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DRY DEPOSITION OF ATMOSPHERIC NITROGEN IN LARGE RESERVOIRS AS DRINKING WATER SOURCES: A CASE STUDY FROM THE DANJIANGKOU RESERVOIR, CHINA

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Abstract

This paper aims to pioneer the research into the dry nitrogen deposition in reservoirs serving as drinking water sources. For this purpose, several field experiments were performed to investigate the dry deposition in Xiaotaipingyang, Danjiangkou Reservoir (Henan) from September 2015 to August 2017. Sample concentrations were measured for NH_4^+-N, NO_3^--N and total dry deposition (TDN). Then, dry deposition flux of NH_4^+-N, NO_3^--N and TDN were estimated based on the measured concentrations and sample volumes. The results show that the TDN at four monitoring sites ranged from 15.2 kg/ha·a to 22.5 kg/ha·a and averaged 19.2 kg/ha·a. The annual dry depositions of NO_3^--N, NH_4^+-N and dissolved organic nitrogen (DON) were respectively 5.7 kg/ha·a, 8.6 kg/ha·a and 5.0 kg/ha·a, indicating that reduced nitrogen dominates the dry deposition and the DON is a non-negligible compound of the TDN. Moreover, seasonal nitrogen dry deposition was ranked in the order of summer, autumn, spring, and winter. Dry deposition mainly occurred from April to September (>75% of TDN), which is consistent with the temperature change and the fertilizing time in local agriculture. These results illustrate the occurrence of nitrogen pollutants input from the atmosphere into Danjiangkou Reservoir (Henan), which may represent a considerable proportion of the total nutrient loading to the reservoir. The research results shed new light on the protection of reservoirs serving as drinking water sources from the dry deposition.

Key words: dry deposition, nitrogen, drinking water source, Danjiangkou Reservoir, variation

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1. Introduction

Nitrogen deposition is an important link of the nitrogen cycle in ecosystems (Csolti et al., 2017; Galloway, 2013; Zhang and Song, 2016). The period between 1850 and 2010 saw the percentage of anthropogenic nitrogen in the total nitrogen production on land surging up from 20% to 75% (Galloway et al., 2013, 2014). The ensuing overload of nitrogen deposition has threatened the biodiversity and productivity of ecosystems (Armitage et al., 2014; Baron et al., 2011; Meunier et al., 2016). In China, it is estimated that the total nitrogen deposition (TDN) roughly increased by 60% from 1980s to 2000s, reaching 54.3kg/ha·a in 2007 (Liu et al., 2013; Ti et al., 2012). The high TDN deposition exerts a significant impact on ecosystems across the country.

Dry deposition is an important, if not dominant, component of nitrogen deposition (Huang et al., 2011; Im et al., 2013). Compared with wet deposition, continuous dry deposition may boost the available nitrogen content in ecosystems. In recent years, China has experienced the greatest increase in dry deposition across the globe (Jia et al., 2016). The statistics show that dry deposition accounts for approximately 62% of the TDN in China (Zheng et al., 2014). However, the existing studies have emphasized wet deposition over dry deposition.

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Over the years, many scholars have collected data on nitrogen deposition and investigated its impacts on ecosystems like forests (Chiwa et al., 2013; Duarte et al., 2013; Waldner et al., 2014), grasslands (Stevens et al., 2004; Zhang and Han, 2012), farmlands (Shen et al., 2014; Zbieranowski and Aherne, 2012), seas (Dai et al., 2018; Kim et al., 2011; Markaki et al., 2010; Shi et al., 2013), lakes (Curtis et al., 2015; Elser et al., 2009; Xin et al., 2015), and marshes (Bragazza et al., 2012; Caporn et al., 2014; Du et al., 2018; Granath et al., 2014), revealing that dry deposition is influenced by pollutant properties, surface features and atmospheric stability (Voldner et al., 1986). Nevertheless, there is no report on the impacts of nitrogen deposition on reservoirs, especially those serving as drinking water sources.

