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Supplement of

Quantifying atmospheric nitrogen deposition through a nationwide monitoring network across China

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Sect. S1. Social-economical and geo-climatic conditions for six regions of China

In this study, we divided the research area into six typical regions: North China (NC), Northeast (NE), Northwest (NW), Southeast China (SE), Southwest China (SW) and Tibetan Plateau (TP). These regions reflect different social-economical and geo-climatic conditions across China (Liu et al., 2013). Basically, NC, SE and SW represent relatively developed regions, NE and NW represent less developed regions while TP represents the least developed region in China.

Statistical data of the gross domestic product (GDP) for the six regions are sourced from the China Statistic Yearbook for the year 2013 (http://www.stats.gov.cn/tjsj/ndsj/2014/indexch.htm).

North China, an intensively managed agricultural region and economically developed area, comprises of Beijing, Tianjin, Hebei, Henan, Shandong, Shanxi and Shaanxi provinces, which is characterized by temperate monsoon climate. The region occupies 10% of the total area of China, and contributes 28% of the total national China GDP. Northeast China includes Liaoning, Jilin and Heilongjiang provinces and the northeast area of Inner Mongolia, and is characterized by a temperate monsoon climate. The region accounts for 6% of the China's total area, and contributes 9% of the total national GDP. Northwest China consists of the northwest region of Inner Mongolia, Xinjiang, Ningxia and Gansu provinces, characterizing by a temperate continental climate. The region occupies 35% of the total area of China, but only contributes 5% of China's GDP. Southeast China covers Shanghai, Jiangsu, Zhejiang, Anhui, Hubei, Hunan, Jiangxi, Fujian, Guangdong, Hong Kong, Macau, Taiwan and Hainan provinces: 13% of China's total area. The region contributes 46% of China's GDP and is characterized by a subtropical monsoon climate. Southwest China consists of Sichuan, Chongqing, Guizhou, Yunnan and Guangxi provinces, and is characterized subtropical and plateau monsoon climates. The region accounts for 14% of China's total area and contributes 12% of China's GDP. The Tibetan Plateau is a remote area of China that consists of Tibet and Qinghai provinces. The region is characterized by an alpine mountain climate. The region occupies 20% of the total area of China but only contributes 0.5% of China's GDP.

Section S2. Other information of the sampling sites

S2.1. Sampling sites in north China (NC)

Thirteen sampling sites, including 3 urban sites (NC1, NC2 and NC3), 8 rural sites (NC4, NC5, NC6, NC7, NC8, NC9, NC10, NC11) and 2background sites (NC12 and NC13), are located in north China.

NC1, located at the west campus of China Agricultural University (CAU, 40.02ºN, 116.28ºE), is near the fifth ring road in Beijing city. NC2, located at the campus of Beijing Normal University (39.96ºN, 116.37ºE), is near the third ring road in Beijing city. NC3 is located in the centre of Zhengzhou city, the capital of Henan province and Henan Academy of Agricultural Sciences (34.75ºN, 113.63ºE). These three urban sites (i.e. NC1, NC2, and NC3) are surrounded by complicated external environments, such as densely occupied residences, industry, small-scale urban agriculture, and roads. NC4is located at Dongbeiwang village (40.04ºN, 116.29ºE), Xibeiwang town, Haidian District, about 20 km the northwest of Beijing city. This site could be polluted by ammonia from agricultural fields and domestic animal farms and emissions from motor vehicles on an adjacent road (c. 0.2 km). NC5 is located at Shangzhuang Agricultural Experimental Station (40.11ºN, 116.20ºE) of CAU, in Shangzhuang town, about 33 km northwest of Beijing city. This site is surrounded by small villages and croplands. NC6 is located at the west campus of Hebei Agricultural University (38.85ºN, 115.48ºE) in Baoding city, Hebei province, about 8 km southwest of the downtown area. This site is surrounded by small villages and croplands, and is close to a main road (c. 0.5 km). NC7 is located at CAU's Quzhou experimental station (36.78ºN, 114.94ºE) in Quzhou town, Hebei province. NC7 is a typical rural agriculture dominated site with a recently constructed industrial district. This site is surrounded by croplands and small villages. A north-south main road is located 0.5 km west of the sampling site. NC8 is located at Yangqu dry farming water-saving demonstration base (38.05ºN, 112.89ºE), Shanxi Academy of Agricultural Sciences, in Hecun village, about 30 km northeast of Taiyuan city, Shanxi province. Hecun is a typical rural village with dryland farming. The sampling site is mainly polluted by ammonia from agricultural fields. NC9 is located at Zhumadian Agricultural Experiment Station (33.02ºN, 114.05ºE), Zhumadian Academy of Agricultural Sciences in Yicheng district of Zhumadian city, Henan province. This site is surrounded by croplands, small villages and small-scale livestock farming. NC10 is located at Wuquan Agricultural Experimental Base (34.31ºN, 108.01ºE), Northwest

Agriculture & Forestry UniversityinYangling city, Shaanxi province. This site is about 8 km northwest of the downtown area, surrounding by croplands and small villages. NC11 is located at Yucheng Experimental Station (36.94ºN, 116.63ºE), Chinese Academy of Sciences, about 8 km southwest of Yucheng city, Shandong province. This site is surrounded by small villages and croplands. At all the above farmland sites, with the exception of NC8, the dominant cropping system is a winter wheat-summer maize rotation (two crops a year). Farmers usually apply nitrogen (N) fertilizers (mainly as urea and ammonium bicarbonate) in March-April, June-August and October to achieve high yields of maize and wheat. Typical application rates of N fertilizer are 500-600 kg N ha^{-1} yr^{-1} at most sites except NC9, where the rate is 100-150 kg N ha^{-1} yr^{-1}. At NC8 site, the dominant cropping system is spring maize (one crop a year). In addition there is small-scale vegetable production (e.g. brussels sprouts, beans and zucchini) throughout the year. Usually, N-fertilizers (compound fertilizer, urea and ammonium bicarbonate) are applied in March-May, with annual application rates of 150-300 kg N ha^{-1}.

NC12 is located at Lingshandao (LSD) island (35.77ºN, 120.18ºE, 514 m a.s.l.) in the Yellow Sea. LSD is the biggest island in north China, about 10 km southeast of Jiaonan city, Shandong province. NC13 is located at Changdao island (37.93ºN, 120.75ºE, 203 m a.s.l.), which is at the join of the Bohai Sea and the Yellow Sea, about 7 km north of Penglai city, Shandong province. There are almost no agricultural and/or industrial activities near these two coastal sampling sites.

S2.2. Sampling sites in northeast China (NE)

Five sampling sites, including 1 urban site (NE1), 2 rural sites (NE2 and NE3), and 2 background sites (NE4, andNE5) are located in northeast China.

NE1 is located at Dalian Jiaotong University (38.92 ºN, 121.58ºE) which lies in the Shahekou district of Dalian city, Liaoning province. Dalian is a sea town (near the Bohai Sea) and is famous for business development. The sampling site is polluted by emissions from industry and transportation. NE2 is located at an experimental station ofthe Soil and Fertilizer Institute (43.53 º N, 124.83ºE), Jilin Academy of Agricultural Sciences, in Gongzhuling city, Jilin province. Arable land accounts for 81% of the total land area in Gongzhuling city, and the other 19% is used for residential, commercial buildings and roads. This site is surrounded by farmland and residences. NE3 is located in Sikeshu town (43.36 ºN, 124.17ºE), Lishu county. This site is about 21 km northwest of Siping city, Jilin province. At NE2 and NE3, the sampling sites

are both surrounded by agricultural fields with a spring maize (single harvest per year) cropping system. Compound fertilizers are the major N source for the crops, and these are usually applied once in April-May, with an annual application rate of 400 kg ha^{-1} yr^{-1} at NE1 and 250 kg $ha^{-1}yr^{-1}$at NE2. NE5 is located in a forested area in Wuying district (48.11 ºN, 129.25 ºE) of Yichun city, Heilongjiang province, about 50 km northeast of downtown. Forest coverage reaches 93% in Wuying district. NE6 (50.78ºN, 121.52ºE) is located in a forested region of Genhe city, Inner Mongolia, where forest cover reaches 92%. Genhe is at the north border of the Greater Hinggan Mountains, and its climate is cold temperate humid monsoon together with the characteristics of continental monsoon, which is similar to that of northeast China, i.e. temperate humid and semi-humid continental monsoon. Given this, NE5 is placed in northeast China in the present study. The forests in Wuyin and Genhe are both natural ecosystems and there are no agricultural and industrial activities near the sampling sites.

