L^{-1}). NH_4^+–N concentration in event-based runoff from wheat fields is much lower than that in rainfall, whereas that of NO_3^-–N is equal to or higher than that in rainfall. N loss through runoff reaches 1.8 kg N ha^{-1}, accounting for 14% of wet deposition N. Results indicate that NH_4^+–N in wet deposition is prone to immobilization by the soil–crop system, whereas NO_3^-–N is relatively easily lost to runoff. The contribution of 11 kg ha^{-1} wet deposition N should be adjusted in fertilizer N recommendations for winter wheat cultivation in this region.

1. Introduction

With the rapid development of the agricultural and industrial sectors in recent decades, the amount of anthropogenic reactive nitrogen (N) has sharply increased, and atmospheric N deposition has become an important component (having reached 70 Tg N yr^{-1}) of the global N cycle as a consequence of increasing anthropogenic reactive N emissions (Zhu and Chen, 2002; Galloway et al., 2004). Elevated levels of wet deposition N, causing eutrophication and acidification, considerably burden various ecosystems, such as forests, grassland, and aquatic ecosystems (Goulding et al., 1998; Bouwman et al., 2002; Stevens et al., 2004).

Wet deposition N primarily exists in the form of ammonium (NH_4^+) and nitrate (NO_3^-) produced by the dissolution of atmospheric NH_3 and HNO_3. Most studies on wet deposition N have focused on its impacts and its flow in natural or semi-natural ecosystems (Providoli et al., 2006; Stevens et al., 2010). For example, an ^{15}N-labeled experiment showed that 2–6% of added deposited NO_3^- and less than 1% of deposited NH_4^+ are present in runoff flow in N-limited forests and meadow ecosystems (Providoli et al., 2005). However, minimal information on the contribution of N deposition to intensive agricultural ecosystems is available (Fahey et al., 1999; He et al., 2010). Deposited N can supplement the supply of available N to agricultural crops—a feature that should be considered when calculating N fertilizer requirements for crop cultivation (He et al., 2007; Mei and Zhang, 2007; Xie et al., 2008; He et al., 2010). Therefore, an evaluation of the contribution of wet deposition N to agricultural fields is important in recommending N fertilizers, improving N use efficiency, and minimizing environmental hazards brought by N loss in croplands.

The Tai Lake Region (TLR) is one of the most important industrially and agriculturally developed regions in China. In this region, winter wheat is a prevalent crop after irrigated rice (Cao and Zhang, 2004; Xiong et al., 2006). The rapid development of the industry, excessive use of chemical N fertilizers, as well as the increasing amount of human and livestock excreta, enhance NO_x and NH_3 emissions; hence, HNO_3 and NH_4^+ deposition may also be enhanced correspondingly (Richter and Roelcke, 2000; Xing and Zhu, 2002). The characteristics and sources of wet deposition N in this region have been documented (Wang et al., 2004; Xie et al., 2008; Zhao et al., 2009). Wang et al. (2004) reported annual wet depositions of up to 27 kg N ha^{-1} in the TLR. However, few studies have focused on the retention of wet deposition N by the soil–crop ecosystem, and which form of wet deposition N is likely to be retained or lost through water flow when precipitation occurs remains unelucidated. The field in the TLR is surrounded by a ridge (15–20 cm high) during the rice growing season and covered by a water layer (3–5 cm) during most of the rice season (Tian et al., 2007). Hence, runoff during the rice season will not occur unless rainfall exceeds 120 mm, which is a low probability event in this region. Therefore,

