On the seasonality of long-range transport of acidic pollutants in East Asia

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Abstract

East Asia currently has the largest SO$_2$ and NO$_x$ emissions in the world. The long-range transport (LRT) of acidic pollutants in this region is of great concern but the extent is not well understood. Here results from combined long-term (≥20 years) atmospheric deposition monitoring and air trajectory analysis in East Asia were reported. The results showed that despite the large decrease of SO$_2$ and NO$_x$ emissions in Taiwan, annual deposition of non-sea-salt sulfate (nss-SO$_{4}^{2-}$) in northern Taiwan showed no decreasing trend during 1994–2020. However, when divided seasonally, both nss-SO$_{4}^{2-}$ and nitrate (NO$_3^-_{}$) deposition had a significant decreasing trend in the summer but not in the winter. Similar patterns were found for Japan and Korea. Air trajectory models in combination with a regional emission map indicate that LRT from eastern China contributed up to 70% of the winter deposition of nss-SO$_{4}^{2-}$ and NO$_3^-$ in Taiwan and up to 50% in Japan and Korea. The results indicate that LRT obscured the efficacy of local pollution control measures in East Asia and suggest that transboundary air pollution regulations are required to combat acid deposition.

1. Introduction

Acid deposition was among the most concerning environmental threats in the 1970s–1990s in North America and Europe [1]. The rapid industrialization in East Asia makes it the new global hotspot of acid deposition following North America and Europe [2]. The annual mean deposition of sulfate and nitrate across China was 11 kg S ha$^{-1}$ yr$^{-1}$ and 3 kg N ha$^{-1}$ yr$^{-1}$, respectively, and could reach 30 kg S ha$^{-1}$ yr$^{-1}$ and 10 kg N ha$^{-1}$ yr$^{-1}$ in eastern China during 2011–2016 [3, 4]. To put it in a global context, the highest mean annual deposition of sulfate and nitrate was 8 kg S ha$^{-1}$ yr$^{-1}$ and 4 kg N ha$^{-1}$ yr$^{-1}$, respectively, in the National Atmospheric Deposition Program (NADP) network and 9 kg S ha$^{-1}$ yr$^{-1}$ and 8 kg N ha$^{-1}$ yr$^{-1}$, respectively, in the European Monitoring and Evaluation Programme (EMEP) network between 1999 and 2018 [5].

Acid deposition is a transboundary environmental issue because acidic pollutants, sulfate and nitrate and their precursors, sulfur dioxide (SO$_2$), and nitrogen oxides (NO$_x$), can be transported and deposited in places far from their origins [6–11]. Long-range transport (LRT) complicates air...
pollution control measures because if LRT is an important source of pollutants, local air pollution control measures would be of limited efficacy in reducing the atmospheric deposition of pollutants. Traditionally, studies used air trajectory models to track/identify origins of air pollutants and/or quantify the contribution of LRT to local or regional atmospheric deposition [9, 12–15]. However, air trajectory analyses are sensitive to the input of meteorological data and large differences could be generated from the model with different input datasets [16]. Thus, although air trajectory analyses provide valuable information on potential sources of air pollutants, the reliability of LRT estimates can be largely improved if they are complemented by ground measurements.

Long-term monitoring is among the most powerful and direct measures to detect changes in precipitation chemistry and validate the effectiveness of pollution control policies [17]. The changes in precipitation acidity at Hubbard Brook Experimental Forest in relation to the enforcement of the Clean Air Act and Clean Air Act Amendments is an excellent example of the power of long-term environmental monitoring [5, 18]. Long-term records of atmospheric deposition tracing back to 1990s are rare in East Asia. This region has been experiencing the most rapid industrialization in the world since the 1990s (e.g. China) and after 2000 it had the world’s largest emission of carbon dioxide [19, 20], SO$_2$ and NO$_x$ [2, 21, 22].

