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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 of nitrogen (N) deposition in agroecosystems have been conducted along landuse and altitude gradients. In an effort to fill this gap of knowledge we selected three typical, interconnected landuse systems (cropland, mountain grassland and plain grassland)at six sampling sites in the transboundary Altay Mountains of NW China and SW Mongolia to compare the dynamics and amounts of wet and dry N deposition. 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. The croplands 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 of NO_3^--N (1.0 mg N L^{-1}in China and 1.2 mg N L^{-1}in Mongolia) in precipitation compared with the other land use types for wet deposition. In contrast, the Mongolian mountain grasslands experienced the highest wet deposition (3.2 kg N

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ha^{-1} yr^{-1}) which was at least partly due to higher summer precipitation (161 mm), the second highest wet deposition occurred on Chinese cropland with 3.1 kg N ha^{-1} yr^{-1} while wet deposition in other landuse types ranged from 1.8 to 2.5 kg N ha^{-1} yr^{-1}. Chinese cropland had the highest NH_3 (3.1μg N m^{-3}) and NO_2 (3.8μg N m^{-3}) concentrations and dry N deposition (15.3kg N $ha^{-1}yr^{-1}$) among all landuse types while Mongolian cropland had dry N deposition of 8.9 kg N $ha^{-1}yr^{-1}$. Chinese cropland (18.4 kg N ha^{-1} yr^{-1}) had the highest total N deposition, followed by the Mongolian cropland with 10.7 kg N ha^{-1} $yr^{-1}$and the Mongolian mountain grassland with 10.5 kg N ha^{-1} yr^{-1}. NH_4^+-N concentration were negatively correlated with precipitation ($P<0.05$). Concentration of NH_3 correlated positively with air temperature ($P<0.05$) likely reflecting promoting effects of temperature on NH_3 emissions whereas NO_2 correlated negatively with temperature. Over all, croplands in China had 72% higher N deposition than in Mongolia whereas the reverse was true for mountain grasslands which received 31% more Nin Mongolia.

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

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Introduction

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

There are several factors responsible for the increases in atmospheric N deposition whereby agriculture and animal husbandry are two important sources (Aber et al. 1997; Granath et al. 2014; Simpson et al. 2014;Yang et al. 2010). The rapid intensification of agriculture in response to the increasing demand for food has led to enhanced use of mineral fertilizers in China, a key factor responsible for high regional Nr deposition (Ju et al. 2009; Zhang et al. 2013). In addition, higher living standards have increased the demand for meat and the development of intensive livestock production systems (Huang et al. 2013; Wang et al. 2010).Overgrazing is also an important problem that has been the subject of new landuse policies and even remote agro-pastoral zones are affected by grazing pressure. This limits the productivity of grasslands, depletes nutrients in grassland soils and may jeopardize the productivity of animal husbandry systems.

Although some N deposition studies have been conducted in agricultural and pastoral areas (Luo et al. 2003; Li et al. 2012; Du et al. 2014; Basto et al. 2015; Huang et al. 2015; Liu et al. 2015; Wasiuta et al. 2015), little research has been conducted to quantify atmospheric N deposition in the agro-pastoral zone. In China over 12 provinces comprising 160 towns and around 117 million people depend on this landuse system which is 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). Monitoring of atmospheric dry and wet N deposition is therefore urgently needed in this region.

The present study was conducted at the Altay Mountains, at the border area of northwest China and western Mongolia representing a typical agro-pastoral transition zone. To compare the effects of exposition and landuse intensity, we choose the same agro-pastoral landuse type on the eastern Mongolian slope of the Altay Mountains. The two nearby study areas have similar landuse types but different cropland/grazing land ratios, intensities of mineral fertilizer input, types of animal husbandry systems and levels of urban development. The combination of these factors may lead to significant differences in N deposition across the year. 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.

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2. Materials and methods

2.1. Sampling sites

The study was conducted in Qinghe county, northwest China and adjacent Bulgan county in Mongolia comprising a study 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 is1218mabove sea level (a.s.l.), with a maximum elevation of 3659 m a.s.l. and a minimum of 900 ma.s.l.. Qinghe county is situated in the continental north temperate arid climatic zone, without four distinct seasons. Year round air humidity is very low 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 and cultivated land amounts to 0.126 million hectares with 1.64 million livestock heads sent to market annually. Most of the agricultural area is planted to spring wheat (*Triticumaestivum* L.), but alfalfa (*Medicago sativa* L.) and sea buckthorn (*Hippophaerhamnoides* L.) are also widely grown. Wheat is sown in early May and harvested at the end of September. The amount of mineral N 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 from July to September while during the winter months they remain for stubble grazing in the oasis croplands and desert margins.

Founded in 1931 Bulgan county (or Soum) neighbors 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 1164 m 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.

The most of the cropland of Bulgan soum is also planted to spring wheat and to a lesser degree to rye (*Secalecereale*L.), but less intensively with 150-250 N kg ha^{-1} yr^{-1}. Similarly to Qinghe, the growing season lasts from May to September and major numbers of sheep, cattle and camels are moved into the mountain grassland from July to September and spend the winters in the lowlands.

