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Key Points:

- Rates of atmospheric wet deposition of N and P were 13.69 and 0.21 kg ha⁻¹ a⁻¹ with a N:P ratio (by mass) of 77 in China
- Regional fertilizer and energy consumption correlated strongly with N deposition but had a limited effect on P deposition
- N:P ratios of atmospheric wet deposition were negatively correlated with soil N:P ratios in ecological regions

Supporting Information:

- Supporting Information S1

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Imbalanced atmospheric nitrogen and phosphorus depositions in China: Implications for nutrient limitation

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Abstract Atmospheric wet nitrogen (N) and phosphorus (P) depositions are important sources of bioavailable N and P, and the input of N and P and their ratios significantly influences nutrient availability and balance in terrestrial as well as aquatic ecosystems. Here we monitored atmospheric P depositions by measuring monthly dissolved P concentration in rainfall at 41 field stations in China. Average deposition fluxes of N and P were 13.69 ± 8.69 kg N ha⁻¹ a⁻¹ (our previous study) and 0.21 ± 0.17 kg P ha⁻¹ a⁻¹, respectively. Central and southern China had higher N and P deposition rates than northwest China, northeast China, Inner Mongolia, or Qinghai-Tibet. Atmospheric N and P depositions showed strong seasonal patterns and were dependent upon seasonal precipitation. Fertilizer and energy consumption were significantly correlated with N deposition but less correlated with P deposition. The N:P ratios of atmospheric wet deposition (with the average of 77 ± 40 , by mass) were negatively correlated with current soil N:P ratios in different ecological regions, suggesting that the imbalanced atmospheric N and P deposition will alter nutrient availability and strengthen P limitation, which may further influence the structure and function of terrestrial ecosystems. The findings provide the assessments of both wet N and P deposition and their N:P ratio across China and indicate potential for strong impacts of atmospheric deposition on broad range of terrestrial ecosystems.

1. Introduction

Nitrogen (N) and phosphorus (P) are generally considered as the two most essential and growth-limiting elements in terrestrial ecosystems [Elser *et al.*, 2007]. In the past few decades, emissions of anthropogenic reactive N and P have been enhanced greatly by growth of global population, industrialization, and agricultural intensification. It is estimated that the production of reactive N will reach 270 Tg N a⁻¹ in 2050, as a result of biological N fixation, burning of fossil fuels, and fertilizer application [Galloway *et al.*, 2004]. Similarly, P mobilization in 2000 was 2.5–5-fold its natural background value on a global scale. In particular, human intensification of the global phosphorus cycle raised P fluxes from about 50 Mt P a⁻¹ in preindustrial time to 155 Mt P a⁻¹ in 2000 [Smil, 2000]. Thus, anthropogenic activities have significantly altered and influenced the N and P cycles in terrestrial ecosystems [Galloway *et al.*, 2004; Liu *et al.*, 2013; Smil, 2000].

This mobilization of the N and P cycles has been manifested in major changes in the rates of nutrient deposition from the atmosphere. It is estimated that N wet deposition accounts for 40–80% of total atmospheric N deposition in most regions [Holland *et al.*, 2005; Jung *et al.*, 2013; Lü and Tian, 2007; Vet *et al.*, 2014]. Recognition of the importance of wet deposition of N has led to the establishment of a number of observational networks and projects quantifying atmospheric wet N deposition in different regions of the globe, including the National Atmospheric Deposition Program, the Clean Status and Trends Network of the United States, the Nitrogen Saturation Experiments, and the Experimental Manipulation of Forest Ecosystems in Europe project. Unlike carbon (C) and N, P does not have a stable gaseous phase in the atmosphere, and thus, atmospheric P deposition has been thought to be relatively small [Smil, 2000]. However, atmospheric P deposition (as direct dust deposition or as dust entrained in precipitation) is a potential source

of new P input to terrestrial ecosystems and could be ecologically important due to frequent limitation of primary productivity by P in many ecosystems [Elser *et al.*, 2007].

N and P cycles are tightly coupled at different trophic levels, and the fertilizing effect of atmospheric N deposition on terrestrial ecosystems may be amplified by concurrent inputs of P deposition [Elser *et al.*, 2007; Harpole *et al.*, 2011]. In ecosystems that are colimited by N and P, primary productivity is determined by both the input intensity and the ratio of N and P inputs [Harpole *et al.*, 2011]. Scientists have widely used the N:P ratio of plant biomass as indicators of N or P limitation in terrestrial ecosystems [Güsewell, 2004]. N:P ratios of vegetation can directly indicate the limiting element (N or P) and reflect the N and P supply ratio of soils at a community level [Güsewell, 2004]. In particular, N:P ratios > 20 (by mass) indicate P limitation, while N:P ratios < 10 indicate N limitation [Güsewell, 2004]. Experimental evidence has shown that changes in N and P availability in terrestrial ecosystems can shift plant N:P ratios as plants adjust to nutrient supplies [Güsewell, 2004]. Furthermore, changes in plant N:P ratios could also influence the structure and function of terrestrial ecosystems, both directly (through plant physiological responses) and indirectly (through the responses of herbivores and decomposers) [Fernández-Martínez *et al.*, 2014; Güsewell, 2004; Harpole *et al.*, 2011].

