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Atmospheric reactive nitrogen concentrations at ten sites with contrasting land use in an arid region of Central Asia

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Abstract

Atmospheric concentrations of reactive nitrogen species (Nr) from 2009 to 2011 are reported for ten sites in Xinjiang, an arid region in Northwest China. Concentrations of NH_3, NO_2, particulate ammonium and nitrate (pNH_4^+ and pNO_3^-) showed large spatial and seasonal variation and averaged 7.71, 9.68, 1.81 and 1.13 $\mu g\,N\,m^{-3}$, and PM_{10} concentrations averaged 249.2 $\mu g\,m^{-3}$ across all sites. Lower NH_3 concentrations and higher NO_2, pNH_4^+ and pNO_3^- concentrations were found in winter, reflecting serious air pollution due to domestic heating in winter and other anthropogenic sources such as increased emissions from motor traffic and industry. The order of increasing total concentrations of Nr species was alpine grassland < desert, desert-oasis ecotone < desert in an oasis < farmland < suburban and urban ecosystems. Lower ratios of secondary particles (NH_4^+ and NO_3^-) were found in the desert and desert-oasis ecotone, while urban and suburban areas had higher ratios, which implies that anthropogenic activities have greatly influenced local air quality and must be controlled.

1 Introduction

Anthropogenic activities have greatly accelerated the emissions of reactive nitrogen (Nr) species worldwide (Galloway et al., 2008), with a consequent increase in atmospheric deposition of Nr species. Elevated N deposition may lead to eutrophication, declining biodiversity, soil acidification and N_2O emission (Clark and Tilman, 2008; Guo et al., 2010; Liu et al., 2011; Song et al., 2011; Li et al., 2012a,b). Nr emissions have resulted in profound deterioration of local air quality (Chan and Yao, 2008; Kulshrestha et al., 2009). Nr deposition has therefore become an important public concern (Sutton et al., 2011).

Arid and semi-arid regions account 47 % of the global land area and play a significant role in global nitrogen cycling. Xinjiang Uygur Autonomous Region is located at the center of the arid and semi-arid regions of Central Asia and is one of the main source

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areas of Asian dust which can be transported from Central Asia to the Pacific Ocean, and occasionally to the west coast of the United States (Li et al., 2008). Rapid economic development has led to significant increases in the combustion of fossil fuels by industry, power plants and vehicles in Xinjiang since 2000, with rates of consumption of diesel oil, crude oil, raw coal and coke increasing by 12, 27, 68 and 92 %, respectively (Mamtimin and Meixner, 2011). In addition, N fertilizer application has also increased steadily since the 1980s due to the expansion of cotton and cereal/fruit/vegetable production in Xinjiang. Increasing atmospheric Nr pollution would therefore be expected in this region. On the other hand, N limitation of plant growth often occurs in arid and semi-arid regions, and atmospheric Nr deposition can be an important source of plant nutrients. Thus, N inputs from Nr deposition can have an important N fertilizer effect in arid and semi-arid regions. However, very little is known about the influence of air pollutants and the dynamics of Nr concentrations in the arid regions of China. The current study attempted to determine the spatial and seasonal characteristics of atmospheric Nr pollution in different ecosystems within the arid Xinjiang region of Northwest China.

2 Materials and methods

2.1 Monitoring sites

The study was carried out at ten sites in the Xinjiang arid region in Northwestern China. Site AKS is located at Aksu Research Station for Farmland Ecosystems, Chinese Academy of Sciences (CAS), with mean annual precipitation of 45.7 mm and a mean annual temperature of 11.2 °C. Site BTH is at an experiment of Xinjiang Academy of Agricultural Sciences and is a typical farmland monitoring site with mean annual precipitation of 44.8 mm and a mean annual temperature of 12.3 °C. Site BYB is at Bayinbuluk Grassland Ecosystem Research Station, CAS. Mean annual precipitation is 265.7 mm and mean annual temperature is −4.8 °C. FKZ is at Fukang Research Station for Desert Ecology, CAS, which is situated at the southern fringe of the Gurbantunggut Desert

