Atmospheric dry and wet nitrogen deposition in agro-pastoral catchments of the China and Mongolia Altay

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Abstract

Very few comparative studies on nitrogen (N) deposition in agroecosystems have been conducted along land use and altitude gradients. In an effort to fill this knowledge gap we selected three typical, interconnected land use types (cropland, mountain grassland and plain grassland) with six sampling sites in the transboundary Altay Mountains of northwest China and western Mongolia. During 12 months from June 2014 to May 2015 dry and wet N deposition, through middle volume total suspended particulates (TSP), passive samplers and precipitation collectors were monitored. Among land use types, cropland had the highest concentrations of NH_4^+-N (1.6 mg N L^{-1} in China and 2.0 mg N L^{-1} in Mongolia) and NO_3^--N (1.0 mg N L^{-1} in China and 1.2 mg N L^{-1} in Mongolia) in precipitation compared to mountain and plain grasslands. In contrast, the Mongolian mountain grasslands (MM) experienced the high wet deposition (3.2 kg N ha^{-1} yr^{-1}) which was at least partly due to high summer precipitation (161 mm), followed by the Chinese cropland (CC) with 3.1 kg N ha^{-1}

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yr^{-1} while wet deposition in other land use types ranged from 1.8 to 2.5 kg N ha^{-1} yr^{-1}. CC had the highest NH_3 (3.1μg N m^{-3}) and NO_2 (3.8μg N m^{-3}) concentrations and dry N deposition (9.5 kg N $ha^{-1}yr^{-1}$) among all land use types ($p<0.05$) while Mongolian cropland (MC) had dry N deposition of 5.4 kg N $ha^{-1}yr^{-1}$. CC (12.6 kg N ha^{-1} yr^{-1}) had the highest total N deposition, followed by the MC with 7.2 kg N ha^{-1} yr^{-1} and the Mongolian mountain grassland (MM) with 6.6 kg N ha^{-1} yr^{-1}. NH_4^+-N concentration in the precipitation were negatively correlated with precipitation ($P<0.05$). Concentration of NH_3 correlated positively with air temperature ($P<0.05$) probably reflecting promoting effects of temperature on NH_3 emissions whereas NO_2 correlated negatively with temperature due to low background value of NO_2 emission. Overall, croplands in China had 76 % higher N deposition than in Mongolia whereas the reverse was true for mountain grasslands which received 26 % more N in Mongolia.

Key words: Agro-pastoral transition zone; Dry deposition; Land-use types; Transborder watershed; Wet deposition

Introduction

During the last three decades China´s industrial development and intensification of agriculture and livestock production have greatly increased the concentration and deposition of atmospheric reactive nitrogen (Liu et al., 2013). Since the 1980s, atmospheric Nr emissions have more than doubled in north and southeast China following 30 years of strong economic development (Liu et al. 2013) and current atmospheric Nr deposition are still very high (Pan et al., 2012; Xu et al. 2015). Total nitrogen deposition of 54.4-103.2 kg N ha^{-1} yr^{-1} have been reported (Luo et al. 2013) in the North China Plain and up to 35 kg N $ha^{-1}yr^{-1}$ in a remote oasis area of Xinjiang, northwest China (Liu et al. 2011; Li et al. 2012) .

Rapid intensification of agriculture in response to the increasing demand for food has led to increased use of mineral fertilizers in China which is one of the main factors responsible for high regional Nr deposition (Ju et al. 2009; Zhang et al. 2013). Also along with the increase of transport, animal breeding and energy extraction, more pollutants swarm into the atmosphere and constitute the main source of atmospheric wet and dry nitrogen deposition. Therefore, there is a complex set of circumstances for the distribution of atmospheric nitrogen deposition in China due to the huge regional differences and rapid economic development (Huang et al. 2013; Wang et al. 2010).

Previous N deposition studies have mainly focused on agricultural and urban areas (Luo et al. 2003; Li et al. 2012; Du et al. 2014; Huang et al. 2015; Liu et al. 2015; Wasiuta et al. 2015). Little research has been conducted to quantify atmospheric N deposition in agro-pastoral zones. Currently approximately 117 million people depend on this land use system which are mainly distributed in the northeastern and northwestern part of the country and in Inner Mongolia (Du et al. 2009; Xu et al. 2014; Zhang et al. 2015) . Grasslands within this agro-pastoral zone in China have been experiencing the use of high amounts of chemical fertilization and overgrazing due to high livestock numbers where atmospheric N deposition may be interacting under such conditions. So, the monitoring of atmospheric dry and wet N deposition is therefore urgently needed in this region. This can provide a basis for a more sustainable utilization of nitrogen fertilizer in farmland and the improvement of deteriorated grassland.

Additionally, the different levels of socio-economic development, cropland management practices and animal husbandry systems could also influence the dry and wet N deposition. However, very few comparative studies for dry and wet N deposition have been conducted in these two regions which have different levels of development. So, the present study was carried out in the Altay Mountains, near the border area of northwest China and western Mongolia which represent a typical agro-pastoral transition zone. The two study areas have similar land use types but different cropland/grazing land ratios, intensities of mineral fertilizer input, animal husbandry systems and levels of urban development due to the different historical and cultural background as well as the significant political and economic transformation processes(Greta et al., 2016). As the combination of these factors may lead to significant differences in N deposition over time, which could also result in a meaningful guidance for the sustainable utilization of nitrogen in both countries, our aims were to quantify seasonal variations in atmospheric N deposition and to compare the difference of dry and wet N deposition in China and Mongolia agro-pastoral catchments.

2. Materials and methods

2.1. Sampling sites

The study was conducted in Qinghe and Bulgan counties located in northwest China and Mongolia respectively, comprising an area ranging from 45-47 °N and 89-91 °E. The topography is characterized by a gradual decline in elevation from north to south and is divided into high, intermediate and low mountains, hills, and the Gobi desert zone. The average altitude of Qinghe county is 1218m above sea level (a.s.l.), with a maximum elevation of 3659 m a.s.l. and a minimum of 900m a.s.l.. Qinghe county is situated in the continental north temperate arid climatic zone. Air humidity is very low all year round with an average annual precipitation of 161 mm and an annual potential evaporation of 1495 mm. Winters are long and cold with an absolute minimum of -53 °C followed by cool and short summers with a recorded maximum temperature of 36.5 °C and an average air temperature of 0 °C (Fig. 2). The pasture area in Qinghe comprises about 14300 km^2 with a cultivated land area of 0.126 million hectares and 1.64 million farm animals traded annually. Most of the agricultural area is cultivated with spring wheat (*Triticumaestivum* L.), though alfalfa (*Medicago sativa* L.) and sea buckthorn (*Hippophaerhamnoides* L.) are also widely grown. Wheat is sown in early May and harvested at the beginning of September. The amount of mineral fertilizer applied to wheat ranges from 300-350 kg N ha^{-1} yr^{-1} in the Chinese croplands. Large numbers of sheep, cattle and camels are moved into the mountain grassland (summer pasture) from July to September while during the winter months they remain for stubble grazing in the oasis croplands and plain grassland (winter pasture).