Drinking water sources are hotspots of nitrogen deposition. To protect the water quality, effective measures must be implemented to control nitrogen pollution in reservoirs serving as drinking water sources. For a gorge-type reservoir, nitrogen deposition is a major nitrogen source, second only to river input, under effective control of nonpoint source pollution.

In general, little attention has been paid to the dry deposition in reservoirs (Lu et al., 2015). To make up for the gap, this paper aims to determine the temporal variation of dry deposition of reservoirs through the field test on the Xiaotaipingyang, Danjiangkou Reservoir (Henan), China.

2. Experimental

2.1. Study area

The study area lies within the Xiaotaipingyang, Danjiangkou Reservoir (Henan). The reservoir is a gorge-type reservoir which serves as a drinking water source of the South-North Water Transfer (SNWT) Project. The project is designed to relieve the water shortage in northern China and serve more than 42 million people. Located in Danjiangkou Reservoir (Henan), the Xiaotaipingyang is the location of Taocha Intake, an important water source of the SNWT project.

To determine the dry deposition of atmospheric nitrogen, four monitoring sites were designed on different types of land-use (Fig. 1), the non-irrigated farmlands (site 1) to the east of the reservoir, the slope farmlands (site 2) to the north of the reservoir, Xianghua village (site 3), home to 28,000 villagers, to the east of the reservoir, and paddy fields (site 4) on the southeast of the reservoir. The monitoring sites were arranged in open areas with no obstacles in the surroundings. Located in the subtropical humid monsoon climate area, the study area has a mean annual temperature of 16.4°C. The main precipitation period lasts from May to October. During the study period, the mean annual precipitation falls in 870~1,130mm according to the rainfall of Nanyang, which is about 91 km away from the reservoir.

