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Wet deposition of sulfur and nitrogen in Jiuzhaigou National Nature Reserve, Sichuan, China during 2015–2016: Possible effects from regional emission reduction and local tourist activities☆

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ABSTRACT

In order to understand the impacts of regional emission changes and local tourism on sulfur and nitrogen wet deposition in Jiuzhaigou National Nature Reserve of southwestern China, wet deposition was monitored at a background site (Rize) and a tourist-affected site (PE: park entrance) in the reserve during 2015–2016. The observation data were compared between Rize and PE and between 2010–2011 and 2015–2016 monitoring campaigns. Also, the observation data were used in the Positive Matrix Factorization (PMF) model to identify the major sources of sulfur and nitrogen wet deposition. The results show that although local tourism emissions had considerable contributions to NH_4^+, NO_2^-, NO_3^-, and SO_4^{2-} concentrations in wet deposition ($p < 0.05$), most of the annual Volume Weighted Mean (VWM) concentrations of these four ions were likely from emissions outside Jiuzhaigou. Annual wet deposition fluxes of the four ions were also affected more by precipitation and regional emissions than by local emissions. Although annual precipitation was higher at Rize (818 mm) during 2015–2016 than at another background site near Long Lake (LL: 752 mm) during 2010–2011, the annual concentrations and fluxes of SO_4^{2-} and NO_3^- wet deposition decreased by 77% and 74% for SO_4^{2-} and by 12% and 19% for NO_3^-, respectively, most likely due to regional emission reductions. Similar large reductions in SO_4^{2-} and NO_3^- concentrations have been also found in some other sites in southwestern China. In contrast, the annual concentration and flux of NH_4^+ wet deposition at Rize during 2015–2016 were 1.4 and 1.2 times of that measured at LL during 2010–2011, respectively. The results of source apportionment analysis and tour bus emission estimates suggest that elevated NH_4^+ wet deposition was possibly related to NH_3 emissions from local tour buses, but additional studies on NH_3 emissions from tour buses in the reserve are needed to confirm this.

1. Introduction

Anthropogenic emissions of sulfur dioxide (SO_2), nitrogen oxides (NO_x), and ammonia (NH_3) have significantly altered global sulfur and nitrogen cycles. Global anthropogenic emissions of NO_x, mainly from fossil fuel combustion, have already exceeded those from natural sources (Mackenzie, 1995). Anthropogenic NH_3 emissions, mainly from agricultural activities such as fertilizer use and livestock husbandry, are significant contributions to overall NH_3 flux to the atmosphere (Doney et al., 2007 and references therein). Globally, SO_2 emissions from anthropogenic activities are about ten times of those from natural sources (Mackenzie, 1995). According to the Representative Concentration Pathway (RCP) 8.5 emission inventories, global anthropogenic SO_2, NO_x, and NH_3 emissions were 110.2, 69.5, and 39.8 Tg in 2005, respectively (Riahi et al., 2007). While global anthropogenic SO_2 and NO_x emissions were predicted to decrease to 48.9 and 57.0 Tg in 2050, respectively, that of NH_3 would increase to 62.1 Tg in 2050 (Riahi et al., 2007).

In the atmosphere, SO_2, NO_x, and NH_3 would be converted into

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different forms (mainly as SO_4^{2-}, NO_3^-, and NH_4^+), transported to downwind locations, and then deposited onto the earth's surface through dry and wet deposition. Thus, anthropogenic emissions of these pollutants can enhance sulfur and nitrogen deposition and cause acid rain (pH < 5.60) not just in urban and suburban areas but also in rural and remote areas (Galloway et al., 1982; Liu et al., 2013; Monks et al., 2009; and references therein). Excessive deposition of sulfur and nitrogen may cause natural vegetation damage (Bytnerowicz et al., 2007 and references therein), soil and water acidification (Schindler, 1988; Sullivan et al., 2005; Zhao et al., 2009; and references therein), accelerated chemical weathering of infrastructures (Kucera and Fitz, 1995), water eutrophication (Camargo and Alonso, 2006), and carbon storage change (Reay et al., 2008).

Mainly due to high SO_2 emissions, South China is identified as one of the world's three largest regions that are most affected by acid rain, which has caused significant adverse effects on China's natural ecosystems (Lydersen et al., 2006). Also, enhanced nitrogen deposition has been reported over China (Liu et al., 2013; Lu and Tian, 2007) and its effects on the country's ecosystems is significant (Liu et al., 2011). In order to control acid deposition and haze, great efforts have been made to reduce SO_2 and NO_x emissions, particularly from energy and industrial sectors, during the period of the 10th to 13th Five Year Plans of China. As a result, China's SO_2 emissions from industrial, domestic, and energy sectors decreased from 25.49 Tg in 2005 to 18.59 Tg in 2015 (Fig. 1a). While the country's NO_x emissions from the aforementioned three sectors decreased from 24.04 Tg in 2011 to 18.51 Tg in 2015 (Fig. 1b), anthropogenic NH_3 emissions increased from 7.18 Tg in 2005 to 7.75 Tg in 2010 and were predicted to be 8.92 Tg in 2050 (Fig. 1c; Riahi et al., 2007). The significant reductions of SO_2 and NO_x emissions in China are expected to lead to reduced concentrations and fluxes of SO_4^{2-} and NO_3^- wet deposition in the country. Mainly due to the large reduction in SO_2 emissions, China's acid rain areas shrunk largely from 1.22 to 0.73 million km^2 from 2010 to 2015 (Ministry of Environmental Protection of China (MEPC), 2011–2016). However, the net change of nitrogen deposition flux remains unknown because of the increase of NH_3 emissions.