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 herdsman and the dates and amounts of rainwater were recorded. In addition, 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 in collecting samples after every rainfall event. Each rain collector comprised a funnel container 40 cm in diameter, a plastic hose and a 10-l plastic bucket. The funnel container was set at a height of around 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 meteorological station nearby Meteorological Bureau of Qinghe Country. The precipitation samples were collected manually once per month at the six sampling points and transferred to plastic bottles followed by storage in a refrigerator at

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-10 °Cuntil analysis. We also sampled snow at the beginning and the end of the snowfall period and combined it with the snowfall data to 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 from the UK Environmental Change Network (Goulding et al., 1998; Bush et al., 2001). The NO_2 samplers consisted of polyethylenetubes (71.0 mm long and 11.0 mm internal diameter), 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 a height of 1.5 m (at least 0.5 m higher than the canopy height) above ground and exposed between15 days 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(Plaisance et al., 2004).

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 placedabout1.5 m above ground. The calculation was made according to Luo et al. (2014) as follows:

$$V=DAt/L$$

Where t represents the time interval; D=2.09×10^{-5}m^{-2}s^{-1} at 10°C, A=3.463×10^{-4}m^{-2}, L=0.006m. 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 whose aerodynamic equivalent diameter is < 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}, and 7-10 daily samples of PM_{10} were collected at QC and BC during each month. Samples at other sites were not taken given lacking power and harsh environmental conditions. The membrane of PM_{10} was 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 refrigerated at 4 °C. Ammonium and nitrate in PM_{10} (pNH_4^+ and pNO_3^-) were measured using a Seal AA3continuous flow analyzer (Seal Analytical Ltd., Southampton, UK).

2.3 Estimation of dry N deposition

Data on dry N deposition are complicated to collect given the effects of variable weather conditions and differences in vegetation types. We did not use micro-meteorological methods because of unavailability of equipment to measure dry N deposition. Rather we estimated dry N deposition by multiplying the measured concentrations of Nr species by their deposition velocities (V_d) obtained from related studies published in the literature (Shen et al. 2013; Yu et al. 2014). 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 (Shen et al. 2009). Because we did not measure V_d, the V_d values of the Nr species under different landuse types were obtained from Flechard et al. (2011) for simplification.

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 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 Mongolian cropland (MC) had the highest NH_4^+-N concentration in the wet deposition compared with the Mongolian Mountain grassland (MM) and the Mongolian Plain grassland (MP)(Table 2, Fig. 3).The NH_4^+-N concentrations of the samples from Chinese sites were relatively low compared with the Mongolian sampling sites. Chinese Cropland (CC) had are relatively high NH_4^+-N concentration compared with the other two sampling sites in the Chinese Mountain grassland (CM) and the Chinese Plain grassland (CP). NO_3^--N concentrations were highest for CP in China, followed by MC in Mongolia.

The different landuse types had different NH_4^+-N and NO_3^--N peaks. Highest cropland NH_4^+-N occurred in May in China and in September in Mongolia and the NO_3^--N peak occurred from March to May in China and from August to October in Mongolia (Fig.3). The two countries had similar mountain pasture (CM and MM)

NH_4^+-N concentration peaks. Both occurred from July to September, and NO_3^--N peaks were recorded from July to September at QP and BM. The Chinese and Mongolian Plain grasslands (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 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 Net NH_3 deposition concentrations

Cropland had the highest NH_3 concentrations of 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). With exception of MM, significantly higher NH_3occurred during the growing season, especially at the croplands (P=0.026) and the mountain grassland had a significantly higher air NH_3 during the non-growing season (Fig. 5).

3.3 NO_2 concentrations

QC 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 power and equipment limitations we chose the croplands in both countries (CC and MC) as the monitoring points for particulate Nr. The monthly pNH_4^+ concentrations were 0.75and 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 of1.38μg N m^{-3}in May.

In addition, the growing season had higher pNH_4^+ concentrations, especially in CC. Average pNH_4^+ concentrations of CC were 60% higher than those of 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 (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 (15.3 kg N ha^{-1}). The second was the MC with 8.9 kg N ha^{-1}. The MM grassland had a higher dry deposition (7.3kg N ha^{-1}) than its Chinese counterpart (5.5 kg N ha^{-1}). Dry deposition rates in plain grasslands were similar across countries.

Total N deposition in CC was 72%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 (7.4 kg N ha^{-1}) than CP grassland (7.7 kg N ha^{-1}). The wet N deposition species (NH_4^+ and NO_3^-) altogether accounted for 16.9-31.2% at the Chinese sites and for 16.8-30%at the Mongolian sites. Dry N deposition accounted for 69-83% at the Chinese sites and 70-83% at Mongolian sites.