In contrast to massive and widespread emissions of reactive N, sources of atmospheric P deposition are rather limited and arise mainly from wind erosion and anthropogenic activities [Mahowald *et al.*, 2008]. However, dust emission has increased significantly because of increased soil disturbance from agricultural activities and land use change [Peñuelas *et al.*, 2012]. As a result of these ongoing and disproportionate changes, the N:P ratio of atmospheric deposition is anticipated to increase in the Northern Hemisphere [Peñuelas *et al.*, 2013]. Thus, it appears likely that increasing atmospheric N and P depositions and the imbalance of N and P input (N:P ratio) may significantly but differentially alter supplies of bioavailable N and P in terrestrial ecosystems.

Over the past few decades, China has experienced some of the largest increases in emissions of reactive N and P globally due to population growth, increased energy consumption, agricultural intensification, and land use change. While some studies have reported atmospheric wet N deposition at local sites, for catchments, across transects, and at the national scale in China [Jia *et al.*, 2014; Lü and Tian, 2007; Zhu *et al.*, 2015], atmospheric P deposition at regional or national scales has not been reported (although some studies have investigated P deposition rates at local sites [Hou *et al.*, 2012; Luo *et al.*, 2011; Parron *et al.*, 2011; Tsukuda *et al.*, 2006]). In this study, 41 stations from the Chinese Ecosystem Research Network (CERN), which was initially established for monitoring N deposition (Figure 1) [Zhu *et al.*, 2015], were further used to assess atmospheric P deposition in rainfall. The main objectives of this study were to (1) evaluate atmospheric P deposition rates and N:P ratios impinging on Chinese terrestrial ecosystems, (2) explore the factors that influence atmospheric P deposition, and (3) evaluate the N:P ratios of atmospheric N and P depositions at different scales and consider their potential ecological effects with regard to ecological stoichiometry.

2. Methods

2.1. Site Description

Forty-one CERN sites were selected to monitor atmospheric wet N and P depositions (Table S1 in the supporting information and Figure 1). The sites cover all major terrestrial ecosystems in China, including forest, grassland, desert, lake, marsh, and karst ecosystems. They are located in 22 provinces of China and encompass eight ecological regions according to climate and vegetation type (Figure 1) [Zhu *et al.*, 2015].

2.2. Sampling and Analysis

Precipitation was collected into plastic buckets installed 1.5 m above the ground at the onset of a rain (or snow) event. Samples were only collected over the duration of rainfall and included rained out soluble and insoluble particulates. In practice, sample collection was carried out 3–5 times per month in 2013. After each rainfall event, the samples were stored in polyethylene plastic bottles at -20°C , and at the end of the month these subsamples were evenly mixed on an equal-volumes-per-collection basis to obtain one sample representing each month. All plastic buckets and polyethylene plastic bottles were cleaned with distilled water 3 times and dried prior to use.

In the laboratory, samples were first filtered by gravity through a $0.45\ \mu\text{m}$ membrane filter to remove insoluble particulates. We then measured dissolved P using inductively coupled plasma optical emission spectrometer

Figure 1. The 41 observation sites of atmospheric N and P depositions in China. The dots with different colors indicate different ecosystems types. Aksu (AKA), Ailao Mountains (ALF), Ansai (ASA), Beijing (BJU), Inner Mongolia Grassland (NMG), Changshu (CSA), Daxinganling (DAF), Dangxiong (DXA), Dinghu Mountain (DHF), Dongling Mountain (BJF), Dongting Lake (DTM), Duolun (DLG), Erdos (ERF), Fengqiu (FQA), Fukang (FKD), Gongga Mountain at 1600 m (GGF1), Gongga Mountain at 3000 m (GGF2), Haibei (HBG), Hailun (HLA), Heshan (HSF), Huzhong (HZF), Huanjiang (HJK), Huitong 1 (HTF1), Huiton 2 (HTF2), Lhasa (LSA), Linze (LZA), Luancheng (LCA), Miaoxian (MXF), Naiman county (NMD), Qianyanzhou (QYA), Sanjiang (SJM), Shapotou (SPD), Shennongjia (SNF), Shenyang (SYA), Taoyuan (TYA), Xishuanbanna (BNF), Yanting (YGA), Yingtan (YTA), Yucheng (YCA), Changbai Mountains (CBF), and Changwu (CWA).