In order to understand the relationship between air pollution and tufa landscape scenery changes in Jiuzhaigou National Nature Reserve (Jiuzhaigou hereafter), which is located in a remote mountainous region in Sichuan Province of southwestern China (Fig. 2a), we observed wet deposition of sulfur and nitrogen from April 2010 to April 2011 at the Long Lake (LL) meteorological station (33.04° N, 103.93° E, 3100 m a.s.l.; Fig. 2b) (Qiao et al., 2015a, 2016). Acid rain was observed and SO_4^{2-} associated with coal combustion was determined to be the major source of acidity (Qiao et al., 2015a). Annual wet deposition fluxes of SO_4^{2-}, NO_3^-, and NH_4^+ were 8.06, 1.29, and 1.39 kg S(N) ha^{-1} during the 2010–2011 monitoring campaign, and these values were much higher than that from the inferred dry deposition fluxes (0.61, 0.14, and 0.22 kg S(N) ha^{-1}, respectively) (Qiao et al., 2015a). By using the Positive Matrix Factorization (PMF) model (Paatero and Tapper, 1994) and a source-oriented Community Multi-scale Air Quality (CMAQ) model (Zhang et al., 2012), anthropogenic sources were identified as the main sources of wet deposition fluxes of SO_4^{2-}, NO_3^-, and NH_4^+ (Qiao et al., 2015a, 2015b). From June to August 2010 (part of the wet season and accounting for about 40% of annual precipitation), wet deposition fluxes of SO_4^{2-} and NO_3^- of the entire watershed were predicted to be mainly from industries (56% and 21%, respectively) and power plants (30% and 30%, respectively), while NH_4^+ wet deposition flux was mainly from fertilizers (50%) and manure management (35%) (Qiao et al., 2015b). However, the NH_4^+ flux was largely under-predicted by the source-oriented CMAQ model probably due to poor emission inventory of NH_3 for Jiuzhaigou and its adjacent regions (Qiao et al., 2015c). At the watershed level, most of the nitrogen and sulfur deposited through wet deposition were long-range transported predicted by using the model, and less than 10% of wet deposition fluxes of SO_4^{2-} and NO_3^- and about 26% of NH_4^+ wet deposition flux during that period were from Jiuzhaigou and its three surrounding counties named Jiuzhaigou, Wen, and Pingwu counties (Qiao et al., 2015b). Long-range transport of atmospheric sulfur and nitrogen was believed to be one of the causes of tufa landscape changes in Jiuzhaigou (Qiao et al., 2016).

From 2010 to 2015, total emissions from industrial, domestic, and energy sectors in Sichuan Province and its three adjacent jurisdictions (Chongqing Municipality and Gansu and Shaanxi provinces) have reduced. Specifically, the total SO_2 emission decreased from 3.2 Tg in 2010 to 2.5 Tg in 2015, and the total NO_x emission decreased from 2.4 Tg in 2011 to 1.9 Tg in 2015 (Fig. 1ab). In contrast, annual tourist number of Jiuzhaigou increased from 1.7 million in 2010 to 5.4 million in 2015 and 5 million in 2016 (Fig. 3a), with over 80% of tourists visiting the reserve during April to October (Data source: Jiuzhaigou Administration Bureau). Subsequently, emissions from tour buses, which carry tourists in the bottom of the three valleys (Rize, Shuzheng, and Zezhawa valleys; Fig. 2) are expected to have increased correspondingly. Particularly, the buses

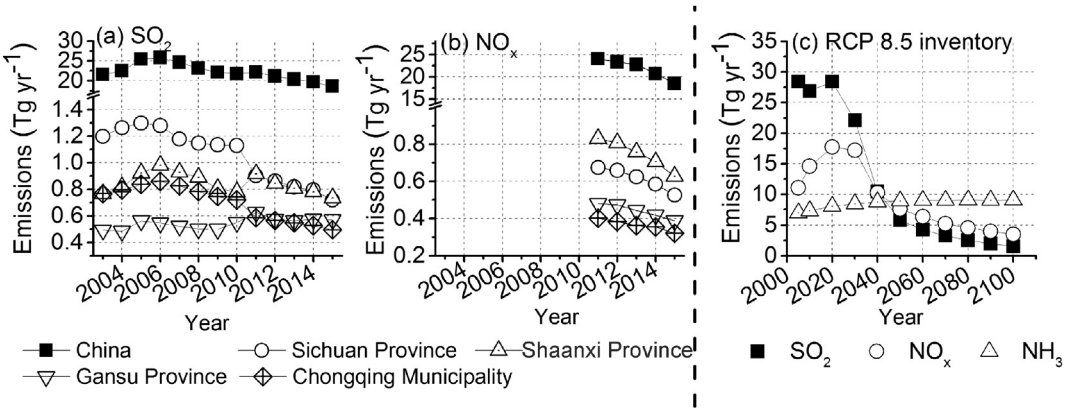

Fig. 1. Annual anthropogenic emissions of SO_2, NO_x, and NH_3 in China, Sichuan, etc. The data of (a) and (b) were obtained from the China Statistical Yearbook 2004–2016 and the anthropogenic sources include industrial, residential, and energy sectors. The data of (c) were obtained from the RCP 8.5 inventory (Riahi et al., 2007) and they include anthropogenic sources of transportation, energy, solvents, wastes, industrial, residential, agricultural, and ship sectors. The RCP 8.5 emission inventory is available at the Global Emission InitiAtive (GEIA) database: http://eccad.sedoo.fr/eccad_extract_interface/JSF/page_login.jsf.