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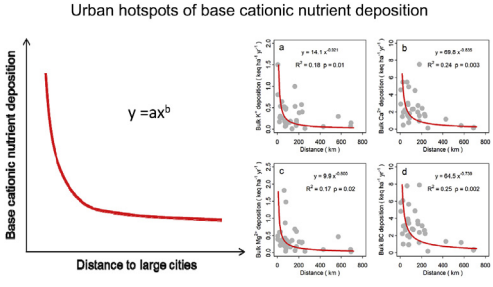
Bulk deposition of base cationic nutrients in China's forests: Annual rates and spatial characteristics

Enzai Du[a,b,*], Wim de Vries[c,d], Steven McNulty[e], Mark E. Fenn[f]

[a] State Key Laboratory of Earth Surface Processes and Resource Ecology, Faculty of Geographical Science, Beijing Normal University, Beijing, 100875, China
[b] School of Natural Resources, Faculty of Geographical Science, Beijing Normal University, Beijing, 100875, China
[c] Environmental Systems Analysis Group, Wageningen University, PO Box 47, 6700 AA Wageningen, The Netherlands
[d] Alterra, Wageningen University and Research Center, PO Box 47, 6700 AA Wageningen, The Netherlands
[e] Southern Research Station, USDA Forest Service, Raleigh, NC 27606, USA
[f] Pacific Southwest Research Station, USDA Forest Service, Riverside, CA 92507, USA

GRAPHICAL ABSTRACT



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ABSTRACT

Base cations, such as potassium (K^+), calcium (Ca^{2+}) and magnesium (Mg^{2+}), are essential nutrients for plant growth and their atmospheric inputs can buffer the effect of acid deposition by nitrogen (N) and sulphur (S) compounds. However, the spatial variation in atmospheric deposition of these base cationic nutrients is less understood compared with N and S deposition. By synthesizing bulk deposition data for K^+, Ca^{2+} and Mg^{2+}, we assessed their annual rates and spatial characteristics at 34 forested sites across China. Our synthesis showed relatively high levels of bulk deposition of base cationic nutrients in China's forests, being an order of magnitude higher than in the USA and Europe. On average, K^+, Ca^{2+} and Mg^{2+} accounted for 13%, 72% and 15% of the bulk deposition of base cationic nutrients, respectively. Surprisingly, base cation deposition was lower at sites near semi-arid regions compared with sites in eastern and southern China, which were far from semi-arid regions. Moreover, elevated base cation deposition was associated with urban hotspots, exhibiting a significant power-law increase with closer distance to the nearest large cities. We estimated that on average base cationic nutrients neutralized a significant proportion (76%) of the potential acid load due to acid deposition. Our findings suggest that in China there is considerable anthropogenic alteration of the regional cycling of base cationic nutrients, which plays an important role in counteracting the risk of soil acidification and base cation depletion in forest ecosystems, especially in the southern regions.

* Corresponding author. State Key Laboratory of Earth Surface Processes and Resource Ecology, Faculty of Geographical Science, Beijing Normal University, Beijing, 100875. China.
E-mail address: enzaidu@bnu.edu.cn (E. Du).

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1. Introduction

Base cations, such as potassium (K^+), calcium (Ca^{2+}) and magnesium (Mg^{2+}), are essential nutrients for plant growth (Mengel et al., 2001). Atmospheric deposition of these base cationic nutrients plays an important role in counteracting soil acidification and nutrient imbalance by replenishing the base cation pool (Draaijers et al., 1997; Larssen and Carmichael, 2000; Watmough et al., 2014; Fenn et al., 2015; Zhu et al., 2016). However, the spatial pattern of base cation deposition on regional and global scales is less understood than nitrogen (N) and sulphur (S) deposition (Dentener et al., 2006; Lamarque et al., 2013; Vet et al., 2014). As a result of rapid industrial and urban development since the 1980s, anthropogenic emissions of acid precursors (NO_x and SO_2) have led to a substantial increase in acid deposition in China and increased concerns about the potential impacts on ecosystem health and function (Larssen et al., 2006; Zhao et al., 2009; Liu et al., 2011; Du et al., 2017). Although previous studies have highlighted the importance of Ca^{2+} deposition in China (Chang et al., 1996; Larssen and Carmichael, 2000), atmospheric deposition of K^+ and Mg^{2+} is relatively unknown. A better understanding of the spatial variation of base cation deposition in China's forests is relevant for risk assessment of soil acidification and nutrient imbalance in view of elevated acid deposition.

Atmospheric particulates of base cations are derived both from natural and anthropogenic sources. Natural sources originate mainly from wind erosion of arid soils and sea salt aerosols (Vet et al., 2014). Anthropogenic emissions are associated with industrial (e.g., cement production, and combustion-induced fly ash), agricultural (e.g., wind erosion of arable land, agricultural tillage practices, and crop residue burning), construction (e.g., construction of buildings and roads) and traffic (e.g., vehicle emissions and traffic on unpaved roads) activities (Draaijers et al., 1997; Lee et al., 1999; Tørseth et al., 2012; Brahney et al., 2013; Vet et al., 2014). However, such anthropogenic sources were not included in previous modelling assessments in China, and as a result the models predicted higher levels of base cation deposition in arid and semi-arid regions, by accounting for wind-blown dust particles, compared with southern and eastern China (Chang et al., 1996; Larssen and Carmichael, 2000). Therefore, base cation deposition has probably been underestimated by these modelling assessments, especially in regions with considerable contribution of anthropogenic sources.