However, unlike the NADP of the US and the EMEP, both were initialized in the 1970s (but under different names), the Acid Deposition Monitoring Network in East Asia (EANET) was not established until 2000 [23, 24]. Some important regional acid deposition patterns have been reported using the EANET dataset. A 15 year (2001–2015) analysis of nss-SO$_{4}^{2-}$ and NO$_3^-$ wet deposition over East Asia based upon EANET indicated that the nss-SO$_{4}^{2-}$ deposition declined after 2006 across the region, however, the NO$_3^-$ deposition did not change significantly over China, Korea and Japan [25]. The decreases in wet deposition of nss-SO$_{4}^{2-}$ over Korea and Japan were related to the reductions of SO$_2$ emissions from China [25]. A 10 year (2003–2012) assessment of nitrogen deposition from eight remote EANET sites in Japan exhibited no significant increasing or decreasing trends [26]. However, the high mean annual total nitrogen deposition (10 kg N ha$^{-1}$ yr$^{-1}$) suggested that the LRT of emissions from China had a significant contribution on nitrogen deposition of these remote sites [26]. Although the two studies described above showed the annual trends of nss-SO$_{4}^{2-}$ and NO$_3^-$ depositions using EANET dataset, the seasonal divergences in the emission and transport of SO$_2$ and NO$_x$ in relation to the deposition of SO$_{4}^{2-}$ and NO$_3^-$ were not thoroughly examined. Moreover, the patterns in the 1990s, the initial stage of rapid industrialization in East Asia, was not explored. To fill this important knowledge gap in acid deposition in East Asia, we combined the rarely available long-term (1994–2020) ground measurements of acid deposition, local emission data, and air trajectory analysis to examine the role of LRT on acid deposition in Taiwan. In addition to combining long-term datasets and trajectory analyses, we also examined the impact of seasonality on LRT, an aspect not discussed in the previous literature. More importantly, to explore whether the patterns observed from our 27 year dataset is unique to our site or common to East Asia, we also used similar data and analysis for four EANET forest sites, two in Japan and two in Korea. The combination of our 27 year dataset and the two decadal dataset from EANET should help to provide a more thorough understanding on the role of LRT on the regional acid deposition of East Asia.

2. Materials and methods

2.1. Study sites

The Fushan Experimental Forest (670–1400 m a.s.l., figure S1) is a subtropical evergreen hardwood rainforest dominated by tree species in Lauraceae and Fagaceae and characterized by high (4200 mm yr$^{-1}$) and frequent (>220 d yr$^{-1}$) rainfall, rough topography (38%), shallow (<60 cm) acidic soils (pH 4.8) [27]. Annual mean temperature was 18.2 °C with the lowest in January (11.8 °C) and highest in July (24.1 °C) and annual mean relative humidity was 96% with the lowest in July (94%) and highest in February (98%). The precipitation chemistry data from four EANET forest sites of Japan and Korea are Ijira and Yusuhara in Japan, and Imsil and Kanghwa in Korea (www.eanet.asia/). These four forest sites were selected because they have continuous data for 20 years, and are categorized as remote or rural sites. Thus, they are less affected by pollution sources in the immediate adjacency, and thus are more comparable with the 27 year dataset of Fushan Experimental Forest and can better reflect the influences of regional pollutions.

2.2. Precipitation chemistry data

Bulk precipitation of the Fushan Experimental Forest site was collected using three collectors mounted on top of a 6 m tower in a forest clearing near the weir of WS1 (figure S1). Each collector consisted of two 20 cm diameter funnels and connected with polypropylene tubing to a 30 l plastic bucket on the ground. Bulk precipitation was collected on an event basis between 1994 and 1996 and thereafter on a weekly basis. Water samples were kept in refrigerators at 4 °C without preservatives prior to chemical analysis. Samples were analyzed for specific conductance, pH, major cations and anions using Dionex ion chromatography (Dionex Corp., Sunnyvale, CA) with the models changed over the two decades. The
good charge balance of samples over the 27 years illustrates the quality of the chemical analysis (figure S2). For the four EANET forest sites of Japan and South Korea, the monthly and annual wet deposition chemistry data including sulfate and nitrate between 2000 and 2019 were obtained from the EANET website (www.eanet.asia/). The precipitation chemistry datasets were based on weekly collections and have been compiled to monthly and annual basis after data processing with standard processes such as ion balance and collection efficiency check of annual rainfall greater than 70% as proposed by World Meteorological Organization [5, 28].

