

Three Decades of Precipitation Chemistry Reveal Changing Sulfate– Nitrate Contributions across Contrasting Air-Mass Regimes in Northwestern Taiwan

Wei-Ti Tseng

National Central University

Ming-Chen Li

National Central University

Thi Thuy Linh Do

National Central University

Chi-Liang Yen

Department of Environmental Protection, Taoyuan City Government

Meng-Fen Yeh

Department of Environmental Protection, Taoyuan City Government

Hsiao-Wan Wu

Department of Environmental Protection, Taoyuan City Government

Moo-Been Chang

Graduate Institute of Environmental Engineering, Graduate Institute of Environmental Engineering

Neng-Huei Lin

nhlin@cc.ncu.edu.tw

National Central University

Research Article

Keywords: Acid deposition, Precipitation chemistry, Sulfur-to-nitrogen transition, Transboundary transport, Long-term trends

Posted Date: October 19th, 2026

DOI: <https://doi.org/10.21203/rs.3.rs-11126633/v1>

License:   This work is licensed under a Creative Commons Attribution 4.0 International License.

[Read Full License](#)

Additional Declarations:

No competing interests reported.

Tables are available in the Supplementary Files section.

Three Decades of Precipitation Chemistry Reveal Changing Sulfate–Nitrate Contributions across Contrasting Air-Mass Regimes in Northwestern Taiwan

Wei-Ti Tseng¹, Ming-Chen Li¹, Linh Thi Thuy Do¹, Chi-Liang Yen⁴, Meng-Fen Yeh⁴, Hsiao-Wan Wu⁴, Moo-Been Chang³ and Neng-Huei Lin^{1,2,*}

¹*Department of Atmospheric Sciences, National Central University, Taoyuan 32001, Taiwan*

²*Center for Environmental Monitoring of Technology, National Central University, Taoyuan 32001, Taiwan*

³*Graduate Institute of Environmental Engineering, National Central University, Taoyuan 32001, Taiwan*

⁴*Department of Environmental Protection, Taoyuan City Government, Taoyuan 320026, Taiwan*

*Corresponding author. Tel: +886-3-422-7151 ext. 65531

E-mail address: nhlin@cc.ncu.edu.tw

Abstract

Understanding how long-term changes in sulfur and nitrogen pollution interact with regional transport is essential for evaluating the evolving controls on acid deposition in East Asia. This study examined three decades (1994–2024) of precipitation chemistry in Taoyuan City, northwestern Taiwan, using a five-station comparison between two monitoring periods, continuous trend analysis at Chungli, and backward-trajectory clustering for 1,461 precipitation samples collected during 2006–2024. Network volume-weighted mean (VWM) pH increased from 4.56 to 5.07, while VWM H^+ decreased by approximately 69%. The most pronounced compositional change was a 54% reduction in non-sea-salt sulfate ($nss-SO_4^{2-}$). In contrast, NO_3^- changed little between the two monitoring periods but exhibited a small significant increase in the continuous Chungli record. The $nss-SO_4^{2-}/NO_3^-$ equivalent ratio consequently declined, indicating weaker sulfate dominance and a greater relative contribution from nitrate. Four transport regimes exhibited contrasting chemical characteristics. The continental/transboundary-influenced C2 regime (37.9% of samples) had the lowest pH and highest acidic-ion concentrations, whereas C3 (23.9%), characterized by predominantly summer southerly transport and limited contact with continental source regions, had lower ionic concentrations. Significant pH increases and $nss-SO_4^{2-}$ decreases occurred in all four regimes. NO_3^- showed no significant trend in C2 but increased significantly in C3; this contrast is consistent with differences in transport, seasonal meteorology, and precipitation characteristics, but

37 does not establish statistically different trend slopes or quantify source contributions. Collectively,
38 the results indicate a broad decline in sulfate dominance and an increasing relative influence of
39 nitrate in precipitation chemistry. Persistent nitrate despite substantial sulfate reductions highlights
40 the need to maintain sulfur controls while strengthening regional and local management of reactive
41 nitrogen.

42

43 **Keywords:** Acid deposition, Precipitation chemistry, Sulfur-to-nitrogen transition, Transboundary
44 transport, Long-term trends.

1. INTRODUCTION

Acid deposition remains an important environmental issue because atmospheric scavenging transfers sulfur- and nitrogen-containing pollutants to terrestrial and aquatic ecosystems, affecting surface-water chemistry, soil nutrient status, vegetation, agricultural productivity, and built materials (de Vries and Kros, 1989; Agrawal, 2008; Wang et al., 2013; Wang et al., 2018; Shi et al., 2021; Prakash et al., 2023; Zhang et al., 2024). Sulfur dioxide (SO_2) and nitrogen oxides (NO_x) are major anthropogenic precursors of sulfuric and nitric acids, and their transformation and removal through precipitation are reflected primarily in sulfate (SO_4^{2-}), nitrate (NO_3^-), and hydrogen ions (H^+). Accordingly, long-term observations of precipitation pH and major ionic species provide an integrated measure of changes in atmospheric composition and acid deposition (Galloway et al., 1987; Schindler, 1988; Anttila et al., 1995; Tang et al., 2005; Vet et al., 2014; Duan et al., 2016; Pascaud et al., 2016; Roy et al., 2016; Pu et al., 2017; Rattigan et al., 2017; Martins et al., 2019; Bhuyan et al., 2020; Lin et al., 2020; Shah et al., 2020). Such records are particularly valuable for evaluating how emission changes and atmospheric transport alter the relative contributions of sulfur and nitrogen to precipitation acidity.

The emission landscape of East Asia has changed markedly during the past several decades. Rapid industrialization and increasing energy demand initially produced substantial increases in SO_2 and NO_x emissions; regional emissions increased by approximately 119% and 176%,

64 respectively, between 1980 and 2003 (Ohara et al., 2007). In China, SO₂ emissions peaked earlier
65 than NO_x emissions, around 2006 and 2011, respectively, and both subsequently declined during
66 the 2010s following increasingly stringent emission controls (Lu et al., 2011; Zheng et al., 2018;
67 Kurokawa and Ohara, 2020). Because sulfur and nitrogen emissions have followed different
68 temporal trajectories, their relative contributions to atmospheric deposition may also have evolved
69 differently. Long-term precipitation chemistry therefore provides an opportunity to determine
70 whether reductions in sulfur pollution have been accompanied by comparable changes in nitrogen
71 deposition, or whether nitrate has become relatively more important as sulfate inputs have declined.

72 Taiwan is a useful downwind receptor for examining these changes because its air quality is
73 influenced by both local emissions and long-range transport from the East Asian continent. During
74 the northeasterly monsoon, particularly in winter and spring, continental air masses can transport
75 pollutants toward Taiwan through monsoonal flows and frontal systems (Chang et al., 2000; Chen
76 et al., 2013; Chen et al., 2014; Chuang et al., 2020; Griffith et al., 2020). Similar transboundary
77 influences have been documented in South Korea and Japan (Aikawa et al., 2010; Kim et al., 2012;
78 Koo et al., 2012; Lee et al., 2013; Kaneyasu et al., 2014; Oh et al., 2015; Ohizumi et al., 2016;
79 Vellingiri et al., 2016; Kim et al., 2017a; Kim et al., 2017b; Itahashi et al., 2018; Inomata et al.,
80 2019; Yim et al., 2019; Kato et al., 2025; Zhou et al., 2025). Consequently, changes observed in
81 Taiwanese precipitation may reflect the combined effects of regional emission evolution, long-

82 range transport, and local pollution, making transport-resolved analysis essential for interpreting
83 long-term chemical trends.