Bulgan county (or Soum) neighbors the Bayan-Olgii Province in the north, Northwest China in the east, and Uyench and Altay soum of Khovd province in the south. Its territory comprises 8105 km^2 at an average altitude of 1164m a.s.l.. It has a continental climate with four seasons: April and May are the windiest months, January is the coldest (-40 °C) and July is the warmest (+35 °C) month of the year.

Annual precipitation averages 120-140 mm, with pronounced spring and autumn seasons allowing some cultivation of arable crops. Most of the cropland of Bulgan soum is also planted with spring wheat and to a lesser extent with rye (*Secalecereale*L.) with 150-250 N kg ha^{-1} yr^{-1}. Similarly to Qinghe, the growing season extends from May to September and large numbers of sheep, cattle and camels are moved into the mountain grassland from July to September to spend the winters in the lowlands.

In addition, the three similar land use types that were investigated (six sampling sites) are described as follows: Chinese cropland (CC), Chinese mountain grassland (CM), Chinese plain grassland (CP), Mongolian cropland (MC), Mongolian mountain grassland (MM) and Mongolian plain grassland (MP).

2.2. Measurement of N deposition and analytical procedures

From June 2014 to May 2015, wet (i.e. bulk) and dry N concentrations and deposition were monitored and quantified at six sites in the border area of Altay Mountains(Fig. 1).

2.2.1 Rainwater collection and calculation of wet N deposition

Rainwater samples were collected with precipitation collectors directly after every rainfall event in Chinese (CC) and Mongolian (MC) croplands by local farmers and/or herdsmen and the dates and amounts of rainwater recorded. Also, some special rainfall collectors (three sub-samples per site) were employed at, mountain grasslands and plain grasslands in both China and Mongolia due to the difficulty of collecting samples after every rainfall event. Same equipments were deployed in the cropland site in order to compare values against rainfall events in a month. For the cropland and mountain grassland, we collected rainfall or snow samples every month throughout the year. For the plain grassland, however, we just collected samples every month during the growing season(from May to October). Outside the growing season samples were collected just for the first time and last snowfall event to compute average values. Each rain collector comprised a funnel container of 40 cm in diameter, a plastic hose and a 20 Liter capacity plastic bucket. The funnel container was set at approx 1.5 m above the ground to avoid dust or leaf contamination from the ground surface. The plastic kettles were grounded at 30 cm depth, stick out 5cm of the ground and were covered with a lid to prevent entry of dust or other pollutants connected to the funnel with a plastic hose. Chloroform ($CHCl_3$, 1-2 ml) was added to each bucket to inhibit N transformations in the rainwater samples. The amount of precipitation was measured by an automatic weather station located close to the Meteorological Bureau of Qinghe Country. The precipitation samples were collected manually once per month at the six sampling points and transferred to plastic bottles and subsequently stored in a refrigerator at -10 °C until analysis. We also sampled snow at the beginning and end of the snowfall period to combine the snowfall data and calculate winter wet N deposition. All samples were analyzed for NH_4^+-N and NO_3^--N (inorganic N) concentrations using an AA3 continuous flow analyzer (Seal Analytical Ltd., Southampton, UK). Wet deposition of inorganic N was calculated according to Luo et al. (2014) as follows:

Wet N (every rainfall event, kg N ha^{-1}) = inorganic N concentration (mg N $L^{-1)}$ ×precipitation (mm) ×0.01

Wet N (every month, kg N ha^{-1}) = 0.001×Σ N (every rainfall event or month)

2.2.2 NH_3 and NO_2 collection and N calculation

Atmospheric NO_2 was collected with passive samplers using Gradko diffusion tubes (Goulding et al., 1998). The NO_2 samplers consisted of polyethylene tubes (71.0 mm long and 11.0 mm internal diameter) with two caps and stainless steel mesh disks. Two dry disks were placed in the caps and 30 ml of a 20% aqueous solution of triethanolamine was pipetted into the gray cap. The samplers were suspended at 1.5 m (at least 0.5 m higher than the canopy height) above ground and exposed between 15 and 30 days in the air every month. The disks were extracted with a solution containing sulphanilamide, H_3PO_4 and N-1-naphthylethylene-diamine dihydrochloride to estimate the NO_2 concentration determined by colorimetry at a wavelength of 542 nm. NH_3 samples were collected using ALPHA passive samplers (Adapted Low-cost High Absorption, Center for Ecology and Hydrology, Edinburgh, UK). This equipment included a tube, a plastic filter and a membrane (absorbed citric acid) and was placed approximately 1.5 m above ground. For the NO_2 and NH_3, we collected the samples one month one time. The cropland and mountain grassland samples were collected from June 1, 2014 to May 31, 2015, and the plain grassland samples were just collected in growing season due to harsh environmental conditions. The calculation was made according to Luo et al. (2014) as follows:

$$V=DAt/L$$

Where t represents the time interval; $D=2.09\times10^{-5}\ m^{-2}\ s^{-1}$ at 10°C, $A=3.463\times10^{-4}\ m^{-2}$, $L=0.006$ m. The following equation was then derived:

$$V(m^3)= 0.004343363(m^3)\times t(h)$$

The concentration of NH_3 (μg N m^{-3}) was obtained as follow:

$$C= (m_e-m_b)/V$$

Where m_e represents the amount of NH_3 in the experimental sample and m_b represents the amount of NH_3in the blank sample.

2.2.3 pNH_4^+ and pNO_3^-

Airborne PM_{10} particles (particulate matter with a aerodynamic equivalent diameter of < 10 μm) were sampled using a middle flow particulate sampler (Tian hong Instruments Co. Ltd., Wuhan, China) with a flow fluxes of 1.05 $m^3\ min^{-1}$. Seven to ten daily samples of PM_{10} were collected at QC and BC during each month. Samples from the other sites were not taken due to lack of electrical power and harsh environmental conditions.

The membrane of PM_{10} consisted of a glass fiber and it was placed in an incubator at constant temperature and humidity (22°C, relative humidity 50%) for 24 hours before and after sampling and weighed on an electronic balance. Finally, the samples were placed in beakers containing 50ml ultrapure water and ultrasonicated for 30 min. The extracts were filtered through 47-mm Whatman GF/F membrane syringe filters

(GE Healthcare Bio-Sciences, Pittsburg, PA, USA). The filtrates were stored and refrigerated at 4 °C. Ammonium and nitrate in PM_{10} (pNH_4^+ and pNO_3^-) were measured using a Seal AA3 continuous flow analyzer (Seal Analytical Ltd., Southampton, UK).