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in Central Asia, with mean annual precipitation of 150 mm, a highest recorded temperature is 46 °C and a lowest of −41.6 °C. TLF denotes Turpan Eremophyte Botanic Garden, CAS, where the mean annual temperature is 13.9 °C, the extreme maximum is 47.6 °C, and mean annual precipitation is 16.4 mm. SDS is located in Urumqi city, the most remote city in the world from the sea at a distance of 2500 km from the Arctic Ocean. It is surrounded by the Tianshan Mountains (3000–4500 m a.s.l.) to the north, east and west and has a mean annual temperature of 7.2 °C and mean annual precipitation of 264.8 mm. The city covers a total area of approximately 140 km^2 with a population of 2.2 million and is a typical urban monitoring site. TFS is located at an experimental farm of Xinjiang Academy of Agricultural Sciences and is a typical suburban monitoring site. SDS and TFS are only about 11 km apart and have similar air temperatures, precipitation, relative humidity and wind speeds. Site CLZ is located at Cele Desert Research Station, CAS, near the southern fringe of the Taklimakan desert, one of the three most extreme arid zones in the world. Mean annual temperature is 12.1 °C and mean annual precipitation 35.3 mm. TZZ is at the Taklimakan Desert Research Station, CAS, in the center of the Taklimakan desert with a mean annual temperature of 12.4 °C and mean annual precipitation of 36.6 mm. The Taklimakan desert is in the arid center of Eurasia and covers a total area of 337 600 km^2. YPH is located at an experimental farm of Xinjiang Academy of Agricultural Sciences and is a typical farmland monitoring site. Additional characteristics of the sites are shown in Table 1 and Fig. 1.

2.2 Sampling procedure and sample analysis

NH_3 and NO_2 samples were collected using Radiello® passive samplers (Aquaria Italy, Trident Equipments Pvt. Ltd., Mumbai, India). At each site three NH_3 and three NO_2 samplers were exposed during each measurement period inside a PVC shelter (2 m above the ground) to protect the samplers from precipitation and direct sunlight. NH_3 and NO_2 concentrations were measured monthly by exposing the samplers for two weeks in the middle of the month. After sampling, absorption cartridges of the passive

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samplers were placed in airtight plastic tubes and stored in a refrigerator at 4 °C until analysis with an automated segmented flow continuous flow analyzer (Seal AA3, Norderstedt, Germany) within two months. Detailed information about the passive samplers has been provided by Shen et al. (2009). Airborne PM_{10} particles (particulate matter whose aerodynamic equivalent diameter is < 10 µm) were sampled using a particulate sampler (BGI, Omni, Waltham, MA) with a flow rate of 5 l min^{-1} and 7–10 daily samples of PM_{10} were collected at each site during each month. The sampler was placed about 2 m above the ground and ran for 24 h to obtain a particulate matter sample on 47 mm quartz filters (Whatman, Maidstone, UK). Before and after sampling, each filter was conditioned for at least 24 h inside a chamber at a relative humidity of 50 % and a temperature of 25 °C, and then weighed (Sartorius, Göttingen, Germany; precision 1 µg). PM_{10} mass concentrations were determined from the mass difference and the sampled air volume. Each sampling filter was extracted with 10 ml deionized water by ultrasonication for 30 min and the extract solution was filtered through a syringe filter (0.45 mm, Tengda Inc., Tianjin, China) and stored in a refrigerator. Chemical analysis of PM_{10} was conducted within two months. Ammonium and nitrate in PM_{10} (pNH_4^+ and pNO_3^-) were measured by continuous flow analyzer (Seal AA3). Monthly mean air temperature, precipitation, wind speed and relative humidity from August 2009 to November 2011 at all sites except TFS and YPH are shown in Fig. 2.

2.3 Statistical analysis

Values of NH_3, NO_2, PM_{10}, pNH_4^+ and pNO_3^- concentrations each month at both sites are means ± standard errors (s.e.). Pearson correlation analysis at a significance of 95 % between N species and air temperature, precipitation, wind speed and relative humidity was calculated. All statistical analysis was performed using the SPSS 13.0 statistical package (SPSS Inc., Chicago, IL).