S2.3. Sampling sites in northwest China (NW)

Five sampling sites, including 2 urban sites (NW1 and NW2), 1 rural sites (NW3) and 2 background sites (NW4 and NW5), are located in northwest China.

NW1 is located at Xinjiang Institute of Ecology and Geography (43.87ºN, 87.57ºE), Chinese Academy of Science, in the urban region of Urumqi city, Xinjiang province. This site may be polluted by emissions from motor vehicles and home heating in winter. NW2 is located at Xinjiang Academy of Agricultural Sciences, about 11 km away from NW1. This site has similar pollution sources as NW1.

NW3is located in Yongchang town (38.07ºN, 102.60 ºE), Wuwei city, Gansu province. The sampling site is surrounded by agricultural fields, small scale livestock housing and small villages. Maize is the major crop grown. Usually cropland receives chemical N fertilizers (urea, ammonium bicarbonate and compound fertilizers) in April, June and July, with an annual application rate of 350 kg N ha^{-1} yr^{-1}. NW5 is located at the meteorological observation site belonging to the Chinese Academy of Sciences, which is situated in temperate grassland in Duolun county (42.20ºN, 116.49ºE), Inner Mongolia. NW6is located at Bayinbuluke Grassland Ecosystem Research Station (42.88 ºN, 83.71ºE), Chinese Academy of Sciences, in the southern Tian Shan Mountains (3000-4500 m a.s.l.) of central Asia. Those two grassland sites (i.e. NW4 and NW5) are very unlikely to be subjected to influences from anthropogenic activities.

S2.4. Sampling sites in southeast China (SE)

Eleven sampling sites, including 2 urban sites (SE1 and SE2), 6 rural sites (SE3, SE4, SE5, SE6, SE7 and SE8), and 3 background sites (SE9, SE10 and SE11), are located in southeast China.

SE1 is located in the southern urban area of Nanjing city (31.84ºN, 118.85ºE). The sampling site is surrounded by complicated external environments, such as housing, industry, small-scale urban agriculture, and is close to a highway (c. 0.5 km). SE2is located at Baiyun Experimental Station (23.20ºN, 113.31ºE), Guangdong Academy of Agricultural Sciences, in Guangzhou city, Guangdong province. This site is surrounded by densely occupied housing and traffic roads. SE3 is located at Wuxue Modern Agricultural Demonstration Base (30.01ºN, 115.79ºE) in Huaqiao town, about 28 km northeast of Wuxue city, Hubei province. The sampling site is surrounded by residential land and paddy fields, where the cropping system is double rice harvests per year. Compound fertilizer and urea are the major N nutrient sources for rice, and these are usually applied in May-July, at a rate of about 180 kg ha^{-1} yr^{-1}. SE4 is located in Santangjie town (28.61ºN, 111.97ºE), which is a remote rural site in Taojiang county, Hunan province. The sampling site is surrounded by mountains and paddy fields. The paddy fields are cultivated with a double rice rotation system, which receives compound fertilizer and urea twice per year (in May, June or July) at about 150 kg N ha^{-1} yr^{-1}. SE5is located at Anhui Tobacco Institute (32.88ºN, 117.56ºE), Anhui Academy of Agricultural Sciences in Fengyang county, Anhui province. The sampling site is surrounded by croplands and residences. In the croplands, the major annual cropping system is a winter wheat-summer maize rotation (two crops per year). Compound fertilizer and urea are the major N nutrient sources for the croplands, and these are usually applied in April, June and October, at a rate of 380-500 kg N ha^{-1} yr^{-1}. SE6 is located in Leizhou peninsula, Zhanjiang city (21.26ºN, 110.33ºE), Guangdong province. This area grows tropical fruit and farmers use urea at a rate of 400 kg N ha^{-1} yr^{-1} for fruit production. SE7 is located on a mountain in the remote rural region of Fuzhou city (26.17ºN, 119.36ºE), Fujian province. This site is surrounded by some agricultural land growing tobacco and paddy rice. Usually, about 200 kg N ha^{-1} yr^{-1} compound fertilizers are used as the major nitrogen source for the crops, applied in January-March, and July and August. SE8 is located in Chunhu town (29.61ºN 121.53ºE), Fenghua city, Zhejiang province. The sampling site is surrounded by a residential area, a very small amount of agricultural land and mountains. SE9, located

in Huinong village (28.52ºN, 113.41ºE), Hunan province, is a rural background site. In a 2-km radius around the sampling site, there is forest and a reservoir, which contribute little to anthropogenic NH_3 and NO_x emissions. SE10, a rural background site is located in Feiyue village (28.56ºN, 113.34ºE), Hunan province. There were no typical emission sources around the site in a 2 km radius except forest and a residential area. SE11 is located at the centre of a 3 km^2 forest in Xishan village (28.61ºN, 113.31ºE), Hunan province. The forest has an average elevation of 250 m and is composed of masson pine, fir and shrubs. For convenience of access (e.g., the accessibility to road and the availability of main power), N deposition monitoring was conducted in the valley between two small hills in the forest. The valley is 100 m wide and 2000 m long in the main wind direction. The sampling site was selected in the centre of the valley, without human activity and anthropogenic emission sources of NH_3 and NO_x. The nearest N_r emission sources are more than 2 km away from the monitoring site, and consist of some small paddy fields distributed at the edge of the forest, accounting for no more than 5% of the total forest area.

S2.5. Sampling sites in southwest China (SW)

Seven sampling sites, including 1 urban site (SW1), 5 rural sites (SW2, SW3, SW4, SW5 and SW6) and 1 background site (SW7), are located in southwest China.

SW1is located in a residential area (30.55ºN, 103.84ºE) which lies in the Wenjiang district of Chengdu city, Sichuan province. The sampling site is surrounded densely occupied housing, roads and small-scale urban agriculture. SW2 is located at Xiangshui village (30.13ºN, 104.63ºE), Ziyang city, Sichuan province. This site is surrounded by paddy fields with a single rice cropping system (single harvest per year) and mountains. N fertilizers (urea and compound fertilizer) are applied mainly in June and July, at a rate of 270 kg N ha^{-1} yr^{-1}. SW3 is located at the Purple Soil Ecological Experiment Station (31.28ºN, 105.47ºE), Chinese Academy of Sciences, in Yanting county, Sichuan province. Agriculture is the dominant source of N at this site. In the agricultural fields, rice-wheat and maize-oilseed rape (canola) are the major double cropping systems for the paddy and upland fields in conventional farming practice, respectively. Ammonium bicarbonate is the major N nutrient sources for the croplands, usually applied at about 300 kg N ha^{-1} yr^{-1} for summer and winter crops together. SW4is located in Huangzhuang village (29.06ºN, 106.18ºE) in Jiangjin district of Chongqing, a city characterized by heavy industry production. The sampling site is surrounded by paddy fields and dry farmland with sorghum or oilseed rape. Farmers

apply N fertilizers mainly in April-July, October and November. Paddy fields at this site usually receive compound fertilizers including 180 kg N ha^{-1} yr^{-1}, whereas the dryland areas planted with sorghum usually receive ammonium phosphate and ammonium bicarbonate (altogether adding about 200 kg N ha^{-1} yr^{-1}), and the drylands used for oilseed rape production usually receive compound fertilizers at 135 kg N ha^{-1} yr^{-1}. SW5 is located in the suburbs of Kunming, a city characterized by flower and vegetable production, and about 500 m away from the Dian Lake (24.97ºN, 102.67ºE). This site is surrounded by agricultural fields and small villages, and is near a road (c. 0.2 km) with few vehicles. Leek is the most important cash crop in the region, and N fertilizer (urea) is usually applied every two months, at a rate of 300-400 kg N ha^{-1} yr^{-1}. SW6, located at the campus of Yunnan Agricultural University (25.13ºN, 102.75ºE), is in the northern rural area of Kunming city. This site is surrounded by housing, commercial buildings and roads without large agricultural sources. SW7 is a typical rural background site located in Kunyang town (24.67ºN, 102.61ºE), and is surrounded by small agricultural land (c. 62 ha), phosphate fertilizer plants and mountains.

