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

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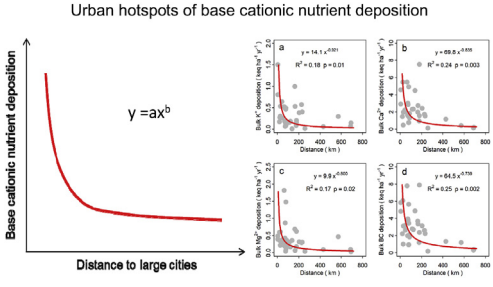
GRAPHICAL ABSTRACT



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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.

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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