84 Long-term monitoring networks, including the National Atmospheric Deposition Program
85 (NADP), the European Monitoring and Evaluation Programme (EMEP), and the Acid Deposition
86 Monitoring Network in East Asia (EANET), have demonstrated the value of multidecadal
87 observations for detecting changes in atmospheric deposition and evaluating environmental
88 policies. Experience in Europe and North America shows that sustained emission reductions can
89 substantially reduce sulfur deposition and promote ecosystem recovery (Grennfelt et al., 2020).
90 However, the response of nitrogen deposition can differ from that of sulfur, and changes in the
91 sulfate-to-nitrate balance may alter the chemical controls on precipitation acidity. Resolving these
92 divergent responses is particularly important in East Asia, where rapid changes in emissions coexist
93 with strong seasonal transport and intensive local anthropogenic activity.

94 Taoyuan City, located in northwestern Taiwan, provides an especially suitable setting for
95 investigating these interactions. The Taoyuan Tableland faces the Taiwan Strait and is exposed to
96 northeasterly monsoonal flow during the cool season, favoring the influence of long-range-
97 transported pollutants. In summer, southwesterly flow and frequent afternoon convection increase
98 the importance of locally and regionally influenced precipitation. Taoyuan is also a rapidly
99 urbanized and industrialized metropolitan area containing major transportation corridors, Taiwan

Taoyuan International Airport, and dense industrial clusters, including the Dayuan, Guanyin, and Pingzhen industrial parks. These activities provide substantial local sources of NO_x, SO_x, and particulate matter. The coexistence of strong local emissions and seasonally varying regional transport makes Taoyuan well suited for separating long-term chemical changes associated with contrasting air-mass regimes.

Despite extensive studies of acid deposition and transboundary air pollution in East Asia, relatively few studies have combined a multidecadal, multi-site precipitation chemistry record with event-based air-mass classification to determine whether the long-term responses of sulfur and nitrogen are consistently observed among transport regimes. This distinction is critical because an overall recovery in precipitation acidity may mask divergent behavior of sulfate and nitrate and, consequently, a changing balance between sulfur- and nitrogen-related acidification. Here, we analyze precipitation chemistry at five monitoring stations in Taoyuan City during 1994–2005 and 2014–2024, supplemented by a continuous 30-year record at Chungli. Period-level chemistry was summarized across all five stations, station-specific trend comparisons used the four non-Chungli stations to avoid duplicating the continuous Chungli analysis, and the Chungli record was used for 1994–2024 and transport-resolved analyses. We further combine backward-trajectory clustering with Mann–Kendall and Sen’s slope analyses to (1) quantify long-term changes in precipitation acidity and the relative contributions of sulfate and nitrate, (2) determine whether these changes

are consistently observed under contrasting regional transport regimes, and (3) assess whether weakening sulfate influence has been accompanied by an increasing relative importance of nitrogen-related pollution. By linking multidecadal chemical trends with transport pathways, this study aims to clarify the evolving controls on acid deposition in northwestern Taiwan and to provide evidence relevant to future sulfur and nitrogen pollution management in Taiwan and other East Asian receptor regions.

2. MATERIALS AND METHODS

2.1 Site Description

Precipitation chemistry samples were collected at five acid-rain monitoring stations in Taoyuan City, northwestern Taiwan (Fig. 1). Three stations—Longtan (N01), Taoyuan (N02), and Chungli (N03)—were established in late 1993, followed by Fuxing (N04) and Xinwu (N05) between late 1996 and 1997. Sampling continued through 2005, after which the monitoring program was suspended. The network was reactivated in 2014. Where feasible, stations were re-established at or near their original locations; when the original sites were no longer suitable, notably at Taoyuan (N02) and Fuxing (N04), nearby alternative locations were selected. Accordingly, 1994–2005 and 2014–2024 are hereafter referred to as Phase I and Phase II, respectively.

To extend the Chungli record across the network hiatus, wet-deposition observations collected during 2006–2013 by the Ministry of Environment at National Central University were

incorporated into the N03 time series. The Ministry site and the Chungli monitoring station are separated by approximately 650 m. Although the two sites were separated by approximately 650 m, the same sampling, handling, and analytical protocols were used, and the supplementary observations were subjected to the same QA/QC procedures before integration. The five stations were used for the network-wide Phase I and Phase II comparison. Because Chungli was independently analyzed using the reconstructed continuous 1994–2024 record, station-specific annual trend analyses were performed for the four non-Chungli stations to avoid duplication. This distinction allows the multi-site decadal comparison and the continuous 30-year trend analysis to provide complementary evidence for long-term changes in precipitation chemistry.

Taoyuan City covers approximately 1,220.95 km² and is characterized by a southeast-to-northwest topographic gradient from mountainous terrain to rolling hills and coastal plains. The western plains contain the major urban and industrial districts, including Taoyuan and Chungli, while large industrial parks are distributed across Chungli and neighboring districts. The region also contains major transportation corridors and Taiwan Taoyuan International Airport (IATA code: TPE). These local anthropogenic sources coexist with seasonally varying regional transport: northeasterly monsoonal flow during the cool season favors the arrival of continental air masses, whereas southwesterly flow and frequent convective rainfall are more important in summer. The resulting contrast makes Taoyuan well suited for examining precipitation chemistry under different

local and regional transport influences. The current locations and characteristics of the five stations are shown in Fig. 1 and summarized in Table 1.

2.2 Rainwater Sampling and Chemical Analysis

Rainwater was collected using a daily sampling interval from 09:00 local time to 09:00 the following day, with a minimum sample volume of 50 mL. Each station was equipped with an automatic wet-only precipitation collector. The collector lid opened automatically when rainfall was detected and closed after rainfall ceased, thereby minimizing contamination by dry deposition (Sheu and Lin, 2013). Following collection, samples were weighed, double-bagged, stored at 4 °C, and transported to National Central University for chemical analysis (Do et al., 2021).

An approximately 20-mL aliquot was used for pH and electrical-conductivity measurements with Mettler Toledo instruments (SevenMulti and MPC227). The remaining sample was passed through a 0.45-µm PVDF filter to remove insoluble material and analyzed for the major water-soluble ions Cl^- , NO_3^- , SO_4^{2-} , NH_4^+ , Na^+ , K^+ , Mg^{2+} , and Ca^{2+} using Dionex ion chromatography systems (DX-100 or ICS-1500/1600/2100).

Analytical quality assurance and quality control followed protocols adopted from the Global Atmosphere Watch/World Meteorological Organization (GAW/WMO) and the European Monitoring and Evaluation Programme (EMEP). Data quality was evaluated using total ionic concentration ($\mu\text{eq L}^{-1}$) and ionic charge balance (Lin and Peng, 1999). After QA/QC screening,

summed anion and cation equivalents across the five stations were strongly correlated ($R = 0.99$), with a regression slope of 1.06 and $R^2 = 0.98$ (Fig. S1), indicating good ionic balance and analytical consistency.