S2.6. Sampling sites in the Tibetan plateau (TP)

Two sampling sites, including 1 urban site (TP1) and 1background site (TP2), are located in the Tibetan plateau.

TP1, located at a meteorological station belonging to Xining Weather Bureau, is in the northern suburbs of Xining city, Qinghai province. Surrounding this site are schools, housing and commercial buildings. TP2 is located at an ecological experimental station belonging to the Agricultural and Animal Husbandry College of Tibet University, which lies in the wide valley area of the middle and lower reaches of the Niyang River in Linzhi county, Tibet province. TP2 (Linzhi site) is relatively undisturbed remote site, surrounded by grassland free from human activity.

Sect. S3. Information on measuring methods, sample replications and collection

S3.1. Description of the DELTA system and the ALPHA and Gradko passive samplers

The DELTA system comprises a denuder filter sampling train, a low-volume pump (D210, TCP Micropumps Ltd., UK) and a high sensitivity dry gas meter (SK25, Kimmon Manufacturing Co., Ltd., Japan). Briefly, the sampling train consists of two potassium carbonate plus glycerol (1 % (m/v) K_2CO_3 + 1 % (m/v) glycerol in methanol) coated denuders in series for the simultaneous collection of HNO_3,

followed by two citric acid (5% (m/v) citric acid in methanol) coated denuders for NH_3 and finally by a filter-pack assembly with a first K_2CO_3/glycerol impregnated filter to capture particle phase anions (NO_3^-, SO_4^{2-}, Cl^-) and cations (NH_4^+, Na^+, Mg^{2+}, Ca^{2+}), and a second filter coated with citric acid to collect any volatilized particulate NH_4^+. The empirically determined effective size cut-off for particle sampling is of the order of 4.5 µm (E. Nemitz, personal communication). The air was drawn through the sampling train at a rate of 0.2-0.4 L min^{-1} and directly into the first denuder with no inlet line to avoid sampling losses. The total sampled air volume of the DELTA system was recorded by the gas meter which was checked every month for data reading, performance and maintenance.

The ALPHA passive sampler, described in detail by Puchalski et al. (2011) is made up of a 26 mm long, 27 mm outer diameter polyethylene tube with one open end. The open end contains a 5 µm PTFE membrane allowing gaseous NH_3 to diffuse through, which is then absorbed onto a citric acid-coated collection filter located at the other end of the diffusion path. The Gradko passive sampler consists of a 71.0 mm long × 11.0 mm internal diameter acrylic tube with coloured and white thermoplastic rubber caps.NO_2 is absorbed into a 20% triethanolamine/deionised-water solution coated onto two stainless steel wire meshes within the coloured cap. Both types of passive samplers at each site were installed at 2 m height above the ground and deployed as three replicates for monthly sampling. For the ALPHA passive sampler, the NH_3 concentration was calculated by considering a temperature dependent diffusion coefficient (Puchalski et al., 2011; Xu et al., 2014), while a constant gas diffusion coefficient based on an assumption of 25 ℃ was used for the calculation of NO_2 concentration, in accordance with the Gradko introduction manual and previous studies (Luo et al., 2013; Shen et al., 2013).

The DELTA sampling trains and all the passive samplers were prepared and measured in the laboratory at China Agricultural University (CAU), Beijing. The prepared samplers were sealed in individual airtight storage bags and shipped in cool boxes, sent out to the monitoring sites for monthly field exposure, then sent back in the same bags and boxes to the laboratory for extraction and analysis. All the samplers were exposed to ambient air for one month at each site and thus provided monthly mean ambient N_r concentrations. Laboratory and field (travel) blanks were prepared and transported together with the field-exposed samples. Three laboratory blanks were prepared for each batch of field exposed samples and represent extracts from samples

prepared in the laboratory. Three field blanks were prepared for each batch of field exposed samples and represent extracts from unexposed samplers that were transported to and from the monitoring sites with the field exposed samplers.

S3.2. Description of particulate sampler

For pNH_4^+ and pNO_3^- sampling, particulate samplers equipped with solar panels were used to collect 7-10 days of PM_{10} samples per month. The average of these 7-10 samples is assumed to represent the monthly average concentration. The sampler was placed about 2 m above the ground and ran for 24h to obtain a particulate matter sample on 47 mm quartz filters (Whatman, Maidstone, UK). Each filter was equilibrated for 24-h in a room at 40 % relative humidity and 25 °C and then weighed on an electronic balance (Satorius, GÖTTINGEN, Germany, precision: 10 μg) before and after sampling. After collection, the loaded quartz filters were sealed in aluminum foil and stored in on-site refrigerators at about 3 °C until analysis in the coordinating laboratories of local universities or institutes. Field blank measurements were made each month at the corresponding sites.

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Sect. S4. Validation of ALPHA passive sampler

The ALPHA passive samplers were used to monitor atmospheric concentrations of NH_3 at seven sites (NC2, NE4, NE5, SW5, SW7, TP1 and TP2) of the network at which the DELTA systems could not be used due to absence of an external power supply. To ensure that differences across sites are due to the pollution climate rather than an artifact caused by methodological choices, we performed simultaneous and continuous 1-month measurements of NH_3 concentrations by using ALPHA passive samplers and the active DELTA denuders at three urban (NC1, NC3 and NE1) and three rural (NC8, SE3 and SE7) sites from Nov. 2013 to Oct. 2014.

Monthly mean concentrations of NH_3 provided by ALPHA passive samplers and the DELTA denuders were reasonably comparable at all site (**Fig S1a-f**), with overall averaged values of 6.7 ± 5.0 (standard deviation) and 7.0 ± 5.1 μg N m^{-3}, respectively. In addition, the correlation between the two methodologies was highly significant (R^2=0.919, p<0.001). Thus any differences in the data can be ascribed to the pollution climate.

Sect. S5. Introduction to the calculation of V_d for all N_r species as well as the estimation of dry deposition

The model calculation of dry deposition of N_r species follows a standard big-leaf resistance-in-series model as described by Wesely (1989) for gases and Zhang et al. (2001) for aerosol. V_d is calculated as the function $V_d=(R_a+R_b+R_c)^{-1}$ determined by local meteorological condition and surface type as described in Zhang et al. (2012). Here R_a is the aerodynamic resistance to turbulent transfer from the lowest model layer (70 m above the surface) to the roughness height, R_b is the boundary layer resistance to molecular diffusion, and R_c is the canopy or surface uptake resistance. In the present study we have run the model calculation of dry deposition velocities for the whole of 2012 and archived the hourly values for both gases and aerosols over the model domain. Then, the monthly V_d at each site was averaged based on the hourly dataset for further estimation of dry deposition flux of each N_r species during the

observation, which was statistically summarized according to land use type and is presented in **Table S3**. Annual mean dry deposition velocities of N_r species for six land use types in this study, averaged from monthly mean values, were fit well into range of annual values calculated and used for similar land use types in other studies (**Table S4** of Supplement).

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Figure captions

Fig. S1. Comparison between simultaneous measurements of NH_3 concentrations by ALPHA passive samplers and the DELTA systems at six sampling sites in the network (a. NC1, b. NC3, c. NE1, d. NC8, e. SE3, f. SE7, g. ALPHA_NH_3 vs. DELTA_NH_3 across all sites). Site names for codes can be found in Table S1.

Fig. S2. Monthly mean atmospheric concentrations of N_r species (a. NH_3, b. NO_2, c. HNO_3, d. pNH_4^+ and e. pNO_3^-) at the forty-three monitoring sites in China. The boundary of the box indicates the 25^{th} and 75^{th} percentile. The black lines and red squares within the box mark the median and the mean, respectively. Whiskers above and below the box indicate the maximum and minimum values. Site names for codes can be found in Table S1.

Fig. S3. Monthly volume-weighted N_r concentrations in precipitation at the forty-three monitoring sites in China (a. NH_4^+-N, b. NO_3^--N). The black lines within the box denote the median. The blue circles denote the volume-weighted mean concentrations of N_r species in rainwater, calculating from the water amount and deposition fluxes of all precipitation events at each site during the sampling period. Whiskers above and below the box indicates the maximum and minimum values. Site names for codes can be found in Table S1.