4 Discussion

4.1 Methodological evaluation of dry and wet deposition collection

Due to the difficult infrastructural conditions (long distances, high altitude and cross-border problems), compromises needed to be made with respect to sampling equipment and collection intervals. While the self-made equipment for rainfall sampling was established at six sites (Fig 3) from June 2014 to May 2015, for the cropland and mountain grassland, we collected rainfall or snow samples every month across the year. For the plain grassland, however, we just collected samples every month during the growing season. Outside the growing season samples were collected just for the first time and last snowfall event to compute average values. The middle flow particulate sampler was just established in the cropland given human surveillance there but not in the mountain and plain grassland where power was lacking For the NO_2 and NH_3, we collected the samples 20 days one time in six sampling points. 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.

In addition the use of the passive sampling devices may have led to severe underestimations of deposition given the hyper arid conditions of our study zone. Relative air humidity was at around 30% in summer and 80-90% in winter, and lowest winter temperatures were -40℃ in winter. Under these conditions the NH_3 (ALPHA) and NO_2 samplers were to the best of our knowledge never tested before. Similar studies show that Palmes NO_2 diffusion tubes (Gradko 7.1 cm open diffusion tubes) may be used over a temperature range from -50℃ to 40℃and a relative humidity range from 30% to 95% (Bush et al. 2001; Plaisance et al. 2004; Gerboles et al., 2005). For the ALPHA samplers the hygroscopic nature of the citric acid may allow for reliable measurements even at 30-35% RH (Perrinoet al., 2002). However, our data certainly merit methodological verification under laboratory and field conditions.

4.2 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 landuse types, mainly due to the large area of cropland on the Chinese side of the border, together with the excessive inputs of mineral fertilizer N, which likely led to large losses *via* NH_3 volatilization and soil NOx emissions.

The Chinese cropland also had the highest inorganic N concentrations and thus had higher dry deposition than the Mongolian cropland. Moreover, cropland had higher dry deposition than the other landuse types. There were usually higher NH_3 emissions in the growing season, mainly due to the fertilizer or manure applications during the growing season. The MM grassland had higher NO_2 depositions than the CM grassland, presumably because the Mongolia site had many herdsmen living in the area over most of the year and, especially in winter, large amounts of coal, wood and cattle manure are burned for home heating from October to May. Many of the herdsmen in China move to the mountains only from July to mid-September in summer, with very few people living there during the winter. Similar conclusions hold for the wet deposition.

The monthly concentrations of NH_3 showed significant positive correlations with temperature (P=0.009) but no correlation with RH (P=0.491) or NO_2 (P=0.580; Fig.10). A similar trend was also found in Guangzhou in south China and in an agricultural catchment in subtropical central China (Ju et al. 2009; Shen et al. 2013). This indicates that increasing temperature promotes the emission of NH_3. Gaseous NO_2 was also positive correlated with temperature (P=0.018) but not with RH or NH_3

(P=0.153).This conclusion is consistent that of with Luo et al. (2013) who studied dry deposition in northern China. 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.

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 (Fig.9), especially in the case of NH_4^+ which was significantly correlated with the precipitation (P=0.039). NH_4^+ and NO_3^-werenot significantly correlated with one another (P=0.143), which indicated that the results for the wet deposition are greatly influenced by the dry deposition. All in all, the different landuse types did not differ significantly in their wet deposition in either country.

4.3 The uncertainty of the compensation point between the NH_3 emission and deposition in three landuse styles

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, on the other hand, part of NH_3 volatilize from the surface soil. Therefore, it is very hard to accurately estimate net NH_3 deposition under our conditions. 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 experiment, the landuse styles included 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_3volatilization. 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). In addition we observed in our study area that N deposition was spatially very unevenly distributed, particularly between mountain pastures and plain pastures.

Nitrogen deposition was possible 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 landuse 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 the in the winter months. Compared with Mongolia, Chinese grassland faces more pronounced Nr losses due to additional N deposition and overgrazing. Thus, it is necessary to reduce the application of N-fertilizers to croplands as well as herd numbers.

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Reference

Aber, J. D., Ollinger, S. V., Driscoll, C. T.: Modeling nitrogen saturation in forest ecosystems in response to land use and atmospheric deposition, Ecol Model, 101, 61-78,1997.

Azati, A., Abudushal, N.: The influence of chemicalfertilizers on the net income of farmers and the environment., Northern Horticulture, 5, 197-200, 2014.

Bush, T., Smith, S., Stevenson, K., Moorcroft, S., Validation of nitrogen dioxide diffusion tube methodology in the UK, Atmos Environ, 35:289-296, 2011.

Du, E. Z., De , Vries. W., Galloway, J. N., Hu, X. Y., Fang, J. Y.: Changes in wet nitrogen deposition in the United States between 1985 and 2012, Environ Res Lett, 9: 095004, 2014.

Du, Z. Q., Shen, Y. D., Wang, J., Cheng, W. S.: Land-use change and its ecological responses: a pilot study of typical agro-pastoral region in the Heihe River, northwest China, Environmental Geology, 58: 1549-1556, 2009.

Flechard, C. R., Nemitz, E., Smith, R. I., Fowler, D., Vermeulen, A. T., Bleeker, A., Erisman, J. W., Simpson, D., Zhang, L., Tang, Y. S., Sutton, M. A.: Dry deposition of reactive nitrogen to European ecosystems: a comparison of inferential models across the NitroEurope network., Atmos Chem Phys, 11, 2703-2728, 2011.