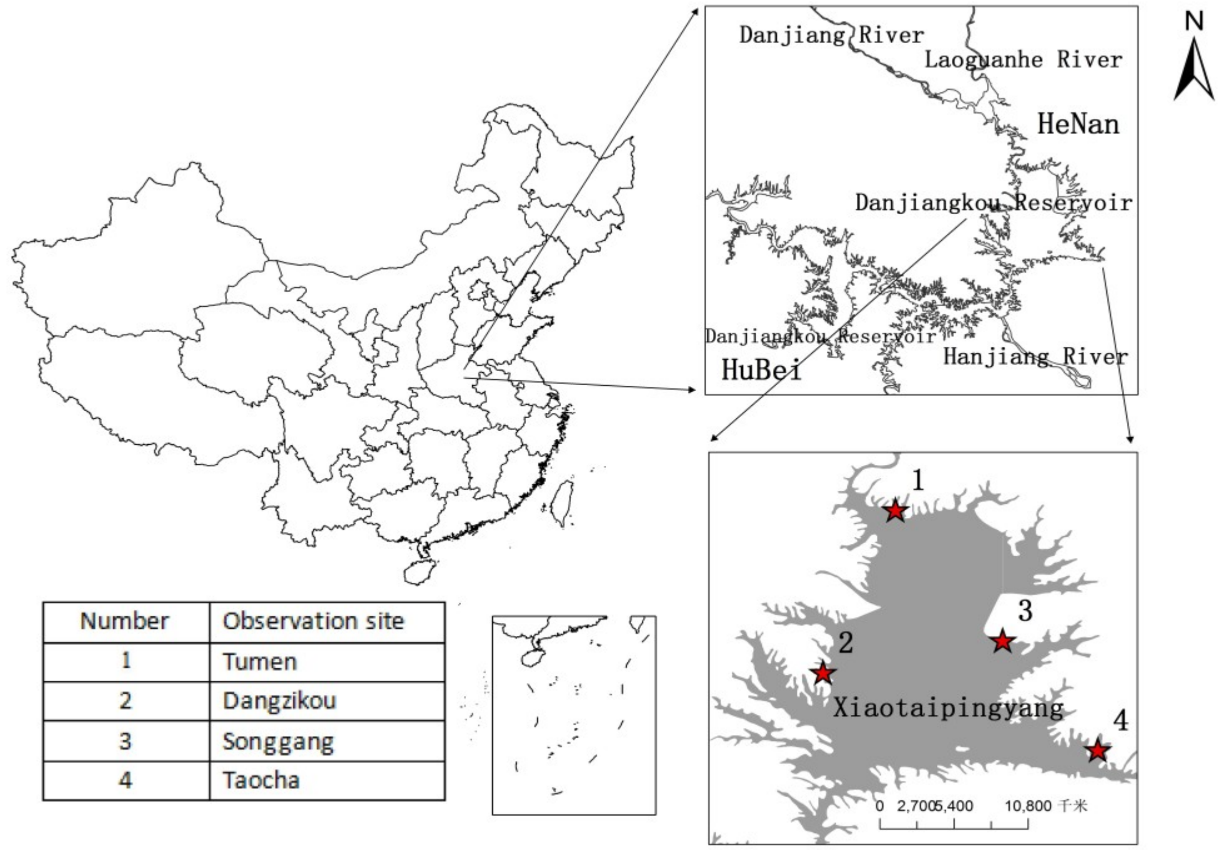

Fig. 1. Study area and monitoring sites

2.2. Sampling and analytical procedures

SYC-2 wet-dry automatic collectors were purchased from Qingdao Laoshan Electronic Instrument General Factory Co., Ltd., installed at each monitoring site on June 29, 2015, and washed with deionized water after every collection. Each collector consists of a 177cm^2 polyethylene bucket for dry deposition collection and a 707cm^2 stainless steel basin for wet deposition collection.

The collection process was regulated by a cover, which is controlled by a precipitation sensor with a thermostatic device exposed in the air. This device controls the evaporation of raindrops falling onto it, such that the cover could close quickly once the rain stops. When the precipitation on the device exceeds 0.1 mm, the precipitation sensor moves the cover to the bucket to collect wet deposition; when the rain stops, the device moves the cover to the basin in 5min to collect dry deposition.

The dry deposition were sampled from Taocha (TC), Dangzikou (DZK), Songgang (SG) and Tumen (TM) four times a month from September 2015 to August 2016. Before each dry collection, the collector was washed with 600mL water for two times, and then filled with a height of 5cm distilled water. 1mL $CuSO_4$ (2mol/L) was added to the sampling water to wipe out the microbes. The unfiltered samples were immediately refrigerated at −8°C. Prior to analysis, the water samples were shaken for 2~3min to agitate the sunken particles at the bottom of the containers. Then, the samples were filtered through 0.45μm cellulose membrane filters before analyzing the concentrations of NH_4^+-N, NO_3^--N and TDN.

The NH_4^+-N and TDN concentrations were determined by a type 5000 continuous flow analyzer (Skalar, Netherlands), which the NO_3^--N concentration was measured by Dionex ICS-1100 ion chromatography system (Thermo Fisher Scientific, US). Then, the NH_4^+-N concentration was subjected to spectrophotometry with salicylic acid, the TDN concentration by continuous flow analysis and N-(1-naphthyl)ethylenediamine dihydrochloride spectrophotometry, and NO_3^--N concentration by ion chromatography, and the detection limits was lower than 0.05 0.1 μ mol/L. Moreover, dissolved organic nitrogen (DON) concentration was calculated by the concentration difference of TDN and DIN (NH_4^+-N + NO_3^--N).