(Optima 5300DV, PerkinElmer, USA). In detail, dissolved P includes phosphate and dissolved organic P. Duplicate blank and standard reference materials (monopotassium phosphate, KH_2PO_4) methods were used to assure data quality.

2.3. Data Calculation and Analysis

The fluxes of P deposition were calculated as follows:

$$D = \sum_{i=1}^n \frac{C_i \times P_i}{100}$$

where D (kg ha^{-1}) is the P deposition flux per year, which is the sum of monthly P deposition flux (kg ha^{-1}); C_i is the monthly P concentration in rainfall (mg L^{-1}); P_i is the monthly precipitation (mm); and 100 is the unit conversion factor. All the N deposition data in this study were obtained from Zhu *et al.* [2015]. N:P ratios are reported as mass ratios.

Due to the high spatial heterogeneity of atmospheric N deposition in China and differing areas relevant for each sampling site, Kriging interpolation was applied to construct national-scale wet N and P deposition maps. Taiwan was not evaluated because the data were insufficient. Wet P deposition fluxes in Linze (LZA) and Haibei (HBG) were noticeably higher than the values around these sites; therefore, they were not included in the Kriging interpolation. Prior to the interpolation, a geostatistical method tool from ArcGIS 10.0 software (Environmental Systems Research Institute, Redlands, CA) was employed to conduct data analyses, including an analysis of the data distribution and identification of outliers and trends. The optimal variogram model and parameters were then determined. The wet P deposition data were log transformed to produce a normal distribution for the original data. The normal distribution plots and tests are shown in Figure S1 and Table S2, respectively. In addition, a cross validation was implemented to evaluate the results

of the Kriging interpolation. The cross-validation analysis and prediction errors are shown in Figure S2 and Table S3, respectively. The predicted values were not perfect due to the limited number of observation sites which had high root-mean-square (10.19 for N deposition and 0.18 for P deposition) and relatively low regression slope values (0.36 for N deposition and 0.32 for P deposition, both $p < 0.01$) (Figure S2 and Table S3).

In this study, only the annual deposition fluxes of China and the eight ecological regions were obtained from Kriging interpolation. Atmospheric wet N and P deposition in the provinces (22 provinces) were the averages of the observation sites that were located in those same provinces. All data are reported with average $\pm$ standard deviation (SD) or average $\pm$ standard error (SE) in the figure. SD could be obtained from ArcGIS software directly after we used Kriging interpolation methods or calculated by the values of sample duplicates in the same sites or ecosystems types. SE could be calculated through the equation: $SE = SD/\sqrt{n}$, where n is the number of duplicates.

Data for chemical fertilizer use in the provinces were obtained from the China Statistical Yearbook (2001–2010, Table S4) [Yearbook, 2001–2010b]. Data for energy consumption in the forms of coal, crude oil, natural gas, and their by-products in the provinces were obtained from the China Energy Statistical Yearbook (2001–2010, Table S4) [Yearbook, 2001–2010a]. Linear and curvilinear regressions were used to evaluate the relationships between N or P depositions and precipitation (at 41 sites scales), chemical fertilizer use (at 22 provinces scales), and energy consumption (at 22 provinces scales). Data for soil N:P ratios in different regions were derived from Tian *et al.* [2010] in which collected 2473 soil profiles and developed mean values for various soil groups across China. Moreover, they calculated the average soil N:P ratio in five regions based on climate differences which were similar to our study. A significance level of $p < 0.1$ was used for all tests. All analyses were conducted using the Statistical Package for the Social Sciences 12.0 program.

3. Results

3.1. Spatial Patterns of N and P Depositions

Average annual wet N deposition was $13.69 \pm 8.69 \text{ kg N ha}^{-1} \text{ a}^{-1}$ in China (Figure 2); i.e., the total N deposition in China was $12.52 \text{ Tg N a}^{-1}$. Central, south, and north China regions had higher N deposition, with averages of 24.94 ± 6.79 , 23.88 ± 4.36 , and $22.75 \pm 5.57 \text{ kg N ha}^{-1} \text{ a}^{-1}$, respectively. N deposition fluxes in the northwest, Qinghai-Tibet, northeast Inner Mongolia, and southwest regions were relatively low, with averages of 7.48 ± 2.61 , 8.36 ± 4.19 , 10.49 ± 7.30 , 11.45 ± 4.10 , and $12.05 \pm 1.68 \text{ kg N ha}^{-1} \text{ a}^{-1}$, respectively.