Gerboles, M., Buzica, D., Amantini, L.: Modification of the Palmes diffusion tube and semi-empirical modeling of the uptake rate for monitoring nitrogen dioxide. Atmos Environ 39, 2579-2592, 2005.

Goulding, K. W. T., Bailey, N. J., Bradbury, N. J., Hargreaves, P., Howe, M. M., Murphy, D. V., Poulton, P. R., Willison, T. W.: Nitrogen deposition and its contribution to nitrogen cycling and associated soil processes, New Phytologist, 139, 49-58, 1998.

Granath, G., Limpens, J., Posch, M., Mucher, S., De, Vries. W.: Spatio-temporal trends of nitrogen deposition and climate effects on Sphagnum productivity in European peatlands, Environ Pollut, 187, 73-80, 2014.

Huang, J., Zhang, W., Zhu, X. M., Gilliam, F. S., Chen, H., Lu, X. K., Mo, J. M.: Urbanization in China changes the composition and main sources of wet inorganic nitrogen deposition, Environ Sci Pollut R, 22: 6526-6534, 2015.

Huang, J. Y., Nkrumah, P. N., Appiah-Sefah, G., Tang, S. J.: Authentication of Pure L-Leucine Products Manufactured in China by Discriminating between Plant and Animal Sources Using Nitrogen Stable Isotope Technique, J Food Sci, 78, H490-H494, 2013.

Ju, X. T., Xing, G. X., Chen, X. P., Zhang, S. L., Zhang, L. J., Liu, X. J., Cui, Z. L., Yin, B., Christie, P., Zhu, Z. L., Zhang, F. S.: Reducing environmental risk by improving N management in intensive Chinese agricultural systems., P Natl Acad Sci USA, 106, 3041-3046, 2009.

Li, K. H., Song, W., Liu, X. J., Shen, J. L., Luo, X. S., Sui, X. Q., Liu, B., Hu, Y. K., Christie, P., Tian, C. Y.: Atmospheric reactive nitrogen concentrations at ten

Biogeosciences Discuss., doi:10.5194/bg-2017-55, 2017
Manuscript under review for journal Biogeosciences
Discussion started: 10 April 2017

sites with contrasting land use in an arid region of central Asia, Biogeosciences, 9, 4013-4021, 2012.

Liu, X. J., Duan, L., Mo, J. M., Du, E. Z., Shen, J. L., Lu, X. K., Zhang, Y., Zhou, X. B., He, C. N., Zhang, F. S.: Nitrogen deposition and its ecological impact in China: An overview, Environ Pollut, 159, 2251-2264, 2011.

Liu, X. J., Zhang, Y., Han, W. X., Tang, A. H., Shen, J. L., Cui, Z. L., Vitousek, P., Erisman, J. W., Goulding, K., Christie, P., Fangmeier, A., Zhang, F. S.: Enhanced nitrogen deposition over China, Nature, 494, 459-462, 2013.

Liu, Y. W., Xu, Ri., Wang, Y. S., Pan, Y. P., Piao, S. L.: Wet deposition of atmospheric inorganic nitrogen at five remote sites in the Tibetan Plateau, Atmos Chem Phys, 15, 11683-11700, 2015.

Luo, X. S., Tang, A. H., Shi, K., Wu, L. H., Li, W. Q., Shi, W. Q., Shi, X. K., Erisman, J. W., Zhang, F. S., Liu, X. J.: Chinese coastal seas are facing heavy atmospheric nitrogen deposition, Environ Res Lett 9, 095007, 2014.

Luo, Y. Z., Yang, X. S, Carley, R. J, Perkins, C.: Effects of geographical location and land use on atmospheric deposition of nitrogen in the State of Connecticut, Environ Pollut, 124, 437-448, 2003.

Luo, X. S., Liu, P., Tang, A. H., Liu, J. Y., Zong, X. Y., Zhang, Q., Kou, C. L., Zhang, L. J., Fowler, D., Fangmeier, A., Christie, P., Zhang, F. S, Liu, X. J.: An evaluation of atmospheric N_r pollution and deposition in North China after the Beijing Olympics, Atmos Environ74, 209-216, 2013.

Maaroufi, N. I., Nordin, A., Hasselquist, N. J., Bach, L. H., Palmqvist, K., Gundale, M. J.: Anthropogenic nitrogen deposition enhances carbon sequestration in boreal soils, Global Change Biol, 21, 3169-3180, 2015.

Pan, Y. P., Wang, Y. S., Tang, G. Q., Wu, D.: Wet and dry deposition of atmospheric nitrogen at ten sites in Northern China, Atmos Chem Phys, 12, 6515-6535, 2012.

Perrino, C., Catrambone, M., DiBucchianico, A. D. M., Allegrini, I. Perrino.: Gaseous ammonia in the urbanarea of Rome, Italy and its relationship with traffic emissions, Atmos Environ, 36, 5385-5394, 2002.

Plaisance, H., Piechocki-Minguy, A., Garcia-Fouque, S., Galloo, J. C.: Influence of meteorological factors on the NO_2 measurements by passive diffusion tube, Atmos Environ, 38, 573-580, 2004.