2.3. Dry deposition calculation

Weekly TDN, NO_3^--N, and NH_4^+-N dry deposition rates (henceforth referred to as TDN, NO_3^--N, and NH_4^+-N dry deposition, respectively) for each study site were calculated using Eq. (1):

$$D=C_i\times V_i/(10\times S) \quad ....(1)$$

where:

D are the weekly dry depositions of TDN, NO_3^--N, and NH_4^+-N (kg/ha);
C_i are the weekly concentrations of TDN, NO_3^--N and NH_4^+-N (mg/L);
V_i are the weekly sample volume (L);
S are the sampling areas of the dry deposition sampler.

Monthly and annual TDN, NO_3^--N and NH_4^+-N dry depositions (kg/ha; kg/ha·a) were derived based on the weekly dry depositions.

2.4. Statistical analysis

The sample differences in NH_4+-N, NO_3^--N, DON and TDN of the four sites were tested with one-way analysis of variance on IBM SPSS Statistics for Windows. The statistical significance was measured against the p value<0.05.

3. Experimental results

3.1. TDN dry deposition

The TDN dry deposition was monitoring for 12 months (Fig. 2). Throughout the monitoring period, the monthly dry deposition of the TDN varied between 0.3 and 4.9kg/ha, putting the average at 1.6kg/ha. The dry deposition reached the peak values in June and July May (3.0 and 2.7 kg/ha, respectively) and the minimum values in December, January, and February (0.9, 0.9 and 0.8 kg/ha, respectively).

The annual dry deposition of the TDN ranged from 15.2 kg/ha·a to 22.5 kg/ha·a, and averaged 19.2 kg/ha·a across the monitoring sites. Comparable to the annual wet deposition of the TDN (21.2 kg/ha·a), the annual dry deposition of the TDN takes up 48% of the total deposition (wet deposition + dry deposition).

An obvious seasonal variation was observed in the TDN dry deposition (2.6~7.3 kg/ha). Out of the annual TDN dry deposition, the spring, summer, autumn and winter respectively accounted for 24% (4.8 kg/ha), 38% (7.3 kg/ha), 25% (4.6 kg/ha) and 14% (2.6 kg/ha). Seasonal TDN dry deposition varied with time (2.6~7.3 kg/ha). The dry deposition observed in summer accounted for 38% (7.3 kg/ha), that in winter accounted for 14% (2.6 kg/ha), that in spring accounted for 24% (4.8 kg/ha), and that in autumn accounted for 25% (4.6 kg/ha) of the annual TDN dry deposition.

The lowest annual dry deposition was observed at the TC site, with the mean value of 15.7 kg/ha·a. This value was 10~33% lower than that of the other three sites. The highest annual dry deposition was observed at the DZK site, with the mean value of 22.5 kg/ha·a. This value was 1~48% higher than that of the other three sites. No significant differences were observed between the four sites (p>0.05), indicating the absence of a geographic distribution of dry deposition in Danjiangkou Reservoir (Henan).

3.2. NH_4^+-N, NO_3^--N, and DON dry deposition

The monthly mean dry depositions of NH_4^+-N, NO_3^--N and DON of the four sites are illustrated in Fig. 3. As shown in the figure, the dry deposition peaks concentrated between May and September. The peak of NH_4^+-N dry deposition appeared in June and July. This is because these two months are the main growing season, during which 450~600 kg/ha·a of nitrogen fertilizers are applied to farmlands (Huang Wenmin et al., 2012); the evaporation of NH_3 is accelerated by the high temperature. Besides, the peak of NO_3^--N dry deposition was observed in May and June, and that of DON in June, July and August. All these months were featured by significant fluctuations in the dry deposition of nitrogen species. Besides, significant temporal variation was observed among NH_4^+-N and DON monthly dry depositions ($p<0.05$).