Linear and non-linear regression models were utilized to examine the trends of SO₂ and NOₓ emission and nss-SO₄²⁻ and NO₃⁻ deposition over time, and the best-fit models was ascertained based on the coefficient of determination (R²).

2.3. Local emission and ground air pollution

The emission data of SO₂ and NOₓ of northern Taiwan were acquired from Taiwan Emission Data System (TEDS v11.0; https://air.epa.gov.tw/EnvTopics/AirQuality_6.aspx) [29], which is the only official emission database available in Taiwan. Emission data for northern Taiwan were used in this study because our long-term bulk precipitation monitoring site (Fushan Experimental Forest) is located in northern Taiwan.

There are 27 air quality monitoring stations (AQS) in northern Taiwan (figure S1) established and maintained by Taiwan Environmental Protection Administration (TEPA; www.epa.gov.tw). The monitoring instruments in each station are installed 4.5 m above ground. Each AQS is equipped with an instrument that measures concentrations of CO, SO₂, NOₓ, PM and O₃. The monthly mean concentrations of SO₂ and NOₓ (the precursors of sulfate and nitrate, respectively) from the 27 AQSs between 1994 and 2020 were used in this study (https://data.epa.gov.tw/en/dataset/aqx_p_08). The annual and seasonal (summer [June–August] and winter [December–February]) means were then calculated and used to examine their temporal trends [30, 31].

2.4. Regional emission

To study the general trend of LRT in East Asia, the annual SO₂ and NOₓ emission data of Asia were obtained from Emissions Database for Global Atmosphere Research (EDGAR v6.1: https://edgar.jrc.ec.europa.eu/dataset_ap61) [32, 33]. From the annual time-series of SO₂ and NOₓ emission datasets at country level and annual grid maps during 1999–2018, we created the patterns of SO₂ and NOₓ emission in Asia. The 2008–2020 emission data of China was also obtained from the annual simulation of multi-resolution emission inventory for China (MEIC: http://meicmodel.org; refers also to [34, 35]).

2.5. HYSPLIT backward trajectory analysis

To identify the origins of air masses and the regions they passed through before arriving at the Fushan Experimental Forest, the backward trajectories were computed using NOAA Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (www.arl.noaa.gov/HYSPLIT_info.php) [36, 37]. The National Centers for Environmental Prediction reanalysis dataset was selected as meteorological input because it covered the time span, 1994–2020, examined in this study. We calculated 5 d backward trajectories with an altitude setting of 1000 m above the ground level, using the daily data sampled at UTC06 and UTC18 to present daily conditions to trace the possible sources of air pollutants as suggested by a number of studies [13, 38–40]. The 5 d backward trajectories of Ijira (Japan) and Kanghwa (South Korea) were also conducted with same criteria during 2000–2019, consistent with the time span of precipitation chemistry data. We conducted 4970 and 4860 simulations for summer and winter, respectively, for the time period of 1994–2020 for Fushan, and used 3680 and 3600 simulations for summer and winter, respectively, during 2000–2019 for Ijira (Japan) and Kanghwa (South Korea). The frequencies of 5 d backward trajectories were merged and the density of samples were mapped in ArcGIS v10.8.1 with 1 × 1 km resolution for both seasons to illustrate their major patterns of trajectory paths spread for three sites [41, 42]. Based on the 1 × 1 km density maps, we divided them by the numbers of seasonal simulations (e.g. density map of summer (winter) divided by 4970 (4860) in Fushan), to obtain the percentage of long-term pattern of trajectories. Our air trajectory maps were based upon long-term average of seasonal patterns and it is possible that the meteorological conditions might be changed over the 20 year period. However, when we divided the 25 years into four 5 year intervals, the seasonal air trajectory patterns were very similar (figure S3). Thus, only the whole view of trajectories was used in this study.