2.3 Data Processing

To reduce the influence of differences in precipitation amount and associated dilution, the volume-weighted mean (VWM) concentrations were calculated for each species as:

$$\text{VWM} = \Sigma(C_i P_i) / \Sigma P_i,$$

where C_i is the concentration of the target species in sample i and P_i is the corresponding precipitation amount (Galloway et al., 1987). VWM concentrations were used for period- and cluster-level comparisons because they weight observations according to the precipitation amount represented. VWM H^+ was calculated using the same equation, and VWM pH was then derived as $-\log_{10}(\text{VWM } \text{H}^+)$, with H^+ expressed in mol L^{-1} ; pH values were not averaged arithmetically.

Because sulfate in precipitation includes both marine and non-marine contributions, non-sea-salt sulfate (nss-SO_4^{2-}) was estimated using Na^+ as the sea-salt tracer (Keene et al., 1986):

$$\text{nss-SO}_4^{2-} = \text{SO}_4^{2-} - 0.121 \times \text{Na}^+ \quad (\mu\text{eq L}^{-1}),$$

where 0.121 is the equivalent $\text{SO}_4^{2-}/\text{Na}^+$ ratio in seawater. This correction was applied to focus the long-term sulfur analysis on sulfate not attributable to sea salt (Itahashi et al., 2015; Seinfeld and Pandis, 2016). The $\text{nss-SO}_4^{2-}/\text{NO}_3^-$ equivalent ratio was subsequently used as an indicator of

the changing relative importance of sulfate and nitrate in precipitation chemistry.

2.4 Trend Analysis

Long-term trends in precipitation chemistry were evaluated using the non-parametric Mann–Kendall (M–K) test, which is widely applied to atmospheric-deposition datasets because it is robust against outliers, accommodates irregular sampling intervals, and requires no distributional assumptions (Gilbert, 1987; Marchetto et al., 2013; Pascaud et al., 2016). The M–K test was applied to pH, major ionic species, and the $\text{nss-SO}_4^{2-}/\text{NO}_3^-$ ratio. Daily ionic concentrations were log-transformed before trend testing to reduce the influence of the strongly right-skewed concentration distributions (Do et al., 2021).

For statistically evaluated trends, Sen’s slope estimator was used to quantify the direction and magnitude of temporal change (Sen, 1968), with significance assessed from the M–K Z statistic following Pascaud et al. (2016). Because the tests were applied to chronologically ordered daily samples, Sen’s slopes are expressed per sampling observation ($\text{pH unit observation}^{-1}$ for pH and $\log_{10}[\mu\text{eq L}^{-1}] \text{ observation}^{-1}$ for log-transformed concentrations). Trend analyses were conducted for the continuous Chungli record and, where applicable, within the trajectory clusters described below. This approach evaluated whether long-term decreases in sulfate-related acidity and nitrate behavior were consistently observed across contrasting air-mass regimes.

2.5 Backward Trajectories Calculation and Clustering

Air-mass transport associated with precipitation at Chungli was characterized using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model developed by the NOAA Air Resources Laboratory (Stein et al., 2015). Meteorological fields were obtained from the National Centers for Environmental Prediction Global Data Assimilation System (NCEP GDAS; Kalnay et al., 1996) at $1.0^\circ \times 1.0^\circ$ spatial resolution. Seventy-two-hour backward trajectories were calculated for precipitation sampling periods during 2006–2024 to characterize the major pathways of air masses arriving at Chungli (N03). Previous studies have shown that low-altitude continental transport can elevate pollutant concentrations in northern Taiwan and that precipitation associated with trajectories passing through highly polluted regions of East Asia can exhibit enhanced acidity (Sheu et al., 2010; Klemm et al., 2015; Sheu et al., 2019).

For each 72-h backward trajectory arriving at N03 (09:00 LST), hourly horizontal coordinates (72 x–y endpoint pairs) were clustered using an automated K-means algorithm. The optimal number of clusters ($k = 4$) was objectively determined via Principal Component Analysis (PCA) diagnostics (Fig. S2). Specifically, the scree plot exhibited a distinct "elbow" at the fourth principal component (Fig. S2a), where the leading four components accounted for over 91% of the cumulative explained variance, exceeding the standard 90% threshold (Fig. S2b). These four identified transport regimes were subsequently used for precipitation chemistry comparisons and long-term trend analyses. Given the inherent temporal growth of trajectory errors, potential source

regions were interpreted cautiously, particularly for earlier segments of the 72-h paths (Stohl, 1998).

The combined analytical framework—network-wide Phase I/Phase II comparisons, the continuous Chungli trend record, and transport-resolved trajectory analysis—was designed to address the study objectives stated in Section 1: to quantify long-term changes in precipitation acidity and sulfate–nitrate balance, determine whether these changes differ among transport regimes, and evaluate whether the weakening influence of sulfate has been accompanied by an increasing relative importance of nitrogen-related pollution.

3. Results and Discussion

3.1 Changes in Precipitation Chemistry Composition

Precipitation chemistry in Taoyuan City differed markedly between Phase I (1994–2005; Table 2) and Phase II (2014–2024; Table 3). The volume-weighted mean (VWM) pH increased from 4.56 to 5.07, while the corresponding H^+ concentration decreased from 27.6 to 8.5 $\mu\text{eq L}^{-1}$ (approximately 69%). Together, these changes indicate a substantial reduction in precipitation acidity over the past three decades. Comparable increases in precipitation pH have been reported in Europe and North America following reductions in SO_2 and NO_x emissions; East Asia has generally shown a smaller but directionally similar improvement (Chang et al., 2022). The Taoyuan record is therefore consistent with the broader response of precipitation chemistry to regional emission controls, although the respective contributions of local emissions, transboundary transport,

and alkaline neutralization are examined below.

Among the major acidic ions, SO_4^{2-} showed the largest decrease. Its VWM concentration declined from 59.5 to 29.6 $\mu\text{eq L}^{-1}$, while non-sea-salt sulfate (nss-SO_4^{2-}) decreased from 53.3 to 24.4 $\mu\text{eq L}^{-1}$, a reduction of approximately 54%. Because nss-SO_4^{2-} is principally derived from the atmospheric oxidation of anthropogenic SO_2 , this decline is consistent with reductions in sulfur emissions associated with fuel-quality regulations, lower coal use in some sectors, and the implementation of flue-gas desulfurization in Taiwan and other upwind regions (Holland et al., 1998; Zhang et al., 2018; Zheng et al., 2018; Likens et al., 2021; Akimoto et al., 2022; Kato et al., 2025). By contrast, NO_3^- changed little, from 25.8 to 24.8 $\mu\text{eq L}^{-1}$. Consequently, the $\text{nss-SO}_4^{2-}/\text{NO}_3^-$ equivalent ratio decreased, indicating a shift from sulfate-dominated acidity toward a greater relative contribution from nitrate. This pattern agrees with EANET observations showing increasing $\text{NO}_3^-/\text{SO}_4^{2-}$ ratios in China, South Korea, and Japan during 2001–2015 (Itahashi et al., 2018).

The fractional ionic composition (Fig. 2) further illustrates this transition. SO_4^{2-} accounted for approximately 22.0% of the total measured ionic equivalents in Phase I but only 15.2% in Phase II, whereas the NO_3^- fraction increased from approximately 12.3% to 14.2%. Thus, although the VWM NO_3^- concentration remained nearly unchanged, its relative importance within the ionic mixture—and hence its potential contribution to precipitation acidity—increased as sulfate

declined.