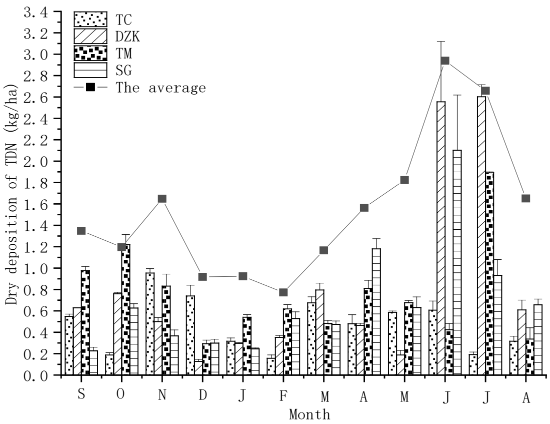

Fig. 2. Monthly mean and standard deviation of dry deposition rates

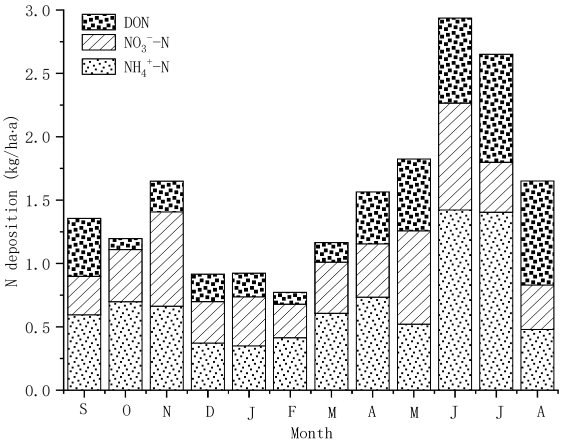

Fig. 3. Monthly mean dry depositions of NH_4^+-N, NO_3^--N, and DON of the four monitoring sites

The correlations between the annual mean dry depositions of NH_4^+-N, NO_3^--N and DON are displayed in Fig. 4 below. It can be seen that the annual mean dry depositions of NH_4^+-N, NO_3^--N and DON were 8.6, 5.7, and 5.0 kg/ha·a, respectively, taking up 44%, 30% and 26% of the TDN dry deposition. Meanwhile, the annual mean wet depositions of these nitrogen species were 11.4, 6.7 and 4.6 kg/ha·a, respectively, taking up 50%, 30% and 20% of the TDN wet deposition. Overall, the NH_4^+-N (44%) dominated the dry deposition of TDN at all sites. NO_3^--N (29%) and DON (25%) were two important parts of the TDN. Based on the annual mean dry depositions of the four sites, it can be concluded that there was a moderate correlation between NH_4^+-N and DON, a week correlation between NH_4^+-N and NO_3^--N but no correlation between DON and NO_3^--N (Fig. 4).

3.3. NH_4^+-N/NO_3^--N ratio

The NH_4^+-N/NO_3^--N ratio was introduced to identify the source of nitrogen deposition. According to the monthly ratios in Fig. 5, the highest ratio of 3.6 occurred in July, while the lowest ratio of 0.7 emerged in May. Notably, the seasonal NH_4^+-N/NO_3^--N ratios were all greater than 1: summer (2.2)>autumn (1.7)>spring (1.3)>winter (1.2). The peak value is attributable to the positive correlation between temperature and NH_3 volatilization from fertilizer and animal waste. However, the lowest value should be further investigated. Overall, the NH_4^+-N/NO_3^--N ratios reveal that the nitrogen pollution of the study area mainly comes from farming and livestock breeding.

4. Discussions

4.1. Dry deposition in the study area and comparison with other regions

Dry deposition is an often-ignored but important type of nitrogen deposition. According to the TDN data across China, dry deposition varied widely from 1.9 kg/ha·a (Qi et al., 2013) to 56.2 kg/ha·a (Zheng et al., 2014). Our research obtains some initial results on the dry deposition and its variation pattern of Danjiangkou Reservoir (Henan). The initial results indicate that the study area received a large amount of dry deposition from October 2015 to September 2016 (19.2 kg/ha·a), and the percentage of dry deposition in the TDN reached the highest level in summer, followed by autumn, spring and winter.