3. Results and discussion

3.1. Declines of SO₂ and NOₓ emissions and concentrations

Annual emission of SO₂ in northern Taiwan significantly decreased approximately 80% from 108 kton in 1994–1998 to less than 22 kton in 2016–2020, and that of NOₓ significantly decreased approximately 58% from 187 kton in 1994–1998 to 79 kton in 2016–2020 (figures 1(a) and (b)). Similarly, the annual mean atmospheric concentration of SO₂ of northern Taiwan decreased approximately 63% from 7.0 ppb in 1994–1998 to 2.6 ppb in 2016–2020, and that of NOₓ decreased approximately 40% from 37 ppb in 1994–1998 to 22 ppb in 2016–2020 (figures 1(c)
Figure 1. Annual emissions and air quality in northern Taiwan, and annual and seasonal sulfate, and nitrate deposition at the Fushan Experimental Forest. (a) Annual SO$_x$ emission, (b) annual NO$_x$ emission, (c) annual air SO$_2$ concentration and nss-SO$_2^-$ deposition, (d) annual air NO$_x$ concentration and NO$_3^-$ deposition, (e) winter air SO$_2$ concentration and nss-SO$_2^-$ deposition, (f) winter air NO$_x$ concentration and NO$_3^-$ deposition, (g) summer air SO$_2$ concentration and nss-SO$_2^-$ deposition, and (h) summer air NO$_x$ concentration and NO$_3^-$ deposition.

and (d)). Because AQS measure pollutant concentrations near the ground (4.5 m), the measured concentrations mostly reflect local emissions. Thus, the concurrent decreases in emissions of SO$_x$ and NO$_x$ and their concentrations in the near-ground atmosphere suggest that pollution control measures in Taiwan have effectively reduced their emissions and improved air quality. The effectiveness of air pollution control in Taiwan is also reflected by the decline of depositions of nutrient cations, i.e. the Ca$^{2+}$ and NH$_4^+$ (figures 2(a) and (b)), which are commonly utilized to indicate acid neutralizing capacity [5, 43, 44].
Figure 2. Annual and seasonal trends of Ca\(^{2+}\) (a), NH\(_{4}^{+}\) (b), and volume weighted mean (VWM) pH (c) in precipitation of the Fushan Experimental Forest of northern Taiwan.

As a result of the concurrent declines of SO\(_{4}^{2-}\) and NO\(_{3}^{-}\) and Ca\(^{2+}\) and NH\(_{4}^{+}\) \([5]\), the pH of precipitation remained relatively stable, mostly between 4.4 and 4.8 but seemed to increase gradually to 5.0 between 2015 and 2020 (figure 2(c)).