2.3 Estimation of dry N deposition

The effects of changing weather conditions and differences in vegetation types make the collection of Data on dry N deposition complicated as(Yu et al. 2014; Simpson et al., 2014). So, we used micro-meteorological methods and estimated dry N deposition by multiplying the measured concentrations of Nr species by their deposition velocities (V_d) in our experimental sites. The V_d of NH_3, NO_2 and TSP can be calculated in accordance with the method recommend by Shen (Shen et al. 2013). The following equations were used:

$F=C\times V_d$

Where by V_d can be expressed by

$V_d= (R_a+R_b+R_c)^{-1}$

Where R_a is the aerodynamic resistance, R_b is the quasi-laminar boundary layer resistance, and R_c is the surface or canopy resistance.

2.4 Statistical analysis

Linear regression was used to analyze interactions among the different Nr species. For Pearson ́s correlation and linear regression analyses, significance was defined at $P<0.05$. T-tests and one way analysis of variance (ANOVA) were employed to compare N deposition among monitoring sites, land-use types and seasons. All statistical analyses were performed using the SPSS 18.0 software package (SPSS Inc., Chicago, IL, USA). Figures were prepared using the Origin 8.0 software package (Origin Lab Corporation, Northampton, MA, USA).

3 Results

3.1 Wet deposition of NH_4^+-N and NO_3^-—N

The NH_4+-N concentration in rainwater collected from MC was highest compared to the MM and MP (Table 2, Fig. 3).The NH_4^+-N concentrations of the samples from Chinese sites were relatively low compared to the Mongolian sampling sites. CC had relatively higher NH_4^+-N concentration compared to CM and CP. NO_3^--N concentrations were highest for CP in China, followed by MC in Mongolia.

The different land use types had different NH_4^+-N and NO_3^--N peaks. Highest NH_4^+-N concentration occurred in May in CC and in September in MC and the highest NO_3^--N peak occurred from March to May in CC and from August to October in MC (Fig.3). Except the cropland, the NO_3^--N peaks were recorded from July to September at CP and MP. The CP and MP had different NH_4^+-N concentration dynamics, CP had a low value with no clear peak throughout the year while MP had a significantly higher NH_4^+-N concentration and a peak occurring from June to September. However, NO_3^-N showed the opposite trend, with CP having a significantly ($P<0.05$) higher peak concentration from June to October and MP having its maximum value in June and similar values in other months (Fig. 4).

3.2 Atmospheric NH_3 concentrations

The NH_3 concentrations in the air collected over cropland was highest in all land use types whereby CC had a maximum NH_3 value of 7.41μg N m^{-3} in May and an average of 3.1μg N m^{-3} throughout the rest of the year. The mountain grasslands (CM and MM) had the lowest NH_3, with average concentrations of 1.07and 1.08μg N m^{-3}, respectively. Plains grasslands had NH_3 concentrations during the key growing season (June to October) of 1.53μg N m^{-3} at CP and 1.94μg N m^{-3} at MP (Table 2). The NH_3 values during the growing and non-growing seasons were significantly different ($P=0.008$). Except for MM, significantly higher NH_3 occurred during the growing season, especially at the croplands ($P=0.026$; Fig. 5).

3.3 Atmospheric NO_2 concentrations

CC had the highest NO_2 concentration with an average value of 3.8μg N m^{-3}over the year and a maximum value of 8.1μg N m^{-3}in June. MC had lower NO_2 with an average value of 2.4μg N m^{-3}over the year. MM had a significantly higher NO_2 (2.6μg

N m^{-3}) than CM (1.6μg N m^{-3}). The CP grassland had a slightly higher NO_2 concentration of 2.2μg N m^{-3} in the key growing season (June to October) than did MP with 1.5μg N m^{-3} (Table 2). The NO_2 values in the growing and non-growing seasons were significantly different ($P<0.001$) for CC and MM. However, NO_2 concentrations were similar for the CM grassland and MP ($P>0.322$; Fig. 6).

3.4 Particulate Nr species in the air

Because of technical limitations, we chose only cropland in both countries (CC and MC) as the monitoring points for particulate Nr. The monthly pNH_4^+ concentrations were 0.75 and 0.53μg N m^{-3} for CC and MC, respectively (Table 2). The CC had a significantly higher pNH_4^+ concentration than the MC ($P=0.033$). The pNH_4^+ concentration peaked from July to August and the highest value (2.66 μg N m^{-3}) was attained in July (Fig.7). Monthly pNO_3^- concentrations were 0.37μg N m^{-3} at CC and 0.11μg N m^{-3} at MC (Table 2). The CC had a significantly higher pNO_3^- concentration than the MC ($P=0.008$) with peaks from July to August and April to May and a maximum value of 1.38μg N m^{-3} in May.

In addition, the growing season of cropland had higher pNH_4^+ concentrations, especially in CC. Average pNH_4^+ concentrations of CC were 60% higher than those for MC. For pNO_3^- concentrations values were similar between growing season and non-growing season in both countries ($P=0.302$). However, average pNO_3^- concentrations of CC was three times higher than for MC in a whole year (Fig.8).

3.5 Wet, dry and total N deposition

Annual wet N deposition amounted to 2.0-3.1 kg N ha^{-1} yr^{-1}at the Chinese sites and 1.8-3.2kg N ha^{-1} yr^{-1} at Mongolian sites. Among the six sampling sites, the highest wet deposition occurred at the MM and CC reflecting high precipitation or high NH_4^+-N and NO_3^--N concentration, with values of 3.1 and 3.2 kg N ha^{-1} yr^{-1} for the Mongolian and Chinese sites, respectively. Wet Deposition was smallest at MC given lowest precipitation. Wet deposition rates at other sites fell in-between. The CC had the highest N dry deposition rate (9.5 kg N ha^{-1}). The second was the MC with 5.4 kg

N ha^{-1}. The MM grassland had a higher dry deposition (3.4 kg N ha^{-1}) than its Chinese counterpart (2.7 kg N ha^{-1}). Dry deposition rates in plain grasslands were similar across countries.

Total N deposition in CC was 75 % higher than in MC, but MM grassland had a higher total N deposition than CM grassland. The MP grassland had a similar total N deposition (5.7 kg N ha^{-1}) compared to CP grassland (5.7 kg N ha^{-1}). In addition, the wet N deposition species (NH_4^+ and NO_3^-) altogether accounted for 25-48% at the Chinese sites and 25-49 % at the Mongolian sites. Dry N deposition accounted for 52-75% at the Chinese sites and 51-75% at Mongolian sites.

4 Discussion

4.1 Atmospheric dry and wet N deposition

Dry deposition includes gas emissions and particulate Nr deposition (Shen et al. 2013; Granath et al. 2014; Maaroufi et al. 2015). In our experiment CC had significant higher NH_3 and NO_2 concentration than the other land use types, mainly due to the large area of cropland on the Chinese side of the border, together with the excessive inputs of mineral N fertilizer, which likely led to large losses *via* NH_3 volatilization and soil NOx emissions. Compared to MM, we found that the CM had a significant higher NH_3 concentration from June to September than MM, which was mainly due to more livestock and excrements in the mountain per unit area ($p<0.05$). However, NH_3 concentration showed the opposite trend in winter in MC, which could have been a as result of having more livestock numbers staying in the Mongolian mountain in winter time due to the difference of traditional grazing practices. Except for NH_3 deposition, the MM had higher NO_2 depositions than the CM grassland, probably as a consequence of many herdsmen staying in the Mongolian mountain site, especially in winter when large amounts of coal, wood and cattle manure are burned for home heating from October to May. However, herdsmen in China move to the mountains only from July to mid-September in summer, with very few people living there during the winter. A similar explanation could be suggested for the wet deposition.