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3 Results

3.1 Spatial variation of Nr concentrations

Annual average NH_3 concentrations ranged from 1.72 to 14.19 $\mu g N m^{-3}$ at the ten sites, with an overall average value of 7.71 $\mu g N m^{-3}$. The NH_3 concentrations followed the following sequence: alpine grassland (BYB, 1.72 $\mu g N m^{-3}$) and desert (TZZ, 2.61 $\mu g N m^{-3}$) < desert in oasis (TLF, 5.61 $\mu g N m^{-3}$), desert-oasis ecotone (CLZ, 6.23 $\mu g N m^{-3}$), farmland near desert (FKZ, 6.29 $\mu g N m^{-3}$; AKS, 7.2 $\mu g N m^{-3}$) and urban (SDS, 7.92 $\mu g N m^{-3}$) < farmland near suburban (TFS, 11.44 $\mu g N m^{-3}$) and farmland (BTH, 13.9 $\mu g N m^{-3}$; YPH, 14.19 $\mu g N m^{-3}$). Annual average NO_2 concentrations at BYB, TZZ, CLZ, AKS, TLF, YPH, FKZ, BTH, TFS and SDS sites were 1.01, 3.74, 4.39, 6.69, 7.31, 7.32, 7.98, 10.55, 17.72 and 30.07 $\mu g N m^{-3}$, respectively, with an overall mean value of 9.68 $\mu g N m^{-3}$. The annual average PM_{10} concentrations at BYB, FKZ, SDS, BTH, TFS, AKS, TLF, YPH, TZZ and CLZ sites were 22.5, 80.9, 166.3, 189.7, 211.3, 229.2, 237.0, 265.4, 498.0 and 591.8 $\mu g m^{-3}$, respectively, and overall averaged 249.23 $\mu g m^{-3}$. The annual mean pNH_4^+ concentrations at the ten sites ranged from 0.08 to 6.6 $\mu g N m^{-3}$ (1.81 $\mu g N m^{-3}$ on average); and the annual average pNO_3^- concentrations ranged from 0.22 to 2.58 $\mu g N m^{-3}$ (1.13 $\mu g N m^{-3}$ on average). Total concentrations of reactive nitrogen species were, respectively, 3.09, 6.81, 11.9, 14.34, 15.28, 19.08, 22.8, 26.21, 36.72 and 46.99 $\mu g N m^{-3}$ at BYB, TZZ, CLZ, AKS, TLF, YPH, BTH, FKZ, TFS and SDS sites (Fig. 5).

3.2 Seasonal variation in Nr concentration

NH_3 exhibited distinct and significant temporal variation, with lowest concentrations in winter than in spring, summer or autumn (Fig. 3a). Monthly mean NH_3 concentrations varied within the range 0.58 to 21.67 $\mu g N m^{-3}$ across all sites. Monthly mean concentrations measured for NO_2 are given in Fig. 3c, and the values ranged from 0.29 to 57.25 $\mu g N m^{-3}$ across all sites. In contrast to NH_3, high monthly mean NO_2

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concentrations were found in winter (except site BYB). Monthly mean PM_{10} concentrations ranged from 3.06 to 876.2 $\mu g\,m^{-3}$, and PM_{10} concentrations reached their maximum values in April at TZZ (876.2 μg^{-3}) and CLZ (852.7 $\mu g\,m^{-3}$) (Fig. 4). PM_{10} concentrations were higher in winter and spring than that during summer and autumn (except site BYB). Monthly mean NH_4^+ concentrations in PM_{10} (pNH_4^+) ranged from 0.02 to 25.79 $\mu g\,N\,m^{-3}$, and pNH_4^+ concentrations peaked in winter with lower values in summer across almost all sites (Fig. 3b). Monthly mean particulate concentrations of NO_3^- in PM_{10} (pNO_3^-) ranged from 0.02 to 11.08 $\mu g\,N\,m^{-3}$, and were higher in winter than that in spring, summer or autumn (Fig. 3d).

3.3 Effect of environmental factors on atmospheric Nr concentrations

NH_3 concentration showed significant positive correlations with air temperature and wind speed and negative correlations with relative humidity (Fig. 6f–h). No significant correlations were found between NO_2 concentration and environmental factors. PM_{10} concentrations showed significant negative correlations with relative humidity (Fig. 6e). Both pNH_4^+ and pNO_3^- concentrations showed significant negative correlations with air temperature and positive correlations with relative humidity (Fig. 6a–d).

3.4 Mass concentrations of inorganic ions in PM_{10}

Mass concentrations of PM_{10} and particulate NH_4^+ and NO_3^- in PM_{10} at the ten sites are shown in Table 2. Mean concentrations of particulate NH_4^+ and NO_3^- in PM_{10} ranged from 0.18 to 8.23 and from 0.96 to 11.24 $\mu g\,m^{-3}$, and the concentrations of pNH_4^+ and pNO_3^- in PM_{10} accounted for 0.8–5.2 and 0.3–8.4 % of PM_{10} across all sites. Concentrations of total inorganic ions (pNH_4^+ and pNO_3^-) in PM_{10} accounted for 0.35–13.55 % of PM_{10} across the ten sampling sites.

