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Trends of two decadal precipitation chemistry in a subtropical rainforest in East Asia

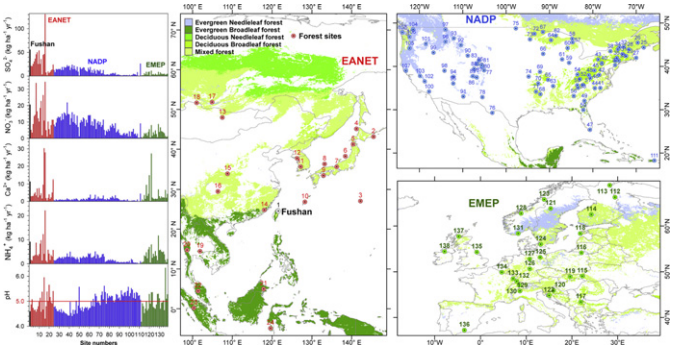
Chung-Te Chang [a], Chiao-Ping Wang [b], Jr-Huang Chuan [a], Lih-Jih Wang [c], Chiung-Pin Liu [d], LinTeng-Chiu [e,*]

[a] Department of Geography, National Taiwan University, No. 1, Sec. 4, Roosevelt Rd., Taipei 10617, Taiwan
[b] Taiwan Forestry Research Institute, 53 Nanhai Rd., Taipei 10066, Taiwan
[c] School of Forestry & Resource Conservation, National Taiwan University, No. 1, Sec. 4, Roosevelt Rd., Taipei 10617, Taiwan
[d] Department of Forestry, National Chung Hsing University, 250, Kuo Kuang Rd., Taichung 40254, Taiwan
[e] Department of Life Science, National Taiwan Normal University, 88 Ting-Chow Rd., Sec. 4, Taipei 11677, Taiwan

HIGHLIGHTS

- VWM concentrations (μeq L^{-1}) of SO_4^{2-} (29) and NO_3^- (10) are high compared to other forest regions
- Decline of both acidic pollutants and base cations led to relatively stable precipitation pH.
- Air pollution control measures contributed to the summer decline in concentrations of many ions.
- The high stable acid depositions in winter were attributed to long-range transport from East Asia.

GRAPHICAL ABSTRACT



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ABSTRACT

Long-term monitoring of precipitation chemistry provides a great opportunity to examine the evolution of air pollutant emissions and effectiveness of air pollution control measures. We evaluated the characteristics and trends of precipitation chemistry at both annual and seasonal scales based on the records of 1994–2013 at Fushan Experimental Forest (FEF) of northeastern Taiwan. The results showed that 77% of the weekly precipitation had pH < 5.0. The two-decadal average annual pH was 4.62, without a significant inter-annual trend, possibly due to the concurrent declines of both acidic pollutants and base cations. There was a significant positive relationship between $[SO_4^{2-} + NO_3^-]$ and $[Ca^{2+} + NH_4^+]$ indicating that their deposition was likely dominated by NH_4NO_3, $(NH_4)_2SO_4$, $Ca(NO_3)_2$, and $CaSO_4$. There was a significant negative relationship between precipitation pH and the difference between $[SO_4^{2-} + NO_3^-]$ and $[Ca^{2+} + NH_4^+]$, not just $[SO_4^{2-} + NO_3^-]$, suggesting that precipitation acidity was not solely determined by acidic pollutants but by the balance between acidic pollutants and base cations. We also found temporal decreases of Ca^{2+} and NH_4^+ concentrations in precipitation which contributed to the low acid neutralization capacity of precipitation. Annual deposition of NO_3^- and SO_4^{2-} was 23 and 55 kg ha^{-1} yr^{-1}, which is much higher than most forest sites in the industrialized countries suggesting that acid deposition is still a major environmental issue in Taiwan. Annual deposition of NH_4^+, Ca^{2+} and NO_3^- showed significant decreasing trends during the 20-year period, which was mostly due to the decreases in the summer deposition associated with air pollution mitigation strategies. Winter deposition showed no decreasing patterns for the same period. The high contribution to annual acid deposition from autumn-winter and spring rains (50%) associated with northeast monsoon implies that long-range transport of anthropogenic emissions from East Asia

* Corresponding author.
E-mail address: tclin@ntnu.edu.tw (T.-C. Lin).

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played a key role on acid depositions at FEF and possibly many areas in the region. Therefore, intergovernmental cooperation is urgently needed to effectively mitigate the threat of acid deposition in East Asia.

1. Introduction

Although acid deposition has been extensively studied over the past several decades, it is still a critical environmental concern, particularly in regions experiencing rapid industrialization such as East Asia. Acid deposition has profound impacts on terrestrial and aquatic ecosystems. Elevated acid deposition may increase base cation leaching, lead to decline of forest growth (Likens et al., 1996; Aber et al., 1998; Tomlinson, 2003; Jonard et al., 2012), losses in biodiversity (Clark and Tilman, 2008; Chen et al., 2013), and eutrophication of downstream and coastal waters (Rabalais, 2002). Over the past three decades, the emissions of sulfur dioxide (SO_2) and nitrogen oxides (NO_x) have decreased substantially in many North American and European countries through the efforts on emission regulations (Fowler et al., 2005; Monks et al., 2009; Sickles and Shadwick, 2015). However, SO_2 and NO_x emissions in China increased more than two times between 1990 (20 and 6 Tg yr^{-1}) and 2003 (37 and 14 Tg yr^{-1}) and have exceeded those of North America (14 and 18 Tg yr^{-1}) and Europe (9 and 12 Tg yr^{-1}) (Akimoto, 2003; Monks et al., 2009). Several emission reduction policies have been implemented since 2001 in China, and the total national NO_x and SO_2 decreased by 21% and 14% between 2005 and 2010 (Fang et al., 2013; Liu et al., 2016). The precipitation pH in China had increased from 4.8 in 2006 to 5.6 in 2014 (Duan et al., 2016). However, sulfate deposition in China (20–70 kg ha^{-1} yr^{-1}) is still 2–3 times higher than the levels in South Korea and Japan and nitrate deposition in China (8–20 kg ha^{-1} yr^{-1}) was also higher than the levels in South Korea (8–15 kg ha^{-1} yr^{-1}) and Japan (6–15 kg ha^{-1} yr^{-1}) (Duan et al., 2016). China has become the global hotspot of SO_2 and NO_x emissions contributing approximately 20% and 13% of global emissions of SO_2 and NO_x, respectively (Monks et al., 2009; Lu et al., 2010; Smith et al., 2011; Vet et al., 2014). In contrast, a recent study in Taiwan revealed that domestic contributions to total SO_2 and NO_x emissions decreased from 82% and 90% in 1994 to <27% and 60% in 2010, respectively (Chen et al., 2015).