Shen, J. L., Li, Y., Liu, X. J., Luo, X. S., Tang, H., Zhang, Y. Z., Wu, J. S.: Atmospheric dry and wet nitrogen deposition on three contrasting land use types of an agricultural catchment in subtropical central China, Atmos Environ, 67, 415-424, 2013.

Shen, J. L, Tang, A. H., Liu, X. J., Fangmeier, A., Goulding, K., Zhang, F. S.: High concentrations and dry deposition of reactive nitrogen species at two sites in the North China Plain, Environ Pollut, 157, 3106-3113, 2009.

Simpson, D., Andersson, C., Christensen, J. H., Engardt, M., Geels, C., Nyiri, A., Posch, M., Soares, J., Sofiev, M., Wind, P., Langner, J.: Impacts of climate and emission changes on nitrogen deposition in Europe: a multi-model study, Atmos Chem Phys, 14, 6995-7017, 2014.

Wang, F. H., Dou, Z. X., Ma, L., Ma, W. Q., Sims, J. T., Zhang, F. S.: Nitrogen mass flow in China's animal production system and environmental implications. J Environ Qual 39, 1537-1544, 2010.

Wasiuta, V., Lafreniere, M. J., Norman, A. L., Hastings, M. G.: Summer deposition of sulfate and reactive nitrogen to two alpine valleys in the Canadian Rocky Mountains, Atmos Environ, 101, 270-285, 2015.

Xu, M. Y., Xie, ., Wang, K.: Response of Vegetation and Soil Carbon and Nitrogen Storage to Grazing Intensity in Semi-Arid Grasslands in the Agro-Pastoral Zone of Northern China, Plos One, 9, e96604, 2014.

Xu, W., Luo, X. S., Pan, Y. P., Liu, X. J.: Quantifying atmospheric nitrogen deposition in China through a nationwide monitoring network, Atmospheric Chemistry and Physics, 15, 12345–12360, 2015.

Yang, R., Hayashi, K., Zhu, B., Li, F. Y., Yan, X. Y.: Atmospheric NH_3 and NO_2 concentration and nitrogen deposition in an agricultural catchment of Eastern China, Sci Total Environ, 408, 4624-4632, 2010.

Yu, J. B., Ning, K. , Li, Y. Z., Du, S. Y., Han, G. X., Xing, Q. H., Wu, H. F., Wang, G. M., Gao, Y. J.: Wet and Dry Atmospheric Depositions of Inorganic Nitrogen during Plant Growing Season in the Coastal Zone of Yellow River Delta, Sci World J, 15, 369-378, 2014.

Zhang, W. F., Dou, Z. X., He, P., Ju, X. T., Powlson, D., Chadwick, D., Norse, D., Lu, Y. L., Zhang, Y., Wu, L., Chen, X. P., Cassman, K. G., Zhang, F. S.: New technologies reduce greenhouse gas emissions from nitrogenous fertilizer in China, P Natl Acad Sci USA, 110, 8375-8380, 2013.

Zhang, X. H., Sun, T., Xu, J. P.: The relationship between the spread of the Catholic Church and the shifting agro-pastoral line in the Chahar Region of northern China, Catena, 134, 75-86, 2015.

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

	Site	Landuse	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 landuse types as shown by the Tukey's multiple range test (P<0.05).

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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	QC	123	1.91	1.23	7.3	7.1	0.6	0.3	3.1	15.3	18.4
	QM	149	1.49	1.04	2.5	3.0			2.5	5.5	8.0
	QP	78	0.47	1.56	3.6	4.1			2.0	7.7	9.7
Mongolia	BC	56	1.12	0.67	3.9	4.5	0.4	0.1	1.8	8.9	10.7
	BM	161	1.93	1.29	2.5	4.8			3.2	7.3	10.5
	BP	83	1.49	0.58	4.6	2.8			2.1	7.4	9.5

[a] Dry deposition velocities of NH_3, NO_2were 0.74 and 0.59, respectively, as cited from Shen et al.(2011)

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

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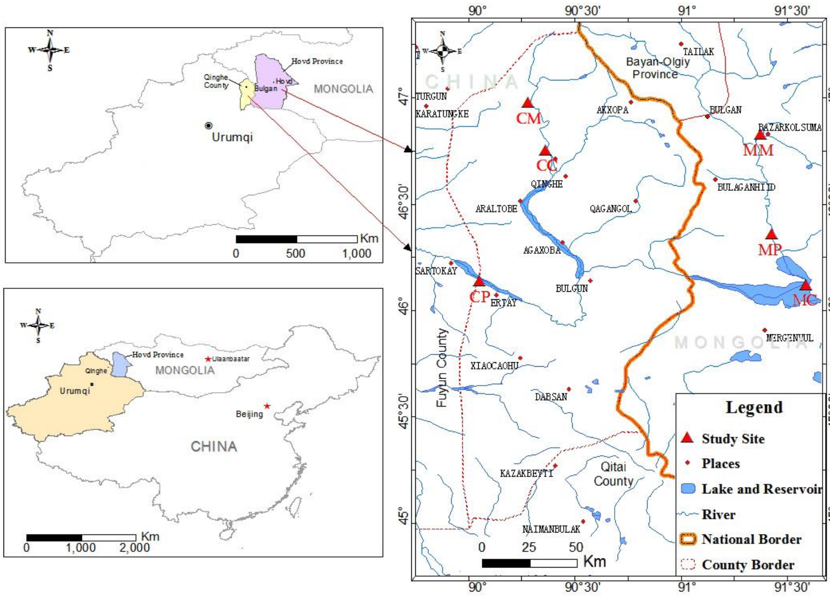

Fig.1. Map of the six sampling sites in the agro-pastoral catchment of the Chinese and Mongolian Altay Mountains.