Previous studies have indicated the occurrence of urban acid islands in China due to elevated S and N position as a result of elevated anthropogenic SO_2 and NO_x emissions in and nearby urban areas (Du et al., 2015). In recent decades, rapid industrial, agricultural and urban development have, however, also increased the anthropogenic emissions of mineral particulates particularly in and nearby large cities (Yang et al., 2011; Zhang et al., 2012). Specifically, concentrations of base cations (e.g., K^+, Ca^{2+}, and Mg^{2+}) in atmospheric aerosols have been observed to be higher at urban sites in comparison with rural sites (Zhang et al., 2012). Due to high levels of anthropogenic emissions and alteration in precipitation regime (e.g., increasing rainless days), aerosol pollution frequently occurred during recent decades, especially in developed regions in eastern and southern China (Rohde and Muller, 2015; Zhang et al., 2015; Li et al., 2016). Base cations in atmospheric aerosols can be transported over long distances, resulting in a decrease of base cation deposition from the emission sources to remote regions (Zhang et al., 2012). We thus expect higher atmospheric deposition of alkaline base cationic nutrients near large cities, which may substantially reduce the risk of urban acid islands and alter the regional cycling of base cationic nutrients.

Forest covers more than one-fifth of the national land area in China and provides fundamental ecosystem services (State Forestry Administration, http://english.forestry.gov.cn). Base cations such as K^+, Ca^{2+} and Mg^{2+} are important nutrients for forest ecosystems, particularly in the context of high-level acid deposition in China. Although base cations are not reported routinely by monitoring networks in China (e.g., Nationwide Nitrogen Deposition Monitoring Network, Xu et al., 2015; Chinese Ecosystem Research Network; Zhu et al., 2015), observational studies on base cation deposition are emerging at individual sites in recent years (See Table S1). The present study synthesized bulk deposition data for base cations (K^+, Ca^{2+} and Mg^{2+}) from published literature to assess the spatial pattern of base cation deposition in China's forests. Specifically, we tested the hypothesis of urban hotspots of base cation deposition and the declining spatial trend in base cation deposition from arid and semi-arid regions. Moreover, we evaluated the role of base cations in neutralizing the potential acid load due to N and S deposition.

2. Data and method

2.1. Data sets

By conducting a survey of the online library of China National Knowledge Infrastructure (http://www.cnki.net/) and ISI Web of Science (http://isiknowledge.com), we collected data from published literature on bulk deposition of K^+, Ca^{2+}, Mg^{2+} from 2001-2015 in China's forests, together with information on site location (latitude and longitude) and forest type. Samples of bulk precipitation were collected in forest environments using buckets that were continuously open. Quality control of chemical analysis was based on the electrical conductivity and ion balance method. Reported data without quality control information were excluded from our database. We only included data on bulk deposition of base cationic nutrients when K^+, Ca^{2+} and Mg^{2+} measurements were conducted simultaneously. Furthermore, we recorded total inorganic N (as the sum of NO_3^- and NH_4^+) and inorganic S (sulphate) concentrations if reported simultaneously to estimate the potential acid load induced by S and N compounds and compared this with the neutralizing capacity of base cationic nutrient deposition (see section 2.2).

Observational data 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 bulk deposition at one site was measured for more than one forest stand or available for more than one year, a volume-weighted mean based on bulk precipitation 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). Overall, our database included bulk deposition of K^+, Ca^{2+} and Mg^{2+} at 34 sites, which were evenly distributed across the main forested regions in China (Fig. 1). Observational data on both total inorganic N and sulphate were available at 16 sites (see Table S1 for detailed information).

2.2. Statistical analysis

Bulk deposition of K^+, Ca^{2+}, Mg^{2+}, inorganic N and S were calculated according to the volume-weighted mean concentration and annual bulk precipitation. We conducted Student's t-test to examine the difference of bulk base cation deposition between sites near the semi-arid regions (n = 14) and far from the semi-arid regions (n = 20), which were classified by using a criterion of a distance of approximately 400 km to the nearest edge of the semi-arid regions (e.g., grassland). We used the criterion of 400 km to the source regions because a previous analysis of phosphorus deposition suggests that bulk deposition showed no change with further distance from the sources (Du et al., 2016).

To test the urban hotspot hypothesis, we used a power-law model (Du et al., 2014, 2015 & 2016) to explore changes in base cation deposition with the distance between the sampling site and the nearest large city. The potential acid load of N and S compounds (n = 16), indicating the potential for soil acidification due to atmospheric