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4 Discussion

4.1 Concentrations of Nr compounds in different ecosystems

The annual NH_3 concentration showed significant spatial variation from alpine grassland to farmland in the Xinjiang arid region of Northwest China. The highest NH_3 concentration found was in farmland (YPH) which would be affected by agricultural pollution sources. The lowest NH_3 concentration was found in alpine grassland (BYB) where the absence of fertilizer applications and the lowest air temperatures may have contributed to the lower NH_3 concentration. Although agriculture is the main source of NH_3, motor vehicles can be important sources of ammonia in urban environments (Ianniello et al., 2010). In the current study annual NO_2 concentrations also exhibited significant spatial variation. The highest NO_2 concentration was found at an urban site (SDS), which would be subject to anthropogenic pollution sources, and the lowest NO_2 concentration was found in alpine grassland which is relatively undisturbed and free from anthropogenic activities. The highest PM_{10} concentrations were found in the desert-oasis ecotone (CLZ) and desert (TZZ), and the values were much higher than reported from numerous other sites (Kumar and Joseph, 2006; Li et al., 2006; Namdeo and Bell, 2005). Higher annual pNH_4^+ concentration were found at SDS and TFS and were much higher than reported at many other urban and suburban sites (Hayashi et al., 2007), possibly due to anthropogenic influences. Higher annual pNO_3^- concentrations were also found at SDS and TFS. We assume that Bayinbuluk alpine grassland (BYB, 3.09 $\mu g\,N\,m^{-3}$) is representative of background conditions because it is remote from urban areas and relatively undisturbed by anthropogenic activities. Such high Nr concentrations (SDS, 46.99 $\mu g\,N\,m^{-3}$) reflect substantial air pollution due to increased emissions by traffic, industry and domestic heating, with consequent effects on the eutrophication of water bodies and soil acidification in neighboring natural and semi-natural ecosystems.

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4.2 Seasonal variation in concentrations of Nr compounds

The lowest seasonal NH_3 concentration was found in winter at almost all sites and this is consistent with results reported from the North China Plain (Shen et al., 2011). Higher NH_3 concentrations were found in summer and autumn in farmland, most likely due to high NH_3 emissions from N fertilizer applications, rapidly increasing livestock production, and motor vehicles. In contrast to other sites, summer NH_3 concentrations were low in alpine grassland. This is most likely due to the transfer of almost all livestock to higher mountain areas (over 3000 m a.s.l.) and lower air temperatures. In North and Northwest China NH_3 concentrations are relatively high in summer and autumn, mainly due to N fertilization and higher air temperatures, both of which promote NH_3 volatilization from arable soils. For example, the N fertilizer application rate can reach 240 kg N ha^{-1} per crop to achieve high yields in spring and summer at site TFS. However, less than 30 % of applied N fertilizer may be taken up by the crops (Zhang et al., 2008) and much of the unaccounted-for fertilizer N may have been lost by NH_3 emission. Source strength and removal efficiency can explain seasonal variation in NH_3 concentrations (Hong et al., 2002). Moreover, air temperature and wind speed determine the NH_3 concentrations. NO_2 concentrations exhibited distinct and significant seasonal variation, with higher concentrations in winter at almost all sites (except BYB) and reached their highest value at site SDS. These NO_2 values were very similar to the high concentrations observed in other regions in China (Aas et al., 2007; Shen et al., 2009). For example, Urumqi is only 140 km^2 in area and had a population of 2.2 million and a total number of motor vehicles of 140 000 units in 2003. However, motor vehicles had reached 240 000 units and were increasing at a rate of 260 units per day in 2009. Therefore the higher NO_2 concentrations are likely related to anthropogenic sources such as vehicle emissions at the urban site, and the use of coal for heating in North and Northwest China in winter may be the main factor responsible for the increased NO_x concentrations in the atmosphere. In addition, due to the paucity rainfall or snowfall in winter in these arid areas, scavenging of atmospheric NO_2 by wet