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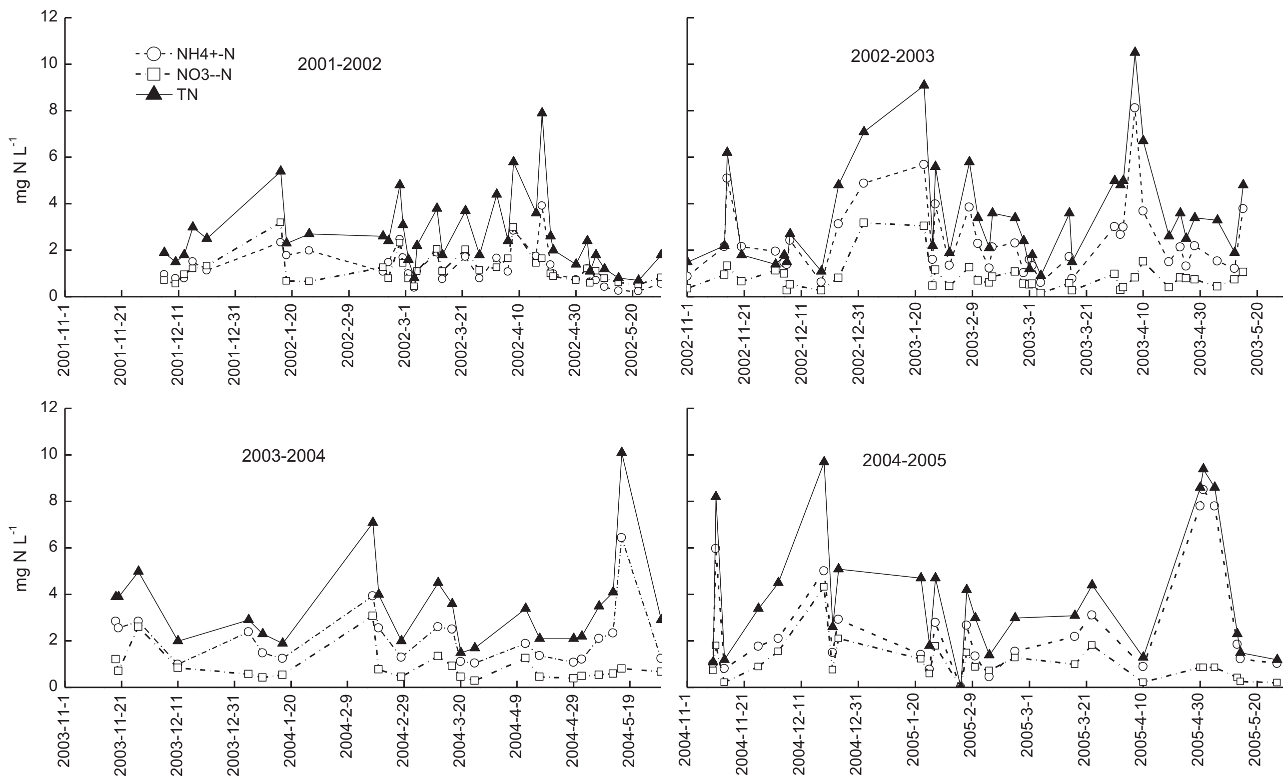

Fig. 1. Changes in $NH_4{}^+$–N, $NO_3{}^-$–N, and TN concentrations in rainwater during the 2001–2002, 2002–2003, 2003–2004, and 2004–2005 wheat seasons.

the winter wheat season was chosen in this study to investigate the wet deposition of N and its rough flow by runoff. This study aims to (1) monitor wet deposition N input and the strength of $NH_4{}^+$ and $NO_3{}^-$ during the winter wheat-growing season; (2) trace the flow of wet deposition N in runoff to preliminarily evaluate which forms of wet deposition N are easily absorbed by the soil-crop ecosystem and which form are prone to be lost to runoff; and (3) assess the contribution of wet deposition N to winter wheat cultivation and provide recommendations for the use of N fertilizers.

2. Materials and methods

2.1. Study site

Experiments were conducted at Changshu Agro-Ecological Experiment Station, Chinese Academy of Sciences, Jiangsu Province (31°32′45″N and 120°41′57″E). Located in the TLR, the study site belongs to the northern subtropical humid climatic zone, with an annual mean temperature of 15.5 °C, rainfall of 1038 mm, and frost-free period of 224 days. The soil is developed from lacustrine deposits and is classified as Gleyi-stagnic Anthrosols, according to the FAO soil taxonomy system. The topsoil (0–15 cm; pH 7.3) contains 35 g kg^{-1} of organic matter, 2.09 g kg^{-1} of total N, and 0.39 g kg^{-1} of total P.

