# Reply to comments on "Imbalanced phosphorus and nitrogen deposition in China's forests" by Anonymous Referee #1

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Review

This is still a potentially publishable manuscript, however there are a few edits that need to be made.

*Reply: Thank you. We have revised the manuscript according to your suggestions.*

I would like to heavily encourage the authors to submit a revised manuscript with all the changes highlighted and indicate in the response to reviewers exactly the text changed. Because this was not done, I had to reread the entire manuscript, and thus, perhaps I have found elements that were in the first version, but would have missed if the authors has highlighted their changes more effectively.

*Reply: We are sorry that we didn't submit a revised manuscript with highlighted changes last time. This time we include a marked-up manuscript version showing the changes we have made.*

The question of what kind of deposition the authors are looking at is better explained in the methods section: I see there the highlighted changes. However, I am still not sure that these aren't the soluble N and P, instead of total deposition. Because of the methods shown, and the uncertainty about what exactly this deposition is, I would like to encourage the authors to be more clear that they are using wet deposition in both the abstract, introduction and

conclusions.

*Reply: We believe that we describe in the methods section how we approximated total P and N deposition. As shown in the first part of the method section, our analysis is based on datasets of total P and total N contents in bulk precipitation and throughfall, which are assessed after digestion and include dissolved inorganic forms, dissolved organic forms and particulate forms. We then mention (see page 3 line 23-26) that on an annual basis, total deposition equals the sum of N or P fluxes in throughfall, stemflow and canopy exchange but that we approximate total deposition by multiplying the volume-weighted mean concentration in throughfall with the sum water fluxes of throughfall and stemflow. We then mention (see top of page 4) that this approach causes a slight underestimate of total deposition since tree foliage can take up a small proportion of soluble N and P and because throughfall concentrations of N or P are generally were lower than those in stemflow. We thus believe that the method section clearly shows how we used total (not soluble) N and P concentrations in bulk precipitation and throughfall to estimated bulk deposition, total deposition and canopy-captured dry deposition.*

In the response to the reviewers, the authors indicate that they are very interested in ecological impacts in managed forests, which is really interesting and important and should be added to the introduction, and more so to the results and conclusions. If they are interested in ecological impacts, aren't they more interested in N:P ratios in the deposition? Following the ideas of Brahney et al., 2016, shouldn't they look at how N:P ratios in vegetation relate to the deposition ? The P deposition is much more effective (14x), because less P is required. How does that change the conclusions of this study?

*Reply: Thanks for the suggestions. The introduction section has included some information on the possible effects of imbalanced P and N deposition. We now added information on the paired analysis by Brahney et al. (2015) on N:P ratios in remote alpine lakes with N:P ratios in atmospheric deposition. We mention however, that such a relationship can expected in remote lakes where atmospheric deposition is the dominant nutrient source, whereas in forest ecosystems atmospheric P and N deposition is only a relatively small part of the total nutrient supply, most of which is derived from mineralization and/or weathering. A lack of corresponding datasets of N:P ratios in forest vegetation hinders us to test this correlation, but we expect a less clear relationships. Nevertheless, based on an analysis of literature, we expect that "the role of P deposition is relevant because high-level N deposition, initially stimulating forest growth, is likely shifting large areas of China's forests towards P limitation". In the discussion section, further analysis has been made on the imbalance of P and N deposition. The N:P ratios in bulk precipitation and throughfall is more than twice of that in tree leaves (geometric mean N:P ratio = 15) (Han et al.,*

*2005) and three times of that in current-year twigs (geometric mean N:P ratio = 10.6) (Yao et al., 2015). It is also much higher than critical N:P ratios related to relative P limitation in view of forest growth, which are near 15 for coniferous forests and near 25 for deciduous forests (after Mellert and Gättlein, 2012).*

"Based on measurements of P content in various fuels and estimates of the P partitioning during combustion processes, a recent assessment indicates that anthropogenic sources, such as combustion-related emissions, can contribute to over 50% of the global atmospheric P and make a substantial contribution to global P deposition (Wang et al., 2015)." This is not correct. They get this large number by ignoring size differentiation between their model (<20um) and locally produced primary biogenic aerosols, and assuming that deposition in forests generated by locally produced biogenic aerosols (in Tipping et al., 2013) comes from combustion instead. Using different assumptions, Brahney et al., 2015 shows that one can match the Tipping et al., 2013 data, as well as other data constraining the size of the combustion sources (as done in Mahowald et al., 2008, but not done in Wang et al., 2015). Please rewrite to "Based on assumptions of the P partitioning during combustion processes and local deposition, a recent assessment hypothesizes that anthropogenic sources, such as combustion-related emissions, can contribute to over 50% of the global atmospheric P budget (Wang et al., 2015), although this result is very sensitive to assumptions about size distribution (Brahney et al., 2016)."

*Reply: Thank you for this correction. We have done according to the suggestion.*

"Anthropogenic sources have been traditionally thought to make only a small contribution to P deposition (Okin et al., 2004; Mahowald et al., 2008). In contrast, our results emphasize that anthropogenic sources near large cities can have significant impacts on the spatial patterns of regional P deposition. The urban hotpot of P deposition might be derived from intensive combustion-related emissions near urban areas (Wang et al., 2015) and a short-distance transfer of P-containing aerosols from P-rich farmland soils (Anderson et al., 2006)." Again, as in the first version, the Mahowald et al., 2008 study also has hotspots close to cities, so it is not correct that you need huge P sources from combustion to get this result. Also, you get maximum values close to the arid regions. Please be more clear on this. I would suggest something like: "Anthropogenic sources have been traditionally thought to make only a small contribution to P deposition (Okin et al., 2004; Mahowald et al., 2008), and yet can still result in local hotspots of deposition (e.g. Mahowald et la., 2008: Wang et al., 2015) along with P from arid regions, which is consistent with our study. The urban hotpot of P deposition might be derived from intensive combustion-related emissions near urban areas (Wang et al., 2015) and a short-distance transfer of P-containing aerosols from P-rich farmland soils

(Anderson et al., 2006)."