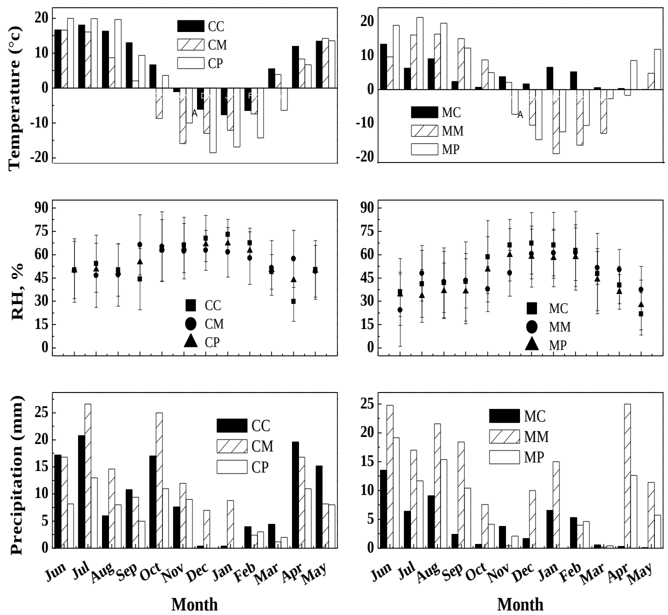

Fig. 2. Monthly mean air temperature and relatively humidity (RH) at six sampling sites of the Chinese and Mongolian Altay Mountains.

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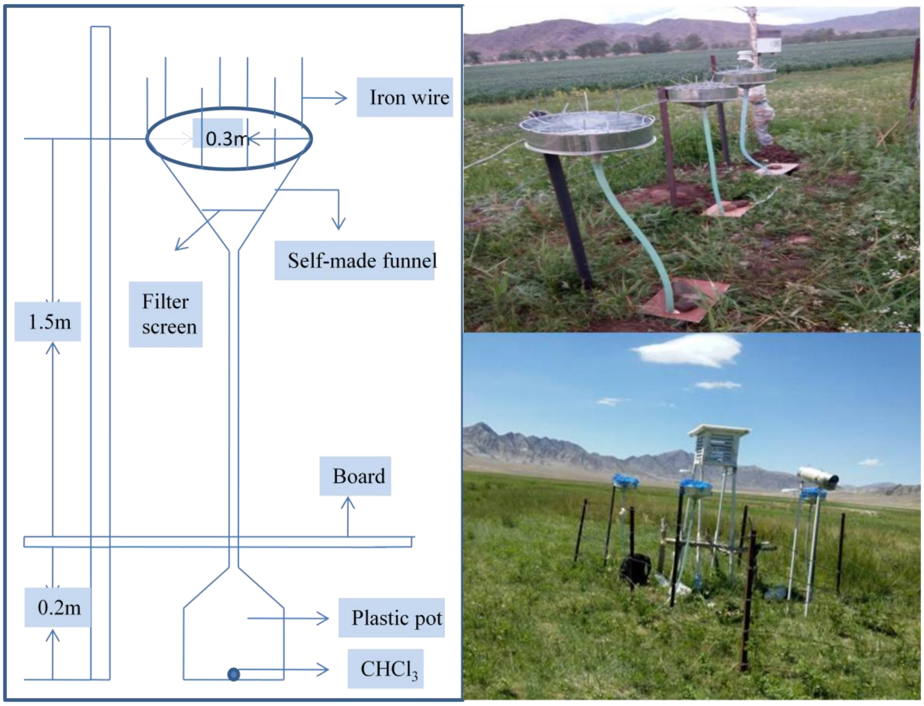

Fig. 3. The self-made wet collection equipment at the sampling sites in the Chinese (up right) and Mongolian Altay Mountains (down right)

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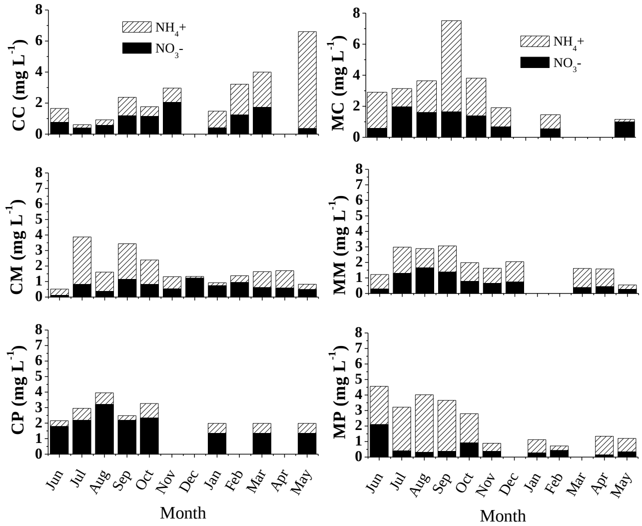