The monthly concentrations of NH_3 showed significant positive correlations with temperature (P=0.009) but no correlation with either RH (P=0.491) or NO_2 (P=0.580; Fig.10).This result was consistent with other findings, for example, a similar trend was also found in Guangzhou in south China and in an agricultural catchment in subtropical central China (Yang et al. 2010; Shen et al. 2013). This suggests that increasing temperature promotes the emission of NH_3. Gaseous NO_2 was also positive correlated with temperature but neither with RH nor NH_3. This may also imply that NO_2 emissions mostly occur as a consequence of human activities, especially the combustion of fossil fuels and automobile exhausts with similarly results in other places but a relatively low value in agro-pastoral areas. The amount of rainfall had a significant effect on the concentration of inorganic N. The higher amount of precipitation, the lower the inorganic N concentration, especially in the case of NH_4^+ which was significantly correlated with the precipitation (P=0.039). NH_4^+ and NO_3^- were not significantly correlated with one another (P=0.143), which indicated that the results for the wet deposition are greatly influenced by the dry deposition in our research area. All in all, the different land use types did not differ significantly in their wet deposition in either country.

4.2 Effects of N deposition on N cycling in the agro-pastoral transition zone between China and Mongolia

The effects of N deposition in the agro-pastoral transition zone are different from other areas. Nitrogen deposition in agriculture areas is mainly affected by fertilizer application rates (Li et al. 2012). However, in the agro-pastoral transition zone, a part of fertilizer applications, the seasonal migration of livestock (transhumance) leads to the general distribution of large amounts of animal manures and which represents the second N emission source. So in this study we found that the dry deposition had higher percentage than wet deposition in agro-pastoral catchments in cropland. However, a different result occurred in the mountain grassland with almost equal proportions for wet and dry deposition which may be explained by higher rainfall with low NH_3 and NO_2 concentrations in mountain grassland and relatively lower rainfall with higher NH_3 and NO_2 concentrations in cropland in this catchment. The different proportion of atmospheric wet and dry N deposition in mountain grassland and cropland appears to be the important feature in agro-pastoral area with different altitudinal gradients.

Furthermore, as well as other studies (Aber et al. 1997; Flechard et al., 2011; Azati et al. 2014), we found that the grasslands face serious N losses especially in mountain areas where large numbers of grazing animals move into the mountain grassland and remain for 3-4 months leading to substantial loss of nutrients under long-term migration conditions. In China, during the last decades, in parts of the Altay- Dzungarian region, laws and policies were implemented to intensify livestock production while reducing the rate of land degradation (Greta et al., 2016), however, the overgrazing phenomenon is also very common, which still leads to the loss of nutrients with livestock being transported to other cities. Instead in Mongolia a larger proportion of the meat is consumed locally and the majority of livestock continue to graze in the mountains throughout the year, leading to more closed N cycles in grassland areas. Thus, N cycles in grasslands are different in China and Mongolia, with Chinese grasslands facing a more pronounced risk of N losses. Our study shows that N deposition in cropland differs between Mongolia and China, mainly due to higher application rates of fertilizer N in China. Therefore China and Mongolia exhibit different N deposition and N cycling rates reflecting different land use management intensities, grazing systems and trading conditions.

4.3 The uncertainty of the compensation point between the NH_3 emission and deposition

The concentration of NH_3 in the air is susceptible to be affected by meteorological and anthropogenic factors. On the one hand, part of atmospheric ammonia settled onto the soil surface, And part of NH_3 volatilize from the surface soil. Therefore, it is difficult to accurately estimate net NH_3 deposition under the conditions of this study.

In order to better estimate the NH_3 deposition value, it is common practice to calculate the deposition velocity rate by means of meteorological factors to get the appropriate deposition compensation point. In our study, the land use included mountain grassland (alpine meadow), plain grassland and farmland. In the farmland, 5.0μg N m^{-3} was assumed as the compensation point of dry deposition of NH_3 in the growing season (Shen et al. 2013), and 0 μg N m^{-3} was assumed as the compensation point of dry deposition of NH_3 in the no-growing season due to low NH_3 volatilization. In the mountain and plain grassland, 0 μg N m^{-3} was chosen as the compensation point of dry deposition of NH_3 due to low NH_3 volatilization (Li et al., 2012; Shen et al. 2013). Except the NH_3 compensation point, the value of wet and dry deposition for different land use styles had the interannual variation due to the change of cropland area and number of livestock with climatic variation in local area. Beyond that, we observed that N deposition was spatially very unevenly distributed, particularly between mountain pastures and plain pastures. Nitrogen deposition was possibly higher next to herdsmen's houses, roads or sheepfolds due to more pronounced NH_3 or NO_X releases. Farm- and grasslands are intertwined in our research areas. Therefore, much uncertainly for wet and dry N deposition remain.

5 Conclusions

The agro-pastoral area around Qinghe (China) and Bulgan (Mongolia) differed in atmospheric N deposition across land use types. The mountain grasslands had relatively higher wet deposition reflecting much higher rainfall and Nr emissions. Chinese croplands had higher wet and total N deposition than Mongolian croplands due to higher population and chemical fertilizer input, but higher N deposition were found in the Mongolian mountain grassland than Chinese mountain grassland due to different grazing systems. Nearly all land use types had higher N deposition in the (warm) growing season than in the winter months. Compared to Mongolia, Chinese grassland faces more pronounced Nr losses due to additional N deposition and overgrazing, suggesting that a reduction of the application of N-fertilizers to croplands as well as livestock numbers would help to decrease N deposition.

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Table 1 Description of the six sampling sites in the Chinese and Mongolian Altay Mountains.

	Site	Land use	Latitude	Longitude	Elevation (masl)	Annual mean temperature (℃)	Annual precipitation (mm)	Sampling period
China	Qinghe (CC)	Cropland	46°44'	90°19'	1126	5.4	123	Jun 2014 -May 2015
	Huzert (CM)	Mountain grassland	46°40'	90°24'	1605	1.2	149	Jun 2014 -May 2015
	Guojiazhan (CP)	Plain grassland	46°08'	89°58'	1284	4.3	78	Jun 2014-Oct 2015
Mongolia	Bulgan Sum (MC)	Cropland	46°6'	91°34'	1184	3.9	56	Jun 2014 -May 2015
	Turgen (MM)	Mountain grassland	46°49'	91°21'	1889	-0.5	161	Jun 2014 -May 2015
	Bayangol (MP)	Plain grassland	46°20'	91°25'	1323	4.2	83	Jun 2014 -Oct 2015

Table 2 Annual volume-weighted mean concentrations of NH_4^+-N and NO_3^--N in rainwater and the annual mean concentrations (standard deviation) of gaseous and particulate Nr species in the air at the six sampling sites in the Chinese and Mongolian Altay Mountains.