The annual mean dry depositions of the study area were compared with many other regions (Table 1). The results in Table 1 show that the study area had a higher annual mean dry deposition than foreign lakes like Lake Victoria, owing to the high nitrogen emissions in China. The NH_3 and NO_x emissions in China ballooned from 1.4 Tg/a to 6.3 Tg/a between 1980 and 2010.

In terms of annual mean dry deposition, the study area was extremely close to Dahekou Reservoir, which lies in the sand source area of Beijing and Inner Mongolia. However, the two reservoirs differed greatly in the variation pattern of dry deposition. For Dahekou, the dry deposition peaked in spring and autumn, particularly in April, which reflects the influence of sandstorms (Lu et al., 2015). By contrast, the variation pattern of dry deposition in our research demonstrates the impacts of temperature and agricultural activities.

Compared with that (7.8 kg/ha·a) of the watershed in Danjiangkou Reservoir (Hubei), the annual mean dry deposition of the study area was relatively high but in accord with the same variation pattern, revealing that the dry deposition could be completely different between reservoir surface and its watershed.

To sum up, the dry deposition is severely affected by the local environment, such as the nitrogen form (gaseous or particulate). Combined with the findings in Dahekou Reservoir and Danjiangkou Reservoir watershed, the comparisons indicate that the dry deposition amount is affected by the local environment, such as the existing form of N (gaseous or particulate), precipitation and pollution sources, and its variation pattern is influenced by the macro-environment, such as type of land use and climate.

4.2. Dry depositions of NH_4^+-N, NO_3^--N and DON

The experimental results show that the annual mean dry depositions of NH_4^+-N, NO_3^--N and DON were 8.6 kg/ha·a (interval: 4.2~17.1 kg/ha·a), 5.7 kg/ha·a (interval: 3.2~10.1 kg/ha·a) and 5.0 kg/ha·a (interval: 1.0~10.2 kg/ha·a), contributing 44%, 30% and 26% of the TDN dry deposition, respectively. Hence, NH_4^+-N is the leading dry deposition species for all sites, which agrees well with the other studies (Huang et al., 2011; Zhang et al., 2013). China consumes 550~600 kg/ha·a of nitrogen fertilizers each year, especially in the hot growing season of maize. As the major pathway of nitrogen loss, NH_3 volatilization is extremely active in summer, leading to the high nitrogen content in air and the high dry deposition (Ju et al., 2009; Cai et al., 2011).

The NO_3^--N dry deposition of our research exceeds the average value in China (Zhao et al., 2017) and obeys the NH_4^+-N/NO_3^--N ratio (>1) in studies on forests in Hubei, which are close to the study area (Xu et al., 2015). Besides, the NO_3^--N dry deposition was relatively stable in this research, with a valley in winter. This means the NO_3^--N dry deposition in Danjiangkou Reservoir (Henan) is not influenced by heating in winter, which contradicts several studies in northern and south-eastern China (Cui et al., 2011; Shen et al., 2008). The pollution sources of NO_3^--N may include vehicles, soil and biomass burning (Lee et al., 1997). HNO_3 (one form of NO_y), which is oxidized from NO_x, is the main dry deposition component in China (Zheng, 2014). In winter, NO_y remains as NO_x due to the reduced photochemical oxidation (Dolislager et al., 2012), dragging down the dry deposition ratio of HNO_3.