Fig.4. Concentration of NH_4^+-N and NO_3^--N of wet deposition at six samples sites in the Chinese and Mongolian Altay Mountains

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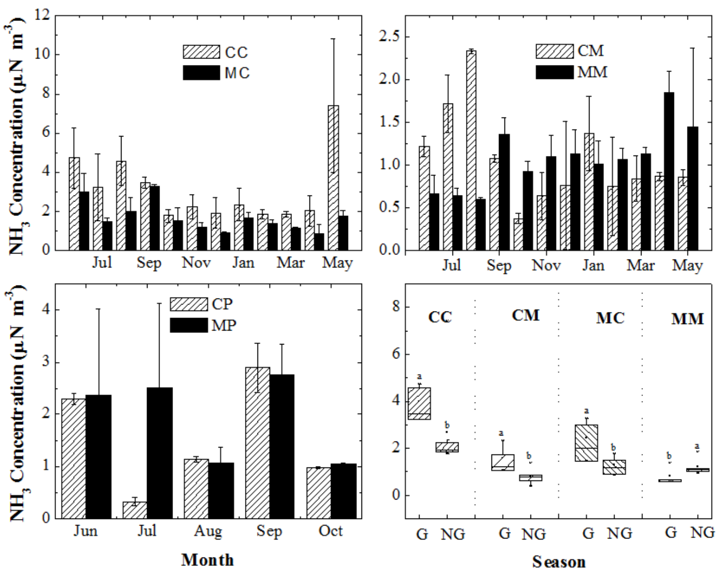

Fig.5. Monthly concentrations of NH_3-Nin the growing season (G) and the non-growing season (NG) at six sites in the Chinese and Mongolian Altay Mountains

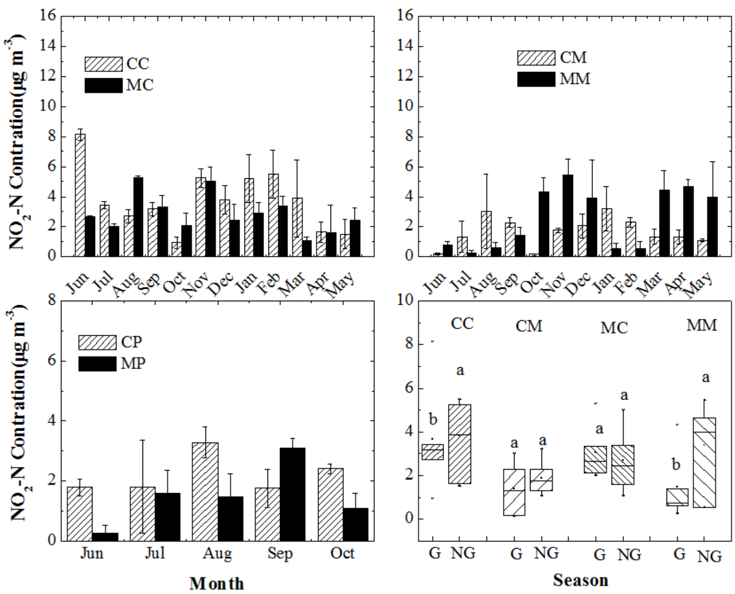

Fig.6.

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

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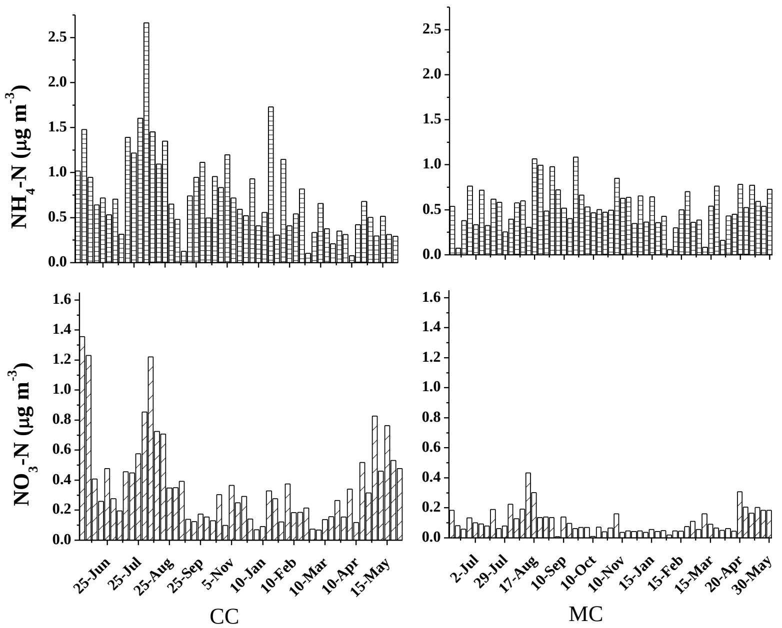