2.2. Sampling and analysis

Rainfall was collected using an automated rainfall collector (APS-3, Wuhan Tian-Hong Intelligent Instrument Plant, China). The rainfall-sensitive sampling system enables the top cover of the rainfall collector to open automatically and collect rainwater samples when it rains, and close automatically when rain stops, protecting the rainwater samples from contamination. The circular rain collecting equipment has an area of 0.38 m^2 and is made of polyvinyl plastic with a filtering system. Following rain events, rainwater samples were immediately recorded (mm) and sampled into 100 ml plastic bottles, and then stored at −18 °C in the refrigerator pending $NH_4{}^+$–N, $NO_3{}^-$–N, and TN concentration analyses.

To trace the flow of wet deposition N by runoff, four replicated field plots ($7 \times 6 = 42\,m^2$) in the area were adopted for investigation during the experimental period. At the beginning of the wheat-growing season, deep (15-cm depth) and narrow (10-cm width) ditches were dug in the plots to quickly drain water in the field and lower the groundwater table, preventing the damage of wheat roots from water logging. This technique is widely adopted in the TLR (Cao and Zhang, 2004; Tian et al., 2007). Therefore, runoff flow during the wheat season includes both surface runoff flow and part of the subsurface flow. Bottom pipe mouths were installed at the bottom level of the ditches and runoff was piped into concrete wells around the experimental area. After each rainfall event, with sufficient runoff volume for analysis, 500 ml water samples were obtained after runoff volume was measured and mixed. The rest of the water was discharged to a nearby canal. Thereafter, the empty wells were washed with tap water, ready for the next runoff collection. Winter wheat seeds were broadcast by the end of October. No irrigation and N fertilizer application were implemented during the winter wheat-growing season.

The runoff samples were filtered with a 0.45-μm membrane and stored at −18 °C in the refrigerator until analysis. Both TN contents in runoff and rainwater were digested using potassium peroxodisulfate, and analyzed using a Shimadzu UV-1601 spectrophotometer. $NH_4{}^+$–N in the runoff and rainfall samples was determined using the indophenol blue method, and $NO_3{}^-$–N was analyzed according to the standard method using a UV-1601 spectrophotometer.

Wet deposition N was calculated using the following equation:

Table 1
Wet deposition fluxes and runoff loss (kg N ha^{-1}) of NH_4^+–N, NO_3^-–N, and TN during the 2001–2002, 2002–2003, 2003–2004, and 2004–2005 wheat seasons.

	2001–2002		2002–2003		2003–2004		2004–2005		Average	
	Wet deposition	Runoff	Wet deposition	Runoff	Wet deposition	Runoff	Wet deposition	Runoff	Wet deposition	Runoff
NH_4^+–N	5.35	0.08	9.69	0.13	8.14	0.26	9.15	0.05	8.08	0.13
	(51.0)[a]	(3.6)	(65.8)	(8.0)	(60.3)	(11.1)	(66.8)	(5.2)	(61.7)	(7.3)
NO_3^-–N	4.42	1.65	3.50	1.08	3.08	1.67	3.48	0.84	3.5	1.31
	(42.1)	(74.7)	(23.8)	(66.7)	(22.8)	(71.1)	(25.4)	(87.5)	(26.7)	(73.2)
TN	10.5	2.21	14.7	1.62	13.5	2.35	13.7	0.96	13.1	1.79

[a] Numbers in parentheses denote the percentages of NH_4^+–N or NO_3^-–N fluxes/loss to TN flux/loss.

TN, NH_4^+–N or NO_3^-–N deposition fluxes during the wheat season

$$(\text{kg N ha}^{-1}) = \sum_{i=1}^{n} Ci \times hi \times 10^{-2}$$

where Ci is the TN, NH_4^+–N, or NO_3^-–N concentration (mg N L^{-1}) pertaining to an individual rain event with rainfall hi (mm), n is the frequency of rain events, and 10^{-2} is the conversion factor.