Annual wet P deposition was $0.21 \pm 0.17 \text{ kg P ha}^{-1} \text{ a}^{-1}$, and the P input through deposition in China was approximately 0.19 Tg P a^{-1} . Atmospheric wet P deposition had a spatial distribution similar to that for N deposition (Figures 2, 3a, and 3b). Southwest, central, and south China regions had higher levels of P deposition and P deposition fluxes that were 0.36 ± 0.04 , 0.43 ± 0.15 , and $0.63 \pm 0.10 \text{ kg P ha}^{-1} \text{ a}^{-1}$, respectively. P deposition fluxes were relatively low in the northwest, Inner Mongolia, Qinghai-Tibet, northeast, and north China regions, where P deposition fluxes were 0.093 ± 0.01 , 0.11 ± 0.02 , 0.13 ± 0.04 , 0.15 ± 0.03 , and $0.18 \pm 0.07 \text{ kg P ha}^{-1} \text{ a}^{-1}$, respectively.

Atmospheric N and P depositions varied greatly among the 41 observation sites (Figure 2). N deposition fluxes ranged from 0.47 to $47.71 \text{ kg N ha}^{-1} \text{ a}^{-1}$, while P deposition fluxes ranged from 0.002 to $2.53 \text{ kg P ha}^{-1} \text{ a}^{-1}$. Specifically, N deposition rates in Daxinganling (DAF) and Huzhong (HZF) were lower than $1.5 \text{ kg N ha}^{-1} \text{ a}^{-1}$, while N deposition exceeded $40 \text{ kg N ha}^{-1} \text{ a}^{-1}$ in Changshu (CSA) and Fengqiu (FQA). Atmospheric wet P deposition was the lowest in Ansai (ASA) and the highest in Changshu (CSA) and Xishuangbanna (BNF), where the values exceeded $2 \text{ kg P ha}^{-1} \text{ a}^{-1}$.

There were no significant differences among the four main ecosystem types for both wet N and P depositions (Figure 4, means test, all $p > 0.1$). However, agriculture and forest ecosystems had relatively higher wet N and P deposition, while grassland and desert ecosystems had relatively lower wet N and P deposition.

3.2. Seasonal Dynamics of Atmospheric Wet N and P Depositions

The monthly concentration of dissolved N first decreased and then increased from January to December ($R^2 = 0.658$, $p = 0.008$, Figure S3). However, monthly P concentrations showed no significant variation with season. Atmospheric wet N and P depositions showed similar seasonal dynamics (Figure S3). Monthly N ($R^2 = 0.801$, $p = 0.001$) and P ($R^2 = 0.808$, $p = 0.001$) deposition both increased and then decreased over the course of a year, with the maximum value in summer and the minimum values in winter.

Figure 2. Atmospheric N and P depositions from rainfall and their mass ratio in different scales (mean $\pm$ SD). Data from the following 41 observation sites with the same color located in the same regions. The eight regions are northwest China (NW), Inner Mongolia (IM), northeast China (NE), north China (NC), central China (CC), south China (SC), southwest China (SW), and Qinghai-Tibet (QT), respectively.

3.3. Factors Influencing Atmospheric Wet N and P Depositions

Mean annual N concentration and N deposition at the 41 observation sites showed significant linear relationships with precipitation ($R^2 = 0.120$, $p = 0.0027$ for N concentration; $R^2 = 0.217$, $p = 0.002$ for N deposition) (Table 1). With increasing precipitation N concentration decreased and annual N deposition fluxes increased. Similarly, as precipitation increased P concentration decreased and annual P deposition fluxes increased (Table 1, $R^2 = 0.165$, $p = 0.008$).

Furthermore, monthly N concentration and N deposition fluxes of the 41 observation sites were significantly related with monthly precipitation (MP) ($R^2 = 0.693$, $p = 0.001$ for N concentration; $R^2 = 0.893$, $p < 0.0001$ for N deposition), respectively (Table 1). However, monthly P concentration was not significantly related to MP, but monthly P deposition did have a positive linear relationship with MP ($R^2 = 0.937$, $p < 0.0001$).

Atmospheric wet N ($R^2 = 0.684$, $p < 0.0001$) and P ($R^2 = 0.159$, $p = 0.066$) depositions showed significant linear positive correlation with chemical fertilizer use, i.e., N and P deposition increased with the increasing of chemical fertilizer use. In addition, N deposition had a significant logarithmic relationship with energy consumption (logarithmic base number $a = 5.93$, $R^2 = 0.422$, $p = 0.001$), whereas there was no significant relationship between P deposition and energy consumption.

3.4. N:P Ratios of Atmospheric Wet Deposition

The N:P ratios of atmospheric wet deposition varied considerably among the 41 observation sites, with an average of 77 ± 40 (by mass) and ranging from 7 to 5418 (Figure 2). The average N:P ratio (by mass) in the eight ecological regions ranged from 35 to 138. The spatial pattern of N:P ratios of deposition is shown in Figure 3c. N:P ratios in the north China and Inner Mongolia regions were relatively high, with

Figure 3. Spatial patterns of atmospheric N and P deposition from rainfall in China. The grids in the right hand corner of the figures represent the South China Sea.