*Reply: Thank you. We have revised the text almost completely as suggested, i.e. "Anthropogenic sources have been traditionally thought to make only a small contribution to P deposition (Okin et al., 2004; Mahowald et al., 2008), and yet can result in local hotspots of deposition (e.g. Mahowald et al., 2008: Wang et al., 2015). This is in accordance with our study, which indicates that anthropogenic sources near large cities can have significant impacts on the spatial patterns of regional P deposition. The urban hotpot of P deposition might be derived from intensive combustion-related emissions near urban areas (Wang et al., 2015) and a short-distance transfer of P-containing aerosols from P-rich farmland soils (Anderson et al., 2006)".*

# Imbalanced phosphorus and nitrogen deposition in China's forests

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**Abstract.** Acceleration of anthropogenic emissions in China has substantially increased nitrogen (N) deposition during last three decades and may result in an imbalance of atmospheric N and phosphorus (P) inputs in terrestrial ecosystems. However, the status of P deposition in China is poorly understood. This study synthesized data on total P and total N concentrations in bulk precipitation and throughfall from published literature to assess the characteristics of P deposition, N deposition and N:P deposition ratio in China's forests. Our results show relatively high mean rates of bulk P deposition (0.38 kg P ha$^{-1}$ yr$^{-1}$) and total P deposition (0.69 kg P ha$^{-1}$ yr$^{-1}$), but they were accompanied by even much more elevated N inputs via bulk deposition (16.5 kg N ha$^{-1}$ yr$^{-1}$) and total deposition (21.6 kg N ha$^{-1}$ yr$^{-1}$), resulting in high N:P ratios in bulk deposition (44.4) and total deposition (32.8), respectively. Based on the difference between total deposition and bulk deposition, canopy-captured dry P and N deposition ~~is~~was estimated to be 0.31 kg P ha$^{-1}$ yr$^{-1}$ and 5.1 kg N ha$^{-1}$ yr$^{-1}$, respectively. We found significantly higher P deposition and lower N:P ratios at sites nearby than those far from semiarid regions. The estimated bulk and total deposition of P and N both showed a significant power-law increase with closer distance to the nearest large cities either in the areas nearby or far from semiarid regions. Our results suggest an anthropogenic alternation of regional P and N cycling, which might shift large areas of China's forests towards human-induced P limitation especially in southern China.

# 1 Introduction

Nitrogen (N) and phosphorus (P) are essential macronutrients, both of which widely limit primary productivity across terrestrial ecosystems (Elser et al., 2007; Vitousek et al., 2010). ~~New~~External N inputs to terrestrial ecosystems are driven by biological N fixation and atmospheric deposition, the latter being increasingly important (Cleveland et al., 2013). Acceleration of anthropogenic N emissions has substantially increased N deposition in China during last three decades and aroused widespread concerns about the consequent impacts on various ecosystems (Liu et al., 2011 & 2013; Cui et al., 2013). Enhanced N deposition often stimulates forest growth and hence carbon sequestration (Högberg, 2007; De Vries et al., 2009;

Thomas et al., 2010), but the expected growth acceleration might be diminished when the accompanied P supply is deficient (Braun et al., 2010; Li et al., 2016). Soil P availability in terrestrial ecosystems is primarily driven by mineral weathering and atmospheric deposition (Vitousek et al., 2010; Cleveland et al., 2013). Newman (1995) reviewed P deposition and weathering in global terrestrial ecosystems and estimated a range of 0.07 – 1.7 kg P ha$^{-1}$ yr$^{-1}$ for P deposition and 0.01 – 1.0

5  kg P ha$^{-1}$ yr$^{-1}$ for P weathering, indicating that both fluxes are in the same order of magnitude. More recent assessment on observed and modelled P deposition rates on a global scale (Mahowald et al., 2008; Wang et al., 2015) show a range of 0.01 − 1.0 kg P ha$^{-1}$ yr$^{-1}$ on land. Despite the significant role of P deposition, its status and characteristics are poorly understood in China.

Considering the importance of atmospheric deposition for N and P availability, the variation in N and P deposition may

10  make a difference in N or P-limited ecosystems (Peñuelas et al., 2013). Spatial patterns and temporal trends of N deposition have recently been well characterized in China (Liu et al., 2013; Du and Liu, 2014; Lu and Tian, 2014; Jia et al., 2014; Du et al., 2014; Xu et al., 2015; Zhu et al., 2015). However, most assessments on N deposition in China have been based on measurements of bulk deposition (Liu et al., 2013), which substantially underestimates the levels of total deposition because it consists mainly of wet deposition (Kulshrestha et al., 1995; Staelens et al., 2005; Chantara and Chunsuk, 2008). The N flux

15  in throughfall has been used as a more precise estimate of total deposition (Du et al., 2014 & 2015), although it is still an underestimate due to neglect of N input by stemflow and the occurrence of canopy N uptake, especially at lower N deposition levels (Draaijers et al., 1996; Sparks, 2009). Unlike N, there is a lack of evaluation on P deposition across China. Furthermore, global scale P deposition data, including scarce measurements in China, are limited to open-field sites, thus including bulk deposition only (Tipping et al., 2014). Forest canopies increase atmospheric deposition inputs of nutrients to

20  forest ecosystems by trapping dust and particulates ~~elevating the deposition inputs~~ (e.g. DeLonge et al., 2008; Das et al., 2011). In the context of low soil P contents in terrestrial ecosystems across most areas of China (Jiang et al., 1986; Han et al., 2005), the role of P deposition is relevant because high-level N deposition, initially stimulating forest growth, is likely shifting large areas of China's forests towards P limitation.