The VWM concentration of NH_4^+ , a major neutralizing ion, decreased moderately from 39.5 $\mu\text{eq L}^{-1}$ in Phase I to 29.4 $\mu\text{eq L}^{-1}$ in Phase II. This change may reflect shifts in agricultural activity, livestock-waste management, and regional NH_3 emissions (Chang et al., 2017; Su et al., 2024), although source-specific attribution cannot be established from precipitation chemistry alone. Declines in precipitation NH_4^+ have likewise accompanied reductions in European NH_3 emissions at several EMEP forest sites (Chang et al., 2022). Because the decrease in NH_4^+ was smaller than that in nss-SO_4^{2-} , the observed reduction in acidity is more consistent with diminished acidifying inputs than with an increase in ammonium-based neutralization, in agreement with long-term observations across EANET (Vet et al., 2014).

In contrast, Ca^{2+} increased from 15.9 to 23.6 $\mu\text{eq L}^{-1}$, and its contribution to the total measured ionic equivalents rose from approximately 7.1% to 13.7%. Ca^{2+} in precipitation is commonly associated with crustal dust and resuspended soil and road material. The increase may therefore reflect some combination of long-range East Asian dust transport during the winter–spring monsoon (Lin et al., 2007; Chang et al., 2010; Kong et al., 2022) and local fugitive-dust emissions associated with urbanization, construction, and infrastructure development (Huang et al., 2009; Zeng et al., 2022). Without source tracers, their individual contributions cannot be quantified. Nevertheless, the higher Ca^{2+} level in Phase II indicates greater alkaline neutralization potential

and suggests that the increase in precipitation pH is consistent with both lower acidifying inputs and changes in alkaline material.

3.2 Decadal Shifts in the Spatial Distribution of Precipitation Chemistry

Fig. 3 to Fig. 5 show pronounced spatial changes in precipitation acidity and ionic composition between Phase I and Phase II. VWM pH increased at all five monitoring locations, from a range of 4.51–4.65 in Phase I to 4.89–5.25 in Phase II. The increase was especially evident at the urban Taoyuan (N02) and Longtan (N01) sites, indicating that the reduction in precipitation acidity was spatially widespread rather than confined to a single station.

The clearest spatial change occurred for nss-SO_4^{2-} . High-concentration areas in the Taoyuan–Chungli–Longtan corridor ($53.0\text{--}56.0\ \mu\text{eq L}^{-1}$ in Phase I) decreased to $26.8\text{--}27.8\ \mu\text{eq L}^{-1}$ in Phase II. This reduction of more than 50% is consistent with the substantial decline in sulfur emissions associated with fuel controls and desulfurization in Taiwan and upwind East Asian regions. Because the spatial analysis does not isolate source regions, however, it should be interpreted as evidence of a regional-scale response rather than direct attribution to any single policy or jurisdiction.

By comparison, the spatial range of VWM NO_3^- concentrations changed relatively little. In Phase I, the highest concentrations occurred along the inland corridor between Longtan and Fuxing (up to approximately $31.4\ \mu\text{eq L}^{-1}$); in Phase II, the apparent high-concentration area shifted toward

Taoyuan, Chungli, and Longtan, while the regional range remained approximately 20.0–32.0 $\mu\text{eq L}^{-1}$. These areas coincide with dense population and transportation corridors near National Freeways 1 and 3, making traffic and other urban NO_x sources plausible contributors, although the concentration maps alone cannot resolve source attribution. The contrast between the pronounced sulfate decline and the persistence of nitrate supports an increasing relative role for nitrate in regional precipitation acidity.

Spatial changes in Ca^{2+} also contributed to the altered acid–base balance. During Phase II, elevated Ca^{2+} extended northward from Chungli (N03), with peak values exceeding 31.0 $\mu\text{eq L}^{-1}$, whereas most of the region remained below 18.0 $\mu\text{eq L}^{-1}$ in Phase I. NH_4^+ showed the opposite broad tendency, with the spatial maximum decreasing from 38.2 to 25.9 $\mu\text{eq L}^{-1}$ and the high-concentration area contracting away from the southern rural sites. This divergence indicates that the greater neutralization potential in Phase II was associated more strongly with Ca-containing material than with NH_4^+ . Possible contributors include long-range dust transport and local fugitive dust from construction and infrastructure development in northern Taoyuan (Grider et al., 2023), but these sources cannot be separated with the available ionic data. Overall, the spatial patterns are consistent with two concurrent influences on pH recovery: reduced anthropogenic sulfur loading and increased neutralization by alkaline particulate matter.

3.3 Decadal Shifts and Continuous 30-Year Trend Analysis of Precipitation Chemistry

To evaluate long-term changes in precipitation chemistry across highly urbanized and industrialized Taoyuan City, we combined a network-wide comparison of two multi-year periods with a continuous 30-year trend analysis. Monotonic trends were evaluated using the Mann–Kendall test and Sen’s slope estimator, with statistical significance assessed at $\alpha = 0.05$. Trend analyses were conducted at the individual precipitation-event level, with valid samples arranged chronologically according to their sampling dates. The Mann–Kendall test therefore evaluated monotonic changes across the complete temporal sequence of precipitation observations rather than trends in annual means. Sen’s slope was estimated from the chronological observation order and represents the median change per observation. The period-level analysis summarized VWM chemistry across all five stations (Tables 2 and 3). To avoid duplicating the continuous Chungli analysis, station-specific annual trends were then evaluated at the four non-Chungli stations during Phase I and Phase II (Figs. S3–S7); the corresponding Mann–Kendall results, together with the continuous Chungli results, are summarized in Table 4.

3.3.1 Network-wide decadal shifts (Phase I vs. Phase II)

The network-wide comparison showed a clear reduction in precipitation acidity. Annual VWM pH at the four non-Chungli stations was generally below 5.0 in Phase I (4.14–5.02) but shifted upward in Phase II (4.62–5.82). Among the principal acidifying anions, non-seq-SO_4^{2-} exhibited the

dominant change: all stations showed significant downward trends during Phase II. By contrast, the five-station period-level VWM NO_3^- concentration changed little (25.8 to 24.8 $\mu\text{eq L}^{-1}$), and station-specific NO_3^- trends during Phase II were small and inconsistent. These results identify the decline in nss-SO_4^{2-} , rather than a comparable decline in NO_3^- , as the main compositional change associated with lower precipitation acidity.

Changes in neutralizing cations may also have influenced the long-term pH response (Park et al., 2015; Chang et al., 2017; Pu et al., 2017; Zhang et al., 2018). For NH_4^+ , only Fuxing showed a significant downward trend during Phase I; the other stations were stable. During Phase II, NH_4^+ decreased slightly at all stations, but none of these trends were statistically significant. Ca^{2+} was stable or decreased slightly during Phase I and decreased significantly at all stations during Phase II ($p < 0.001$). Nevertheless, Phase II concentrations remained higher than those in Phase I. This combination—higher Phase II levels but declining trends within Phase II—suggests that Ca^{2+} inputs increased during the unobserved interval (2006–2013) or early in Phase II and subsequently declined. Because observations are discontinuous during the interval, the timing and magnitude of that change cannot be determined directly from the network data.

3.3.2 Continuous 30-year evolution at the Chungli representative station

The network comparison is based on two separate monitoring periods (1994–2005 and 2014–2024) and therefore cannot resolve changes during 2006–2013. To examine the intervening

evolution, validated observations from the Ministry of Environment site near Chungli, subjected to routine quality assurance and quality control, were integrated with the N03 record to construct a continuous series for 1994–2024. Individual precipitation-event observations were arranged chronologically according to sampling date for trend analysis, whereas annual means, medians, interquartile ranges, and 10th–90th percentiles were used only to visualize interannual variability (Fig. 6). Thus, the Mann–Kendall test and Sen’s slope were calculated from the complete sequence of individual precipitation observations rather than from annual means.