Figure 4. Atmospheric N and P depositions in main terrestrial ecosystem types (mean $\pm$ SE). The ratios in figure indicate the N:P ratios in four ecosystems types.

values of 138 ± 30 and 112 ± 41 , respectively; N:P ratios in the south-west and south China regions were lower (35 ± 7 and 38 ± 7 , respectively). Moreover, N:P ratios of atmospheric wet deposition were surprisingly negatively correlated with soil N:P ratios (Figure 5, $R^2 = 0.80$, $p = 0.007$). Meanwhile, deposition N:P ratios in desert, grassland, forest, and agriculture were 25 ± 8 , 67 ± 42 , 91 ± 195 and 422 ± 1336 , respectively (Figure 4). There were no significant differences in N:P ratio among the four main ecosystem types (means test, $p = 0.69$).

4. Discussion

4.1. Atmospheric Wet N and P Depositions and Their N:P Ratios in Chinese Terrestrial Ecosystems

Atmospheric wet N deposition in Chinese terrestrial ecosystems in 2013 was $13.69 \text{ kg N ha}^{-1} \text{ a}^{-1}$ (including $7.25 \text{ kg N ha}^{-1} \text{ a}^{-1}$ for NH_4^+-N , $5.93 \text{ kg N ha}^{-1} \text{ a}^{-1}$ for NO_3^--N , and $0.52 \text{ kg N ha}^{-1} \text{ a}^{-1}$ for dissolved organic nitrogen, reported in Zhu *et al.* [2015]). Previous studies have also evaluated atmospheric wet N deposition for China using different data sources and approaches. Lü and Tian [2007] obtained estimates of $\sim 9.88 \text{ kg N ha}^{-1} \text{ a}^{-1}$ for 1980 to 2005. Jia *et al.* [2014] collected published monitoring data concluding that atmospheric wet N deposition in China increased from $11.11 \text{ kg N ha}^{-1} \text{ a}^{-1}$ in the 1990s to $13.87 \text{ kg N ha}^{-1} \text{ a}^{-1}$ in the 2000s. These results clearly indicate that atmospheric N deposition in China increased over the past few decades [Jia *et al.*, 2014; Liu *et al.*, 2013]. While such compilation studies imply that summarizing annual monitoring data from publications are likely to result in large uncertainties because the data were obtained from different researchers that used different methods and at different times. Moreover, in our study we could document the dynamics of N deposition directly at the same time scale by using the observation network based on CERN.

Atmospheric wet P deposition in Chinese terrestrial ecosystems was approximately $0.21 \text{ kg P ha}^{-1} \text{ a}^{-1}$. Previous studies have reported P deposition only from scattered sites; e.g., Luo *et al.* [2011] reported that wet P deposition was about $0.33 \text{ kg P ha}^{-1} \text{ a}^{-1}$ in Lake Taihu, while Sun *et al.* [2013] reported rates of $1.1 \text{ kg P ha}^{-1} \text{ a}^{-1}$ in Nanjing. Other studies in different parts of the world also have recorded levels of atmospheric wet P deposition around $0.028\text{--}0.7 \text{ kg P ha}^{-1} \text{ a}^{-1}$ (Table 2) [He *et al.*, 2011; Herut *et al.*, 1999; Koelliker *et al.*, 2004; Morales *et al.*, 2001; Parron *et al.*, 2011; Pollman *et al.*, 2002; Rolff *et al.*, 2008; Tsukuda *et al.*, 2006]. Meanwhile, Tipping *et al.* [2014] conducted a metaanalysis of P deposition (included 246 sites and covered

Table 1. Regression Analyses Between Atmospheric Wet Deposition and Its Influencing Factors

Independent Variable	Variable	Type	a	b	n	R ²	p
Annual precipitation	N concentration	Linear	−0.0014	3.92	41	0.120	0.027
	P concentration	Linear	−0.00004	0.11	41	0.060	0.123
	N deposition	Linear	0.011	6.97	41	0.217	0.002
	P deposition	Linear	0.0005	0.12	41	0.165	0.008
Monthly precipitation	N concentration	Linear	−0.0099	3.47	12	0.693	0.001
	P concentration	Linear	0.00005	0.072	12	0.044	0.511
	N deposition	Linear	0.015	0.34	12	0.893	<0.0001
	P deposition	Linear	0.0006	−0.0011	12	0.937	<0.0001
Fertilizer use	N deposition	Linear	1.01	8.30	22	0.684	<0.0001
	P deposition	Linear	0.022	0.37	22	0.159	0.066
Energy consumption	N deposition	Logarithm	5.93	−15.26	22	0.422	0.001
	P deposition	Logarithm	0.057	0.27	22	0.018	0.546