3.2. LRT and seasonal pattern of SO\(_{4}^{2-}\) and NO\(_{3}^{-}\) deposition

In contrast to the steady decreases in the emissions and atmospheric concentrations of SO\(_{2}\) and NO\(_{x}\), the deposition of non-sea-salt sulfate (nss-SO\(_{4}^{2-}\)) and NO\(_{3}^{-}\) of the Fushan Experimental Forest of northern Taiwan showed a weak decreasing pattern during the same period and the trend was only significant for NO\(_{3}^{-}\) (figures 1(c) and (d)). When divided seasonally, neither nss-SO\(_{4}^{2-}\) nor NO\(_{3}^{-}\) deposition showed a significant decreasing trend in the winter (December–February) (figures 1(e) and (f)), during which air masses originate mostly from Mongolia and northwest China and pass through heavily industrialized eastern China before arriving in Taiwan (figures 3(a)–(c)). According to a recent report, 87% of the winter air masses arriving at a small islet approximately 60 km from northern Taiwan originated from northern China and contributed to lower precipitation pH and higher ionic concentrations in precipitation compared to air masses from southern China or the Pacific Ocean \([45]\). Overall during 1994–2018, the emissions of SO\(_{2}\) and NO\(_{x}\) increased from slightly more than 20 million tons yr\(^{-1}\) to more than 40 and 30 million tons yr\(^{-1}\) in China respectively (figure 4). While the emissions showed significant decreases in Japan and South Korea during the same period, the emission quantities of the two countries were one order of magnitude smaller than that of China (figures 5(a) and (b)). Although there were sharp declines of SO\(_{2}\) and NO\(_{x}\) of China over last decade based on the MEIC emission dataset, their more recent of SO\(_{2}\) and NO\(_{x}\) emissions in 2018 are 8 and 20 million tons yr\(^{-1}\) which remain one order of magnitude of the emissions of Japan and South Korea (figures 4, 5(a) and (b)). Thus, the lack of concurrent decreasing trends between emission (and air concentration) and deposition in the winter could be, at least, partially attributed to LRT from eastern continental Asia. In contrast, both nss-SO\(_{4}^{2-}\) and NO\(_{3}^{-}\) decreased significantly in the summer (figures 1(g) and (h)) when LRT from eastern continental Asia was minimal (June–August) (figures 3(f)–(h)), because the summer monsoon originates mostly from the Pacific Ocean south and southwest of Taiwan and passes through no major emission sources before arriving in Taiwan (figure 3(f)).

Summer deposition of nss-SO\(_{4}^{2-}\) and NO\(_{3}^{-}\) in 2016–2020 of the Fushan Experimental Forest was approximately 30% and 24% of that in 1994–1998 (figures 1(g) and (h)). Assuming that all the nss-SO\(_{4}^{2-}\) and NO\(_{3}^{-}\) deposition in the summer were from local emissions, and similar air pollution control measures between summer and winter, the proportional decreases in nss-SO\(_{4}^{2-}\) and NO\(_{3}^{-}\) deposition originating from local emissions should also
Figure 3. The spatial distributions of pollutant (SO$_2$ and NO$_x$) emissions, and air trajectories of three distinct locations in East Asia in the winter and summer times. (a) Annual average of SO$_2$ emission, (b) annual average of NO$_x$ emission. The patterns of 5 d backward trajectory in the winter (December–February) in (c) Fushan (1994–2020), (d) Kanghwa (2000–2019), (e) Ijira (2000–2019), and the patterns of 5 d backward trajectory in the summer (June–August) in (f) Fushan (1994–2020), (g) Kanghwa (2000–2019), and (h) Ijira (2000–2019) for each day (UTC06 and 18) during 1994–2020 for Fushan Experimental Forest of northern Taiwan, and during 2000–2019 for Kanghwa and Ijira (see detail in section 2). The pattern of SO$_2$ and NO$_x$ emissions was created using EDGAR v6.1 grid map (https://edgar.jrc.ec.europa.eu/index.php/dataset_ap61#p2).

Figure 4. The annual trends of SO$_2$ (a) and NO$_x$ (b) emissions during 1994–2018 in China derived from EDGAR v6.1 (red dots: https://edgar.jrc.ec.europa.eu/dataset_ap61), and during 2008–2020 acquired from MEIC (black dots; http://meicmodel.org/; refers also to [34, 35]).
be similar between the two seasons. Based on these assumptions, winter nss-SO$_2^{2−}$ and NO$_3^−$ deposition originating from local emissions would be 4.0 and 2.0 kg ha$^{-1}$ yr$^{-1}$ instead of the observed 15.4 and 3.6 kg ha$^{-1}$ yr$^{-1}$ in 2016–2020. Attributing the differences between the measured deposition and the estimated deposition from local emissions to LRT would mean that approximately 74% and 44% of the winter nss-SO$_2^{2−}$ and NO$_3^−$ deposition, respectively, were due to LRT from eastern continental Asia. However, because the LRT contribution in summer is unlikely to be zero, this assumption would result in underestimating the LRT contribution in winter. In the winter when air quality is poor, power plants are required to reduce their emissions so that the nss-SO$_2^{2−}$ and NO$_3^−$ deposition originating from local emissions in the winter is likely to be smaller. Thus, our simplified calculations should be considered as conservative estimates.