Fig. S4. Relationships between monthly precipitation and volume-weighted concentrations of NH_4^+ and NO_3^- in precipitation at the sampling sites in the six regions. NC, NE, NW, SE, SW, and TP are the region codes and denote North China, Northeast China, Northwest China, Southeast China, Southwest China, and the Tibetan Plateau, respectively.

Fig. S5. Annual mean precipitation amounts at the forty-three sites in China. Site names for codes can be found in Table S1.

Fig. S6. Relationship between total annual dry N deposition fluxes and total annual atmospheric N_r concentrations across all forty-three sites. The plots with same color denote the same land use type.

Fig. S1

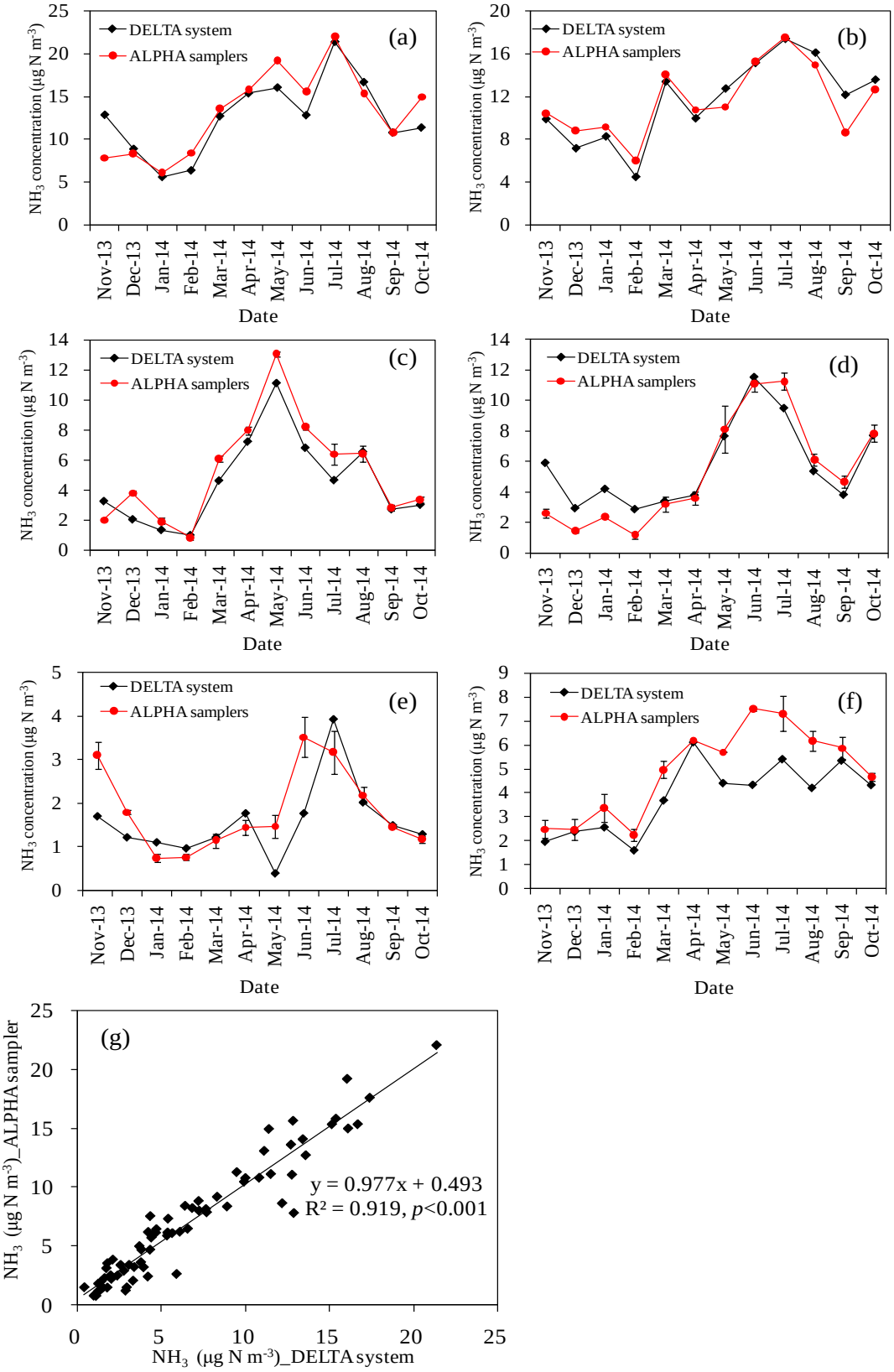

(a)
(b)
(c)
(d)
(e)
(f)
(g)
DELTA system
ALPHA samplers
NH3 concentration (µg N m-3)
Date
Nov-13
Dec-13
Jan-14
Feb-14
Mar-14
Apr-14
May-14
Jun-14
Jul-14
Aug-14
Sep-14
Oct-14
NH3 (µg N m-3)_ALPHA sampler
NH3 (µg N m-3)_DELTA system
y = 0.977x + 0.493
R² = 0.919, p<0.001