Fig.7. Monthly concentrations of NO_2-N at six sampling sites in Chinese and Mongolian Altay Mountains

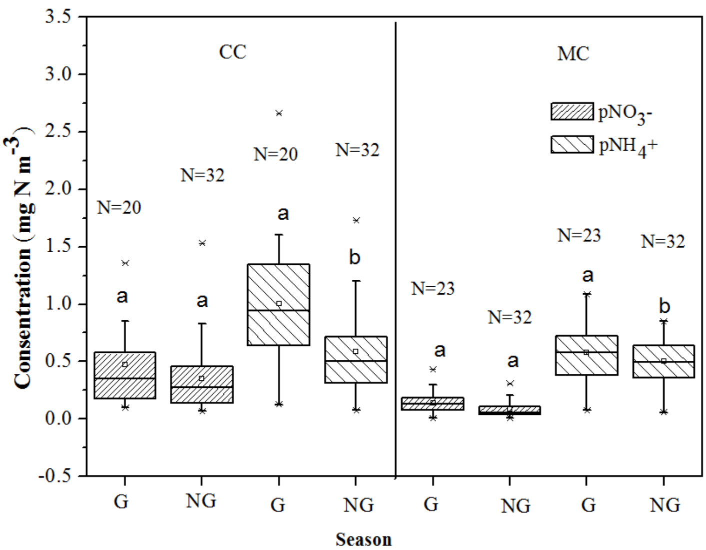

Fig.8. Concentrations of NO_2-N in the G (growing season) and the NG (non-growing season) at six sites in the Chinese and Mongolian Altay Mountains

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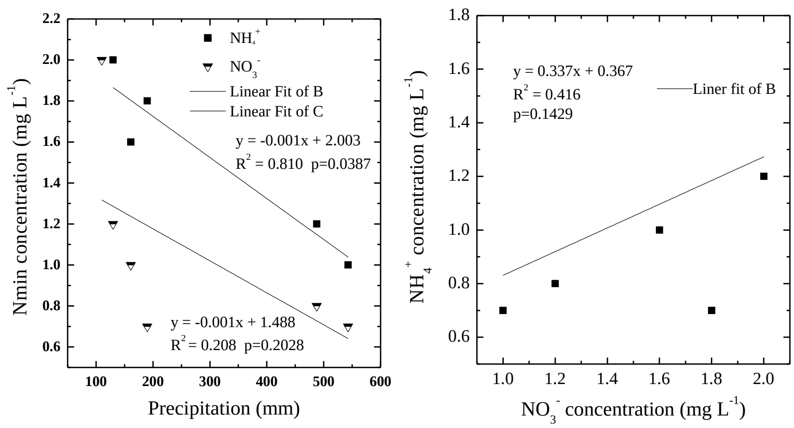

Fig.9. Relationship between monthly precipitation and NH_4^+-N and NO_3^--N in rainwater at six sampling sites in the Chinese and Mongolian Altay Mountains

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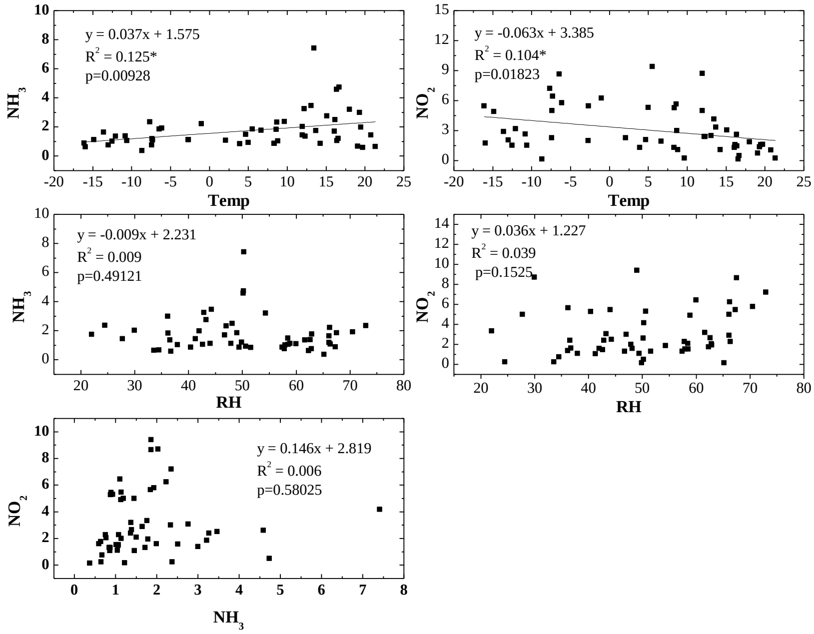

Fig.10. Relationship between atmospheric NH_3 and NO_2 and temperature (Temp) and relatively humidity (RH) in the Chinese and Mongolian Altay Mountains.