Site	NH_4^+-N mg N L^{-1}	NO_3^--N mg N L^{-1}	NH_3 μg N m^{-3}	NO_2 μg N m^{-3}	pNH_4^+ μg N m^{-3}	pNO_3^- μg N m^{-3}
CC	1.6 (0.2-6.2)[*]	1.0 (0.4-2.1)	3.1(1.8-7.4)[b]	3.8(0.9-8.1)	0.8(0.3-2.8)[a]	0.4(0.1-1.38)[a]
CM	1.0 (0.1-3.1)	0.7 (0.1-1.2)	1.1(0.3-2.3)	1.6 (0.1-2.6)		
CP	0.6 (0.3-0.8)	2.0 (1.4-3.2)	1.5(0.3-2.8)	2.2(1.8-3.3)		
MC	2.0 (0.2-5.9)	1.2 (0.5-2.0)	1.7 (0.9-3.3)	2.4(0.6-5.8)	0.5(0.1-1.2)[b]	0.1(0.03-0.43)[b]
MM	1.2 (0.4-5.5)	0.8 (0.3-1.7)	1.1(0.6-1.8)	2.6(0.2-5.5)		
MP	1.8 (0.3-3.7)	0.7 (0.2-3.4)	1.9(1.1-2.8)	1.5(0.2-3.1)		

[*] Values in the parentheses indicate the variation range of the Nr of the rain across the whole year.

[a, b] Different letters within the same column indicate statistical differences in variables mean among land use types as shown by Tukey's multiple range test ($P<0.05$).

Table 3 Wet and dry N deposition (kg N ha^{-1} yr^{-1}) at the sampling sites in the Chinese and Mongolian Altay Mountains from June 2014 to May 2015.

	Site	Rainfall (mm)	Wet deposition		Dry deposition[a]				WD[b]	DD	TD
			NH_4^+	NO_3^-	NH_3	NO_2	pNH_4^+	pNO_3^-			
China	CC	123	3.1	1.2	5	3.6	0.6	0.3	3.1	9.5	12.6
	CM	149	0.7	0.3	1.4	1.3			2.5	2.7	5.2
	CP	78	1	0.4	1.9	1.8			2.0	3.7	5.7
Mongolia	MC	56	1.8	0.5	2.9	2.0	0.4	0.1	1.8	5.4	7.2
	MM	161	0.7	0.3	1.4	2.0			3.2	3.4	6.6
	MP	83	1.3	0.2	2.5	1.1			2.1	3.6	5.7

[a] Dry deposition velocities of NH_3 was 0.4, 0.55 and 0.42 cm s^{-1} for CM, CC and CP, 0.41, 0.52 and 0.41 cm s^{-1} for MM, MC and MP respectively. Dry deposition velocities of NO_2 was 0,26, 0.3 and 0.26 cm s^{-1} for CM, CC and CP, respectively. 0.24, 0.26 and 0.26 cm s^{-1} for MM,MC and MP respectively. Dry deposition velocities of pNH_4^+ and pNO_3^- was 0.2-0.22 cm s^{-1} for CC and 0.20- 0.22 cm s^{-1} for MC, the method was from Shen et al.(2013)

[b] WD: total wet N deposition, DD: total dry N deposition, TD: total N deposition

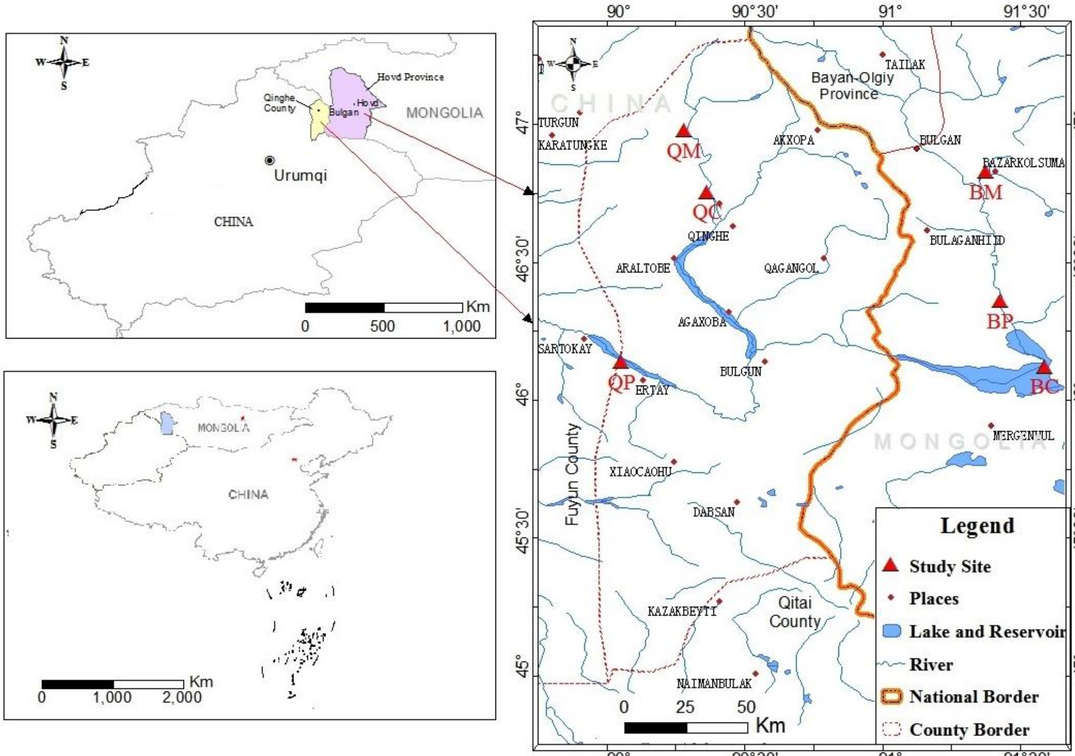

Fig.1. Map of the six sampling sites in the agro-pastoral catchment of the Chinese and Mongolian Altay Mountains.