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deposition will be greatly limited. Higher PM_{10} concentrations in this study were found in spring and winter and were much higher than reported from numerous other sites (Kumar and Joseph, 2006; Li et al., 2006). We observed PM_{10} concentrations reaching maximum values due to dust storm events in spring, notably 852.7 and 876.2 $\mu g\,m^{-3}$ at sites CLZ and TZZ, and they were even higher than in Lanzhou, a city with some of the highest air pollution worldwide with a peak PM_{10} concentration (541.9 $\mu g\,m^{-3}$) occurring in April due to dust events (Wang et al., 2009). Higher PM_{10} concentrations occurred in winter due to domestic heating. Moreover, increasing numbers of vehicles and low precipitation and relative humidity in the arid regions resulted in considerable PM_{10} pollution. The lowest pNH_4^+ concentrations were found in summer at almost all sites while the highest pNO_3^- concentrations were found in winter, especially at sites SDS, TFS and FKZ where the values were much higher than reported for numerous other sites (Hayashi et al., 2007). This was most likely generated from coal combustion in winter and from other anthropogenic sources such as vehicles, manufacturing and industrial processes at other times of year. Meteorological parameters such as low temperature and high relative humidity point towards poor dilution of pollutants during the winter period. Regarding the presence of nitrate associated with ammonium, it was more likely to form secondary particulate ammonium nitrate during periods of low temperatures and relatively high humidity in the arid areas.

4.3 Ratios of concentrations of secondary particles to PM_{10}

Secondary particles (NH_4^+ and NO_3^-) have been regarded as the main contributor to fine particulate matter (PM) which is harmful to human health and reduces visibility (Erisman and Schaap, 2004; Pinder and Adams, 2007) and is also implicated in climate change due to its effects on direct and indirect radiative forcing. In North China the ratios of secondary inorganic nitrogen particles to PM_{10} were relatively high. For example, concentrations of secondary particles (NH_4^+ and NO_3^-) accounted for 14.6–22.3 % of the PM_{10} concentrations (Shen et al., 2011). Lower ratios were found in desert (TZZ, 0.35) and desert-oasis ecotone (CLZ, 0.63) sites, while urban (SDS, 11.7) and suburban

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(TFS, 8.3) areas had higher ratios. This indicates that anthropogenic activities have greatly increased the emissions of reactive nitrogen species and influenced local air pollution. Therefore, a decline in the concentration of secondary inorganic particles would be a worthwhile target to improve air quality in these arid regions.

5 Conclusions

Overall annual mean concentrations of NH_3, NO_2, pNH_4^+ and pNO_3^- were 7.71, 9.68, 1.81 and 1.13 $\mu g\,N\,m^{-3}$, and the annual average PM_{10} concentration was 249.2 $\mu g\,m^{-3}$ in the Xinjiang arid region of Northwest China. The order of total concentrations of Nr species were, respectively, alpine grassland < desert, desert-oasis ecotone < desert in an oasis < farmland < suburban and urban ecosystems. NO_2 concentrations exhibited distinct and significant seasonal variation with higher concentrations in winter. Lower pNH_4^+ concentrations were found in summer at almost all sites while the highest pNO_3^- concentrations were found in winter. Lower ratios of secondary particles (NH_4^+ and NO_3^-) were found in desert and the desert-oasis ecotone, while urban and suburban sites had higher ratios, indicating that anthropogenic activities have greatly influenced local air quality and pollution and require the introduction of effective control measures.

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Table 1. Characteristics of the ten monitoring sites in the Xinjiang arid region of Northwest China.

Site No.	Site Name	Land Use Type	Latitude (N)	Longitude (E)	Altitude (m)	Sampling Period
1	AKS (Akesu)	Famland	40°37.107′	80°49.722′	1019	Apr 2011–Nov 2011
2	BTH (Baotouhu)	Famland	41°40.620′	85°51.650′	893	Apr 2011–Nov 2011
3	BYB (Bayinbuluk)	Alpine grassland	42°53.079′	83°42.528′	2470	Sep 2009–Sep 2011
4	CLZ (Cele)	Desert-oasis ecotone	37°00.938′	80°43.749′	1363	Apr 2011–Nov 2011
5	FKZ (Fukang)	Famland near a desert	44°17.574′	87°56.025′	455	Aug 2009–Sep 2011
6	SDS (Shengdisuo)	Urban	43°51.182′	87°33.747′	775	Aug 2009–Oct 2011
7	TFS (Tufeisuo)	Suburban	43°56.482′	87°28.240′	576	Aug 2009–Oct 2011
8	TLF (Tulufan)	Desert in an oasis	42°51.245′	89°11.424′	−106	Aug 2009–Sep 2011
9	TZZ (Tazhong)	Desert	38°58.317′	83°39.548′	1095	Apr 2011–Nov 2011
10	YPH (Yuepuhu)	Famland	39°00.187′	77°16.057′	1175	Apr 2011–Nov 2011

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Table 2. Concentrations of PM_{10} and secondary particles (NH_4^+ and NO_3^-) in PM_{10} and ratios of concentrations of secondary particles to PM_{10}.