Fig. S2

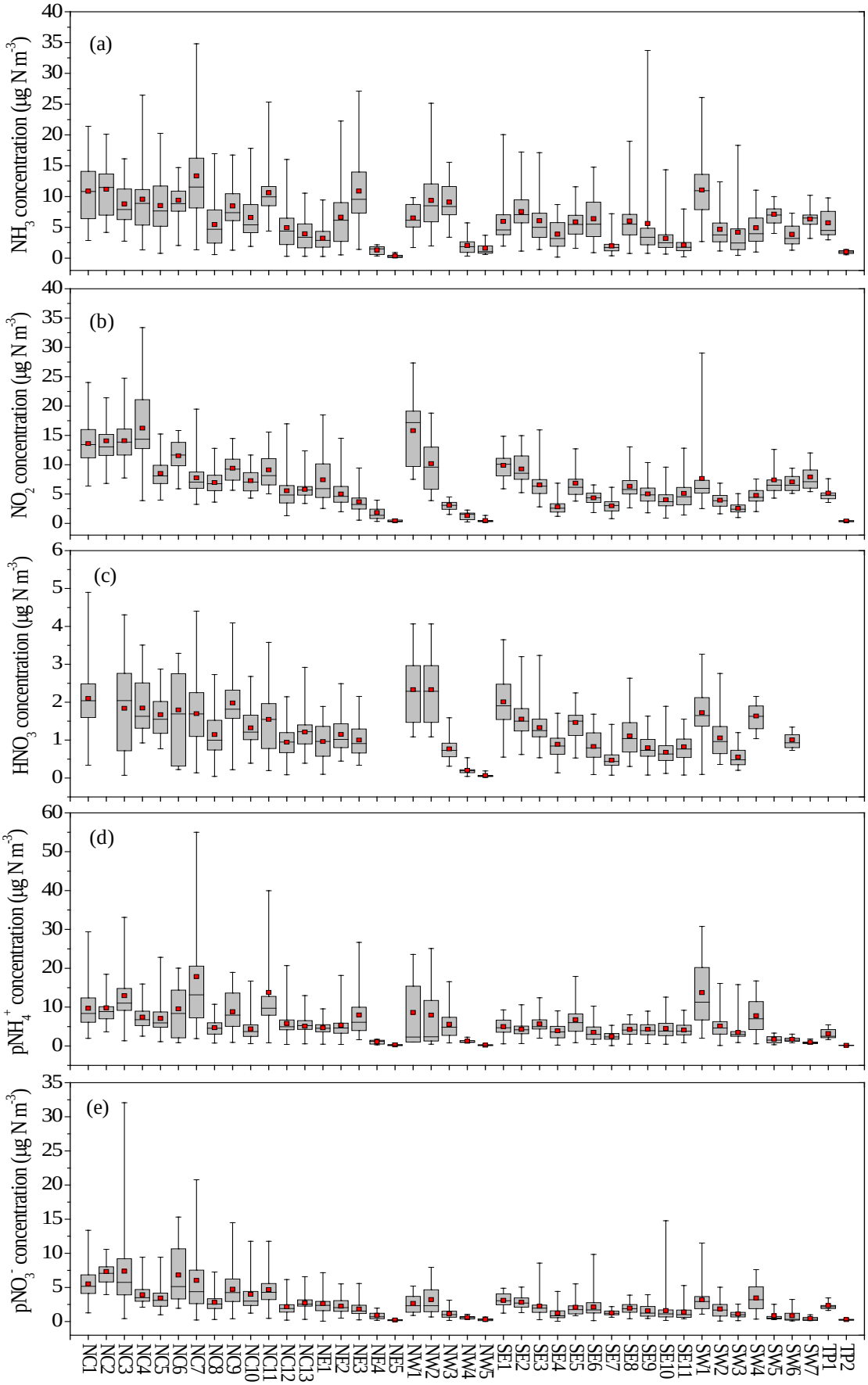
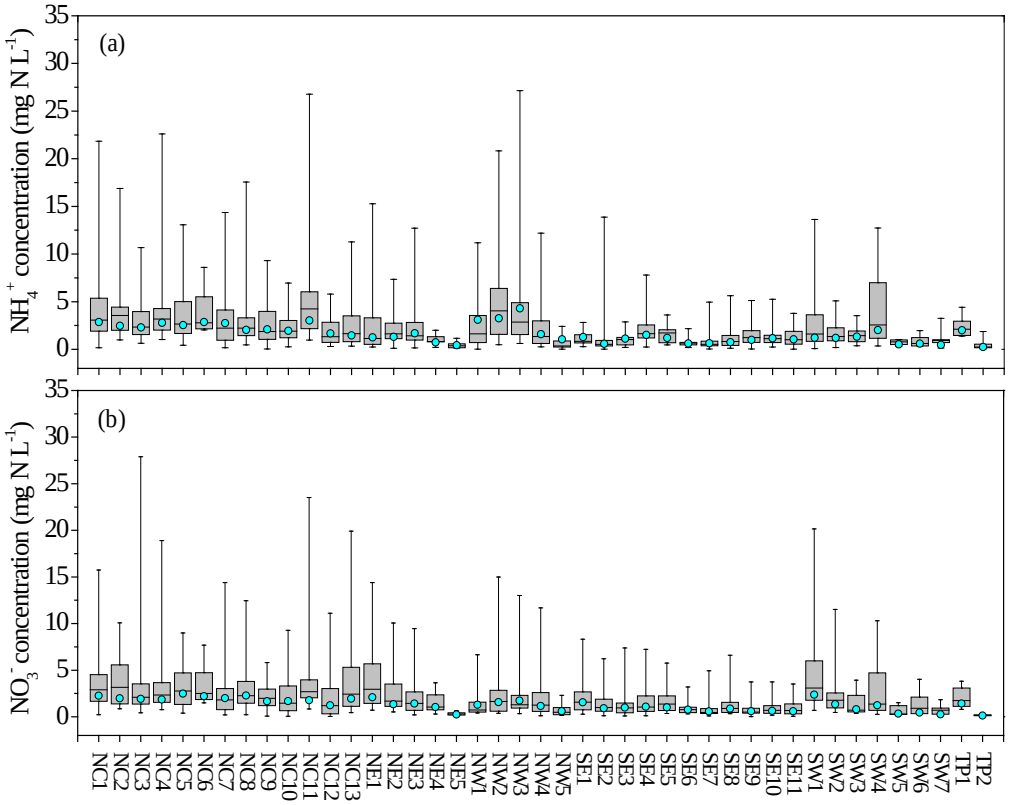
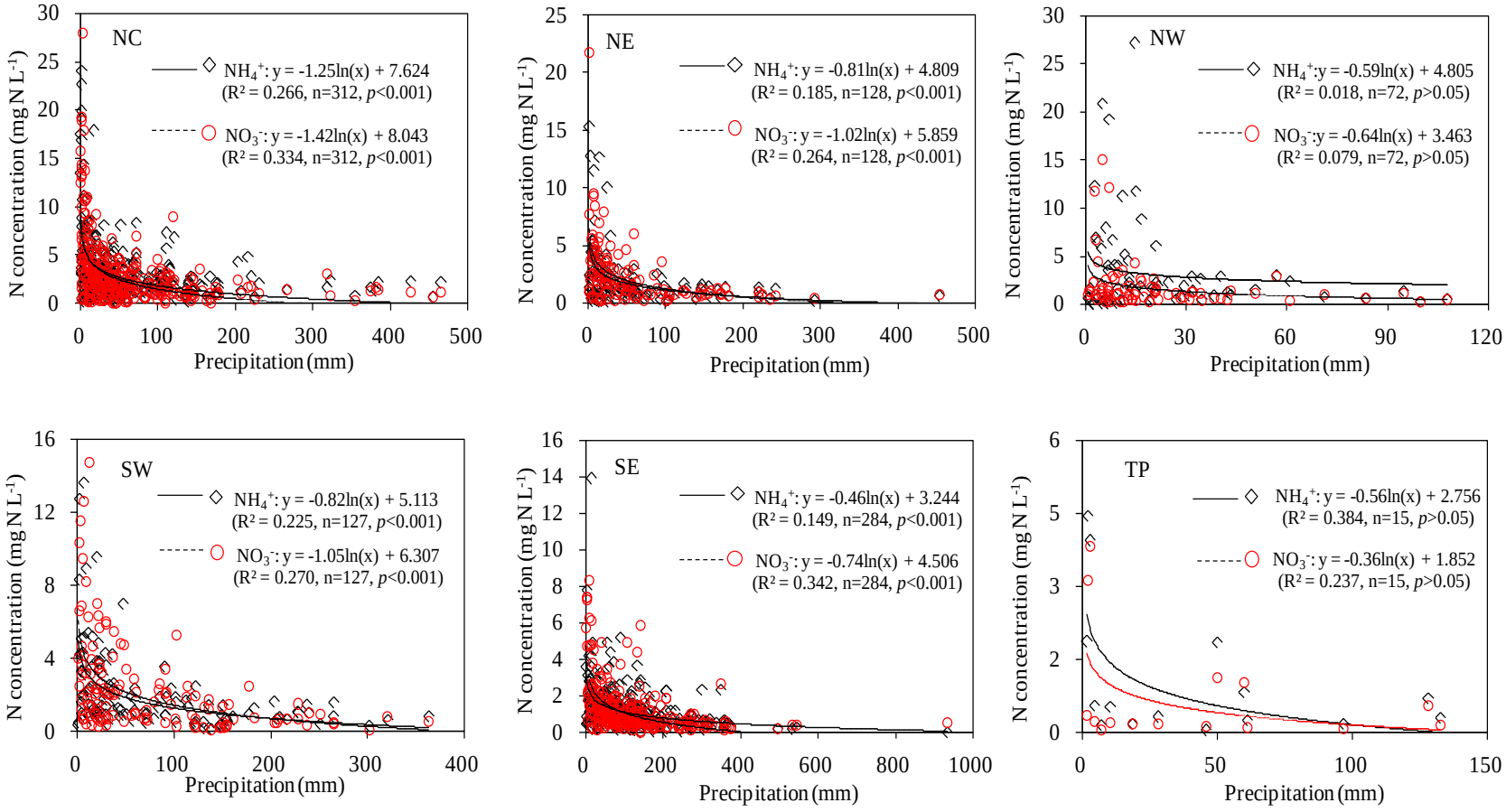

(a)
NH3 concentration (μg N m-3)
(b)
NO2 concentration (μg N m-3)
(c)
HNO3 concentration (μg N m-3)
(d)
pNH4+ concentration (μg N m-3)
(e)
pNO3- concentration (μg N m-3)
NC1 NC2 NC3 NC4 NC5 NC6 NC7 NC8 NC9 NC10 NC11 NC12 NC13 NE1 NE2 NE3 NE4 NE5 NW1 NW2 NW3 NW4 NW5 SE1 SE2 SE3 SE4 SE5 SE6 SE7 SE8 SE9 SE10 SE11 SW1 SW2 SW3 SW4 SW5 SW6 SW7 TP1 TP2

Fig. S3


(a)
NH_4^+ concentration (mg N L^{-1})
(b)
NO_3^- concentration (mg N L^{-1})
0
5
10
15
20
25
30
35
NC1
NC2
NC3
NC4
NC5
NC6
NC7
NC8
NC9
NC10
NC11
NC12
NC13
NE1
NE2
NE3
NE4
NE5
NW1
NW2
NW3
NW4
NW5
SE1
SE2
SE3
SE4
SE5
SE6
SE7
SE8
SE9
SE10
SE11
SW1
SW2
SW3
SW4
SW5
SW6
SW7
TP1
TP2