N loss through runoff was calculated with the following equation:

TN, NH_4^+–N or NO_3^-–N runoff loss during the wheat season

$$(\text{kg N ha}^{-1}) = \sum_{i=1}^{n} Ci \times Vi \times 2.45 \times 10^{-4}$$

where Ci is the TN, NH_4^+–N, or NO_3^-–N concentration (mg N L^{-1}) pertaining to an individual runoff event with volume Vi (L), n is the frequency of runoff events, and 2.45×10^{-4} is the conversion factor.

3. Results

3.1. Temporal variations in nitrogen wet deposition during winter wheat seasons

3.1.1. Temporal variations in NH_4^+–N and NO_3^-–N concentrations in wet deposition

Inorganic N was the dominant component of wet deposition N, and the volume-weighted concentrations of NH_4^+–N and NO_3^-–N in rainwater showed distinct temporal variations during the wheat season (seven months, from November to May of the following year). NH_4^+–N and NO_3^-–N concentrations ranged from 0.3 to 8.5 (weighted average, 1.7) and 0.2 to 4 (weighted average, 0.8) mg N L^{-1}, respectively (Fig. 1). NH_4^+ concentrations were higher and exhibited greater variability than did NO_3^- concentrations, and the average molar ratio of NH_4^+/NO_3^- was 2.3.

3.1.2. Seasonal patterns of NH_4^+–N and NO_3^-–N deposition

Fig. 2 shows the monthly distribution of inorganic N in precipitation during the investigated wheat seasons. The deposition of inorganic N varied from month to month. Monthly NH_4^+–N and NO_3^-–N deposition ranged from 0.31 to 2.63 kg ha^{-1} and 0.23 to 1.38 kg ha^{-1}, respectively. NH_4^+ wet deposition was concentrated in April–May, and NH_4^+ deposition in this period accounted for 45% of the entire NH_4^+ deposition during the winter-growing season.

3.2. Comparison of event-based runoff loss of N with wet deposition N

3.2.1. Comparison of NH_4^+–N and NO_3^-–N concentrations in runoff and wet deposition

During the four winter wheat seasons, NH_4^+–N in runoff water was less than 0.2 mg N L^{-1}, much lower than that in rainwater (Fig. 3). However, NO_3^-–N in runoff ranged from 0.5 to 4.5 mg N L^{-1}, equal to or a little higher than that in rainwater (Fig. 4).

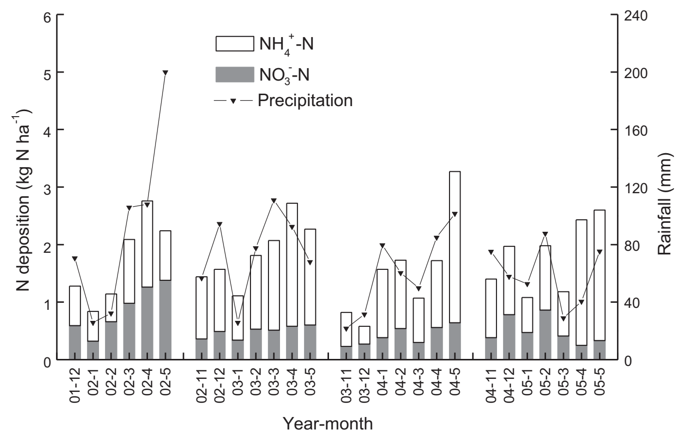

Fig. 2. Monthly distribution of NH_4^+–N, NO_3^-–N, and rainfall during the 2001–2002, 2002–2003, 2003–2004, and 2004–2005 wheat seasons.

3.2.2. Comparison of N loads in runoff and wet deposition N fluxes

The average wet deposition N during the winter wheat-growing season was 13.1 kg N ha^{-1} (Table 1). During the 2001–2002, 2002–2003, 2003–2004, and 2004–2005 wheat seasons, NH_4^+–N wet depositions were computed at 5.4, 9.7, 8.1, and 9.2 kg N ha^{-1} (average of 8.1 kg N ha^{-1}), respectively, accounting for 61% of TN load in wet deposition. NO_3^-–N deposition was less than NH_4^+–N at 4.4, 3.5, 3.1, and 3.5 kg N ha^{-1} (average, 3.6 kg N ha^{-1}), respectively, accounting for 29% of TN load deposition (Table 1).