The seasonal deposition pattern, air trajectory analyses and deposition calculation suggest that the high emission of SO$_2$ and NO$_x$ in East Asia, mainly China, over the past decades (figures 3 and 4) significantly contributed to the deposition of nss-SO$_2^{2−}$ and NO$_3^−$ in Taiwan through LRT. The results suggest that although air pollution control measures adopted in Taiwan effectively reduced the emissions of SO$_2$ and NO$_x$, and their concentrations in the atmosphere, they have a limited effect on reducing acid deposition in winter when the contribution from LRT is high. Because Taiwan is located only 150 km off southeast coast of China, the local air conditions are affected by anthropogenic emissions from East Asia during the winter, particularly when northeast monsoon is prevailing [5, 45]. The rainwater chemistry of a high-altitude remote background site (2862 m a.s.l.), at the peak of Mt. Lulin in central Taiwan, also showed seasonal divergences. The nss-SO$_2^{2−}$ and NO$_3^−$ concentrations were high in the precipitation of the Mt. Lulin site during the winter monsoon season due to LRT of pollutants from China whereas summer nss-SO$_2^{2−}$ and NO$_3^−$ concentrations were low as they were primarily influenced by marine air masses [46]. The sulfur and nitrogen isotope analysis also supported the seasonal patterns and the contribution of LRT on winter deposition of nss-SO$_2^{2−}$ and NO$_3^−$ at the Mt. Lulin site [47, 48]. A recent estimation suggested that the domestic fractions of SO$_2$ and NO$_x$ reduced by 55% and 15% in Taiwan between 1994 and 2010, however the local emission control was nullified by LRT from China [10]. Our results from the Fushan Experimental Forest are also consistent with observations of ground-based sites in northern Taiwan [45, 49].

### 3.3. LRT in East Asia

The role of LRT from eastern continental Asia on the deposition of nss-SO$_2^{2−}$ and NO$_3^−$ is not limited to Taiwan. Several modeling studies have reported LRT of sulfur and nitrogen from China to other Asian countries [9, 50–52]. Using Eulerian-type Community Multiscale Air Quality model, it is estimated that LRT from eastern China contributes more than 20% of anthropogenic nitrogen and sulfur deposition in East Asia [50–54]. Results from an aerosol chemical transport model estimated that emissions from China contributed approximately 50%–60% of the total nitrogen deposition in Korea and Japan [9].

Using SO$_2$ and NO$_x$ emission data from EDGAR and precipitation chemistry data from the four EANET forest sites of Japan and Korea, we found that the temporal trends of SO$_2$ and NO$_x$ emission and nss-SO$_2^{2−}$ and NO$_3^−$ deposition also clearly pointed to the role of LRT on the deposition of nss-SO$_2^{2−}$ and NO$_3^−$. The annual emission of SO$_2$ and NO$_x$ as well as the deposition of nss-SO$_2^{2−}$ and NO$_3^−$ showed a steady decreasing trend in the Japan sites between 2000 and 2019 (figures 5(a)–(d)). However, when divided seasonally the trends remained significant in the summer but became not significant in the winter (figures 5(e)–(h)), during which the air trajectories primarily originated from northern China (figure 3(e)). For the Korean forest site, the annual emission of SO$_2$ and NO$_x$ showed a steady decreasing trend during 2000–2018 (figures 5(a) and (b)). The annual deposition of nss-SO$_2^{2−}$ also showed a decreasing trend during the period but when divided seasonally, the deposition trend was only significant in the summer not in the winter (figures 5(c)–(h)). Using the same method used for estimating LRT contribution to the deposition of nss-SO$_2^{2−}$ and NO$_3^−$ in northern Taiwan, we estimated that LRT contributed 24% of the winter deposition of nss-SO$_2^{2−}$ and 50% of the winter NO$_3^−$ deposition to the Japanese sites and 36% of the nss-SO$_2^{2−}$ deposition to the Korea sites. The slower declining trend for NO$_x$ in Korea is not surprising. Many countries showed significant reduction of SO$_2$ emission and concentration but no change or even increasing trends of NO$_x$ emission and concentration due to increases of emission from the transportation sector [55, 56].