Site	NH_4^+	NO_3^-	PM_{10}	Ratio
BYB	0.18	0.96	22.48	5.09
FKZ	4.17	6.80	80.93	13.55
TLF	1.51	5.18	237.02	2.82
SDS	8.23	11.24	166.33	11.70
TFS	6.38	11.21	211.26	8.33
CLZ	0.76	2.99	591.80	0.63
TZZ	0.11	1.65	498.02	0.35
AKS	0.14	1.43	229.25	0.69
BTH	0.95	4.43	189.73	2.83
YPH	0.76	3.06	265.45	1.44

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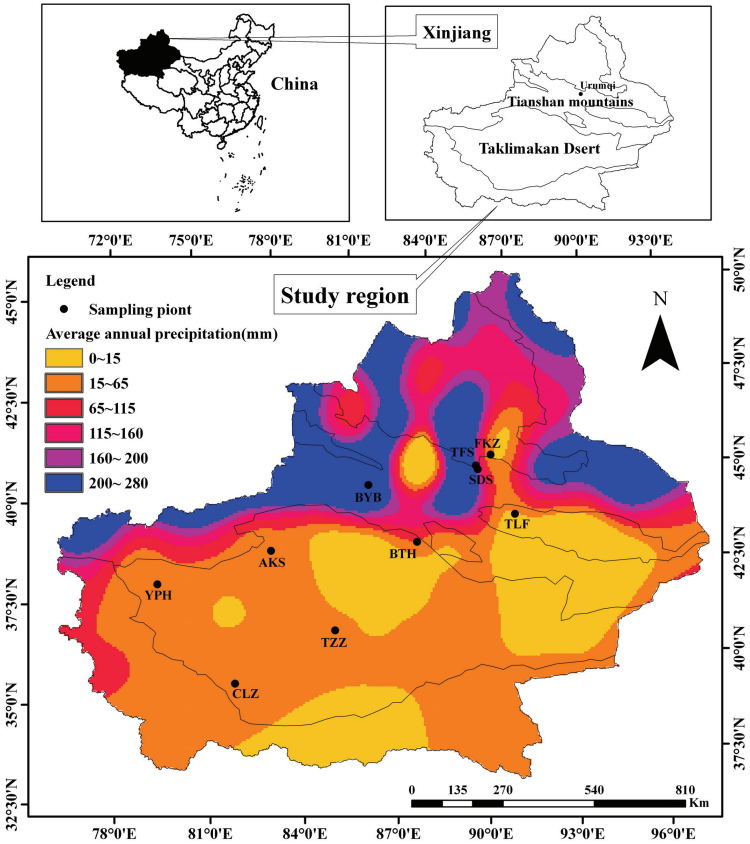

Fig. 1. Distribution of the ten monitoring sites in the Xinjiang arid region of Northwest China.

BGD
9, 6627–6649, 2012

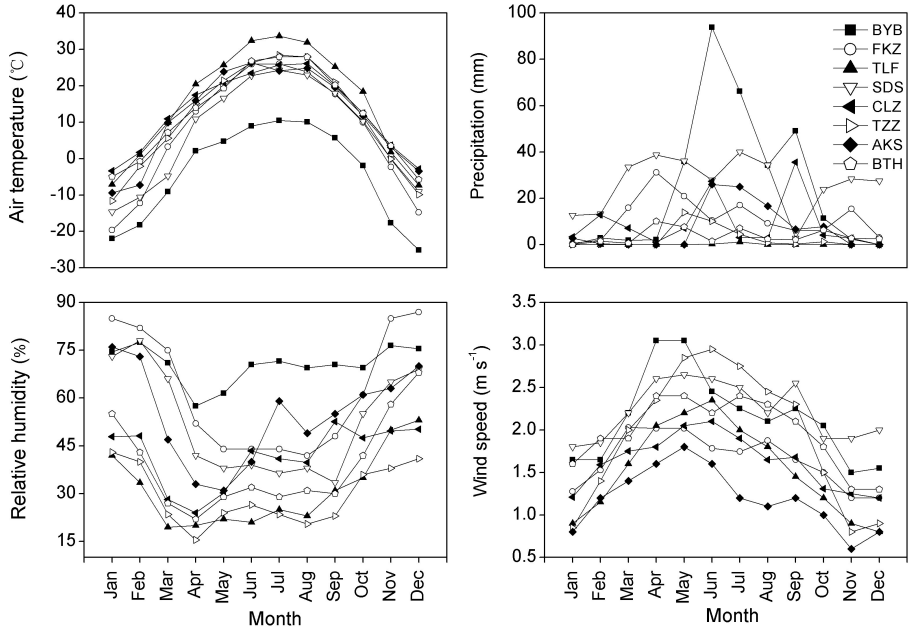

Fig. 2. Air temperature, precipitation, wind speed and relative humidity from August 2009 to November 2011 at the monitoring sites.

