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Global atmospheric sulfur deposition and associated impaction on nitrogen cycling in ecosystems

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ABSTRACT

S has become the dominant contributor of acidic deposition globally, so we conducted a global overview on estimations of wet and dry S deposition patterns and then further analyzed coupling interactions between S and N cycling in this study. Results show that global average S wet deposition rate can reach $141.64 \pm 120.04\ TgS \cdot a(year)^{-1}$, wherein the average S wet deposition rate of $84.5 \pm 79.31\ TgS \cdot a^{-1}$ in Asia could result in a potential risk for soil acidification, contributing 59.7% to global wet S deposition. In comparison, the average S wet deposition flux in Europe is $18.51 \pm 16.06\ kgS \cdot ha^{-1} \cdot a^{-1}$, but North America and South America only have 8.16 ± 4.12 and $4.96 \pm 3.45\ kgS \cdot ha^{-1} \cdot a^{-1}$, respectively. Moreover, the average S wet deposition rate in Europe and North America is only $18.81 \pm 16.32\ TgS \cdot a^{-1}$ and $14.56 \pm 7.36\ TgS \cdot a^{-1}$, respectively. Global average dry S deposition rate is estimated at $80.1 \pm 69.37\ TgS \cdot a^{-1}$, wherein Asia contributes 58.3% of total global dry S deposition. However, the dry S deposition in Africa has reached $9.82 \pm 6.08\ TgS \cdot a^{-1}$, and its contribution exceeds that of both Europe and South America. In addition, the elevated S emissions exhibit biochemical and physicochemical reactions with N under different interfaces. Therefore, the increases in N and S emissions will be important for future acidification levels. The coupling relationships of N and S under different interfaces will potentially reveal any new scientific issues that may arise and provide a better understanding of S and N cycling. Thus, distinguishing between N and S deposition, their sources, and associative critical loads could provide new insight into environmental effects and acid deposition estimations of greenhouse gases.

1. Introduction

Sulfur (S) and nitrogen (N) are released by natural processes and anthropogenic activities, such as the combustion of fossil fuels, the refinement and smelting of sulfide ores, as well as mining and other industrial processes (Hsu et al., 2016). In addition, phytomass combustion also increases significant S and N emissions (Mardoyan and Braun, 2014; Maroušek et al., 2015a). Both S and N deposition will result in soil, freshwater, and marine ecosystem acidification (Longhurst, 1991; Hao et al., 2017; Gao et al., 2017a), making forests and other ecosystems more vulnerable to stress, such as frost, drought, and pests (Bouwman et al., 2002; Vivanco et al., 2017). Accordingly, S and N deposition has become an important source of chemical elements for vegetation via stomatal uptake in forest ecosystems (Fowler et al., 1989).

However, increases in nitrogen dioxide, ammonia, and sulfur dioxide emissions in the twentieth century have led to excessive S and N deposition, which have had effects on terrestrial and aquatic ecosystems (Bergstorm and Jansson, 2006; Gao et al., 2014, 2017b). The dry and wet deposition are the primary mechanisms of S and N deposition to ecosystems. The dry deposition primarily occurs through gaseous compounds (sulfur dioxide, nitric acid, ammonia,

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and nitrogen dioxide) in conjunction with aerosols. The wet deposition occurs by incorporating aerosol particles. It is transported in the form of cloud condensation nuclei and below-cloud scavenging, falling to the ground as precipitation as well as the below-cloud and in-cloud scavenging of soluble gases (Dore et al., 2015; Yang et al., 2015, 2016). The soluble gas species of S and N can be dissolved into falling raindrops, while airborne particles can be collected by raindrops through collisions (Vivanco et al., 2017; Yang et al., 2017).

SO_x are predominantly caused by power generation from elevated point sources, which is strongly impacted by the speed of vertical diffusion and other meteorological conditions (Dore et al., 2015). In addition, vegetation stomatal uptake in forest ecosystem is an important pathway for S deposition to vegetation (Hsu et al., 2016). Although reducing SO_2 emissions to the atmosphere has been arisen to international legislation, such as the European Union National Emissions Ceiling Directive and the United Nations Economic Commission for Europe Gothenburg Protocol, in most of developing country S emission still continue to rapidly increase due to coal used as main fuel and large of power generating plants. Therefore, how to deal with waste from those plants and apply engineering measures to alleviate soil and water acidification, it would become sustainably research topic in the future.

Ecological impacts of S and N deposition have been extensively reported in Asia, Europe, and North America (Hsu et al., 2016; Duan et al., 2016a). In China, the S and N deposition flux has been estimated at approximately 12–160 $kgS \cdot ha^{-1} \cdot a^{-1}$ (Larssen et al., 2006; Duan et al., 2016b) and at approximately 0–29.5 $kgN \cdot ha^{-1} \cdot a^{-1}$ (Liu et al., 2013; Zhu et al., 2015; Gao et al., 2016; Jia and Gao, 2017), respectively, which are close to acid deposition peaks observed in Europe and North America in the 1980s (Larssen et al., 2006; Liu et al., 2013). Vet et al. (2014) have shown that sulfate has been the dominant contributor to acidic deposition in most regions, although nitrate has also been an important source in many areas.