As shown in Fig. 6a, the annual distributions of precipitation pH exhibited a significant upward trend over the three decades, with annual mean pH increasing from 4.46 in 1994 to 5.38 in 2024 (Mann-Kendall $\tau = 0.278$, $p < 0.001$). The positive Sen’s slope indicates a systematic increase in pH across the chronological sequence of precipitation observations. The magnitude of the long-term pH increase is broadly comparable to the upward trend reported for Nanjing, China, between 1999 and 2016 (Zhang et al., 2018), and aligns with similar upward pH trends recorded at various monitoring sites across China, Japan, and South Korea (Pu et al., 2017; Zhang et al., 2018). Furthermore, starting in 2021, the entire interquartile range of annual pH values consistently shifted upward, with annual means pH values exceeded 5.0—a phenomenon similarly observed in neighboring Taipei (Chang et al., 2026)—indicating a substantial long-term recovery in precipitation acidity.

The driving mechanisms behind this pH recovery are explicitly detailed in the annual distributions of the primary acidifying precursors (Fig. 6c,d) and their relative equivalent ratio (Fig. 6b):

- Sulfur-to-Nitrogen Transition (Fig. 6b): The annual distributions of the $\text{nss-SO}_4^{2-}/\text{NO}_3^-$ equivalent ratio showed a significant long-term decline over the 30-year span. This declining ratio highlights a fundamental structural transition in regional precipitation chemistry: decreasing sulfate dominance accompanied by an increasing relative contribution of nitrate.
- Anion Divergence (Fig. 6c,d): The log-transformed nss-SO_4^{2-} concentrations (Fig. 6c) demonstrated a highly significant monotonic downward trend (Mann-Kendall $\tau = -0.204$, $p < 0.001$), with a Sen's slope of $-0.00021 \log_{10}(\mu\text{eq L}^{-1}) \text{ observation}^{-1}$. This result indicates a persistent decline in sulfate concentrations across the chronological sequence of precipitation observations and is consistent with long-term reductions in sulfur-related precipitation inputs reported elsewhere in East Asia (Do et al., 2021). Conversely, the continuous 30-year record of log-transformed NO_3^- at Chungli (Fig. 6d) revealed a gradual but statistically significant upward trend (Mann-Kendall $\tau = 0.04$, $p < 0.01$; Sen's slope = $+0.000005 \log_{10}(\mu\text{eq L}^{-1}) \text{ observation}^{-1}$). This result contrasts with the network-wide comparison, in which NO_3^- concentrations showed no statistically significant

difference between Phase I and Phase II. This discrepancy is not an inconsistency but reflects the different analytical designs: the period-level comparison evaluates differences between two separated monitoring periods, whereas the continuous event-based analysis evaluates monotonic change across the complete chronological sequence of precipitation observations. Despite its small magnitude, the significant positive trend suggests that nitrate concentrations did not decline in parallel with sulfate over the continuous record and may have increased slightly over time.

Long-term changes in alkaline species provide additional context for the evolving acid–base balance (Fig. 6e,f). Log-transformed NH_4^+ showed a small but statistically significant downward trend (Sen’s slope = $-0.00004 \log_{10}(\mu\text{eq L}^{-1}) \text{ observation}^{-1}$, $p = 0.005$; Fig. 6e). The near-zero slope and comparatively stable interquartile range indicate only a modest multidecadal decline rather than a major change in neutralizing capacity. The persistence of NH_4^+ is compatible with continued NH_3 emissions from agricultural, livestock, waste, and urban sources in Taoyuan, but the available measurements do not permit these source contributions to be partitioned.

Log-transformed Ca^{2+} at Chungli decreased during 2014–2024 but increased significantly over the complete 1994–2024 record (Sen’s slope = $+0.00021 \log_{10}(\mu\text{eq L}^{-1}) \text{ observation}^{-1}$, $p < 0.001$; Fig. 6f). This apparent contrast indicates that the long-term record was not monotonic across all subperiods and is consistent with an increase before or near the beginning of Phase II followed by

a recent decline. Greater crustal and terrestrial inputs may be associated with urbanization, airport-area development, infrastructure construction, and soil disturbance in metropolitan Taoyuan. However, in the absence of mineralogical or source-tracer data, the Ca^{2+} trend should be regarded as consistent with, rather than definitive proof of, enhanced fugitive-dust loading.

In summary, the multidecadal increase in precipitation pH across Taoyuan City was associated primarily with the pronounced decline in nss-SO_4^{2-} . Relatively persistent NH_4^+ provided a continuing neutralization reservoir, while higher long-term Ca^{2+} levels may have supplied additional alkaline buffering (Grider et al., 2023). These observations support a combined explanation involving reduced acidic sulfur inputs and changes in neutralizing species, while avoiding attribution of the pH response to any single source or control measure.

3.4 Air-mass Transport and Precipitation Chemistry

3.4.1 Characteristics of precipitation chemistry under different air-mass transport regimes

To assess how air-mass origin and transport pathways influence precipitation chemistry, backward trajectories were calculated for individual precipitation events and grouped using trajectory clustering. This approach has been widely used in acid-deposition studies to identify recurring transport regimes and evaluate their statistical associations with precipitation composition (Sun et al., 2010; Kim et al., 2012; Ge et al., 2016; Fedkin et al., 2019; Guo et al., 2025; Lei et al., 2026). Trajectories indicate potential transport histories rather than source–receptor

causality; therefore, the cluster results are interpreted as compositional contrasts among transport regimes.

Using HYSPLIT, 1,461 backward trajectories were generated at 09:00 local time for precipitation-sampling days with valid samples at Chungli during 2006–2024. Clustering identified four recurring transport patterns (Fig. 7), accounting for 6.8%, 37.9%, 23.9%, and 31.5% of samples for C1–C4, respectively. C1 and C2 represent predominantly continental transport from East Asia. C1 followed the longest northwesterly pathway and generally remained above 1,500 m before approaching the southeastern coast of China. C2, the most frequent cluster (37.9%), arrived from the north to northwest after traversing eastern, northern, and central China; it was therefore classified as the principal continental/transboundary-influenced regime. C3 approached from the southwest over predominantly maritime areas and occurred mainly in summer, when afternoon convection is common in Taiwan. Because these pathways had comparatively limited contact with continental source regions, C3 was treated as a low-transboundary-influence regime in which scavenging of regional and local pollutants may be more evident. C4 approached from the northeast after passing over the East China Sea and waters near Japan and Korea and was classified as a regional marine transport regime. These labels describe dominant pathways and do not imply that any cluster is chemically pristine or exclusively influenced by one source region.

Precipitation composition differed among the four trajectory clusters (Fig. 8). The distributions

454 of acidifying ions were consistent with the pH contrasts. C2 had the highest nss-SO_4^{2-} and NO_3^-
455 concentrations (43 and 30 $\mu\text{eq L}^{-1}$, respectively), followed by C1 (38 and 28 $\mu\text{eq L}^{-1}$).
456 Concentrations were lower in C3 (24 and 23 $\mu\text{eq L}^{-1}$) and intermediate in C4 (32 and 25 $\mu\text{eq L}^{-1}$).
457 The higher acidic-ion load in clusters that traversed continental East Asia is consistent with the
458 transport of anthropogenic sulfur- and nitrogen-containing pollutants (Klemm et al., 2015; Wu et
459 al., 2016; Nguyen et al., 2019). The concurrent occurrence of high acidic-ion concentrations and
460 low pH in C2 supports a substantial continental influence, while recognizing that local emissions
461 and meteorological differences may also contribute.