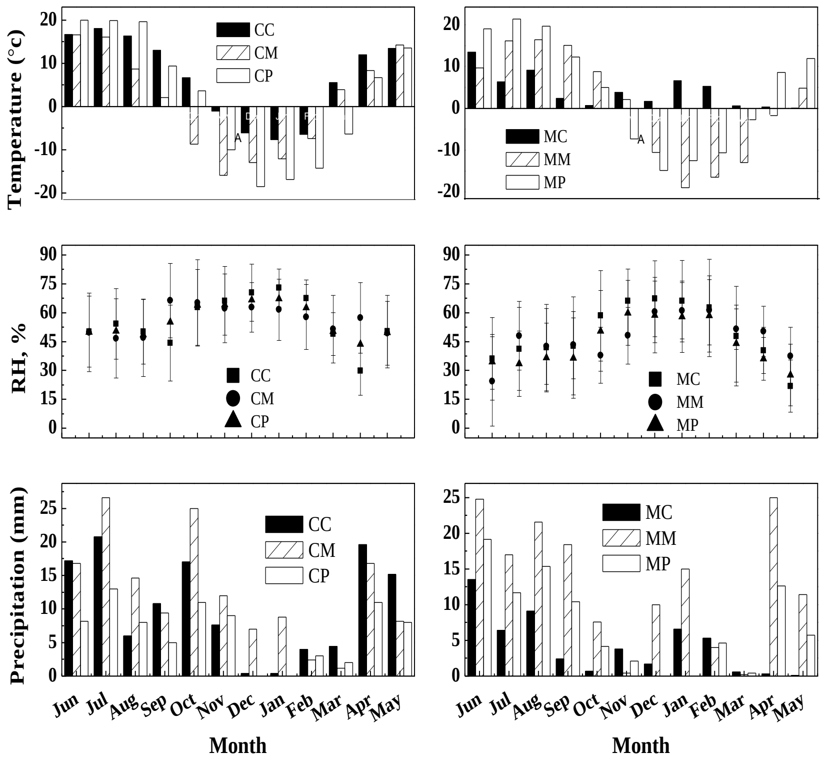

Fig. 2. Monthly mean air temperature and relatively humidity (RH) at six sampling sites of the Chinese and Mongolian Altay Mountains.

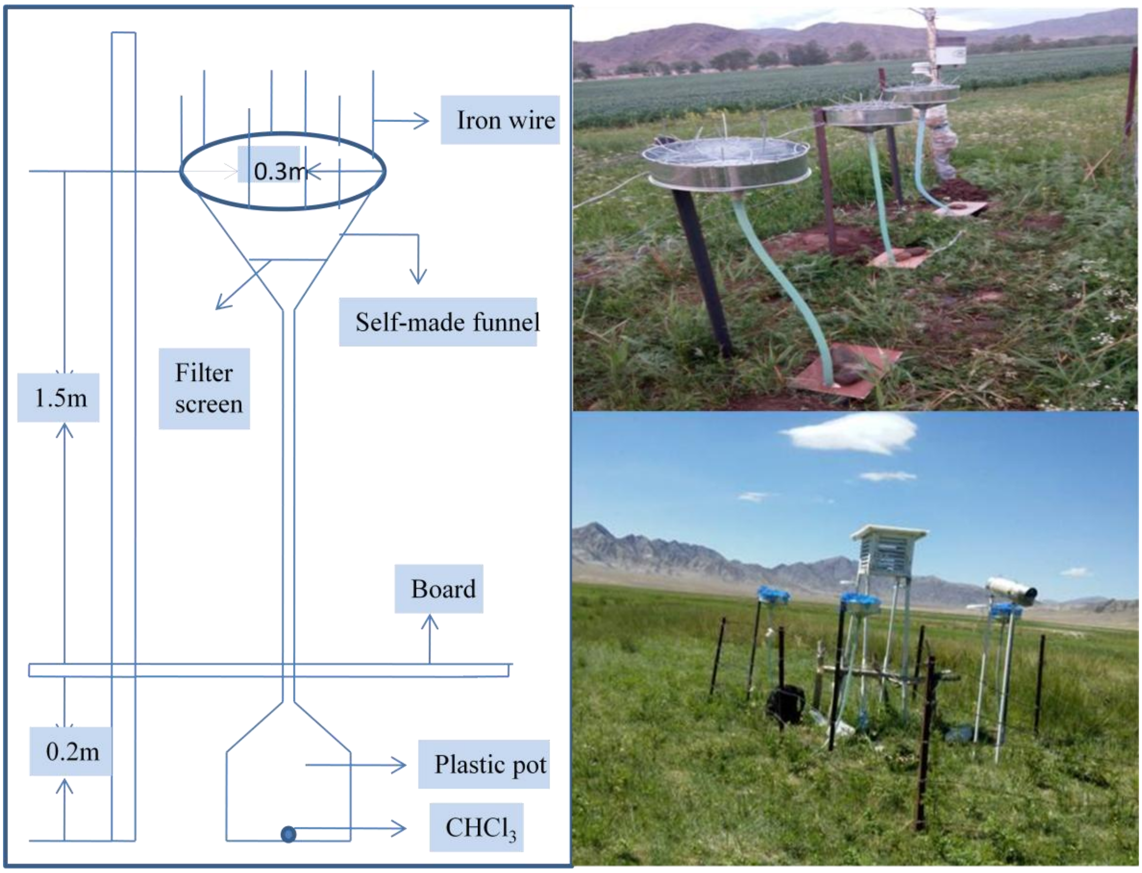

Fig. 3. The self-made wet collection equipment at the sampling sites in the Chinese (up right) and Mongolian Altay Mountains (down right)

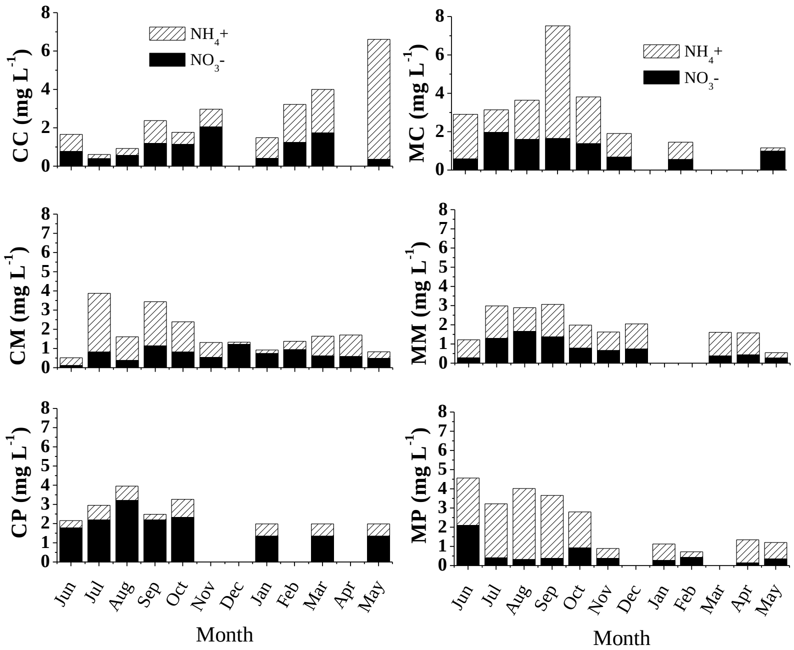

Fig.4. Concentration of NH_4^+-N and NO_3^--N of wet deposition at six samples sites in the Chinese and Mongolian Altay Mountains