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9, 6627–6649, 2012

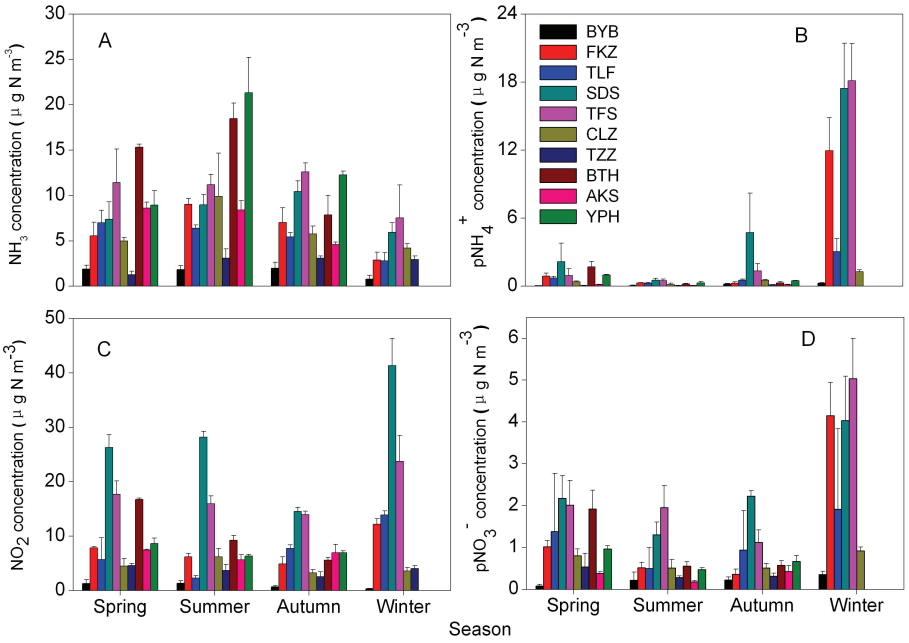

Fig. 3. Seasonal concentrations of **(A)** NH_3, **(B)** pNH_4^+, **(C)** NO_2 and **(D)** pNO_3^- in PM_{10} at the ten sites.

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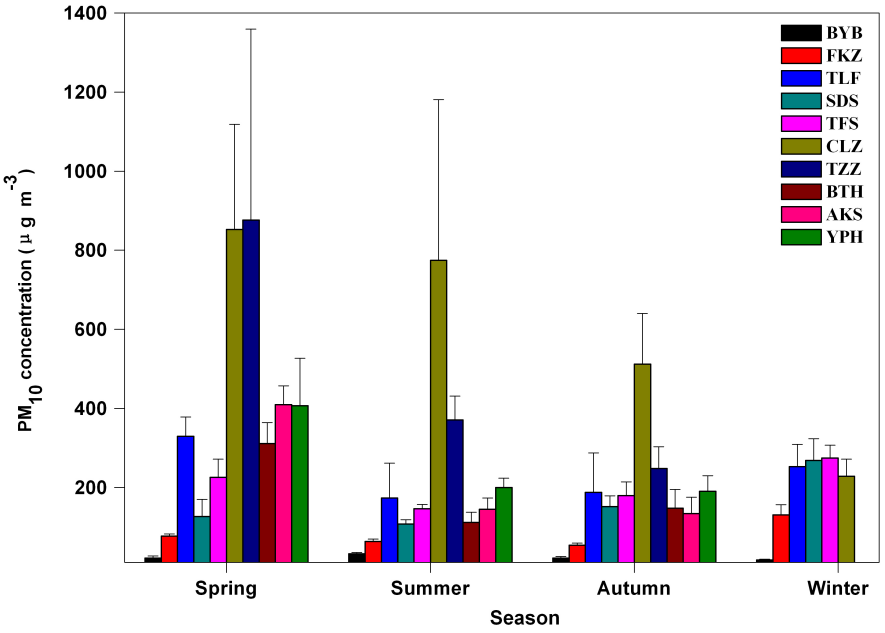

Fig. 4. Seasonal concentrations of PM_{10} at the ten sites.