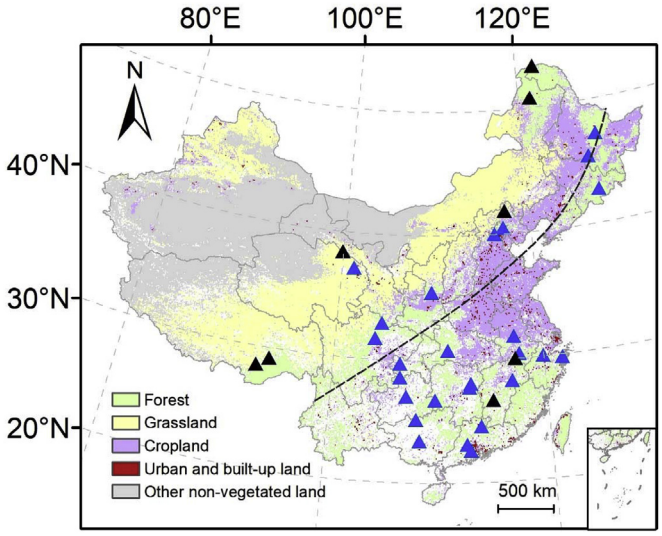

Fig. 1. Locations of 34 observed sites in this study. Black triangles indicate sites (n = 8) with a distance > 200 km to nearest large cities, while blue triangles indicate sites (n = 26) with a distance < 200 km to nearest large cities. The background shadows in light green, light yellow, light purple and light grey indicate the distributions of forest, grassland (semiarid region), cropland and non-vegetated land (mainly arid deserts), respectively. The 14 sites in the northwest of the dashed grey line have a distance approximately less than 400 km to the nearest semi-arid regions (grassland), while the other 20 sites to the southeast of the dashed line were far from the semi-arid regions. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

deposition of these compounds, was estimated as an equivalent sum of nitrate, ammonium and sulphate (Draaijers et al., 1997). The overall role of base cationic nutrients in neutralizing deposition-induced soil acidification was then estimated by using a standardized major axis regression with potential acid load induced by S and N deposition. We also used Student's t-test to compare bulk base cation deposition across forest types (deciduous broadleaved forest, deciduous conifer forest, evergreen broadleaved forest, evergreen conifer forest and mixed forest). Spatial patterns of base cationic nutrient deposition were illustrated using ArcGIS Desktop (version 9.3, ESRI, USA). Data ranges were indicated by the 10th and 90th percentage. All statistical analysis was performed using R software (version 3.4.0; R Development Core Team, 2017; http://www.r-project.org/) with a significance level of $p < 0.05$.

3. Results

3.1. Base cationic nutrient deposition in China's forest

Mean bulk deposition of K^+, Ca^{2+} and Mg^{2+} was 0.34 (0.07–0.63, 10th-90th percentile), 2.26 (0.44–4.82) and 0.39 (0.08–0.67) keq ha^{-1} yr^{-1}, respectively (Fig. 2). The contribution of K^+, Ca^{2+} and Mg^{2+} to the sum of base cationic nutrient fluxes in bulk deposition (2.98, 0.93–5.79 keq ha^{-1} yr^{-1}) was on average 13% (4%-26%), 72% (50%-88%) and 15% (5%-25%), respectively. Bulk deposition of K^+, Ca^{2+}, Mg^{2+} and the sum of base cationic nutrients showed similar spatial patterns (Fig. 3). In contrast to our hypothesis, bulk deposition of K^+ (Student's t-test, $df = 32$, $p = 0.09$), Ca^{2+} ($df = 32$, $p = 0.01$), Mg^{2+} ($df = 32$, $p < 0.05$) and sum of base cationic nutrients ($df = 32$, $p < 0.01$) all showed lower levels at sites (n = 14) near semi-arid regions compared with the sites (n = 20) far from the semi-arid regions (Fig. 4). Moreover, we found no significant difference in bulk deposition of base cationic nutrients between forest types, except significantly lower bulk Ca^{2+} deposition in deciduous conifer forest than deciduous broadleaved forest ($p < 0.05$) (see Table S2).

3.2. Urban hotspots of base cationic nutrient deposition

Consistent with the urban hotspot hypothesis, bulk deposition of K^+ ($R^2 = 0.18$, $p = 0.01$), Ca^{2+} ($R^2 = 0.24$, $p = 0.003$), Mg^{2+} ($R^2 = 0.17$, $p = 0.02$) and the sum of base cationic nutrients ($R^2 = 0.25$, $p = 0.002$) all showed a significant power-law increase with closer distance to the nearest large cities (Fig. 5). Based on the observational bulk deposition of N, S and base cationic nutrients available from 16 sites, we estimated the role of base cationic nutrient deposition in neutralizing potential acid load due to atmospheric deposition of N and S compounds. Our analysis indicates that base cationic nutrients on average neutralized 76% ($R^2 = 0.75$, $p < 0.001$) of the potential acid load due to acid deposition (Fig. 6).