Previous studies have indicated that large cities in China are hotspots of N deposition (Du et al., 2014 & 2015) because

25  the enhancement of N deposition is primarily driven by anthropogenic emissions from power generation, motor traffic and agricultural activities near urban areas (Liu et al., 2013; Jia et al., 2014). Unlike reactive N which can form stable gaseous compounds (e.g. NO$_x$ and NH$_3$), atmospheric P occurs primarily in the form of coarse particles and is dominantly derived from mineral dust in natural conditions (Mahowald et al. 2008; Vet et al., 2014). Higher P deposition is thus expected nearby the semiarid regions, where receive large amounts of P-containing particulates transported from neighbouring deserts or

30  barren lands due to the effect of wind erosion (Okin et al., 2004; Mahowald et al. 2008). Based on ~~measurements~~ assumptions of ~~P content in various fuels and estimates of the~~ P partitioning during combustion processes and local deposition, a recent assessment ~~indicates~~ hypothesizes that anthropogenic sources, such as combustion-related emissions, can contribute to over 50% of the global atmospheric P budget and make a substantial contribution to global P deposition (Wang et al., 2015), although this result is very sensitive to assumptions about size distribution (Brahney et al., 2015). We can ~~thus~~

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potentially expect higher P deposition nearby large cities with intensive anthropogenic P emissions, although the transportation of P-containing dust may lead to distinct urban hotspots of P deposition nearby and far from the semiarid regions. Here we synthesized data on total P and total N concentrations in bulk precipitation and throughfall from published literature to assess the status and characteristics of P deposition, N deposition and N:P deposition ratios in China's forests.

5   Moreover, we also tested whether spatial patterns of P deposition showed an increase with closer distance to large cities.

## 2. Data and methods

### 2.1 Data sets

We collected data from published literatures on total P and total N concentrations in bulk precipitation and throughfall for typical forests in China, as well as information on site location (latitude and longitude) and annual precipitation. Total P and

10   total N concentrations were assessed after digestion and include dissolved inorganic forms, dissolved organic forms and particulate forms. Observational data were selected only when P and/or N concentrations in bulk precipitation and throughfall were measured simultaneously. Data on P and N concentrations for each sampling period were either taken directly from tables or digitized from figures using a GetData Graph Digitizer (Version 2.25, http://www.getdata-graph-digitizer.com). If concentration data at one site were measured for more than one forest stand or available for more than one

15   year, a volume-weighted mean was calculated and used for further analysis. The distance between the sampling site and the geometric centre of the nearest large city (non-agricultural population > 0.5 million) was derived using Google Earth for Microsoft Windows (Version 7.1.5.1557, Google Inc., USA).

    Our database consisted of 33 sites which were distributed across main forest biomes in China (see Fig. S1 and Table S1 for detailed information). Annual precipitation ranged from 500 mm to 2900 mm. The distance from the sampling sites to the

20   nearest large cities ranged from 5 km to 465 km. Using a criteria of a distance of 400 km to the semiarid regions (e.g. grassland), we grouped our datasets into sites nearby semiarid regions (n= 11) and sites far from the semiarid regions (n=22) (Fig. S1).

### 2.2 Calculation of bulk deposition, total deposition and canopy-captured dry deposition

Bulk deposition of total P and total N was calculated by multiplying the volume-weighted mean concentration in bulk

25   precipitation with annual precipitation. Canopy-captured dry deposition accumulates during periods without precipitation and it is washed down during precipitation events. On an annual basis, total deposition equals can be estimated as the sum of N or P fluxes in throughfall, stemflow and canopy exchange (Fig. S2). Due to a lack of measured data on canopy exchange and stemflow concentrations, we approximate total deposition by multiplying the volume-weighted mean concentration in throughfall with the sum water fluxes of throughfall and stemflow water fluxes. To estimate the sum water flux of

30   throughfall and stemflow water fluxes, we establish a database of observed throughfall and stemflow (including data from 28 forested stands from 26 sites, Fig. S3) and explored an empirical equation between precipitation and the sum water flux of

throughfall and stemflow (Fig. S4). The sum of throughfall and stemflow water fluxes for each forested site was then estimated from the available bulk precipitation data based on this empirical equation. This method generally underestimates the factual total deposition. First, tree foliage can take up a small proportion of soluble N and P in rainwater and gaseous N (part of dry deposition) (Reddy and Majmudar, 1983; Draaijers et al., 1996; Sparks, 2009). Moreover, P and N fluxes in

5  stemflow were underestimated because throughfall concentrations of N or P are generally were lower than those in stemflow. However, the total element input by stemflow is mostly less than 5% of that by throughfall (e.g. Talkner et al., 2010) and thus it only introduces a small error of the overall estimate.

Canopy-captured dry deposition, defined as the amounts of dry deposition captured and accumulated in forest canopy during the period without precipitation events, was calculated as the difference between the estimated total deposition and

10  bulk deposition. Our estimate of canopy-captured dry deposition is lower than total dry deposition because a proportion of dry deposition is already included in bulk deposition and the total deposition is likely underestimated.

## 2.3 Statistical analysis

A Shapiro-Wilk normality test was used to test whether data were normally distributed. Data ranges were indicated by the first and third quantile. We used a paired sample t-test to test the differences of concentrations and N:P ratios between bulk

15  precipitation and throughfall. A Student's t-test was used to test the difference of N deposition levels between sites nearby the semiarid regions (n=11) and far from the semiarid regions (n=22).

Previous studies have indicated that N deposition shows a power-law increase with closer distance to the nearest large cities (Du et al., 2014 & 2015). To test the urban hotspot hypothesis of P deposition, we also used a power-law model to explore changes in concentrations and fluxes of total P with the distance between the sampling site and the nearest large city.

20  The analysis was conducted separately for sites nearby the semiarid regions (n=11) and far from the semiarid regions (n=22). Spatial patterns of P deposition, N deposition and N:P ratios were illustrated using ArcGIS Desktop (version 9.3, ESRI, USA). All statistical analysis was performed using R software (version 3.1.0; R Development Core Team, 2014, http://www.r-project.org/) with a significance level of $p < 0.05$.

## 3. Results

25  ### 3.1 Concentrations and deposition of total phosphorus

Phosphorus concentrations in bulk precipitation and throughfall were characterized by a log-normal distribution (Shapiro-Wilk normality test, $p > 0.10$). The geometric mean of P concentration in throughfall (0.068, ranging $0.041 - 0.138$ mg P L$^{-1}$) was significantly higher (paired sample t-test, $df = 32$, $p < 0.01$), on average being 2.18 ($1.40 - 3.11$) times of that in bulk precipitation (0.031, ranging $0.017 - 0.050$ mg P L$^{-1}$). Similarly, bulk P deposition and total P deposition were also

30  characterized by a log-normal distribution (Shapiro-Wilk normality test, $p > 0.10$), showing geometric means at 0.38 ($0.21 - 0.57$) and 0.69 ($0.40 - 1.20$) kg P ha$^{-1}$ yr$^{-1}$, respectively. The canopy-captured dry P deposition, calculated as the difference

between total deposition and bulk deposition, was estimated at 0.31 kg P ha$^{-1}$ yr$^{-1}$, being comparable to bulk P deposition (paired sample t-test, $df = 32$, $p = 0.36$).