After deposition, substantial resultant reductions in sulfur dioxide emissions have subsequently influenced an increase in eutrophication in different ecosystems due to N deposition from both oxidized N and reduced N (Vogt et al., 2013). Additionally, the sulfuric acid is also considered to have a potential effect on climate change given that it can reduce sunshine by acting as cloud condensation nuclei. Therefore, understanding mechanisms of S and N deposition and their interactive effects are critical not only for issues related to acid deposition on a continental scale but also for climate change on a global scale (Ohizumi et al., 2016).

In this study, we think global S deposition exhibit significant spatial pattern accompanied with rapid economic development, and also impact on N process in ecosystem. Therefore, the objective of this study was to evaluate the spatial pattern of the wet and dry S deposition and associative contributions and patterns for different continents by a global overview and meta-analysis of wet and dry S deposition, while further synthesizing current knowledge on the coupling of S the N cycling to disclose how the S deposition impact on different N process in ecosystem.

2. Methods

2.1. Data sources extraction

In this study, we determined that there was a change in data relating to wet and dry S deposition from the 1990s to the 2010s. We selected study sites that had three years of continuous wet S deposition monitoring data and with at least one to two years of monitoring data for dry S deposition. As a result, this study had greater than 150 sites from which to monitor wet and dry S deposition to analyze global interannual S deposition variability (Fig. 1). See the Supplementary Material for more detailed information.

2.2. Dry and wet sulfur deposition calculation

All sites were classified into Asia, Europe, North America, South America, Africa, and Oceania, respectively. Firstly, we need to estimate the annual average wet and dry deposition flux for different continents, which show as mean ± standard deviation by statistics method on the whole continent monitor sites. Secondly, we calculated average annual wet and dry S deposition rate for different continents using the following equation:

$$S_{de} = F \times A \quad (1)$$

where S_{de} is the annual average wet or dry S deposition rate; A is the total area of Asia, Europe, North America, South America, Africa, and Oceania, respectively; and F is the average annual wet and dry deposition flux.

It is difficult to make direct measurements of dry deposition due to the requirements for highly sophisticated methods and instrumentation (Wesely and Hicks, 2000; Vet et al., 2014). Therefore, we only had a limited amount of monitoring sites to estimate dry deposition in this study, and we had to extract most dry S deposition and a proportion of wet S deposition data directly from references in Supplementary Materials.

Differences on annual average wet and dry deposition flux were tested using one-way analyses of variance (ANOVA). The differences with P values < 0.05 were considered significant. All statistical analyses were conducted by SPSS software (Chicago, IL, USA); Graphs on site distribution and S deposition pattern were drawn by using ERSI ArcGIS software (Version 10.1; Redlands, CA, USA).

3. Results and discussion

3.1. Spatial patterns of wet S deposition

Studies have long applied S deposition to deduce whether critical loads have exceeded aquatic and terrestrial thresholds; however, at the present time global S deposition patterns are primarily estimated by chemical transport models (Rodhe et al., 2002; Bouwman et al., 2002; Dentener et al., 2006). Historically, S deposition is regionally variable, and amounts and sources of S emissions have been altering since the Industrial Revolution (Ishida et al., 2015). Globally, Vet et al. (2014) systematically estimated that East Asia produced the highest S deposition, particularly in Southeast China (Duan et al., 2016b), Northeast India, and Bangladesh. This is followed by Central Europe and the northeastern region of North America.

As Fig. 2 shows, East Asia, Central Europe, North Africa, and Eastern China are areas of high wet S deposition. The patterns of high spatial variability in S deposition are likely the result of the long-distance transport of aerosols composed of sulfate (SO_4^{2-}) and base cations or the formation of such aerosols during atmospheric transport (Fenn et al., 2015). Asia remains a region of high wet S deposition flux, ranging from 26 $kgS \cdot ha^{-1} \cdot a^{-1}$ to 40 $kgS \cdot ha^{-1} \cdot a^{-1}$, where there is always a potential risk for soil acidification due to the high S deposition in eastern and southern China and parts of Southeast Asia (Hicks et al., 2008). Over the last decade, it has been reported that the wet deposition is highest in China (>60 $kgS \cdot ha^{-1} \cdot a^{-1}$) (Duan et al., 2016a, 2016b), but in this study, we estimated that wet S deposition in most areas of China ranged from 22 $kgS \cdot ha^{-1} \cdot a^{-1}$ to 30 $kgS \cdot ha^{-1} \cdot a^{-1}$, with the exception of Western China (being lower than 20 $kgS \cdot ha^{-1} \cdot a^{-1}$) and North and East China (being higher than 30 $kgS \cdot ha^{-1} \cdot a^{-1}$) (Fig. 2). For South