4. Discussion

4.1. High rates of base cationic nutrient deposition in China's forests

Our synthesis showed relatively high levels of bulk deposition of K^+, Ca^{2+} and Mg^{2+} in China's forests, which are an order of magnitude higher than the averages (K^+, 0.34 *vs.* 0.02 keq ha^{-1} yr^{-1}; Ca^{2+}, 2.26 *vs.* 0.10 keq ha^{-1} yr^{-1}; Mg^{2+}, 0.39 *vs.* 0.05 keq ha^{-1} yr^{-1}) of 21 forested sites across Europe, the United States and Canada (Watmough et al., 2005). We also found that Ca^{2+} dominated base cationic nutrient flux in bulk deposition, being in line with the results of previous studies (Watmough et al., 2005; Vet et al., 2014). The higher Ca^{2+} deposition was likely due to the fact that both natural (e.g., wind-blown dust from arid and semiarid soils) and anthropogenic (e.g., cement production, wind erosion of arable land, construction of buildings and roads, and traffic on unpaved roads) sources have higher Ca concentrations relative to other base cations (Watmough et al., 2005; Zhang et al., 2012). Different from previous modelling results (e.g., Chang et al., 1996; Lee et al., 1999), our assessment of base cationic nutrient deposition in China's forests indicate lower levels at sites near semi-arid regions compared with sites in eastern and southern China, which were far from the semi-arid regions (Fig. 4). Modelling results for the 1980s and early 1990s predicted a decrease of base cation deposition with increasing distance from the arid and semi-arid regions due to wind-blown dust (Chang et al., 1996; Lee et al., 1999; Larssen and Carmichael, 2000; Rodhe et al., 2002). The difference between our results and these modelling-based assessments might be attributed to the fact that previous models failed to account for anthropogenic sources, whose contribution to base cation deposition was poorly understood.

Two causes are likely responsible for the higher levels of base cationic nutrient deposition in eastern and southern China compared with sites nearby semi-arid regions. First, recent recovery of degraded vegetation and reduction of wind speed in semi-arid regions may have resulted in a decrease in bulk base cation deposition from aeolian sources. In recent decades, the implementation of several ecological restoration projects (e.g., the Three-North Shelterbelt Forest Program, the Beijing–Tianjin Sand Source Control Program, the Natural Forest Conservation Program, and the Grain to Green Program) have effectively increased vegetation coverage and decreased wind speed in the arid and semi-arid regions (Tan and Li, 2015; Zhang et al., 2016). Compared with the 1980s and 1990s (Chang et al., 1996; Lee et al., 1999; Larssen and Carmichael, 2000), the contribution of wind-blown dust particulates from the arid and semi-arid regions to base cation deposition has likely decreased substantially due to lowered wind speed and increased soil protection by vegetation cover. Second, the rapid increase in anthropogenic emissions during urbanization and industrialization has likely contributed substantially to the bulk base cation deposition in developed regions of eastern and southern China (Rohde and Muller, 2015; Zhang et al., 2015; Li et al., 2016). For instance, recent analyses based on monitoring and satellite datasets have indicated high-level concentrations of atmospheric aerosols in eastern and southern China, especially in or near large cities (Henriksson et al.,

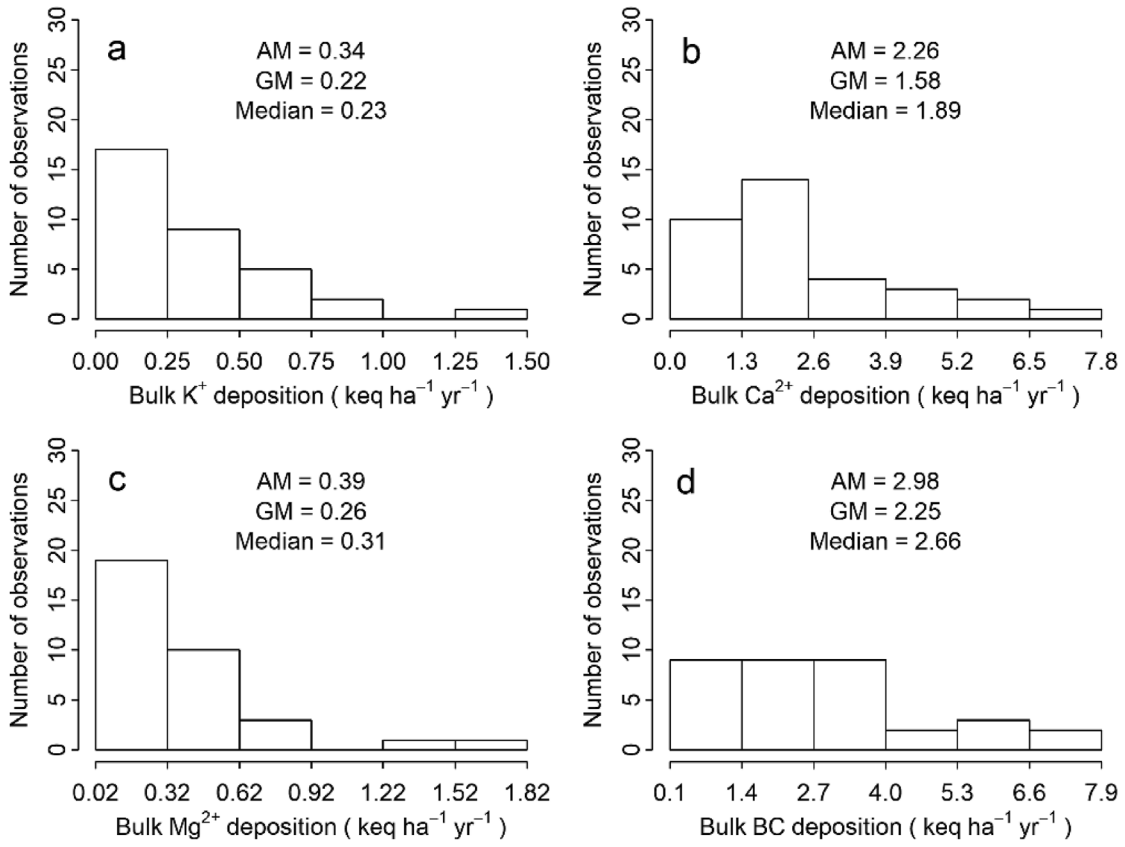

Fig. 2. Histograms showing the frequency distribution of bulk deposition of a) K^+, b) Ca^{2+}, c) Mg^{2+} and d) sum base cations (BC, calculated as the sum of K^+, Ca^{2+} and Mg^{2+}) in China's forests. AM is arithmetic mean; GM is geometric mean.