Spatial patterns of bulk P deposition and total P deposition showed remarkable heterogeneity, varying by a factor exceeding four (Fig. 1a and 1b). Specifically, geometric means of bulk P deposition and total P deposition were estimated to be 0.54 (0.39 – 0.98) and 0.97 (0.74 – 1.27) kg P ha$^{-1}$ yr$^{-1}$ at the 11 sites nearby the semiarid regions. At the other 22 sites far from the semiarid regions, geometric means of bulk P deposition and total P deposition were significantly lower (Student's t-test, $df = 31$, $p < 0.01$), being 0.31 (0.20 – 0.50) and 0.57 (0.26 – 1.05) kg P ha$^{-1}$ yr$^{-1}$, respectively.

## 3.2 Concentrations and deposition of total nitrogen

Concentrations and fluxes of total N were also log-normally distributed (Shapiro-Wilk normality test, $p > 0.10$). The geometric mean of total N concentration in bulk precipitation were 1.34 (0.88 – 2.19) mg N L$^{-1}$ and increased significantly (paired sample t-test, $df = 31$, $p < 0.01$) to 2.13 (1.23 – 3.87) mg N L$^{-1}$ in throughfall. Geometric means of bulk N deposition and total N deposition were 16.5 (9.9 – 24.2) and 21.6 (15.0 – 31.2) kg N ha$^{-1}$ yr$^{-1}$, leading to an estimate of canopy-captured dry N deposition of 5.1 kg N ha$^{-1}$ yr$^{-1}$. The ratio of total deposition versus bulk deposition was 1.31 (0.98 – 1.60) for N, which was significantly lower than that (1.80, 1.19 – 2.56) for P (paired sample t-test, $df = 31$, $p < 0.01$). Spatial patterns of bulk N deposition and total N deposition showed several regional hotspots near large city clusters in central, eastern and southern China (Fig. 2a and 2b).

## 3.3 Ratios of nitrogen deposition versus phosphorus deposition

The N:P ratios in bulk precipitation and throughfall were also characterized by a log-normal distribution (Shapiro-Wilk normality test, $p > 0.10$). The geometric mean of N:P ratio was 44.4 (23.1 – 97.6) in bulk precipitation and decreased to 32.8 (18.7 – 63.6) in throughfall. At the 11 sites nearby the semiarid regions, the geometric mean of N:P ratio in bulk precipitation and throughfall were estimated at 19.3 (9.1 – 39.9) and 16.2 (10.9 – 28.6), respectively. However, the N:P ratios in bulk precipitation (geometric mean = 64.0, ranging 40.9 – 116.7) and throughfall (geometric mean =45.3, ranging 25.8 – 74.6) were significantly higher (Student's t-test, $df = 31$, $p < 0.01$) at the other 22 sites far from the semiarid regions, most of which were located in southern China (Figs. 3a and 3b).

## 3.4 Urban hotspots of phosphorus and nitrogen deposition

Bulk P deposition and total P deposition both showed a significant power-law increase with closer distance to the nearest large cities, either nearby or far from the semiarid regions (Figs. 4a, 4d, 5a & 5d). In line with the urban hotspot hypothesis, bulk N deposition and total N deposition also showed a significant power-law increase with closer distance to the nearest large cities (Figs. 4b, 4e, 5b & 5e). At sites far from semiarid regions, N:P ratios in bulk precipitation and throughfall both showed no significant trend with changing distance to the nearest large cities (Figs. 4c & 4f). However, the N:P ratios in

throughfall at sites nearby semiarid regions showed a significant power-law increase with closer distance to the nearest large cities, while no such trend was found for N:P ratios in bulk precipitation (Figs. 5c & 5f).

## 4. Discussion

### 4.1 Phosphorus deposition in China's forests

5   Atmospheric P-containing aerosols are either scavenged by precipitation or gravitationally deposited to the ground surface during dry weather. Based on very limited measurements of wet deposition around the world, Vet et al. (2014) summarized that total P fluxes in wet deposition range from 0.04 to 0.32 kg P ha$^{-1}$ yr$^{-1}$. However, most reported measurements are based on bulk deposition, which includes wet deposition plus a fraction of dry deposition. A recent synthesis of global datasets with 246 sites indicates that bulk deposition of total P is log-normally distributed and showed a geometric mean of 0.27 kg P

10  ha$^{-1}$ yr$^{-1}$ with total P being nearly double the amount of inorganic P, which equalled 0.14 kg P ha$^{-1}$ yr$^{-1}$ (Tipping et al., 2014). However, large uncertainties remain in regional estimates due to the scarcity of observational data and unevenness of site distribution. For instance, mean bulk deposition of total P was estimated at 0.12 kg P ha$^{-1}$ yr$^{-1}$ in Asia based on measurements from only 7 sites (Tipping et al., 2014). Using observed data from 33 sites in China's forests, here we show much higher bulk deposition of total P with a geometric mean of 0.38 kg P ha$^{-1}$ yr$^{-1}$. Overall, our estimate of bulk P

15  deposition in China is higher than the estimates for Europe (0.22 kg P ha$^{-1}$ yr$^{-1}$), Oceania (0.24 kg P ha$^{-1}$ yr$^{-1}$) and North America (0.29 kg P ha$^{-1}$ yr$^{-1}$) by Tipping et al. (2014), while it is lower than those in South America (0.43 kg P ha$^{-1}$ yr$^{-1}$) and Africa (0.62 kg P ha$^{-1}$ yr$^{-1}$).