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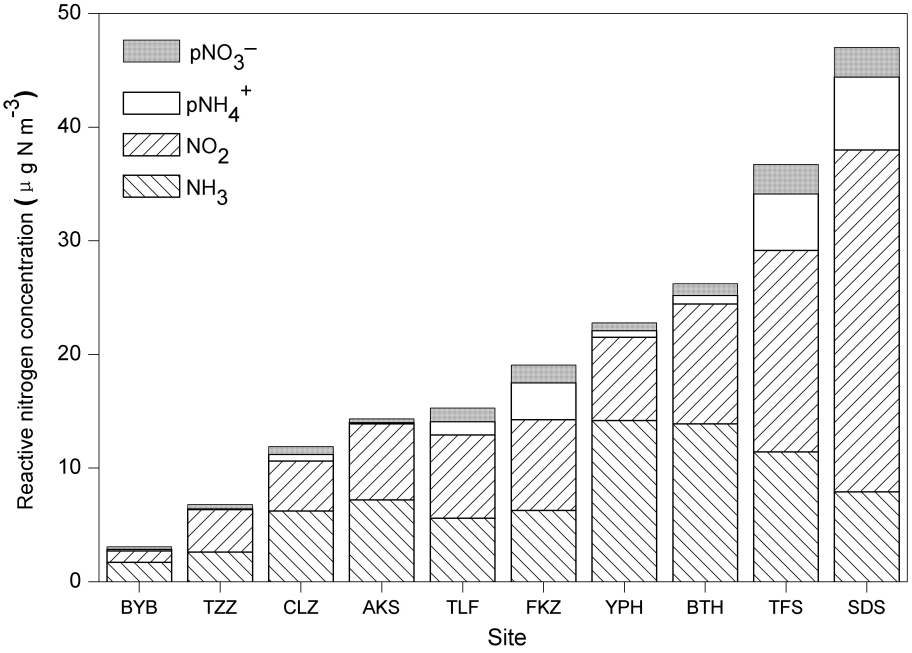

Fig. 5. Total concentrations of reactive nitrogen species at the ten sites.

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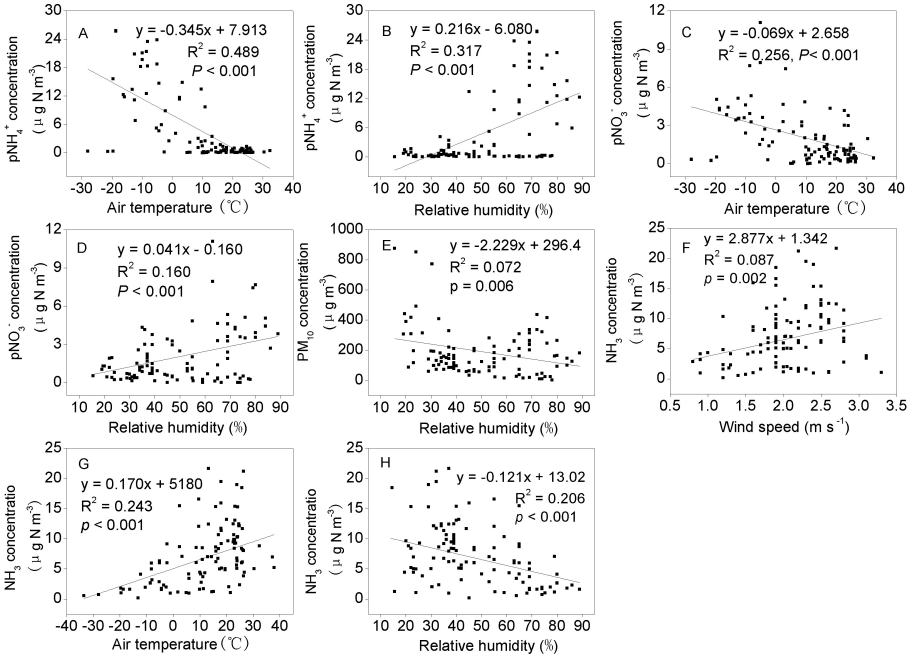

Fig. 6. Relationships between pNH_4^+, pNO_3^-, PM_{10} and NH_3 concentrations and air temperature, relative humidity, precipitation and wind speed.

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