2011; Rohde and Muller, 2015). Considering that Ca^{2+}, K^+ and Mg^{2+} are a considerable component of atmospheric aerosols ((Yang et al., 2011; Zhang et al., 2012), elevated atmospheric deposition of base cationic nutrients thus occurred in eastern and southern China.

4.2. Urban hotspots of base cationic nutrient deposition

In line with our urban hotspot hypothesis, we found a significant power-law increase of base cation deposition with proximity to the nearest large cities (Fig. 5). This is consistent with the observational data that show elevated atmospheric aerosol base cation concentrations at urban sites in China (Zhang et al., 2012). By synthesizing observed data of acid deposition, a recent assessment indicates the occurrence of urban acid islands in China (Du et al., 2015). In this context, the regional urban hotspots of base cation deposition may partially alleviate the risk of soil acidification and the potential deficiency of base cationic nutrients for ecosystems in and near large cities. Our analysis indicates that base cations neutralized a major proportion of potential acid load due to bulk deposition (76%) in China's forests. This may explain why negative impacts experienced in Europe and North America associated with such high levels of acid deposition (Reuss and Johnson, 2012) have not been widely detected in China (Duan et al., 2016).

The urban hotspots of base cationic nutrient deposition are likely induced by intensive anthropogenic emissions. For instance, particulate matter emitted from the cement industry in China in 2009 was estimated to be 3.60 million tons, which accounted for approximately 26.2% of the total national particulate emissions (Chen et al., 2015). The density of road networks and the intensity of traffic activities are generally higher in and nearby large cities, especially for the eastern and southern regions of China (Li et al., 2010). By measuring atmospheric aerosol compositions at 16 sites in typical regions of China, Zhang et al. (2012) found higher concentrations of Ca^{2+}, K^+ and Mg^{2+} in PM_{10} at urban sites than rural sites. Field measurements also indicate that concentrations of base cations were considerable in $PM_{2.5}$ aerosols in megacities across China (Yang et al., 2011). These base cationic nutrients in atmospheric aerosols can be transported from sources in and nearby large cities over long distances, and deposit to natural ecosystems via wet or dry pathways, resulting in urban hotpots of base cation deposition.

4.3. Uncertainties of the current analysis

Several uncertainties remain in our assessment of base cation deposition in China's forests. Unlike SO_2 and NO_x, base cations in the atmosphere occur primarily in the form of particulates and none has significant gaseous precursors (Chang et al., 1996; Vet et al., 2014). Consequently, bulk deposition, consisting of wet deposition and a proportion of dry deposition, may substantially underestimate the total deposition of base cations (Chantara and Chunsuk, 2008). However, the assessment of dry base cation deposition, which requires routine long-term measurements of size-resolved, ion-specific, particle concentrations in air coupled with an inferential size-resolved dry deposition velocity model, is lacking regionally or globally (Vet et al., 2014). Thus, our study has been limited to field measured bulk deposition of base cationic nutrients. Our analysis did not account for the role of particulates in bulk deposition, the amount of which could be considerable, especially for particulate Ca (Avila et al., 1998). Moreover, our analysis of bulk deposition of base cationic nutrients ignores inter-annual variations within the period 2001-2015, because the published datasets of base cationic nutrient deposition in China's forests are seldom longer than two years and do thus not allow to evaluate inter-annual variation.