    Measurements of dry P deposition are rather scarce. Based onBy using limited data of aerosol concentrations and the referential dry deposition velocity, Vet et al. (2014) estimated that annual dry deposition of total P on land ranges from 0.01

20  to 0.62 kg P ha$^{-1}$ yr$^{-1}$. Based on the difference between total deposition and bulk deposition, here we estimated that the mean canopy-captured dry P deposition was as high as 0.31 kg P ha$^{-1}$ yr$^{-1}$ in China's forests. Although canopy-captured dry P deposition underestimates dry deposition because a proportion of dry deposition is already included in bulk deposition and the total N deposition was likely underestimated in this study, it was comparable to bulk deposition (0.38 kg P ha$^{-1}$ yr$^{-1}$) (paired t-test, $df = 32$, $p = 0.36$). This result implies that dry deposition is an important pathway of atmospheric P deposition.

25  Our assessment shows that the geometric mean of total P deposition in China's forests was 0.69 (0.40 – 1.20) kg P ha$^{-1}$ yr$^{-1}$, being comparable to the modelling results ranging from $0.4 - 1.0$ kg P ha$^{-1}$ yr$^{-1}$ in the forested areas in China (Wang et al., 2015). Using the mean total deposition of 0.69 kg P ha$^{-1}$ yr$^{-1}$ and a forested area of $1.76 \times 10^{8}$ ha (Zhang et al., 2010), we further estimated a total atmospheric P input of 0.12 Tg P yr$^{-1}$ in China's forests.

    Previous assessments have suggested that large-scale gradients of P deposition is dust dominated because mineral dust

30  from neighbouring deserts contributes substantially to atmospheric P deposition in semiarid regions (Okin et al., 2004; Mahowald et al., 2008). Accordingly, our results also show that P deposition tended to be higher nearby the semiarid regions (Fig. 1a & 1b). Geometric means of bulk P deposition and total P deposition were estimated to be as high as 0.54 (0.39 –

0.98) and 0.97 (0.74 – 1.27) kg P ha$^{-1}$ yr$^{-1}$ nearby the semiarid regions, while the values were significantly lower (Student's t-test, $df$ = 31, $p < 0.01$) far from the semiarid regions, being 0.31 (0.20 – 0.50) and 0.57 (0.26 – 1.05) kg P ha$^{-1}$ yr$^{-1}$, respectively. This indicates a significant contribution of dust-borne sources to atmospheric P deposition in forest ecosystems nearby the semiarid regions.

## 4.2 Nitrogen deposition in China's forests

Our results show high levels of bulk N deposition and total N deposition in China's forests, on average being 16.5 kg N ha$^{-1}$ yr$^{-1}$ and 21.6 kg N ha$^{-1}$ yr$^{-1}$, respectively. Most previous assessments on N deposition in China were either based on bulk deposition or wet deposition (Liu et al., 2013; Du and Liu, 2014; Jia et al., 2014; Du et al., 2014; Zhu et al., 2015; Xu et al., 2015), showing similar values (13.7 to 19.3 kg N ha$^{-1}$ yr$^{-1}$) as our estimates based on bulk deposition in China's forests. Although our estimate based on total N concentrations in throughfall and annual sum of throughfall and stemflow water fluxes is still lower than the factual total deposition because tree foliage can take up a small proportion of gaseous N (part of dry deposition) and soluble N in rainwater (Draaijers et al., 1996; Sparks, 2009) and the stemflow N flux is likely underestimated, it is a better proxy of total N deposition than bulk deposition. Bulk deposition (16.5 kg N ha$^{-1}$ yr$^{-1}$) only accounts for 76% of our estimate of total deposition (21.6 kg N ha$^{-1}$ yr$^{-1}$) in China's forests. This difference leads to an estimate of canopy-captured dry N deposition of at least 5.1 kg ha$^{-1}$ yr$^{-1}$, which is equivalent to 31 % of bulk deposition. The likely underestimation of this value is confirmed by a recent estimate based on airborne concentration measurements via a nationwide N deposition monitoring network and modelling inferential dry deposition velocity, which indicated that on average dry N deposition even exceeded wet/bulk deposition across China (Xu et al., 2015). Based on the mean total N deposition of 21.6 kg N ha$^{-1}$ yr$^{-1}$ and a forested area of $1.76 \times 10^8$ ha (Zhang et al., 2010), total N inputs via atmospheric deposition was thus estimated to be 3.8 Tg N yr$^{-1}$ in China's forests.

## 4.3 Urban hotspots of phosphorus and nitrogen deposition

Bulk P deposition and total P deposition both showed a significant power-law increase with closer distance to the nearest large cities, either far from (Figs. 4a, 4d) or nearby (Figs. 5a & 5d) the semiarid regions. Anthropogenic sources have been traditionally thought to make only a small contribution to P deposition (Okin et al., 2004; Mahowald et al., 2008). ), and yet can possibly result in local hotspots of deposition (e.g. Mahowald et al., 2008: Wang et al., 2015). This is in accordance with our study, which indicates that anthropogenic sources near large cities can have significant impacts on the spatial patterns of regional P deposition. The urban hotpot of P deposition might be derived from intensive combustion-related emissions near urban areas (Wang et al., 2015) and a short-distance transfer of P-containing aerosols from P-rich farmland soils (Anderson et al., 2006).

Spatial patterns of bulk N deposition and total N deposition showed several regional hotspots near large city clusters in central, eastern and southern China (Fig. 2a and 2b). In line with our previous assessments on inorganic N deposition (Du et al., 2014 & 2015), bulk N deposition and total N deposition were also in accordance to the urban hotspot hypothesis (Figs.

4b, 4e, 5b & 5e). This spatial pattern has been attributed to intensive motor traffic, energy production, waste treatment and agricultural activities (mainly N-fertilizer application and livestock breeding) in and/or near urban areas (Du et al., 2014 & 2015). Overall, our results suggest that rapid urbanization in China may have exerted significant alternation of regional P and N cycling by enlarging anthropogenic emissions and deposition. In addition, the urban hotspot model is an important approach to describe the way in which large cities shape the spatial pattern of P and N deposition. Therefore, it should be incorporated in modelling of P and N deposition at regional scales.