The high levels of sulfur and nitrogen emissions in China, lead to very high deposition of SO_4^{2-} (60 kg ha^{-1} yr^{-1}) and NO_3^- (50 kg ha^{-1} yr^{-1}) in eastern China (Vet et al., 2014). Due to the transboundary nature of air pollutants, large quantities of SO_2 and NO_x emitted in China are transported to neighboring countries and the open ocean (Vet et al., 2014). Through long-term monitoring, it was estimated that the contribution of sulfur emissions from China to total sulfur deposition in East Asia had increased from 65% in 1981 to 77% in 2005 (Kuribayashi et al., 2012). In addition, nitrogen emissions from China accounted for 60–75% of total nitrate deposition downwind in Korea and Japan (Kajino et al., 2013). Under favorable meteorological conditions, the NO_x emissions from Asia could even be transported across the Pacific Ocean reaching western North America and contribute to episodic high ozone concentrations, particularly in springtime (Cooper et al., 2010).

Regional climate systems have major effects on precipitation chemistry, because they strongly affect the source regions that air masses pass over before arriving at the location of deposition (Mkadam et al., 2008; Balestrini et al., 2016; Cheng et al., 2016). East Asia is characterized by distinctive seasonal monsoons. During winter, the northeast monsoon, which originates from Siberia-Mongolia and passes through the heavily industrialized northern and northeastern China, often transports large quantities of pollutants to the east coast of China, as well as to other countries and the Pacific Ocean east off China (Chang et al., 2000; Kuribayashi et al., 2012; Chen et al., 2015). In contrast, in summer the southwest monsoon, which originates from the Pacific Ocean south and southeast from China, does not pass through areas with high sulfur and nitrogen emissions before arriving at Taiwan.

Long-term monitoring is among the most powerful measures to detect changes in precipitation chemistry and atmosphere-biosphere interactions (Fowler et al., 2009). In addition, the required or the effectiveness of pollutions control measures could also be identified or evaluated from long-term observations (Lindenmayer and Likens, 2009). There are many long-term acid deposition monitoring programs in North America (Lehmann et al., 2005) and Europe (Tørseth et al., 2012). The results from these long-term records assisted in the development of air pollution control measures such as the Clean Air Act in the US, Air Quality Daughter Directive and Clean Air for Europe (European Commission, 2005; European Environment Agency, 2006). Compared to North America and Europe, long-term monitoring of acid deposition in Asia started considerably later, although current acid deposition in this region is considered to be among the highest in the world (Dentener et al., 2006; Vet et al., 2014). For example, the Acid Deposition Monitoring Network in East Asia [EANET] was initiated in January 2001 (Duan et al., 2016). Unfortunately, acid deposition monitoring sites in Taiwan are not included the EANET forming a gap in comprehending regional patterns of atmospheric deposition of pollutants. In this study, we analyzed long-term precipitation chemistry data (1994–2013) at the Fushan Experimental Forest (FEF) of northeastern Taiwan. This long-term record provides a great opportunity to examine the temporal trends of the concentration and deposition of major ions, as well as evaluate the effectiveness of air pollution control measures enforced during the period.

In this study, this long-term record was used to test the following hypotheses. First, due to the enforcement of pollution control policies such as the revised Air Pollution Control Act (in 2002) and Air Pollution Control Act Enforcement Rules (in 2003) in Taiwan, we hypothesize that there were decreasing trends of the deposition of acidic pollutants, i.e. NO_3^- and SO_4^{2-} (H_1). Second, as a result of the decreased deposition of acidic pollutants, we hypothesize that there was an increasing trend of precipitation pH (H_2). Third, because different rainfall types are associated with air masses passing over different regions with various levels of air pollution, it is expected that the composition of precipitation and the associated temporal trends were different for different rainfall types (H_3). Specifically, since the northeast monsoon moves from the inland of Mongolia-China and passes through the heavily industrialized eastern China before arriving Taiwan in the winter-spring, long-range transport should have a substantial contribution on the deposition of NO_3^- and SO_4^{2-} at FEF during the period. The examinations of these hypotheses would assist in providing an improved understanding of the patterns and underlying control factors of precipitation chemistry at the FEF. Moreover, these long-term results are critical for evaluating whether local air pollution control measures could effectively mitigate atmospheric deposition of air pollutants and also whether regional collaborative efforts are required due to the transboundary nature of air pollutants.

2. Methods

2.1. Study site

The sampling site was located in the experimental Watershed 1 (W1, 38 ha) of FEF (1000 ha) in northeastern Taiwan (23°34′N, 121°34′E), 30 km east of Taipei (the largest city in Taiwan) and 27 km west of the east coast of Taiwan (Fig. 1a, b). The elevation of W1 ranges