Despite the difficulty of direct measurement of total deposition,

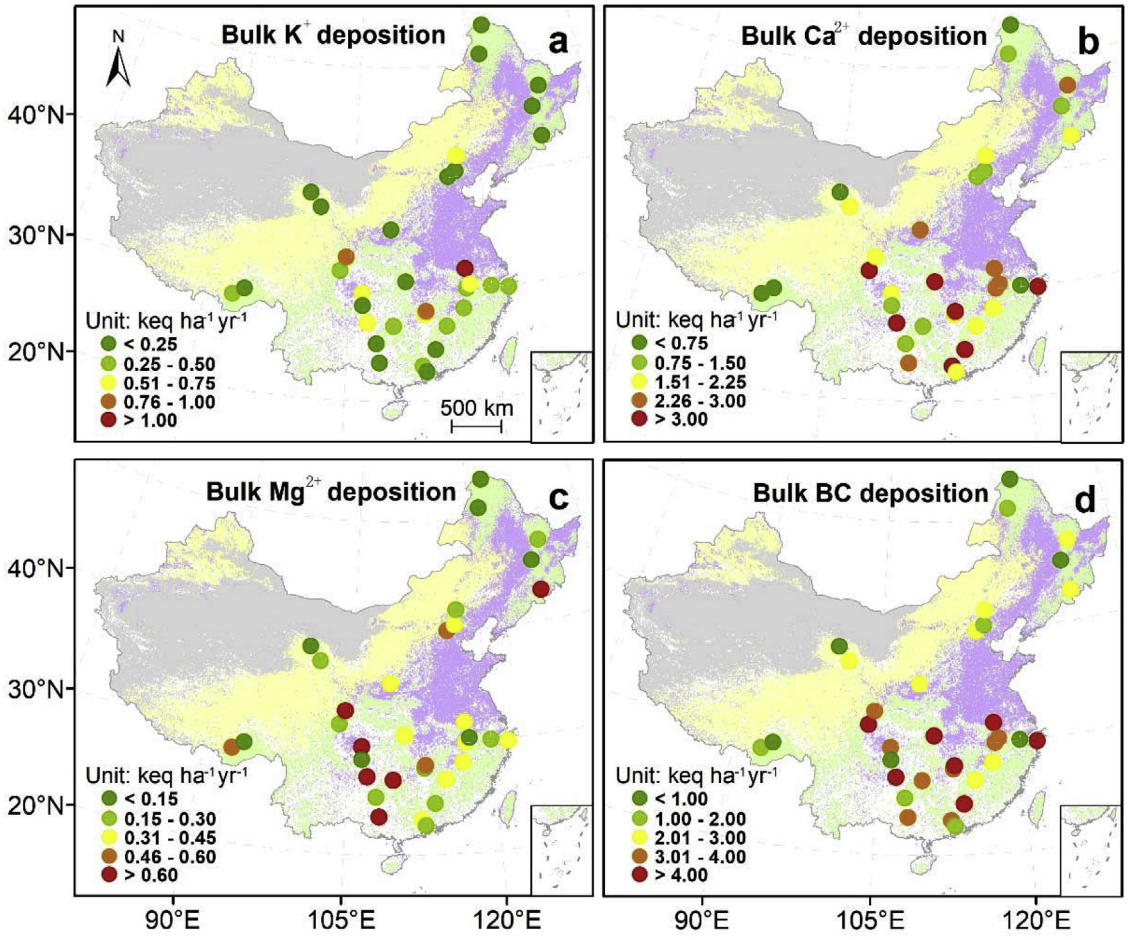

Fig. 3. Spatial patterns of bulk deposition of K^+, Ca^{2+}, Mg^{2+} and the sum of base cations (BC, calculated as the sum of K^+, Ca^{2+} and Mg^{2+}) in China's forests. The background shadows in light green, light yellow, light purple and light grey indicate the distributions of forest, grassland (semi-arid region), cropland and non-vegetated land (mainly deserts), respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

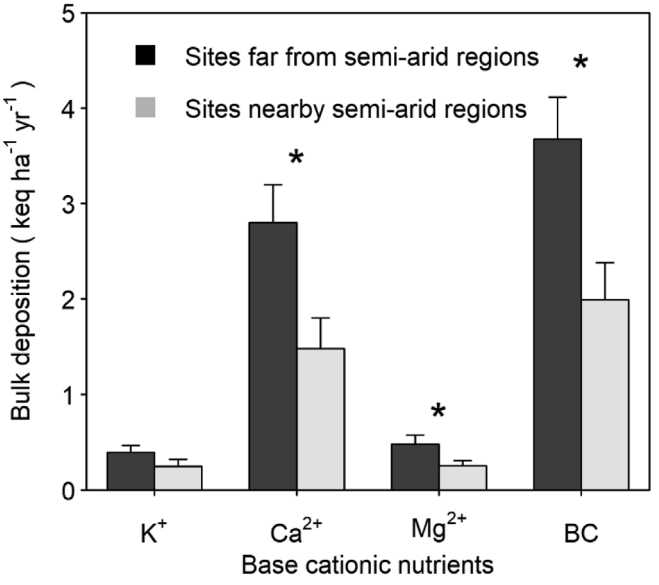

Fig. 4. Mean bulk deposition of K^+, Ca^{2+}, Mg^{2+} and sum of base cations (BC, calculated as the sum of K^+, Ca^{2+} and Mg^{2+}) at forested sites nearby (< 400 km, n = 14) and far from (> 400 km, n = 20) semi-arid regions. Error bars indicate the standard error of the mean. The symbol * indicates a significant difference of $p < 0.05$ (Student's t-test).

several inferential methods have been proposed to estimate total deposition in forest ecosystems. For instance, sodium (Na^+) has been used a tracer to estimate total deposition because it is usually assumed to be neither leached from nor taken up by the forest canopy (see more details in Ulrich, 1983; Draaijers and Erisman, 1995). By using Na^+ in throughfall, stemflow and bulk deposition as a tracer, we roughly estimated that total base cation deposition was on average 1.48 (n = 12) times of bulk deposition (unpublished data). However, this result is based on very few data and further measurement and modelling efforts are needed to provide a more accurate understanding of total base cation deposition in China. This is crucial for assessing the counteracting effects of base cation deposition on soil acidification, which may differ in bulk deposition from total deposition. However, the ratio of Na^+ in throughfall and stemflow compared to bulk deposition was not significantly different from the same ratio for S (on average 1.64, n = 18) and for N (on average 1.33, n = 29) (unpublished data), where the latter lower value may also be influenced by foliar N uptake (Sparks, 2009).