At sites far from semiarid regions, N:P ratios in bulk precipitation and throughfall both showed no significant trend with changing distance to the nearest large cities (Figs. 4c & 4f), indicating an synchronous changes in P and N deposition around these urban hotspots. However, N:P ratios in throughfall at sites nearby semiarid regions showed a significant power-law increase with closer distance to the nearest large cities, while no such trend was found for N:P ratio in bulk precipitation (Figs. 5c & 5f). This distinct trend of N:P ratios in bulk precipitation and throughfall suggest a more rapid increase in dry N deposition than in dry P deposition with closer distance to the nearest large cities.

### 4.4 Implications of imbalanced phosphorus and nitrogen deposition

Our results indicated an imbalance of N and P supply by atmospheric nutrient deposition in China's forests. The N:P ratio generally showed high values (geometric mean = 44.4) in bulk precipitation in China's forests. Although the N:P ratio decreased in the throughfall, the geometric mean (32.8) was still more than twice of that in tree leaves (geometric mean N:P ratio = 15) (Han et al., 2005) and three times of that in current-year twigs (geometric mean N:P ratio = 10.6) (Yao et al., 2015). It is also much higher than critical N:P ratios related to relative P limitation in view of forest growth, which are near 15 for coniferous forests and near 25 for deciduous forests (after Mellert and Göttlein, 2012). Compared to the sites nearby semiarid regions, the imbalance of P and N deposition was more intense in forests of southern China, showing significantly higher N:P ratios in bulk precipitation (geometric mean = 64.0, ranging 40.9 – 116.7) and throughfall (geometric mean =45.3, ranging 25.8 – 74.6) (Figs. 3a & 3b). Moreover, the ratio of total N versus total P may overestimate the relative P supply versus N because a fraction of atmospheric N deposition is most likely already taken up by the canopy (Draaijers et al., 1996; Sparks, 2009) and a fraction of atmospheric P usually is not bioavailable (Mahowald et al., 2008; Tipping et al., 2014).

Based on a paired analysis of N and P concentrations in deposition and lake water, Brahney et al. (2015) showed a clear relationship between N:P ratios in the investigated remote alpine lakes and N:P ratios in atmospheric deposition. A lack of corresponding datasets of N:P ratios in forest vegetation hinders us to test this correlation for forests but we expect a less clear relationship since atmospheric deposition is the dominant nutrient source in remote lakes, while it is only a relatively small part of the total nutrient supply in forests, most of which is derived from mineralization and/or weathering.. Nevertheless, it is likely that tThe imbalance of N and P deposition will may lead to an increase in the N:P ratio of soils and plant tissues and hence a shift towards human-induced P limitation in the long term (Peñuelas et al., 2013). Accordingly, a recent assessment has shown that foliar N concentration of woody plants in China's non-agricultural ecosystems increased significantly between the 1980s and the 2000s, while leaf P concentration did not change significantly over the same period

(Liu et al., 2013). In addition, mean leaf P concentrations of China's plants were found to be significantly lower than the global averages, most likely due to lower soil P content (Han et al., 2005). Lower leaf P may constrain the response of photosynthetic capacity to leaf N as P-deficient plants have limited ribulose-1,5-bisphosphate regeneration (Reich et al., 2009). Although enhanced N deposition often stimulates forest growth and carbon sequestration (Högberg, 2007; De Vries et al., 2009; Thomas et al., 2010), the expected growth acceleration can be diminished in P-limited forest ecosystems (Braun et al., 2010; Crowley et al., 2012; Li et al., 2016). Phosphorus limitation may not only constrain future forest growth in response to N deposition but also lower the projected $CO_2$ fertilization effects on primary productivity (Wieder et al., 2015). Modelling results have also indicated that terrestrial carbon sequestration in China showed a lower response to per unit N deposition in recent years (Tian et al., 2011). Unless efficient measures are taken to reduce anthropogenic N emissions in China, the threats of human-induced nutrient imbalance to the health and function of forest ecosystems may keep increasing in the next decades.

## 5. Conclusions

Our results show elevated atmospheric P deposition in China's forests, but they are accompanied by even much more elevated N deposition. High N:P ratios in bulk precipitation and throughfall suggest an imbalance of P and N deposition, which is likely resulting in a shift towards P limitation in forests especially in southern China. Moreover, spatial patterns of P and N deposition both showed a strong power-law increase with closer distance to the nearest large cities, implying a significant alternation of regional P and N cycling by rapid urbanization in China.

China has started to cut down $NO_x$ emissions since early 2010s (Twelfth Five-year Plan, A full version of the plan is available at http://news.xinhuanet.com/politics/2011-03/16/c_121193916.htm) and a stricter national control of air pollution is expected to reduce emissions of NOx from combustion of fossil fuels and biofuels in the future (http://www.mep.gov.cn/ztbd/rdzl/dqst/201307/t20130709_255093.htm). This will consequently lead to a substantial decrease in anthropogenic deposition of oxidized N (e.g. nitrate). However, the absence of $NH_3$ regulation policy and an increase in meat and dairy consumption may further enhance emissions and deposition of reduced N (e.g. ammonium) in the future. Overall, the issue of increasing imbalance of P and N deposition may further worsen because high levels of N deposition are expected to continue in the next decades. In order to gain a better understanding of the sources, composition and rates of P deposition as well as its ecological effects, monitoring networks of atmospheric deposition (e.g. Nationwide Nitrogen Deposition Monitoring Network, NNDMN, Xu et al., 2015; Chinese Ecosystem Research Network, CERN, Zhu et al., 2015) are encouraged to include measurements of P deposition across the country based on standardized sampling protocols and analytical methods. Field observations and manipulated experiments should be conducted to assess the impacts of nutrient imbalance on the health and function of China's forest ecosystems. Moreover, better forest management strategies should be adopted to avoid loss of forestry production from the human-induced P and N imbalance.