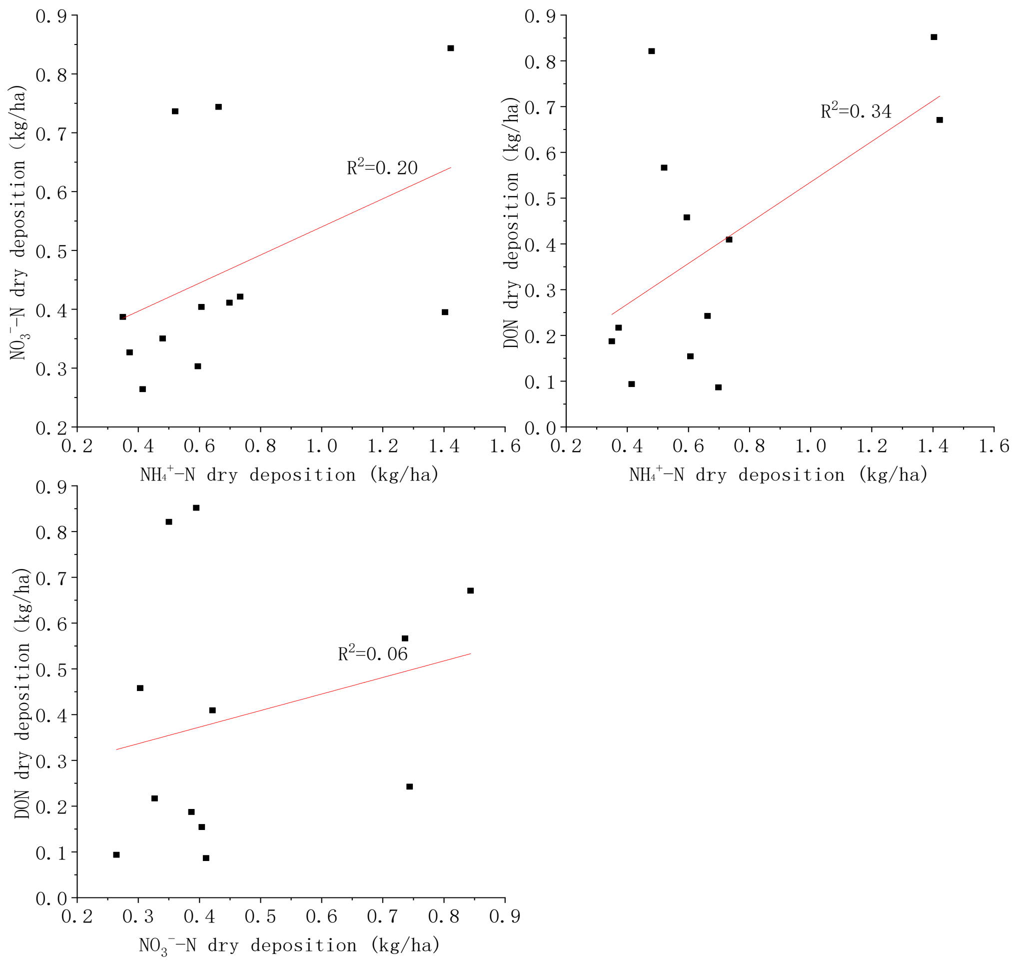

Fig. 4. The correlation between NH_4^+-N, NO_3^--N and DON

Table 1. Comparison of dry deposition of nitrogen among some regions over the world

Location	***Ecosystems***	***Monitoring period***	***Annual mean deposition (kg/ha·a)***	***Data resource***
Danjiangkou Reservior in Henan	reservoir	2015 to 2016	19.05	this study
Danjiangkou Reservior in Hubei	watershed	2009 to 2011	7.8	Liu et al. (2015)
Dahekou Reservoir	reservoir	2014	21.4	Lu et al. (2015)
Lake Taihu	lake	2011	78.3	Liu et al. (2011)
Lake Beili	lake	February to July in 2010	51.3	Li et al. (2010)
National Parks in Washington State	lake	summer and early autumn in 2008	1.4	Sheibley et al. (2014)
Lake Kivu	lake	October 2006 to June 2008	6.8	Nsengimana et al. (2010)
Lake Tahoe	wetland	1997 to 1998	1.2-8.6	Tarnay et al. (2001)
Coastal area of the Yellow River Delta	wetland	May 2012 to November 2012	7.3	Ning et al. (2015)
La Plata River	coastal water	1999 to 2001	0.8-1.4	Andrea et al. (2009)
Jiulong River Estuary - Xiamen Bay	coastal water	2004 to 2005 and 2009 to 2010	4.8	Chen et al. (2011)
Fengqiu	cropland	2008 to 2009	25.2	Huang et al. (2011)
Yingtan city southeast China	cropland	2005 to 2009	73.1	Cui (2011)
Lake Ontario	cropland	2010 to 2011	15-28	Zbieranowski and Aherne (2012)
Xiangjiang river watershed	forest	2010 to 2011	34.7	Shen et al. (2013)
Denali National Park, Alaska	forest	1999 to 2013	8.7	Nagano and Iwata (2016)
Flanders, Northern Belgium	forest	2005 to 2013	3-3.9	Verstraeten et al. (2016)