Our analysis assessed the effect of the dust sources by comparing bulk deposition at the sites near the semi-arid regions with that at sites far from the semi-arid regions. We did so because in practice it is difficult to quantify the distance to dust sources from arid (e.g., deserts) and semi-arid (e.g., grasslands) regions. Specifically, deserts and grasslands spread widely in northwest China and the distribution of both land cover types are in an extremely irregular shape (Fig. 1). The geographical centres of deserts and grasslands are hard to define, and

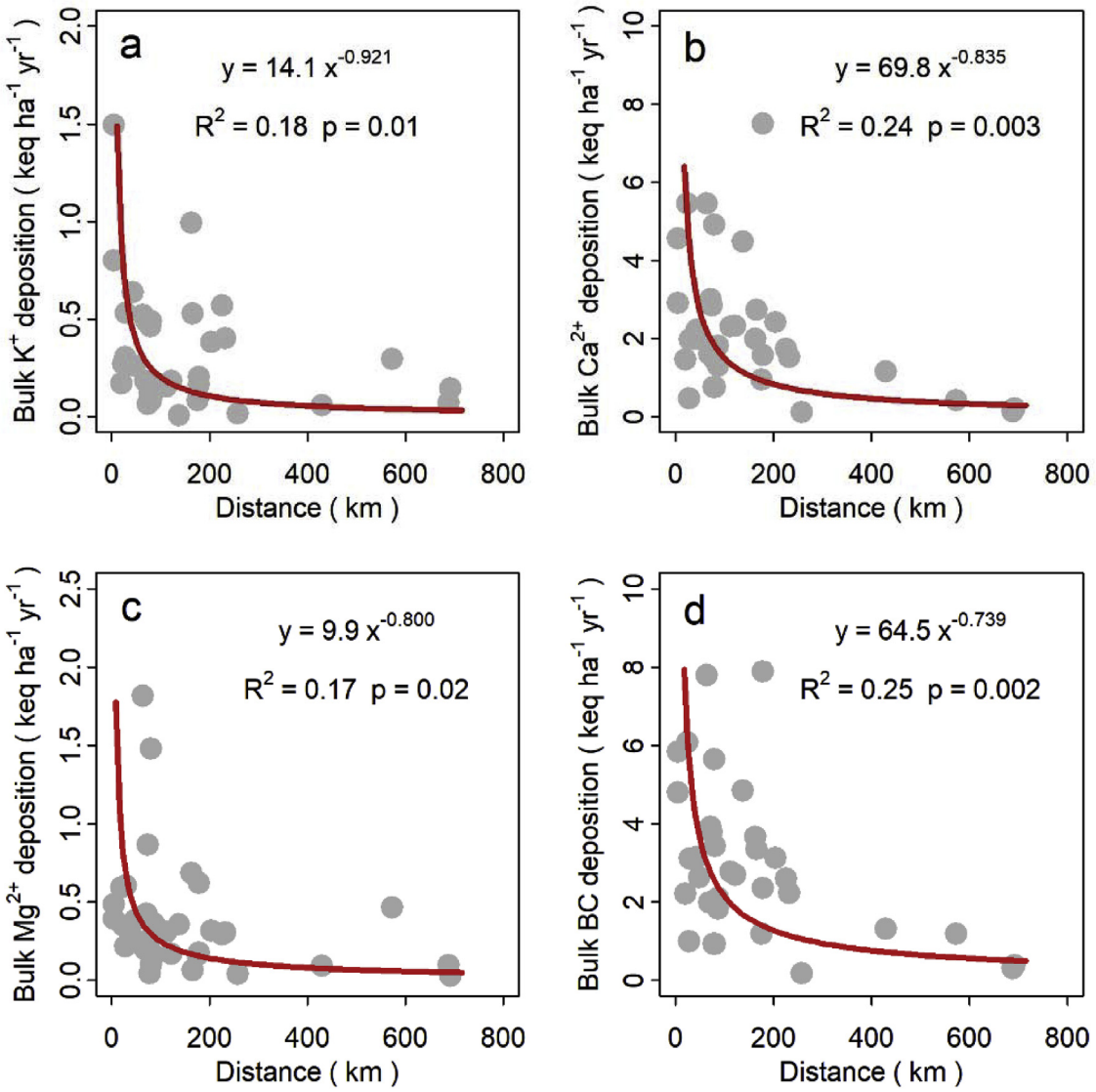

Fig. 5. Changes in bulk deposition of K^+, Ca^{2+}, Mg^{2+} and the sum of base cations (BC, calculated as the sum of K^+, Ca^{2+} and Mg^{2+}) with distance to the nearest large cities.

the distances to both centres are likely not able to indicate the effect of dust sources. The criterion distance of 400 km to the semiarid region is evidenced to be appropriate by our analysis of the changes in bulk base cation deposition with distance to urban sources (Fig. 5). Moreover, it should be noted that the spatial pattern of lower levels of bulk deposition at sites near semi-arid regions than at sites in eastern and southern China was only valid for base cations. If the contribution of particulates (mainly Ca) is accounted for, the spatial pattern will likely differ.

Uncertainties also remain in quantifying the urban hotspots of base cation deposition. For instance, larger cities with a more extensive spatial extent of anthropogenic emissions would result in larger urban hotpots of base cation deposition. The spatial extent of the urban hotspots may also be influenced by prevailing wind direction and wind velocity. Specifically, the amount of anthropogenic base cations that is transported from urban hotpots to forests is likely larger in the downwind direction than in the upwind direction. The effect of wind direction has been evidenced for NO_x based on satellite data (Beirle et al., 2011). Further efforts are needed to quantify the spatial shape and extent of urban hotspots by integrating regional transport modelling of base cation deposition with more detailed datasets on the factors as discussed above.