Figure 5. The relationship between N:P ratios of atmospheric wet deposition and soil N:P ratios in different ecoregions (mean $\pm$ SE). The data of soil N:P ratios in different regions were derived from Tian *et al.* [2010]. The red dot in figure representing northwest regions in China was not considered in the fitting functions due to lowest soil N:P ratios; and if considered it, $R^2 = 0.393$, $p = 0.096$.

with the exception of the driest areas (MAP < 40 cm) in the world (including southwest North America, southwest South America, and southwest and northern Africa), there are roughly equal contributions of dry and wet N deposition (i.e., $50\% \pm 10\%$) at continental scales (including China). Similarly, it was estimated that dry P deposition contributed 40–75% of the total atmospheric P deposition [Hou *et al.*, 2012; Luo *et al.*, 2011; Sun *et al.*, 2013]. Therefore, we assumed that the N:P ratios of total deposition (including wet and dry deposition) still remain at a high level, resulting from high N deposition and low P deposition [Peñuelas *et al.*, 2013]. It is important to point out that dry deposition is likely to be more important for P deposition than N deposition in some regions (i.e., desert) because P is sourced from mineral dust originating from deserts [Mahowald *et al.*, 2008]. Therefore, dry deposition should be more intensively studied to accurate estimate N and P depositions in future.

4.2. Dynamics Related to and Factors Influencing Atmospheric Wet N and P Depositions

Atmospheric wet N and P deposition rates in our data showed substantial seasonal dynamics i.e., both annual and monthly precipitations were negatively correlated with N concentration and positively correlated with N and P deposition fluxes (Table 1). Huang *et al.* [2013] reported similar dynamics for wet N deposition in Shenzhen during 1986–2006, when the lowest N concentrations and the highest N deposition fluxes occurred during the warm season. In contrast, Pollman *et al.* [2002] demonstrated that seasonal changes in P concentration were not evident and that wet P deposition followed a strongly seasonal pattern in response to precipitation. Low N concentration and high N deposition flux occurred during the summer, likely because

the period 1954 to 2012) and reported that the bulk deposition of total P is about $0.27 \text{ kg P ha}^{-1} \text{ a}^{-1}$ on a global scale, indicating that atmospheric wet P deposition in China is comparable to the rest of the world.

The average N:P ratio of atmospheric wet deposition in China was 77:1, ranging between 35 and 138 across the eight ecological regions. These estimates do not consider dry deposition because direct measurements of atmospheric dry deposition at large scale are difficult and require highly sophisticated methods and instruments [Wesely and Hicks, 2000]. Previous studies have reported high N:P ratios of atmospheric wet deposition, such as ~ 10 –45 in Lake Maracaibo [Morales *et al.*, 2001] and ~ 46 –200 in Baltic Sea [Rolff *et al.*, 2008]. Vet *et al.* [2014] estimated that

Table 2. N and P Deposition in Rainfall Reported in Previous Literature and the Present Study ($\text{kg ha}^{-1} \text{ a}^{-1}$)

Site	N Deposition	P Deposition	Mass N:P	Rainfall (mm)	Reference
Western Baltic proper	3.87 ~ 7.27	0.028 ~ 0.158	46 ~ 200	643	Rolff <i>et al.</i> , [2008]
Eastern Mediterranean	2.8	0.09	31	499 ~ 537	Herut <i>et al.</i> , [1999]
Ashiu, central Japan	–	0.24	–	2323	Tsukuda <i>et al.</i> , [2006]
Florida, USA	–	0.075	–	161	Pollman <i>et al.</i> , [2002]
Cerrado, central Brazil	12.6	0.7	18	1400	Parron <i>et al.</i> , [2011]
New Jersey, USA	–	0.052 ~ 0.081	–	–	Koelliker <i>et al.</i> , [2004]
Singapore	15.48	0.51	30	2230	He <i>et al.</i> , [2011]
Lake Maracaibo, Venezuela	6.93 ~ 16.84	0.6	10 ~ 45	1123	Morales <i>et al.</i> , [2001]
Globe	–	0.27	–	–	Tipping <i>et al.</i> , [2014]
China	13.69	0.21	65	653	This study

atmospheric N was removed from the atmosphere and was greatly diluted by frequent, intense precipitation during the rainy season. Clearly, precipitation has a significant effect on overall atmospheric wet N and P deposition fluxes, and the seasonal distribution of precipitation is an important driver of the temporal-spatial patterns of atmospheric wet N and P depositions. Though there is no consensus regarding whether precipitation will increase or decrease globally (including China), a well-accepted feature of forecasted alterations to the global climate is an increase in the frequency and magnitude of climate extremes, such as severe drought and periods of heavy rainfall [Smith, 2011; Solomon, 2007]. Thus, variability in N and P depositions might increase with increases in the fluctuation of precipitation.