An average of 1.8 kg N ha^{-1} season^{-1} was lost to runoff, and NO_3^-–N was the dominant form lost to runoff, accounting for 67–88% (75% on average) of TN loss. NH_4^+–N load in runoff only accounted for 0.6–3.2% of NH_4^+–N deposition fluxes. Conversely, NO_3^-–N in runoff accounted for a considerable proportion of NO_3^-–N deposition fluxes (36.5% on average). In general, an average 14% wet deposition N is lost to runoff during the winter wheat-growing season (Table 2).

4. Discussion

4.1. N wet deposition characteristics during wheat seasons

During the wheat-growing period, 45% of NH_4^+–N deposition was distributed in April and May. The primary source of NH_4^+ in wet deposition is NH_3 volatilized from fertilizers, as well as human and animal excreta, which are influenced by climate and agricultural activities. The gradually rising temperature (monthly mean

Table 2
Percentages (%) of runoff N to wet deposition N for NH_4^+–N, NO_3^-–N, and TN during the 2001–2002, 2002–2003, 2003–2004, and 2004–2005 wheat seasons.

	2001–2002	2002–2003	2003–2004	2004–2005	Average
NH_4^+–N (%)	1.5	1.3	3.2	0.6	1.7
NO_3^-–N (%)	37.3	30.9	54.2	24.1	36.6
TN (%)	21.0	11.0	17.4	7.0	14.1