## Author contribution

E. Du. and W. de Vries conceived the idea. E. Du. conducted data analysis and prepared the manuscript. W. de Vries, Y. Jiang, W. Han, X. Liu and Z. Yan reviewed and edited the manuscript.
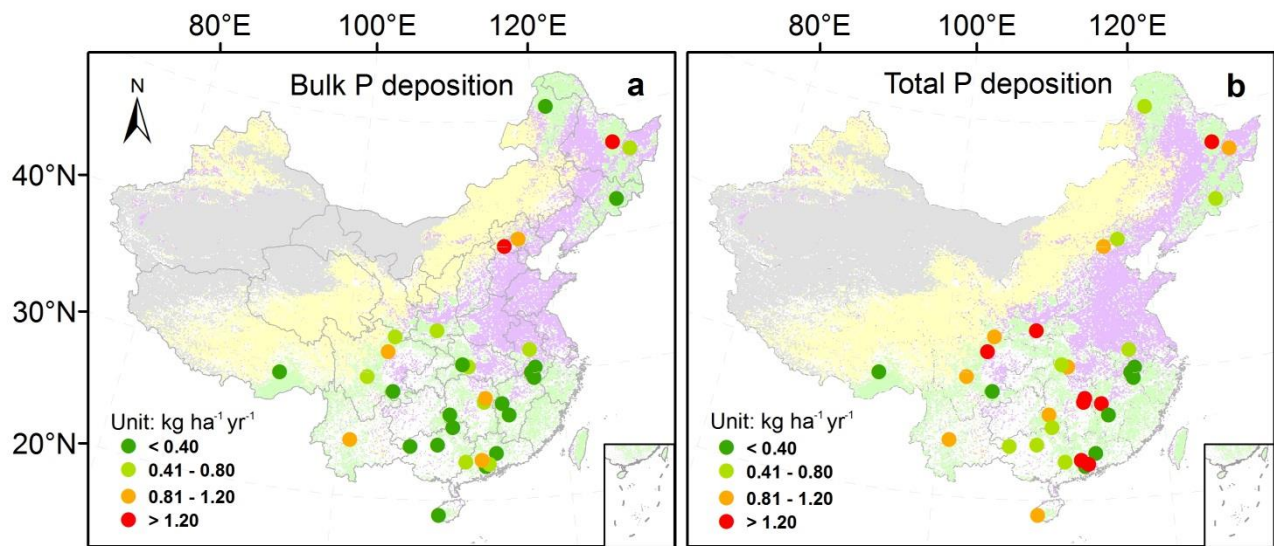
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Figure 1. Spatial patterns of (a) bulk P deposition (kg ha$^{-1}$ yr$^{-1}$) and (b) total P deposition (kg ha$^{-1}$ yr$^{-1}$) in China's forests. The background shadows in light green, light yellow, light purple and light grey indicate the distributions of forest, grassland (semiarid region), cropland and non-vegetated land, respectively.
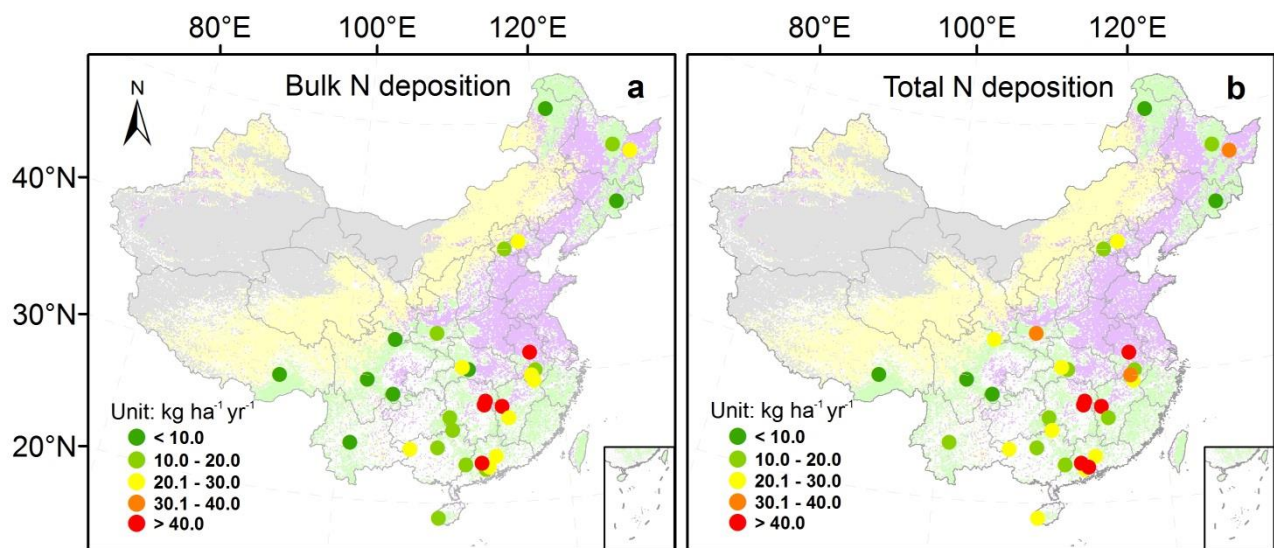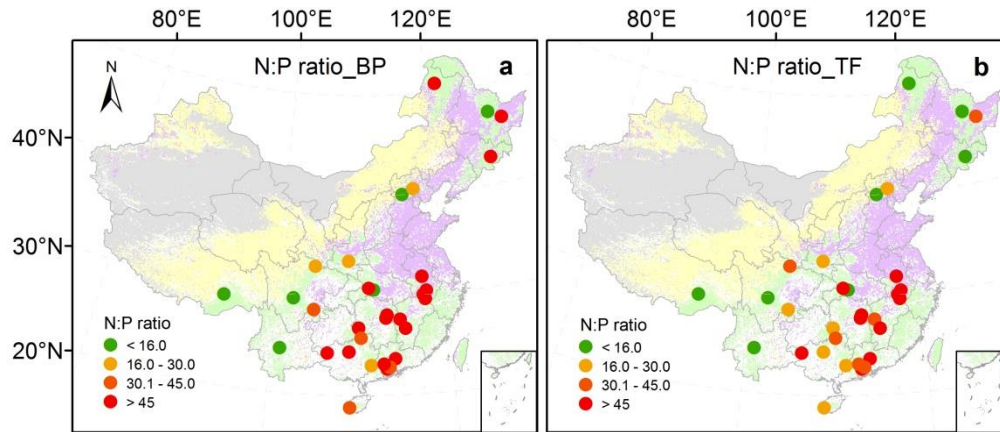
Figure 2. Spatial patterns of (a) bulk N deposition (kg ha$^{-1}$ yr$^{-1}$) and (b) total N deposition (kg ha$^{-1}$ yr$^{-1}$) in China's forests. The background shadows in light green, light yellow, light purple and light grey indicate the distributions of forest, grassland (semiarid region), cropland and non-vegetated land, respectively.