462 The $\text{nss-SO}_4^{2-}/\text{NO}_3^-$ equivalent ratio also varied among clusters: C2 had the highest value (1.67),
463 followed by C4 (1.59), C1 (1.54), and C3 (1.27). Ratios above 1 in all clusters indicate that nss-
464 SO_4^{2-} exceeded NO_3^- on an equivalent basis, but the lower ratio in C3 indicates a greater relative
465 contribution from nitrate. Air-mass history therefore influenced not only the total acidic-ion load
466 but also its sulfate–nitrate balance.

467 Neutralizing ions showed similar cluster dependence. NH_4^+ concentrations were 38 $\mu\text{eq L}^{-1}$ in
468 C1 and C2, compared with 29 $\mu\text{eq L}^{-1}$ in C3 and C4. Ca^{2+} was likewise higher in C2 (23 $\mu\text{eq L}^{-1}$)
469 and C1 (22 $\mu\text{eq L}^{-1}$) than in C3 (16 $\mu\text{eq L}^{-1}$). Continental transport was therefore associated with
470 higher concentrations of both acidic and neutralizing species. Nevertheless, C2 retained the lowest
471 pH, indicating that its greater neutralizing-ion content did not fully compensate for the

472 accompanying acidic load.

473 Sea-salt ions (Cl^- and Na^+) also varied among clusters. C2 had the highest Cl^- and Na^+
474 concentrations (101 and 91 $\mu\text{eq L}^{-1}$, respectively), whereas C3 had the lowest (28 and 26 $\mu\text{eq L}^{-1}$);
475 C1 and C4 were intermediate. These differences show that simple continental-versus-marine labels
476 do not fully describe the chemical history of an air mass. Sea-salt concentrations can depend on the
477 marine segments of the complete pathway, wind speed, boundary-layer mixing, storm development,
478 and precipitation amount. Accordingly, the high sea-salt concentrations in C2 cannot be attributed
479 to its origin alone, and the relatively low values in the maritime C3 cluster may reflect the
480 meteorological and scavenging characteristics of summer convective events.

481 Overall, the four transport regimes had distinct precipitation-chemistry signatures. C2 combined
482 the highest frequency, lowest VWM pH, and highest nss-SO_4^{2-} and NO_3^- concentrations, making it
483 the clearest continental/high-loading regime. C3 had the highest pH and lowest concentrations of
484 most major ions and was dominated by summer convective precipitation. It therefore provides a
485 useful low-transboundary-influence contrast, although it should not be interpreted as a purely local
486 or pollution-free baseline.

487 The contrast between C2 and C3 provides the basis for the following trend analysis. C2
488 represents frequent transport across continental source regions, whereas C3 represents
489 predominantly southerly transport with comparatively limited continental influence. Comparing

their long-term trends allows us to test whether the evolution of sulfate, nitrate, and neutralizing ions differed between these transport contexts. The analysis evaluates consistency with transboundary versus regional/local influences; it does not directly apportion sources.

3.4.2 Contrasting long-term trends under transboundary and regionally influenced transport regimes

Mann–Kendall tests and Sen’s slope estimates were applied separately to the four trajectory clusters (Table 5). VWM pH increased significantly, and nss-SO_4^{2-} decreased significantly in every cluster, showing that the mitigation of precipitation acidity was not restricted to a single transport pathway. Trend patterns for NO_3^- , NH_4^+ , and Ca^{2+} were less uniform among clusters. These within-cluster results describe contrasting temporal patterns; without a formal interaction or pairwise slope-difference test, differences in significance should not be interpreted as proof that cluster slopes differ statistically.

Within C2, the principal continental/transboundary-influenced regime, VWM pH increased significantly (Sen’s slope = $+0.0019 \text{ pH units observation}^{-1}$, $p < 0.001$), while nss-SO_4^{2-} decreased significantly (Sen’s slope = $-0.0008 \log_{10}(\mu\text{eq L}^{-1}) \text{ observation}^{-1}$, $p < 0.001$; Fig. 9) (Itahashi et al., 2018; Zhang et al., 2018). The concurrence of these trends is consistent with declining sulfate loading under transport conditions influenced by continental East Asia (Wang et al., 2014; Duan et al., 2016; Li et al., 2020). Neither NO_3^- nor NH_4^+ showed a decreasing trend (both Sen’s slope =

509 $-0.0001 \log_{10}(\mu\text{eq L}^{-1}) \text{ observation}^{-1}$), whereas Ca^{2+} increased significantly ($+0.0004 \log_{10}(\mu\text{eq L}^{-1}) \text{ observation}^{-1}$, $p < 0.05$). Thus, the pH increase in C2 was closely associated with declining nss-
510 SO_4^{2-} , with increased Ca^{2+} potentially providing additional neutralization.

512 C3 showed a contrasting combination of within-cluster trends. VWM pH increased significantly
513 (Sen's slope = $+0.0026 \text{ pH unit observation}^{-1}$, $p < 0.001$), while nss- SO_4^{2-} decreased significantly
514 (Sen's slope = $-0.0007 \log_{10}(\mu\text{eq L}^{-1}) \text{ observation}^{-1}$, $p < 0.01$), showing that pH recovery also
515 occurred under limited-transboundary-influence conditions. NO_3^- increased significantly within C3
516 (Sen's slope = $+0.0006 \log_{10}(\mu\text{eq L}^{-1}) \text{ observation}^{-1}$, $p < 0.05$), whereas its trend was nonsignificant
517 within C2.

518 Approximately 74% of C3 samples occurred during June–August and 81% during June–
519 September. In Taiwan, summer convective rainfall is strongly influenced by land–sea-breeze
520 circulation and terrain-induced convergence (John Kerns et al., 2010; Huang et al., 2015). Because
521 C3 is predominantly associated with summer southerly transport and afternoon convective
522 precipitation, the observed within-cluster nitrate trend may reflect a combination of long-term
523 changes in precipitation chemistry and the seasonal meteorological conditions characteristic of this
524 transport regime. The present event-based analysis does not explicitly separate seasonal
525 meteorological effects from long-term temporal change. Therefore, the positive NO_3^- trend in C3
526 reflects a regime-specific temporal association rather than evidence of a season-independent

increase in nitrate concentrations.

Furthermore, Ca^{2+} also increased significantly in C3 (Sen's slope = $+0.0012 \log_{10}(\mu\text{eq L}^{-1})$ observation⁻¹, $p < 0.001$) was concurrent with the decrease in nss-SO_4^{2-} and is consistent with the observed pH recovery. NH_4^+ showed no significant trend (Sen's slope = $+0.0004 \log_{10}(\mu\text{eq L}^{-1})$ observation⁻¹, $p > 0.005$). The increase in NO_3^- would act in the opposite direction and may have partly offset that recovery, underscoring that sulfur- and nitrogen-related changes were not synchronous.