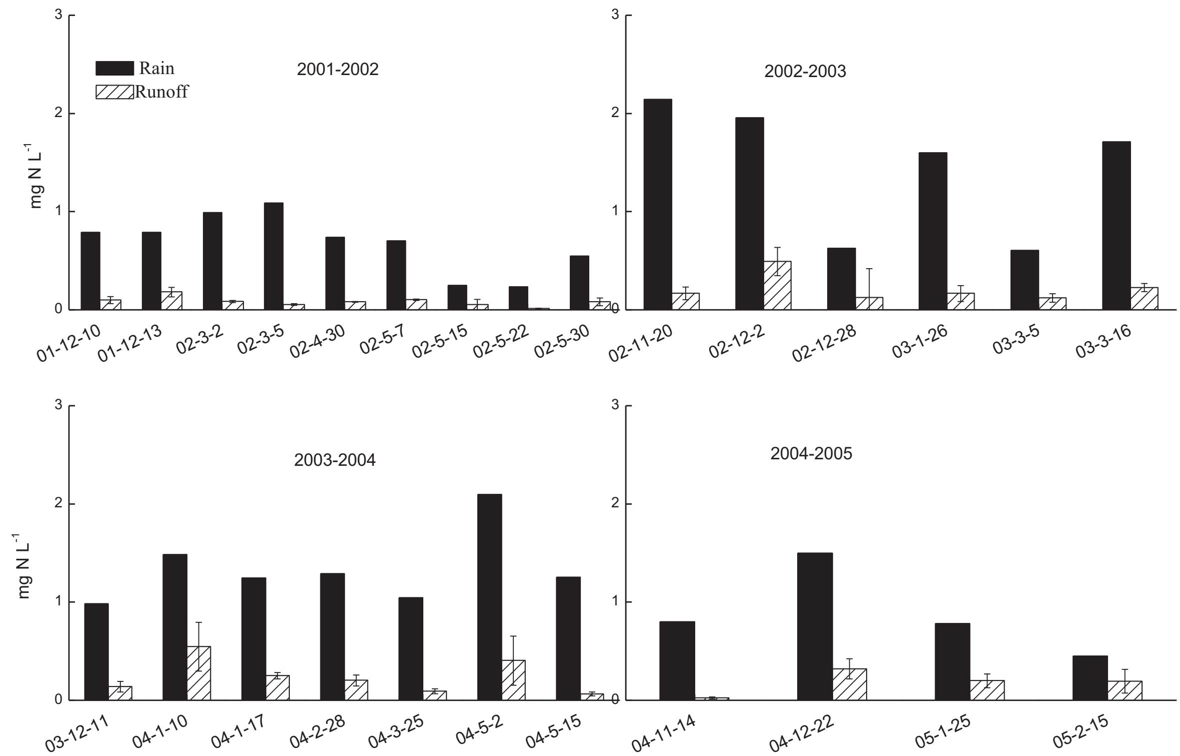

Fig. 3. NH_4^+–N concentrations of rainwater and runoff water in each runoff event during the 2001–2002, 2002–2003, 2003–2004, and 2004–2005 wheat seasons.

temperature in April and May is 17 and 21 °C, respectively, whereas in other months, temperature is lower than 15 °C) and intense biological activities in this period facilitated NH_3 volatilization from human and livestock excreta, which can be revealed by the positive $\delta^{15}NH_4^+$ values in wet deposition (Zhao et al., 2009) because $\delta^{15}N$ values in human and animal excreta are high and positive (Yeatman et al., 2001; Xing and Zhu, 2002). The mean NH_4^+/NO_3^- ratio in the present study was 2.3, lower than those in agricultural regions (Li and Li, 1999; Anderson and Downing, 2006), higher than those in suburban areas (Zhao et al., 2009), and similar to those in other similar areas in China (Liu et al., 2006; Chen and Mulder, 2007; Zhang et al., 2008). These results indicate that human and

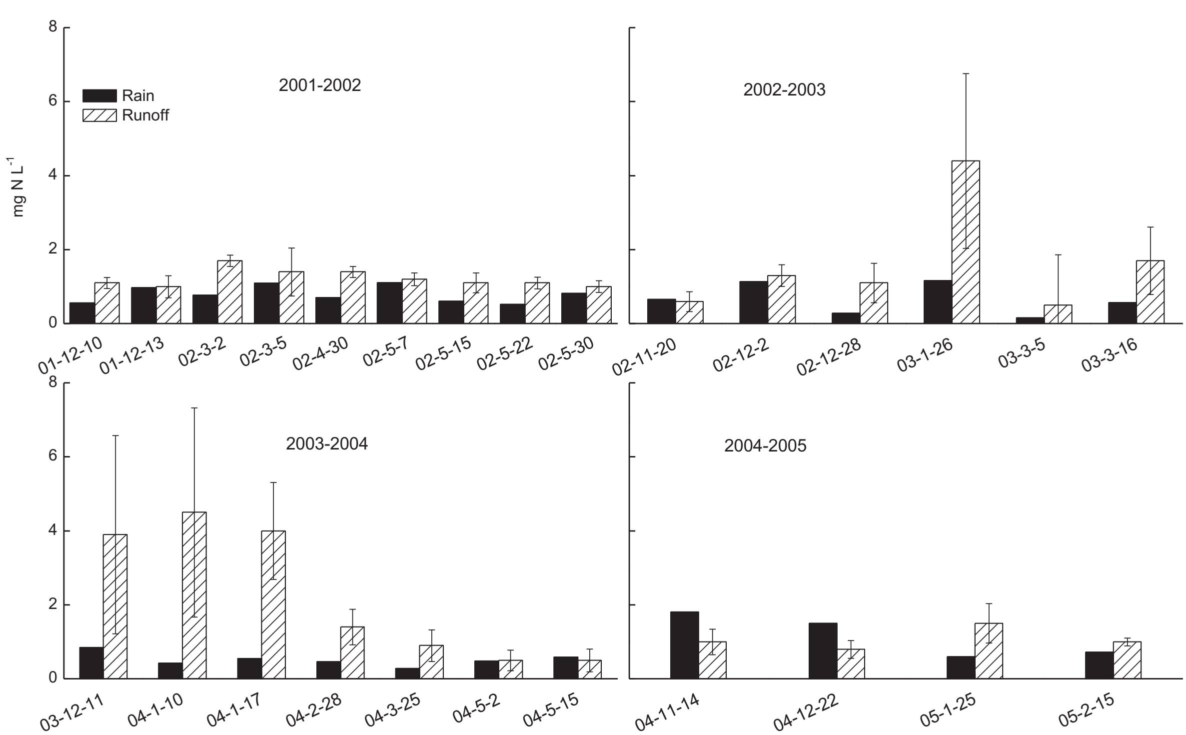

Fig. 4. NO_3^-–N concentrations of rainwater and runoff water in each runoff event during the 2001–2002, 2002–2003, 2003–2004, and 2004–2005 wheat seasons.

animal excreta, as well as agriculture, primarily account for wet N deposition.