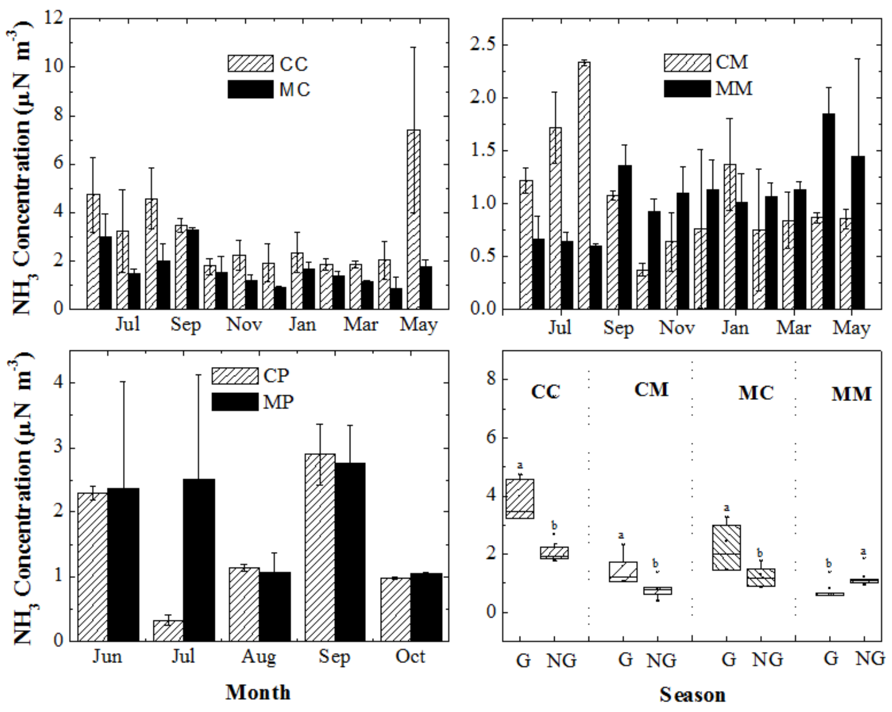

Fig.5. Monthly concentrations of NH_3-Nin the growing season (G) and the non-growing season (NG) at six sites in the Chinese and Mongolian Altay Mountains

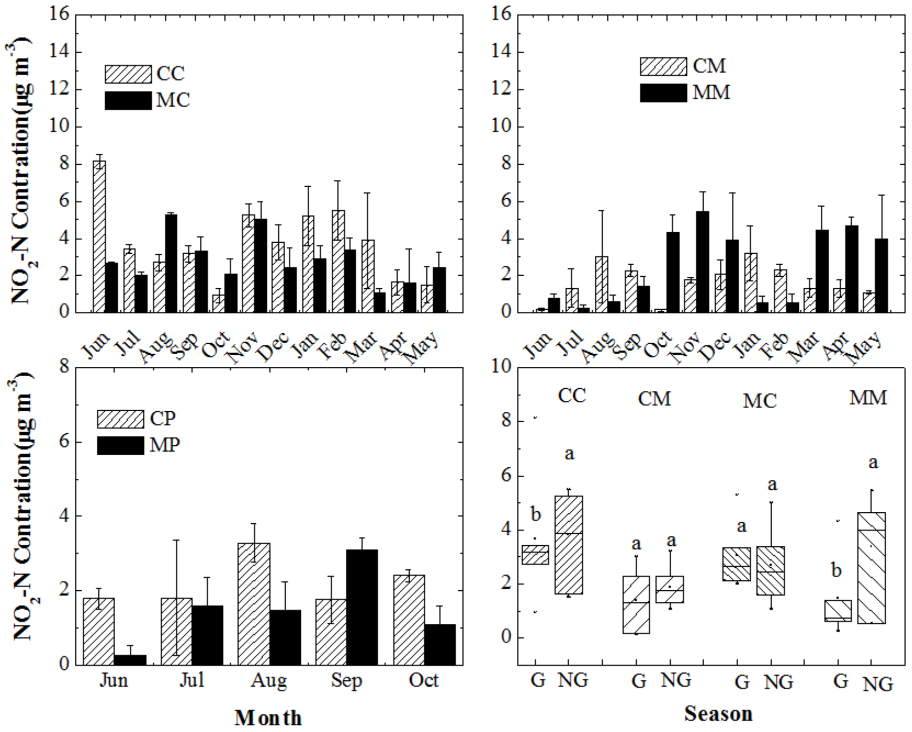

Fig.6. Monthly concentrations of NO_2-N in the growing season (G) and the non-growing season (NG)of six sites in the Chinese and Mongolian Altay Mountains

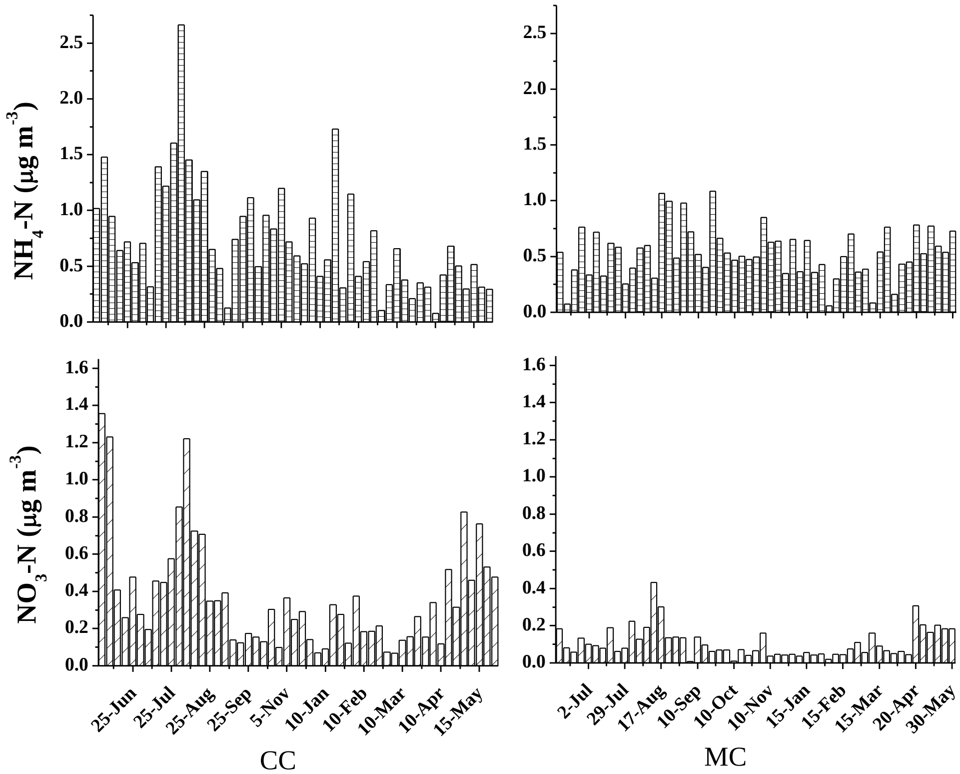

Fig.7. Monthly concentrations of NO_2-N at six sampling sites in Chinese and Mongolian Altay Mountains