435

436 **Fig. S4**



437

Fig. S5

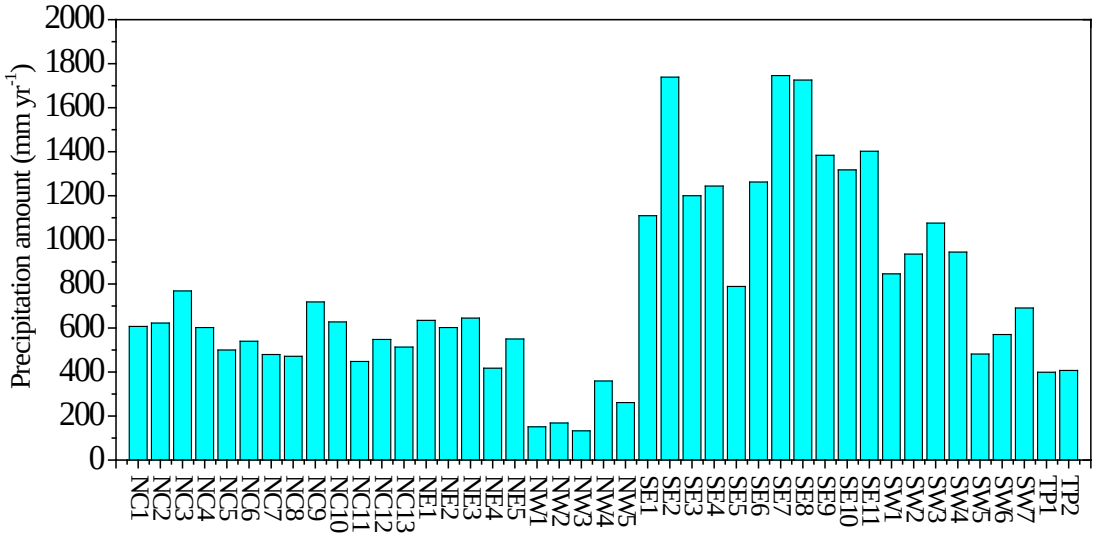
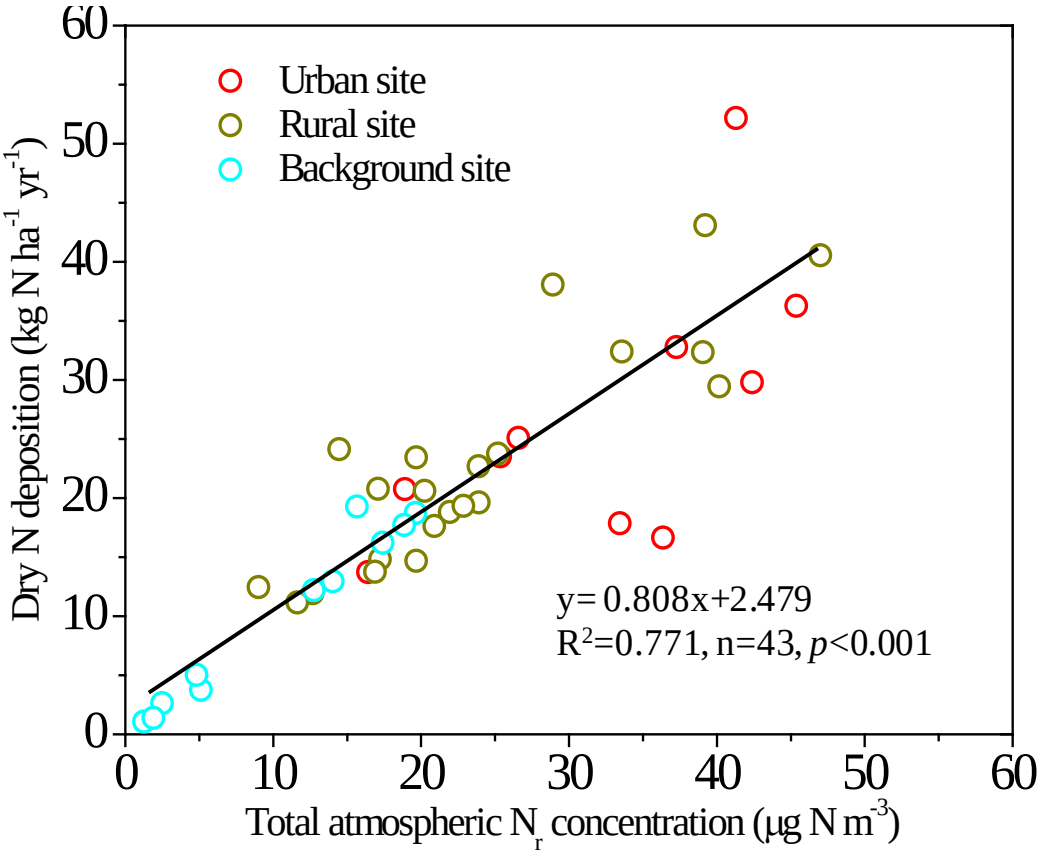

Precipitation amount (mm yr-1)
2000
1800
1600
1400
1200
1000
800
600
400
200
0
NC1
NC2
NC3
NC4
NC5
NC6
NC7
NC8
NC9
NC10
NC11
NC12
NC13
NE1
NE2
NE3
NE4
NE5
NW1
NW2
NW3
NW4
NW5
SE1
SE2
SE3
SE4
SE5
SE6
SE7
SE8
SE9
SE10
SE11
SW1
SW2
SW3
SW4
SW5
SW6
SW7
TP1
TP2

Fig. S6


Urban site
Rural site
Background site
y= 0.808x+2.479
R2=0.771, n=43, p<0.001
Dry N deposition (kg N ha-1 yr-1)
Total atmospheric Nr concentration (μg N m-3)
0
10
20
30
40
50
60

480 **Table S1.** Descriptions of the forty-tree sampling sites in the Nationwide Nitrogen Deposition Monitoring Network (NNDMN)[a]

Site Code	Site Name	Region	Coordinate	Meteorological parameters[b]				Population density (persons km^{-2})	Land use type
				Wind speed (m s^{-1})	Rainfall (mm)	Temp. (°C)	RH (%)		
NC1	China Agric. University	NC	116.28 º E, 40.02 º N	2.5	556	12.4	56	7616	Urban
NC2	BeijingNormal University	NC	116.37 º E, 39.96 º N	2.5	556	12.4	56	7616	Urban
NC3	Zhengzhou	NC	113.63 º E, 34.75 º N	2.5	641	14.6	65	17069	Urban
NC4	Dongbeiwang	NC	116.29 º E, 40.04 º N	2.5	556	12.4	56	587	Rural
NC5	Shangzhuang	NC	116.20 º E, 40.11 º N	2.5	556	12.4	56	327	Rural
NC6	Baoding	NC	115.48 º E, 38.85 º N	2.0	519	12.9	61	3029	Rural
NC7	Quzhou	NC	114.94 º E, 36.78 º N	1.8	517	13.9	61	606	Rural
NC8	Yangqu	NC	112.89 º E, 38.05 º N	2.1	443	10.1	59	58	Rural
NC9	Zhumadian	NC	114.05 º E, 33.02 º N	2.3	958	15.0	72	815	Rural
NC10	Yangling	NC	108.01 º E, 34.31 º N	1.6	599	13.3	71	1763	Rural
NC11	Yucheng	NC	116.63 º E, 36.94 º N	2.5	567	13.1	64	495	Rural
NC12	Lingshandao	NC	120.18 º E, 35.77 º N	4.9	712	12.7	71	482	Background
NC13	Changdao	NC	120.75 º E, 37.93 º N	5.5	562	12.2	68	786	Background
NE1	Dalian	NE	121.58 º E, 38.92 º N	4.6	623	10.8	65	14280	Urban
NE2	Gongzhuling	NE	124.83 º E, 43.53 º N	3.8	573	5.7	63	271	Rural
NE3	Lishu	NE	124.17 º E, 43.36 º N	2.8	611	6.7	64	221	Rural
NE4	Wuyin	NE	129.25 º E, 48.11 º N	2.1	628	1.1	69	33	Background
NE5	Genhe	NE	121.52 º E, 50.78 º N	2.2	518	-6.2	67	5	Background
NW1	Shengdisuo	NW	87.57 º E, 43.87 º N	2.5	265	7.2	57	5122	Urban