Figure 3. Spatial patterns of N:P ratios in (a) bulk precipitation (BP) and (b) throughfall (TF) in China's forests. The background shadows in light green, light yellow, light purple and light grey indicate the distributions of forest, grassland (semiarid region), cropland and non-vegetated land, respectively.
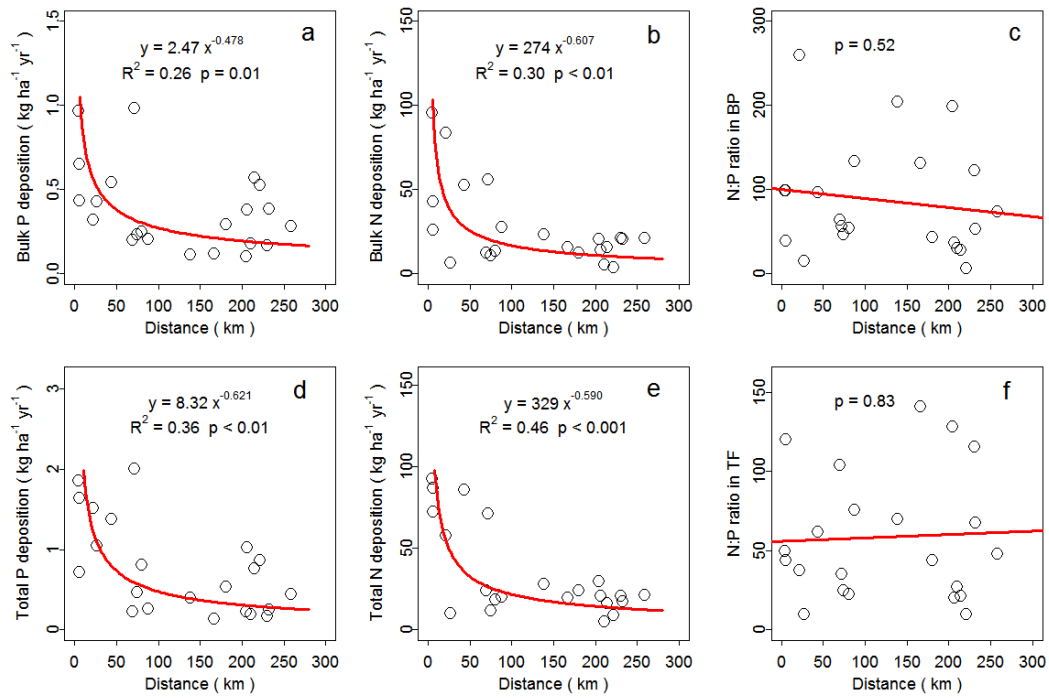
Figure 4. Changes in bulk deposition of P and N (kg ha$^{-1}$ yr$^{-1}$), total deposition of P and N (kg ha$^{-1}$ yr$^{-1}$), and N:P ratios in bulk precipitation (BP) and throughfall (TF) with the distance to the nearest large cities based on datasets of forested sites far from semiarid regions in China.
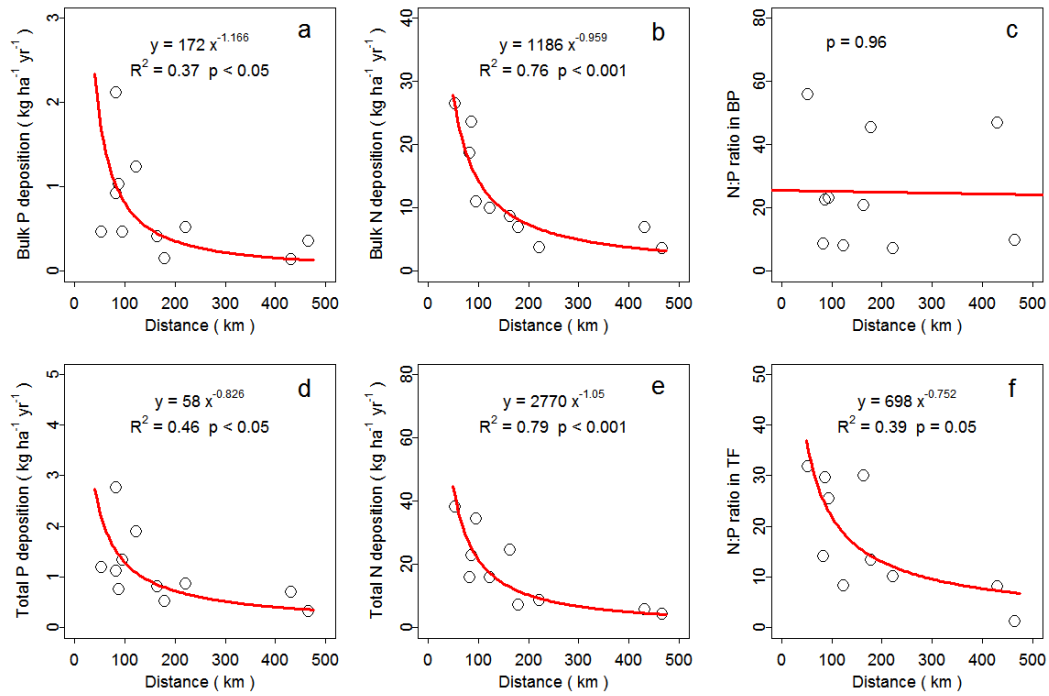
Figure 5. Changes in bulk deposition of P and N (kg ha$^{-1}$ yr$^{-1}$), total deposition of P and N (kg ha$^{-1}$ yr$^{-1}$), and N:P ratios in bulk precipitation (BP) and throughfall (TF) with the distance to the nearest large cities based on datasets of forested sites nearby semiarid regions in China.