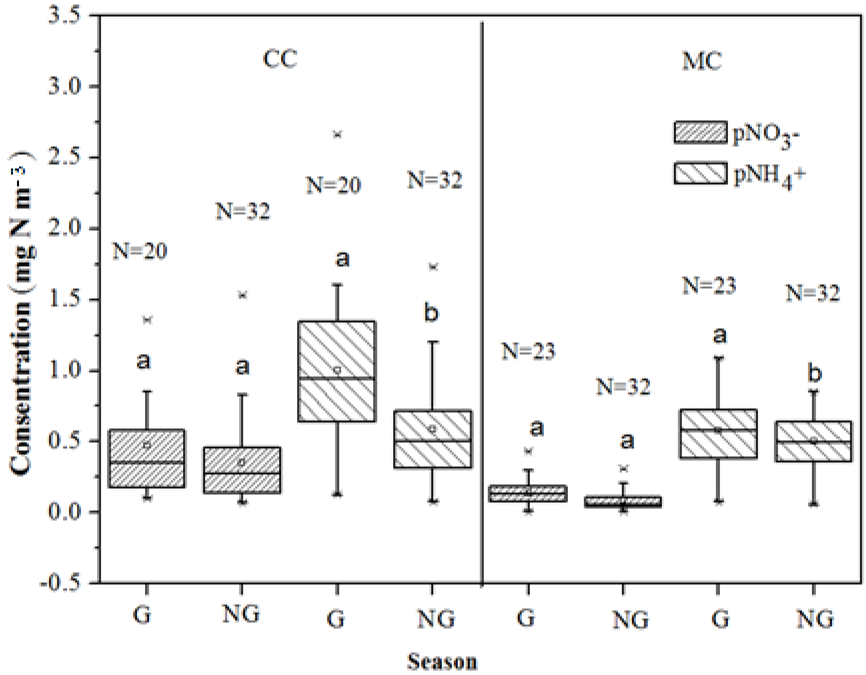

Fig.8. Concentrations of NO_2-N in the G (growing season) and the NG (non-growing season) at six sites in the Chinese and Mongolian Altay Mountains

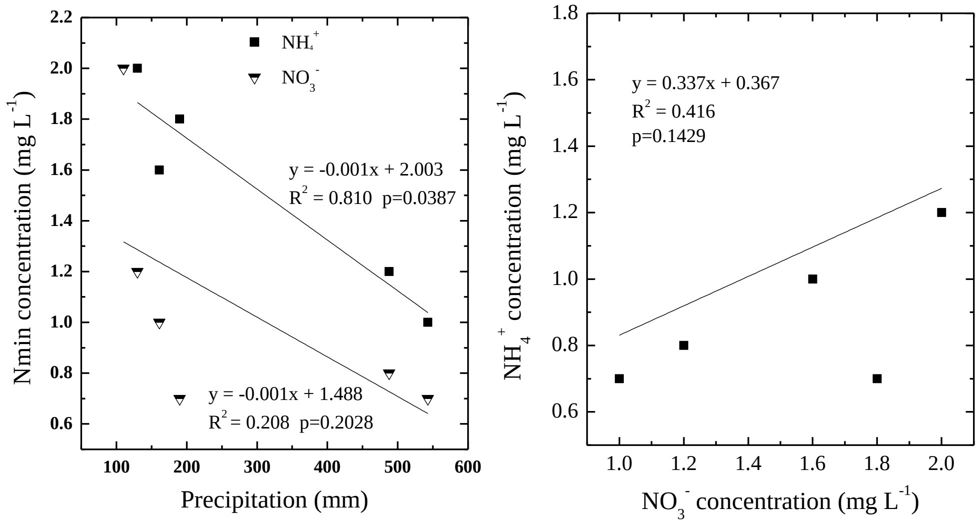

Fig.9. Relationship between monthly precipitation and NH_4^+-N and NO_3^--N in rainwater at six sampling sites in the Chinese and Mongolian Altay Mountains

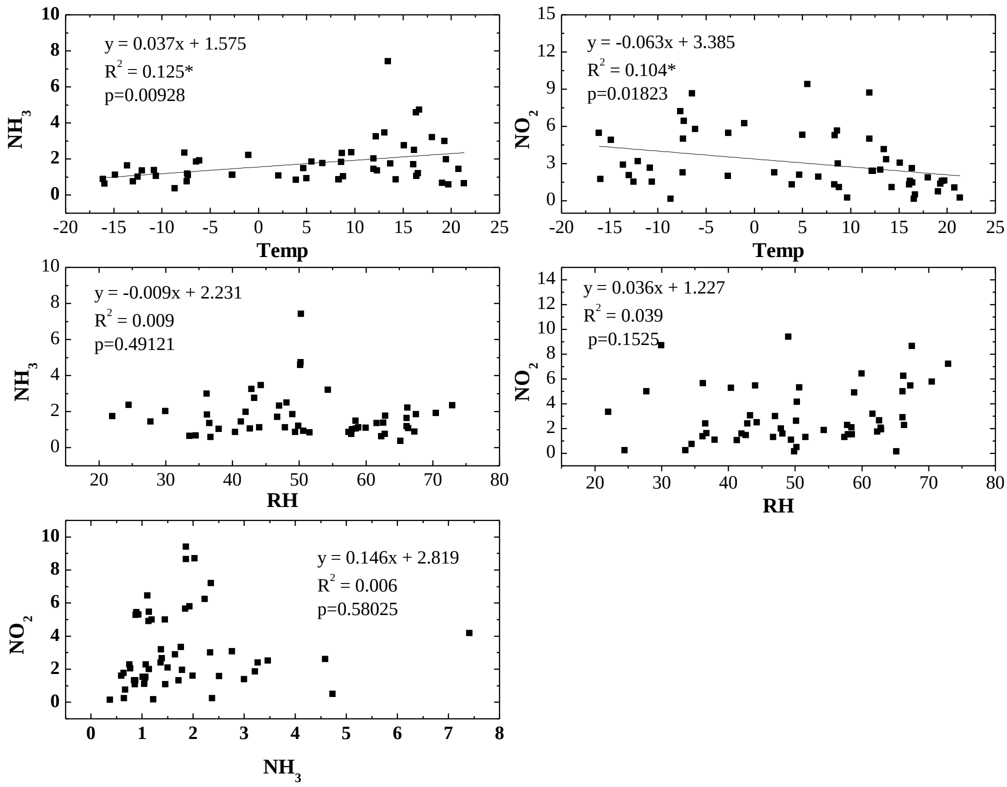

Fig.10. Relationship between atmospheric NH_3 and NO_2 and temperature (Temp) and relatively humidity (RH) in the Chinese and Mongolian Altay Mountains.