NO_3^-–N deposition was lower than NH_4^+–N deposition and relatively steady across months because the main sources of NO_x emission—fossil fuel combustion and motor vehicles (Dise and Stevens, 2005)—are relatively constant in this region, unlike in Northern China where large amounts of coal are consumed in winter for heating purposes (Liu et al., 2006). However, the contribution of NO_3^- has become increasingly important in this region because of the rapid increase in fossil fuel consumption in this region, and NO_3^-–N deposition has become 5 times higher than in the early 1980s (Zhao et al., 2009).

4.2. Runoff flow of wet deposition N during winter wheat season

No irrigation is implemented during the winter wheat season, and a drainage ditch is dug to drain away the excess rainwater. Most of the NH_4^+–N in precipitation was retained by the soil–plant system and could be used as a stable N source for field crops. Aside from utilization by microbes and plants, NH_4^+–N can also be adsorbed onto the soil cation exchange complex for later use by plants (Moldan et al., 2006). However, when runoff occurred, NO_3^-–N concentration in runoff was equal to or a little higher than that in rainwater. Higher NO_3^-–N concentration in runoff water than in rainwater may be attributed to the NO_3^-–N stored in the soil. Parallel results by Schleppi et al. (2004) asserted that aside from deposited NO_3^-–N, NO_3^-–N in runoff may also originate from the flushing of older NO_3^-–N temporarily stored in the soil pores or from NO_3^- produced by nitrification mechanisms.

Fourteen percent of wet deposition N was lost to runoff, a little higher than that in a study on mountain forests to which $^{15}NH_4{}^{15}NO_3$ tracers were added for a period of two years (Schleppi et al., 1999). However, the loss is much higher than the percentage of wet deposition N lost to runoff in forest catchments in South China (Chen et al., 2004). The same was observed in Switzerland where only 0.2% and 0.6% of added $^{15}NH_4^+$ tracers, as well as 2.7% and 6.4% of added $^{15}NO_3^-$ tracers, appeared in runoff in forests and meadow catchments, respectively (Providoli et al., 2005). The soil N content or mineralization rate in wheat fields may be higher than that in forests or meadow catchments, and a considerable amount of NO_3^- in runoff may come from the nitrification of soil mineralized N or deposited NH_4^+–N.

Nevertheless, this study clearly indicates that 11 kg N ha^{-1} can be used as nutrients in croplands during the winter wheat season, and should be taken into account when calculating N fertilizer requirements for winter wheat in this region, with high fertilizer N input (200–250 kg N ha^{-1} for winter wheat) and soil N mineralization rate (Yan et al., 2006). Fertilizer N recommendations should be adjusted accordingly to optimize N use efficiency and avoid substantial N losses either to water bodies or to the atmosphere, with related potential environmental risks (Fan et al., 2005; Tian et al., 2007).

5. Conclusions

During winter wheat-growing seasons in the TLR, one of the most developed and populated regions in China, wet deposition N reaches 13 kg ha^{-1}, mainly in the form NH_4^+–N. NH_4^+–N concentrations show large temporal variations and NH_4^+–N deposition is mainly distributed in April and May, resulting from heightened NH_3 volatilization from human and livestock excreta. Fourteen percent of wet deposition N is lost to runoff, of which 75% is NO_3^-–N. The present study sheds light on the flow of wet deposition N in intensive agricultural croplands. Fertilizer N recommendations for winter wheat cultivation should be adjusted by subtracting 11 kg ha^{-1}, accordingly.

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