Energy consumption and fertilizer use correlated strongly with atmospheric wet N deposition, suggesting that anthropogenic emissions, including fossil fuel combustion, chemical fertilizers, and the burning of biofuels, are main sources of atmospheric N deposition [Galloway *et al.*, 2004; Liu *et al.*, 2013]. However, the sources and formation of atmospheric P were less clear and likely depended on soil disturbances due to farming, soil erosion, pollen spreading, burning of plant material, and burning of fossil fuels [Herut *et al.*, 1999]. Mahowald *et al.* [2008] demonstrated that mineral dust resulting from the wind erosion of soils account for 82% of the total global aerosol P, while 12%, 5%, and 1% of atmospheric P are from primary biogenic sources, combustion of materials, and sea salt and volcanic aerosols, respectively. This is one possible reason that P deposition was less strongly correlated with fertilizer use and energy consumption in the present study. Compared with atmospheric P, atmospheric N is easily transmitted over long distances; therefore, the effect of N deposition was more extensive and regionalized, whereas a large portion of atmospheric P deposition likely reflected local factors [Mahowald *et al.*, 2008; Vet *et al.*, 2014].

4.3. N:P Ratios of Wet Deposition in Different Ecosystems Types and Regions and Its Potential Effects

Human activities play an important role in atmospheric deposition. Our results show that central and south China (the most intensively developed regions) had higher levels of N and P depositions (Figures 3a and 3b), which also contributed to the observation that atmospheric N and P deposition rates were higher in agriculture and forest but lower in grassland and desert ecosystems (Figure 4). Moreover, deposition N:P ratios (mass) in forest (91:1) and agriculture (422:1) were considerably higher than those in desert (25:1) and grassland (67:1). Deposition collectors in forest and agriculture ecosystems were located in areas that also had high population density, while desert and grassland ecosystems are more likely to be present in lightly populated areas in China [Yue *et al.*, 2005]. This indicates that economically developed regions, with their accompanying higher energy consumption and fertilizer application (as well as higher annual rainfall), have strongly elevated levels of N and P depositions in China.

N:P ratios of atmospheric deposition were highest in north China (138:1) and lowest in southwest (35:1) and south China (38:1) regions (Figures 2 and 3c). Despite being highly complex and heterogeneous in vegetation, tropical regions in south China are often considered to be less frequently N limited than temperate regions [Peñuelas *et al.*, 2013]. Tian *et al.* [2010] estimated that soil N:P ratios were about 1.6 in warm temperate zone and 2.9 in subtropical and tropical zones. We found a negative correlation between the N:P ratios of atmospheric wet deposition and soil (Figure 5). The higher N:P ratio of atmospheric inputs might serve as positive feedback to temperate and boreal vegetation by enhancing N availability and alleviating N limitation to some extent. However, long-term human N inputs might be converting originally N-limited ecosystems into a state of N saturation and enhanced P limitation (as seen in lakes, Elser *et al.* [2009]), which indicates the positive feedback of N inputs on productivity might vanish with chronic atmospheric N loading. While the N:P input ratio of atmospheric deposition was relatively low in tropical regions compared to temperate regions, the response of tropical ecosystems to atmospheric deposition is more uncertain due to the higher plant diversity and strong nutrient limitation.

Elser *et al.* [2009] have argued that higher N deposition in ecosystems might result in lower producer diversity by skewing resource supply ratios in favor of one particular nutrient (N) relative to others, such as P, and thus favoring only a small subset of primary producer species. The potential effect of imbalanced N and P on agriculture ecosystems might be very weak even though agriculture ecosystems had the highest N:P ratios because of human management of nutrients and biota. However, for forest and grassland ecosystems, the addition of limiting nutrients might dramatically change the dominant species [Yu *et al.*, 2015] and significantly decrease ecosystem biodiversity [Vitousek and Farrington, 1997; Yu *et al.*, 2015]. Aber *et al.* [1989] also demonstrated that long-term elevated N inputs might also cause a decrease of net primary productivity. Therefore, high

N deposition coupled with relatively low P deposition input might modulate the “hump-shaped” curve between ecosystem productivity and biodiversity but with effects dependent on the biota that exists in those ecosystems.