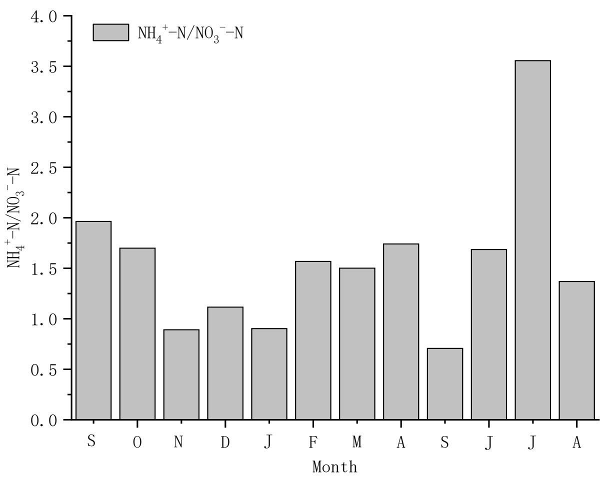

Fig. 5. Monthly NH_4^+-N/NO_3^--N ratios

DON dry deposition took up 11~56% of the TDN across the world (Jiang et al., 2013). Violaki et al. (2010) even put the percentage at 78%. In our research, the annual DON dry deposition in Danjiangkou Reservoir (Henan) was 5.0 kg/ha·a (interval: 1.0~9.9 kg/ha·a), 1.9 kg/ha·a lower than the DON wet deposition in China (Zhang et al., 2012). In the study period, the dry deposition of the DON peaked between June, July and August, while the same trend was not observed in that of NH_4^+-N and NO_3^--N. This phenomenon goes against several other studies (Cape et al., 2012; Li et al., 2012), indicating that the seasonal variation of DON dry deposition is independent from the dry depositions of NH_4^+-N and NO_3^--N (Jiang et al., 2013).

5. Conclusions

Four dry deposition monitoring sites were established around Xiaotaipingyang water area of Danjiangkou Reservoir (Henan), where no dry deposition data were available despite the occurrence air pollution. Through the discussion on the dry deposition from October 2015 to September 2016, the following conclusions were reached.

The dry deposition of the study area was lower than other lakes or reservoirs in China but much greater than many foreign lakes. Thus, the study area is under heavy air pollution, as influenced by the general nitrogen deposition trend in China. Moreover, there was no significant spatial variance of dry deposition in the study area. NH_4^+-N was the dominant dry deposition species because of the nitrogen release from fertilizer application in nearby farmlands. Meanwhile, a seasonal variation of the nitrogen species (peak in summer and valley in winter) was observed to be in good agreement with temperature and local agriculture activities.

The findings show that Danjiangkou Reservoir (Henan), which serves as a drinking water source, is a nitrogen deposition hotspot. Local agriculture activities could be the main pollutant source. Therefore, the ecosystem of farmlands should be given special attention to control the dry deposition of reservoirs serving as drinking water sources.

In future, more data will be collected over a longer period to elucidate the long-term trend of dry deposition in the study area, and the effects of nitrogen deposition on the structure and function of reservoir ecosystems will be investigated in details.

Acknowledgements

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