NW2	Tufeisuo	NW	87.58 º E, 43.82 º N	2.5	265	7.2	57	236	Urban
NW3	Wuwei	NW	102.60 º E, 38.07 º N	1.8	170	8.2	52	58	Rural
NW4	Duolun	NW	116.49 º E, 42.20 º N	3.4	374	2.4	60	5	Background
NW5	Bayinbuluke	NW	83.71 º E, 42.88 º N	2.7	272	-4.3	70	26	Background
SE1	Nanjing	SE	118.85 º E, 31.84 º N	2.5	1062	15.7	75	729	Urban
SE2	Baiyun	SE	113.27 º E, 23.16 º N	1.8	1748	22.1	77	322	Urban
SE3	Wuxue	SE	115.79 º E, 30.01 º N	2.0	1418	17.3	77	469	Rural
SE4	Taojiang	SE	111.97 º E, 28.61 º N	1.8	1332	17.1	78	372	Rural
SE5	Fengyang	SE	117.56 º E, 32.88 º N	2.6	935	15.4	73	331	Rural
SE6	Zhanjiang	SE	110.33 º E, 21.26 º N	2.9	1678	23.2	82	639	Rural
SE7	Fuzhou	SE	119.36 º E, 26.17 º N	2.6	1374	20	75	97	Rural
SE8	Fenghua	SE	121.53 º E, 29.61 º N	2.4	1408	16.8	79	413	Rural
SE9	Feiyue	SE	113.34 º E, 28.56 º N	1.3	1502	16.9	81	228	Background
SE10	Huinong	SE	113.41 º E, 28.52 º N	1.3	1502	16.9	81	313	Background
SE11	Xishan	SE	113.31 º E, 28.61 º N	1.3	1502	16.9	81	313	Background
SW1	Wenjiang	SW	103.84 º E, 30.55 º N	1.4	939	18.6	80	1650	Urban
SW2	Ziyang	SW	104.63 º E, 30.13 º N	1.1	945	17.3	80	555	Rural
SW3	Yanting	SW	105.47 º E, 31.28 º N	1.2	879	16.4	77	253	Rural
SW4	Jiangjin	SW	106.18 º E, 29.06 º N	1.3	855	16.4	77	383	Rural
SW5	Dianchi	SW	102.67 º E, 24.97 º N	2.1	991	15.1	72	858	Rural
SW6	Yunnan Agric. University	SW	102.75 º E, 25.13 º N	2.1	991	15.1	72	2384	Rural
SW7	Kunyang	SW	102.61 º E, 24.67 º N	2.3	903	16.3	75	204	Background
TP1	Xining	TP	101.79 º E, 36.62 º N	1.5	387	6	56	2167	Urban
TP2	Linzhi	TP	94.36 º E, 29.65 º N	1.7	673	8.9	63	5	Background

[a]See Sect. S2 in the online supplement for the details of all the sampling sites in the network.

[b]There were no mean annual meteorological data for the forty-three monitoring sites; the surface climate parameters presented (1961-2012) were obtained from the nearest meteorological observation stations available on the China Meteorological Data Sharing Services website (http://cdc.gov.cn/).

[c] The population density was estimated by dividing population by area of the town/district/county where the monitoring site is located. Population data were sourced from the sixth census of China in 2010 and can be accessed online (http://www.stats.gov.cn).

Table S2. Monitoring periods for atmospheric N deposition and types of underlying surface at the forty-three sites in China.

Site Code	Site Name	Monitoring period		Underlying surface
		Dry deposition	Wet deposition	
NC1	China Agric. University	Apr. 2010-Sep. 2014	Apr. 2010-Sep. 2014	Vegetations
NC2	Beijing Normal University	Oct. 2013-Sep. 2014	Jan. 2012-Sep. 2014	Roof
NC3	Zhengzhou	May. 2010-Sep.2014	Jan. 2010-Dec. 2011	Vegetations
NC4	Dongbeiwang	Aug. 2006-Sep. 2008	Aug. 2006-Sep. 2008	Vegetations
NC5	Shangzhuang	Apr. 2010-Sep. 2014	Apr. 2010-Sep. 2014	Vegetations
NC6	Baoding	Jan. 2011-Dec. 2011	Jan. 2011-Dec. 2011	Vegetations
NC7	Quzhou	Apr. 2010-Sep. 2014	Apr. 2010-Sep. 2014	Vegetations
NC8	Yangqu	Apr. 2010-Sep. 2014	Apr. 2010-Sep. 2014	Vegetations
NC9	Zhumadian	Apr. 2010-Sep. 2014	Jan. 2011-Sep. 2014	Vegetations
NC10	Yangling	Apr. 2010-Sep. 2014	Apr. 2010-Sep. 2014	Vegetations
NC11	Yucheng	Sep. 2012-Sep. 2014	Jan. 2013-Sep. 2014	Vegetations
NC12	Lingshandao	Feb. 2011-Sep. 2014	Feb. 2011-Sep. 2014	Vegetations
NC13	Changdao	Sep. 2010-Sep. 2014	Sep. 2010-Sep. 2014	Vegetations
NE1	Dalian	Sep. 2010-Sep. 2014	Sep. 2010-Sep. 2014	Vegetations
NE2	Gongzhuling	Jul. 2010-Sep. 2014	Jul. 2010-Sep.2014	Vegetations
NE3	Lishu	Jul. 2010-Sep. 2014	Jul. 2010-Sep.2014	Vegetations
NE4	Wuyin	Oct. 2010-Sep. 2011	Oct. 2010-Sep. 2011	Vegetations
NE5	Genhe	Oct. 2010-Sep. 2011	Oct. 2010-Sep. 2011	Vegetations
NW1	Shengdisuo	Sep. 2009-Dec. 2011	Sep. 2009-Aug. 2010	Vegetations

NW2	Tufeisuo	Sep. 2009-Dec. 2011	Sep. 2009-Aug. 2010	Vegetations
NW3	Wuwei	Oct. 2010-Sep. 2014	Oct. 2010-Sep. 2014	Vegetations
NW4	Duolun	Jul. 2013-Sep. 2014	Jan. 2010-Sep. 2014	Grass
NW5	Bayinbuluke	May.2010-Aug. 2011	Nov. 2010-Jun. 2012	Grass
SE1	Nanjing	Sep. 2010-Feb. 2012	Sep. 2010-Feb. 2012	Vegetations
SE2	Baiyun	May. 2010-Sep. 2014	May. 2010-Sep. 2014	Roof
SE3	Wuxue	Aug. 2011-Sep. 2014	Aug. 2011-Sep. 2014	Roof
SE4	Taojiang	Oct. 2010-Sep. 2014	Jan. 2011-Sep. 2014	Roof
SE5	Fengyang	Feb. 2013-Sep. 2014	Oct. 2013-Sep. 2014	Vegetations
SE6	Zhanjiang	Aug. 2010-Sep. 2014	Jan. 2013-Sep. 2014	Vegetations
SE7	Fuzhou	Apr. 2010-Sep. 2014	Apr. 2010-Sep. 2014	Vegetations
SE8	Fenghua	Aug. 2010-Sep. 2014	Jan. 2011-Sep. 2014	Vegetations
SE9	Feiyue	Sep. 2010-Sep. 2014	Sep. 2010-Sep. 2014	Vegetations
SE10	Huinong	Sep. 2010-Sep. 2014	Sep. 2010-Sep. 2014	Vegetations
SE11	Xishan	Sep. 2010-Sep. 2014	Sep. 2010-Sep. 2014	Vegetations
SW1	Wenjiang	Oct. 2010-Sep. 2014	Jan. 2011-Sep. 2014	Vegetations
SW2	Ziyang	Jul. 2010-Sep. 2014	Jan. 2011-Sep. 2014	Vegetations
SW3	Yanting	May. 2011-Oct. 2013	May. 2011-Dec. 2012	Vegetations
SW4	Jiangjin	Jan. 2013-Sep. 2014	Jan. 2013-Sep. 2014	Vegetations
SW5	Dianchi	Apr. 2009-Mar. 2010	Apr. 2009-Mar. 2010	Vegetations
SW6	Yunnan Agric. University	Apr. 2009-Mar. 2010	Apr. 2010-Mar. 2010	Vegetations
SW7	Kunyang	Apr. 2009-Mar. 2010	Apr. 2009-Mar. 2010	Vegetations
TP1	Xining	Dec. 2013-Nov. 2014	Dec. 2013-Nov. 2014	Vegetations

TP2	Linzhi	Jan. 2009-Dec. 2009	Jan. 2009-Dec. 2009	Vegetations

Table S3. Statistics of monthly mean dry deposition velocities of N_r species for the three land use types, basing on the modeled hourly values at the forty-three sites from January 2010 to May 2013[a].