4.4. Potential Risk of P Limitation Caused by High N:P Ratios in Atmospheric Wet Deposition

Effects of high N:P ratios of nutrient input through wet deposition on terrestrial ecosystems are likely to be complex. Responses could include shifts in the N:P ratio of plant biomass via indirect effects of changing the N and P supply ratio of soils [Elser *et al.*, 2009; Koerselman and Meuleman, 1996]. After long-term adaptation to the environment, the N:P in a specific ecosystem or component of ecosystems might achieve a relatively stable value akin to the Redfield ratio in marine plankton. Based on long-term survey data from CERN, Zhao *et al.* [2014] found that the overall biomass N:P ratios (including the plant community, soil, and litter) were about 4.5 in Chinese forest ecosystems and about 1.4 in Chinese agricultural ecosystems. N:P ratios of different components of ecosystems have also been studied separately. These studies report N:P ratios (mass) were ~14:1 in leaves [Reich and Oleksyn, 2004], 6:1 and 3:1 in soil and soil microbial biomass [Cleveland and Liptzin, 2007], 20:1 in forest litter [McGroddy *et al.*, 2004], and 16:1 in plant roots [Yuan *et al.*, 2011] at the global scale. Our results showed that N:P ratios of wet deposition in China centered around 77:1 and were often much higher. N deposition in 2050 will reach 270 Tg yr⁻¹ globally, and Liu *et al.* [2013] reported that N deposition in China has increased significantly in the past 30 years, mainly from fossil fuel combustion from industrial activities and vehicle, synthetic N fertilizers, and livestock cultivation in agricultural intensification. However, our unpublished results showed that dissolved inorganic N of wet deposition is likely to decrease in China due to decreases of ammonium in wet deposition. The current policies, such as “Zero Increase Action Plan” for national fertilizer use [Liu *et al.*, 2016], and the relative stable number of synthetic N fertilizers and livestock cultivation (from China Statistical Yearbook) in the past few years might be responsible for decreasing atmospheric deposition of ammonium. Although nitrate deposition has kept increasing along with energy consumption, nitrate deposition might be controlled in the next decade because of the focus of Chinese government on structural economic change. Nevertheless, N deposition is likely to stay at a high level and contribution to high N:P for a long time. Therefore, the relative balance of N and P in different components of terrestrial ecosystems could be distorted by the very imbalanced nutrient inputs coming from deposition.

Clearly, inputs of N and P through wet deposition would enhance nutrient availability in terrestrial ecosystems, promote gross primary productivity, alter biomass production efficiencies, and affect patterns of carbon allocation in ecosystems [Fernández-Martínez *et al.*, 2014; Wieder *et al.*, 2015]. Based on a compilation of published experimental studies, Elser *et al.* [2007] proposed that N and P limitation are equally prevalent in both terrestrial and freshwater systems. Moreover, simultaneous N and P enrichment often creates positive synergistic responses of primary productivity. Güsewell [2004] proposed that vegetation N:P ratios > 20 (by mass) indicate P limitation, which implies supply N:P ratios > 20 may also induce P limitation. We therefore infer that hugely imbalanced N and P inputs from atmospheric deposition should result in P having an increasingly strong effect on the structure and function of terrestrial ecosystems in China. Specifically, P limitation induced by imbalanced N:P ratios from wet deposition might influence biomass production, species composition, and species richness of terrestrial ecosystems over a long-term period [Braakhekke and Hooftman, 1999; Güsewell, 2004]. Various changes in ecosystem function, such as litter accumulation, sensitivity to drought, increased rate of fungal infection, altered dynamics of herbivores, and impaired reproduction of species, might ensue if N:P ratios increase [Güsewell, 2004]. The ecological effects of N will likely reflect variable N-loading gradients in different deposition regions and ecosystems and depend on the critical N and P concentrations in particular ecosystems. Further research is needed to clarify the validity of these expectations and determine the specific effects of imbalanced N:P ratio on terrestrial ecosystems in China and elsewhere [Elser *et al.*, 2007; Güsewell, 2004; Koerselman and Meuleman, 1996; Peñuelas *et al.*, 2013; Reich and Oleksyn, 2004].

5. Conclusion

In summary, atmospheric deposition of N and P through rainfall in China shows strong temporal-spatial variability. Precipitation has a significant effect on atmospheric wet deposition flux, and the seasonal or spatial distribution patterns of precipitation are important drivers of the temporal-spatial patterns of atmospheric wet N and P depositions. The average N:P ratio for atmospheric deposition in China was 77:1 (by mass), far greater than the N:P ratios of most biotic components in terrestrial ecosystems. Meanwhile,

N:P ratios of atmospheric wet deposition were negatively correlated with soil N:P ratios in different ecoregions. It is likely that high N:P ratios from wet deposition may influence nutrient status and influence terrestrial ecosystem structure and function in the long run. These data document N and P depositions at regional and national scales in China for the first time, demonstrating a highly elevated N:P ratio of atmospheric wet deposition and highlighting considerable potential for ecological effects of these inputs as a result of imbalanced ecological stoichiometry.

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