C2 and C3 shared significant increases in pH and decreases in nss-SO_4^{2-} , whereas their NO_3^- trends differed in statistical significance: NO_3^- was nonsignificant in C2 but increased significantly in C3. This pattern suggests that nitrate may be more sensitive than sulfate to the combination of regional/local emissions, seasonal meteorology, and precipitation characteristics. However, because no formal interaction or pairwise slope-difference test was performed, the difference in statistical significance should not be interpreted as evidence that the C2 and C3 slopes are statistically different. In addition, the strong summer concentration of C3 events means that seasonal effects cannot be separated from the within-cluster temporal trend in the present analysis.

The trend analyses for C1 and C4 provide complementary evidence supporting these interpretations (Fig. S8). Although C1 recorded the lowest occurrence frequency (6.8%), it exhibited a significant upward pH trend (Sen's slope = $+0.0084 \text{ pH units observation}^{-1}$, $p < 0.001$),

545 accompanied by a significant decrease in nss-SO_4^{2-} (Sen's slope = $-0.0038 \log_{10}(\mu\text{eq L}^{-1})$
546 observation^{-1} , $p < 0.01$). This indicates a pronounced long-term increase in precipitation pH under
547 the C1 transport regime. C4 showed evolutionary characteristics similar to C2, with a significant
548 increase in pH (Sen's slope = $+0.0022 \text{ pH units observation}^{-1}$, $p < 0.001$) and a notable decrease in
549 nss-SO_4^{2-} (Sen's slope = $-0.0010 \log_{10}(\mu\text{eq L}^{-1}) \text{ observation}^{-1}$, $p < 0.001$), along with a minor yet
550 significant downward trend in NH_4^+ (Sen's slope = $-0.0004 \log_{10}(\mu\text{eq L}^{-1}) \text{ observation}^{-1}$, $p < 0.01$).
551 Overall, the trajectories of C1 and C4 align consistently with the upward pH and downward sulfate
552 trends observed in C2 and C3, further corroborating that the comprehensive mitigation of
553 precipitation acidification in Taoyuan City during the study period is robust and consistent across
554 diverse air mass transport regimes.

555 Across all four clusters, the most consistent changes were increasing pH and decreasing nss-
556 SO_4^{2-} . This common pattern identifies declining sulfate-related acidity as the principal regional
557 feature of the 2006–2024 record. NO_3^- behavior was less uniform: its trend was nonsignificant in
558 the continental C2 regime but significantly positive in the low-transboundary-influence C3 regime.
559 The cluster analysis therefore refines the network-scale findings in Section 3.3: sulfate declined
560 broadly across transport conditions, whereas nitrate showed greater variation with transport and
561 seasonal context. From a policy perspective, the results indicate substantial benefits from sulfur-
562 emission controls while highlighting persistent—and under some conditions increasing—nitrogen-

related contributions that warrant continued control of NO_x emissions at both regional and local scales.

4. CONCLUSIONS

This study integrated a five-station comparison of period-level chemistry (1994–2005 versus 2014–2024), station-specific trend analysis at the four non-Chungli sites, a continuous 30-year record at Chungli, and trajectory-cluster analysis to characterize the long-term evolution of precipitation chemistry in highly urbanized and industrialized Taoyuan City, northwestern Taiwan. Network VWM pH increased from 4.56 in Phase I to 5.07 in Phase II, while VWM H⁺ decreased by approximately 69%. The continuous Chungli record, based on chronological individual precipitation-event observations, independently confirmed a significant multidecadal increase in pH. Together, the spatially coherent network response and event-based continuous trend analysis demonstrate sustained recovery from the more acidic conditions of the 1990s.

The principal compositional change associated with this recovery was the pronounced decrease in sulfur-related acidity. Network VWM nss-SO₄²⁻ declined by approximately 54%, and significant downward trends were observed across all four trajectory regimes. In contrast, NO₃⁻ changed little between the two network periods but showed a small yet statistically significant upward trend in the continuous Chungli record. The declining nss-SO₄²⁻/NO₃⁻ equivalent ratio therefore identifies

a transition from stronger sulfate dominance toward a greater relative contribution from nitrate. This finding indicates that declining sulfur loading has altered the relative contributions of sulfur- and nitrogen-related acidity, while nitrogen-related inputs may represent an increasingly important constraint on further recovery.

Changes in neutralizing species also modified the precipitation acid–base balance. NH_4^+ declined only modestly over the long term and continued to provide substantial neutralization, whereas Ca^{2+} was higher in Phase II and increased across the complete Chungli record despite decreasing during 2014–2024. These patterns are consistent with additional alkaline inputs from crustal or resuspended material, but the available ionic measurements cannot distinguish long-range dust from local construction, road dust, or soil sources. Accordingly, the increase in pH is best explained by the combined influence of lower sulfate loading and changes in alkaline neutralization, with the sulfate decline providing the most consistent signal across stations and transport regimes.

Trajectory clustering further clarified how this chemical transition was expressed under different transport contexts. The continental C2 regime was the most frequent cluster and had the lowest pH and highest nss-SO_4^{2-} and NO_3^- concentrations, confirming that continental transport remained associated with high acidic-ion loading. Nevertheless, pH increased, and nss-SO_4^{2-} decreased in every cluster, indicating that long-term mitigation was not confined to a single pathway. The

600 within-cluster NO_3^- results contrasted: no significant trend occurred in C2, whereas NO_3^- increased
601 significantly in the predominantly summer, southerly C3 regime with comparatively limited
602 continental influence. This contrast is consistent with an increasing relative influence of regional
603 and local nitrogen sources under convective precipitation conditions, but the current analysis does
604 not establish statistically different C2 and C3 slopes. Moreover, trajectory clusters do not constitute
605 quantitative source apportionment and cannot fully separate transport effects from seasonality,
606 precipitation amount, and atmospheric processing.

607 The combined network, continuous-record, and trajectory evidence provides a policy-relevant
608 benchmark for evaluating changes in East Asian acid deposition. Continued sulfur-emission control
609 remains necessary to preserve the gains already achieved, while further improvement may
610 increasingly benefit from continued reductions in NO_x emissions from transportation, industrial
611 combustion, and other urban sources, together with sustained management of NH_3 emissions.
612 Future studies should combine precipitation chemistry with sulfur and nitrogen isotopes, receptor
613 modeling, emission inventories, and higher-resolution trajectory or chemical-transport modeling to
614 quantify source contributions. Event-based analyses that explicitly account for rainfall amount,
615 season, aerosol composition, and below-cloud scavenging would help test the proposed
616 regional/local nitrate influence. Continued long-term monitoring is also essential for determining
617 whether changing monsoon circulation, extreme precipitation, and urban development modify the

emerging sulfate–nitrate balance and wet-deposition fluxes in northern Taiwan.

ACKNOWLEDGEMENTS

This study was supported by the Department of Environmental Protection, Taoyuan, and the Ministry of Environment, Taiwan, which provided acid rain monitoring data. The authors gratefully acknowledge the assistance and support provided by staff at all Taoyuan Acid Rain Monitoring Network stations.

FUNDING

This research was supported by the Department of Environmental Protection, Taoyuan City Government under Grant 114010610.

COMPETING INTERESTS

The authors declare that they have no competing interests.

AUTHOR CONTRIBUTIONS

T. conceived and designed the study, formulated the research objectives, performed data processing and analysis, and wrote the original manuscript; L. performed data validation and data

processing; D. performed data calculations and visualization; Y. Y. and W. contributed to data collection and provided the precipitation chemistry monitoring data; Moo-Been Chang contributed to the investigation; and Neng-Huei Lin supervised the study and critically reviewed, edited, and revised the manuscript. All authors reviewed and approved the final manuscript.