Land use type		Monthly mean deposition velocities (cm s^{-1})				
		NH_3	NO_2	HNO_3	pNH_4^+	pNO_3^-
Urban	N	410	410	410	410	410
	Min	0.30	0.01	0.12	0.06	0.06
	Max	1.63	0.44	5.78	0.35	0.35
	Mean	0.45	0.12	1.22	0.17	0.17
	Median	0.34	0.10	1.15	0.17	0.17
	SD	0.29	0.10	1.00	0.07	0.07
Rural	N	902	902	902	902	902
	Min	0.09	0.01	0.05	0.07	0.07
	Max	1.10	0.46	5.78	0.37	0.37
	Mean	0.40	0.17	1.49	0.18	0.18
	Median	0.35	0.16	1.49	0.18	0.18
	SD	0.16	0.12	1.10	0.06	0.06
Background	N	451	451	451	451	451
	Min	0.20	0.01	0.06	0.05	0.05
	Max	1.48	0.57	8.88	0.31	0.31
	Mean	0.47	0.17	1.78	0.16	0.16
	Median	0.43	0.13	1.48	0.16	0.16
	SD	0.22	0.15	1.66	0.06	0.06

[a] The forty-three sites consist of 10 urban, 22 rural and 11 background sites. Among the forty-three monitoring sites, 20 farmland, 5 coastal, 6 forest and 2 grassland sites were included in the rural and background sites.

Table S4. Comparison of annual deposition velocities of nitrogen compounds on different land use types in this study with literature.

Land use type	Country	Deposition velocity (cm s^{-1})					References
		NH_3	NO_2	HNO_3	pNH_4^+	pNO_3^-	
Farmland	China	0.40 (0.06)	0.18 (0.08)	1.52 (0.72)	0.19 (0.03)	0.19 (0.03)	This study
		0.26	0.12	0.81	0.16	0.16	Zhu et al. (2010)
		0.74	0.59	2	0.24	0.24	Shen et al. (2009)
		0.13	0.23	—	—	—	Yang et al. (2010)
		0.18	0.10	0.76	—	0.25	Zhang et al. (2004)
		0.71	0.05	—	—	—	Pan et al. (2012)
	West Africa	0.23-0.42	0.20-0.35	0.63-0.73	—	—	Delon et al. (2012)
	Britain	—	0.16	2.06	0.10	0.26	Marner and Harrison. (2004)
	Europe	0.35-0.75	0.05-0.20	0.8-1.0	0.11-0.20	0.10-0.25	Flechard et al. (2011)
Urban	China	0.45 (0.27)	0.12 (0.06)	1.25 (0.57)	0.17 (0.04)	0.17 (0.04)	This study
		0.78	0.06	—	—	—	Pan et al. (2012)
		0.28	0.07	1.77	0.44	0.44	Li et al. (2013)
		0.50	0.60	1.1	0.24	0.24	Luo et al. (2013)
	Greece	0.81	0.26	2.4	0.1	0.2	Anatolaki et al. (2007)
	Britain	—	0.08	7.27	1.02	1.78	Marner and Harrison. (2004)
Coastal	China	0.65 (0.26)	0.16 (0.11)	1.56 (0.54)	0.13 (0.03)	0.13 (0.03)	This study
		—	—	—	0.6	1.15	Zhang et al. (2007)
		0.63	0.00534	0.63	—	—	Zhang et al. (2010)
		0.55	0.01	0.84	0.27	0.27	Zhang et al. (2004)
		—	—	—	0.019	0.088	Qi et al. (2013)

Table S4 (*continued*)

Land use type	Country	Deposition velocity (cm s^{-1})					References
		NH_3	NO_2	HNO_3	pNH_4^+	pNO_3^-	
Forest	China	0.41 (0.05)	0.19 (0.05)	2.23 (1.28)	0.16 (0.01)	0.16 (0.01)	This study
		0.1	0.05	—	—	—	Pan et al. (2012)
		0.20	0.09-0.11	2.43-2.46	0.27-0.33	0.27-0.33	Zhang et al. (2004)
	Belgium	1.1-2.9	0.25	—	—	—	Staelen et al. (2012)
	Switzerland	2.2-3.0	0.3-0.4	1.5	0.2-0.4	0.2-0.4	Schmitt et al. (2005)
	Britain	—	0.26	7.27	1.02	1.78	Marner and Harrison. (2004)
	Europe	0.75-2.4	0.06-0.28	2.3-4.4	0.2-2.0	0.2-2.5	Flechard et al. (2011)
Grassland	China	0.378 (0.002)	0.15 (0.01)	1.09 (0.26)	0.194 (0.002)	0.194 (0.002)	This study
		0.23	0.13	1.16	0.28	0.28	Zhang et al. (2004)
		1.1	0.25	—	—	—	Staelen et al. (2012)
	Britain	—	0.16	1.39	0.06	0.15	Marner and Harrison. (2004)
	Europe	0.45-1.1	0.05-0.25	1.1-1.2	0.12-0.14	0.11-0.19	Flechard et al. (2011)
All land uses	The United States	0.65	0.36	2.70	0.15	0.15	Zhang et al. (2012)

Values in the parentheses are standard deviations of annual mean deposition velocities at sampling sites with the same land use type.

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585
586
587 **Table S5.** Emission inventory of NH_3 and NO_x for thirty-three provinces as well as the six regions of China in 2010 (kt)

Region	Site	Area ($\times 10^3 km^2$)	NH_3 (kt)	NO_x(kt)
North China	Beijing	16.4	61.5	389.2
	Tianjin	11.9	48.4	731.3
	Hebei	188.8	904.1	1685.2
	Shandong	187.1	1027.2	1941.8
	Shanxi	156.7	322.5	779.7
	Henan	167.0	1165.4	1140.0
	Shaanxi	205.8	281.5	452.1
	Total	933.8	3810.5	7120.2
Northeast China	Liaoning	148.0	340.4	1021.6
	Jilin	190.2	329.4	474.5
	Heilongjiang	437.0	465.9	716.5
	Total	612.1	1135.8	2212.6
Northwest China	Xinjiang	1660	374.2	308.9
	Inner Mongolia	1183	380.2	660.0

	Ningxia	66.4	73.9	177.0
	Gansu	453.7	199.0	374.4
	Total	3363.1	1027.2	1520.3
Southeast China	Shanghai	6.3	55.0	1091.2
	Jiangsu	102.6	806.3	1557.0
	Zhejiang	101.8	346.6	1199.8
	Anhui	140.3	631.9	768.1
	Hubei	185.9	668.2	783.5
	Hunan	211.8	742.6	566.2
	Jiangxi	166.9	461.3	439.7
	Fujian	124.0	288.9	439.4
	Guangdong	179.8	618.1	1382.1
	Hong Kong & Macau	1.1	6.7	251.2
	Taiwan	36.2	120.0	431.9
	Hainan	35.4	92.0	89.7
	Total	1254.8	4838.8	8962.7
Southwest China	Sichuan	485.0	925.4	650.1

	Chongqing	82.4	254.3	296.4
	Guizhou	176.2	465.5	312.9
	Yunan	390.0	700.0	453.1
	Guangxi	236.7	547.8	399.5
	Total	1370.3	2893.1	2112.0
Tibetan plateau	Tibet	1200.0	172.2	85.2
	Qinghai	722.3	121.6	85.0
	Total	1922.3	293.8	170.2

588 The emission data of NO_x and NH_3 for the year 2010 are from the **GAINS (Greenhouse Gas and Air Pollution Interactions and Synergies)**
589 Model (available online: http://www.iiasa.ac.at/)
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