Assessing the status of base cationic nutrient deposition in different forest types is crucial to evaluate the corresponding ecological effects. Based on the current database, we found no significant difference in bulk deposition of base cationic nutrients between most forest types (see Table S2). This is most likely because the data are for bulk deposition at forested sites and there is only a difference between forest types when they are distributed in distinct geographic regions of the country. The relatively few observational sites for each forest type may also limit the statistical power of cross-type comparison. Therefore, future monitoring and modelling efforts are needed to provide a more precise mapping of bulk deposition of base cationic nutrients in China's forests.

4.4. Implications for nutrient balance and soil acidification

As driven by long-term acid deposition, progressive depletion of exchangeable base cations in sensitive soils may result in a deficiency of base cationic nutrients in forest ecosystems, causing forests to rely increasingly on base cationic nutrients (especially Ca^{2+}) from atmospheric deposition (e.g., Fernandez et al., 2003; Bowman et al., 2008; Hynicka et al., 2016). A previous assessment on mineral elements in plant leaves demonstrate significant latitudinal trends in leaf contents

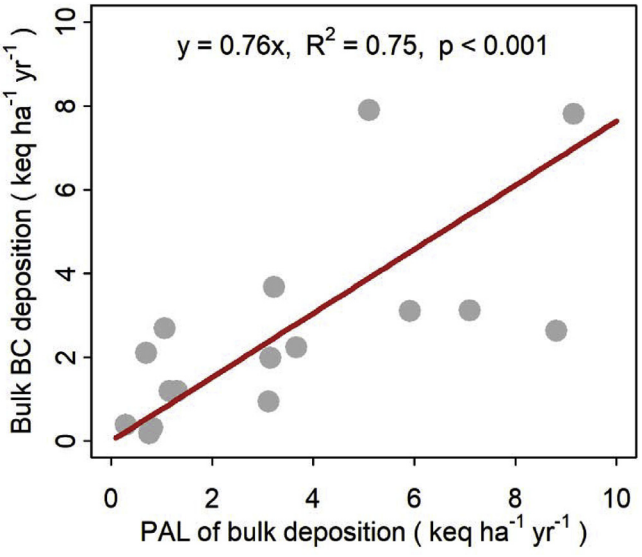

Fig. 6. Relationships between sum base cationic nutrient (BC) deposition and potential acid load (PAL) due to sulphur and nitrogen deposition (n = 16). The regression line has been forced through the origin, and thus the slope indicates the capacity of base cationic nutrient deposition to counteract the potential acid load due to sulphur and nitrogen deposition.

of K, Ca and Mg, generally decreasing from north to south in China (Han et al., 2011). They also demonstrated positive correlations between plant leaf and soil contents of K, Ca and Mg, while leaf contents of K, Ca and Mg were negatively correlated with soil acidity. Higher levels of acid deposition in southern China (Du et al., 2015) may have partially contributed to this latitudinal trend of K, Ca and Mg by leaching soil base cations. Our results indicate that atmospheric inputs can be a quantitatively important source of base cationic nutrients in soils, which may partially alleviate the risk of soil acidification and the potential base cation deficiency in southern China. In this context, the role of base cationic nutrients should be considered in future assessments of forest health and function.

5. Conclusions

By synthesizing observational data on bulk deposition of base cationic nutrients (K^+, Ca^{2+} and Mg^{2+}), our synthesis showed that bulk deposition of base cationic nutrients was relatively high in China's forests compared with those in Europe and North America. Contrary to previous observations (Chang et al., 1996; Larssen and Carmichael, 2000), base cation deposition was higher in eastern and southern China compared with sites near arid and semi-arid regions. In line with the urban hotspot hypothesis, we found a significant power-law increase of base cation deposition with increasing proximity to the nearest large cities, implying that anthropogenic emissions have altered the regional cycling of base cationic nutrients in China. Our analysis also indicates that base cationic nutrient deposition neutralized a major proportion of potential acid load due to acid deposition, suggesting an important role in counteracting the risk of soil acidification, especially in southern China.

Because of increasing vegetation coverage and stricter control of anthropogenic particulate emissions in China (Wang and Hao, 2012; Zhang et al., 2016), we can expect a reduction of base cation deposition in the near future, which follows the trend in Europe since the 1980s (Tørseth et al., 2012). As a result of ongoing elevated N deposition, base cation (mainly Ca^{2+}) depletion might be aggravated in subtropical and tropical forests in southern China (Bowman et al., 2008; Hynicka et al., 2016). Routine measurements and reporting of base cations are thus recommended to be included in existing monitoring networks of atmospheric deposition in China (e.g., Nationwide Nitrogen Deposition Monitoring Network, Xu et al., 2015; Chinese Ecosystem Research Network; Zhu et al., 2015). In conjunction with monitoring results, future modelling efforts are needed to provide a more spatially extensive mapping of total base cation deposition in China, which is important for assessment of critical acid loads and forest health in the context changing acid deposition.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2018.04.042.

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