Our study illustrates that the rapid industrialization in China has a pervasive impact on atmospheric chemistry in East Asia. Although China's anthropogenic emissions of SO$_2$ and NO$_x$ have decreased by 62% and 17% between 2010 and 2017, and the ambient air quality substantially improved especially during COVID-19 pandemic in 2020, but they rapidly returned to levels of 2019 due to accelerated industrial and economic activities afterward [34, 35]. A recent study reported the emission and deposition of SO$_2$ and NO$_x$ in China using a generalized additive model that integrated multiple datasets [57]. The study showed that between 2005 and 2020 the emission of SO$_2$ and NO$_x$ had been steadily decreasing since 2011 but the decline of sulfate, nitrate and ammonium deposition lagged for several years and at a much slower rate [57]. The possible explanations include
Figure 5. Annual emissions of SO$_2$ (a) and NO$_x$ (b) in Japan and South Korea and annual (c), (d) and seasonal (e)–(h) deposition of sulfate and nitrate at two sites in Japan and two sites in Korea. The annual emission of SO$_2$ and NO$_x$ in Japan and Korea were acquired from the EDGAR v6.1 dataset (https://edgar.jrc.ec.europa.eu/dataset_ap61). The long-term acid deposition of the four sites in Japan and South Korea was obtained from EANET (www.eanet.asia/), and the annual and seasonal depositions were average for the two sites of Japan (Ijira and Yusuhara) and the two sites of Korea (Imsil and Kanghwa).

Increases in precipitation and enhanced regional pollution transport and changes in the chemistry of aerosol formation. Slower changes in atmospheric deposition relative to emission has also been reported in other regions [58, 59]. Thus, it is not surprising that the decreases in SO$_2$ and NO$_x$ emission have not been reflected in the winter deposition of nss-SO$_4^{2−}$ and NO$_3^{−}$ in Taiwan and it would be interesting to re-examine the patterns of nss-SO$_4^{2−}$ and NO$_3^{−}$ deposition several years later. The result would further test the role of LRT of SO$_2$ and NO$_x$ from China on the deposition of nss-SO$_4^{2−}$ and NO$_3^{−}$ in East Asia.

Across Europe and North America, there are trans-boundary efforts on air pollution control that...
made contributions to reduce acidification of precipitation and streams. The Clean Air Act [60] has contributed to the acidification of precipitation and streams and benefited society and ecosystems. Following the 1991 Canada-US Air Quality Agreement, total North American emissions were ~40% less than the levels in 1980 by the end of the last century and contributed to the recovery of acidified lakes near smelter sources in southeastern Canada [61]. In Europe, several cross-boundary protocols including the 1985 Helsinki Protocol, the 1994 Oslo Protocol, and the 1999 Gothenburg Protocol have been implemented to abate acid deposition [62]. Following the implementation of the protocols, sulfur emissions had decreased by 90% and NOx emissions by 50% between 1995 and 2010 and widespread surface water recovery from acidification was observed [63]. In East Asia, although individual countries have been making progress to reduce air pollution and EANET has been established, we are unaware of any regional air pollution control regulation being implemented.

Using long-term emissions, air concentrations and deposition of nss-SO$_4^{2-}$ and NO$_3^-$ in combination with back trajectory analysis (figures 1, 3 and 5), our results clearly illustrate that local pollution control measures have improved local air quality, whereas they are insufficient to reduce acid deposition. Inter-governmental collaborations not only on monitoring but also on policies are urgently needed to combat acid deposition in East Asia, the largest nss-SO$_4^{2-}$ and NO$_3^-$ emission center across the globe.

Data availability statement

The data that supporting the findings of this study are available upon reasonable request from the authors. Datasets used are from sources referenced in the manuscript.

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Conflict of interest

The authors declare no competing interests.

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