DATA AVAILABILITY

The datasets generated and analyzed during the current study are available from the corresponding author on reasonable request.

ETHICS APPROVAL

Ethics declaration: not applicable.

CONSENT TO PARTICIPATE

Consent to Participate declaration: not applicable.

REFERENCES

Agrawal, A.S.a.M. (2008). Acid Rain and Its Ecological Consequences. *J. Environ. Biol.* 29(1): 15–24.

662 Aikawa, M., Ohara, T., Hiraki, T., Oishi, O., Tsuji, A., Yamagami, M., Murano, K. and Mukai, H.
 663 (2010). Significant Geographic Gradients in Particulate Sulfate over Japan Determined
 664 from Multiple-Site Measurements and a Chemical Transport Model: Impacts of
 665 Transboundary Pollution from the Asian Continent. *Atmos. Environ.* 44: 381–391.

666 Akimoto, H., Sato, K., Sase, H., Dong, Y., Hu, M., Duan, L., Sunwoo, Y., Suzuki, K. and Tang,
 667 X. (2022). Development of Science and Policy Related to Acid Deposition in East Asia
 668 over 30 years. *Ambio* 51: 1800–1818.

669 Anttila, P., Paatero, P., Tapper, U. and Järvinen, O. (1995). Source Identification of Bulk Wet
 670 Deposition in Finland by Positive Matrix Factorization. *Atmos. Environ.* 29: 1705–1718.

671 Bhuyan, P., Ahmed, M.S., Hopke, P.K. and Hoque, R.R. (2020). Understanding the Chemistry
 672 and Sources of Precipitation Ions in the Mid-Brahmaputra Valley of Northeastern India.
 673 *Aerosol Air Qual. Res.* 20: 2690–2704.

674 Chang, C.-T., Wang, C.-P., Huang, J.-C., Wang, L.-J., Liu, C.-P. and Lin, T.-C. (2017). Trends of
 675 Two Decadal Precipitation Chemistry in a Subtropical Rainforest in East Asia. *Sci. Total*
 676 *Environ.* 605-606: 88–98.

677 Chang, C.-T., Yang, C.-J., Huang, K.-H., Huang, J.-C. and Lin, T.-C. (2022). Changes of
 678 Precipitation Acidity Related to Sulfur and Nitrogen Deposition in Forests across Three
 679 Continents in North Hemisphere over Last Two Decades. *Sci. Total Environ.* 806: 150552.

680 Chang, C.-T., Yang, C.-J., Wang, C.-H., Huang, J.-C., Lin, N.-H. and Lin, T.-C. (2026). Divergent
681 Contributions of Acidic and Neutralization Potential to Precipitation Acidity across Islet,
682 Urban and Forest Sites, Northern Taiwan. *Environ. Pollut.* 406: 128630.

683 Chang, K.-H., Jeng, F.-T., Tsai, Y.-L. and Lin, P.-L. (2000). Modeling of Long-Range Transport
684 on Taiwan's Acid Deposition under Different Weather Conditions. *Atmos. Environ.* 34:
685 3281–3295.

686 Chang, S.-C., Chou, C.C.K., Chen, W.-N. and Lee, C.-T. (2010). Asian Dust and Pollution
687 Transport—a Comprehensive Observation in the Downwind Taiwan in 2006. *Atmos. Res.*
688 95: 19–31.

689 Chen, T.-F., Chang, K.-H. and Tsai, C.-Y. (2014). Modeling Direct and Indirect Effect of Long
690 Range Transport on Atmospheric Pm2.5 Levels. *Atmos. Environ.* 89: 1–9.

691 Chen, T.-F., Tsai, C.-Y. and Chang, K.-H. (2013). Performance Evaluation of Atmospheric
692 Particulate Matter Modeling for East Asia. *Atmos. Environ.* 77: 365–375.

693 Chuang, M.T., Ooi, M.C.G., Lin, N.H., Fu, J.S., Lee, C.T., Wang, S.H., Yen, M.C., Kong, S.S.K.
694 and Huang, W.S. (2020). Study on the Impact of Three Asian Industrial Regions on Pm2.5
695 in Taiwan and the Process Analysis during Transport. *Atmos. Chem. Phys.* 20: 14947–
696 14967.

697 de Vries, W. and Kros, H. (1989). The Long-Term Impact of Acid Deposition on the Aluminum

698 Chemistry of an Acid Forest Soil, In *Regional Acidification Models: Geographic Extent*
699 *and Time Development*, Springer, pp. 113–128.

700 Do, L.T.T., Griffith, S.M., Tseng, W.-T. and Lin, N.-H. (2021). Long-Term Trend of Wintertime
701 Precipitation Chemistry at a Remote Islet Site Influenced by Anthropogenic Emissions
702 from Continental East Asia. *Atmos. Environ.* 262.

703 Duan, L., Yu, Q., Zhang, Q., Wang, Z., Pan, Y., Larssen, T., Tang, J. and Mulder, J. (2016). Acid
704 Deposition in Asia: Emissions, Deposition, and Ecosystem Effects. *Atmos. Environ.* 146:
705 55–69.

706 Fedkin, N.M., Li, C., Dickerson, R.R., Canty, T. and Krotkov, N.A. (2019). Linking
707 Improvements in Sulfur Dioxide Emissions to Decreasing Sulfate Wet Deposition by
708 Combining Satellite and Surface Observations with Trajectory Analysis. *Atmos. Environ.*
709 199: 210–223.

710 Galloway, J.N., Dianwu, Z., Jiling, X. and Likens, G.E. (1987). Acid Rain: China, United States,
711 and a Remote Area. *Science* 236: 1559–1562.

712 Ge, B., Wang, Z., Gbaguidi, A.E. and Zhang, Q. (2016). Source Identification of Acid Rain
713 Arising over Northeast China: Observed Evidence and Model Simulation. *Aerosol Air*
714 *Qual. Res.* 16: 1366–1377.

715 Gilbert, R.O. (1987). *Statistical Methods for Environmental Pollution Monitoring*. John Wiley &

716 Sons.

717 Grennfelt, P., Engleryd, A., Forsius, M., Hov, O., Rodhe, H. and Cowling, E. (2020). Acid Rain
718 and Air Pollution: 50 Years of Progress in Environmental Science and Policy. *Ambio* 49:
719 849–864.

720 Grider, A., Ponette-González, A. and Heindel, R. (2023). Calcium and Ammonium Now Control
721 the Ph of Wet and Bulk Deposition in Ohio, U.S. *Atmos. Environ.* 310.

722 Griffith, S.M., Huang, W.-S., Lin, C.-C., Chen, Y.-C., Chang, K.-E., Lin, T.-H., Wang, S.-H. and
723 Lin, N.-H. (2020). Long-Range Air Pollution Transport in East Asia during the First Week
724 of the Covid-19 Lockdown in China. *Sci. Total Environ.* 741: 140214.

725 Guo, T., Li, M., He, S., Mo, Z., Kang, X., Pei, J., Liao, W., Chang, M. and Wang, X. (2025).
726 Characterization of Atmospheric Arsenic Wet Deposition Transport Pathways and
727 Potential Sources Areas in the Pearl River Delta Region. *J. Environ. Sci.* 158: 372–385.

728 Holland, D.M., Principe, P.P. and Sickles, J.E. (1998). Trends in Atmospheric Sulfur and Nitrogen
729 Species in the Eastern United States for 1989–1995. *Atmos. Environ.* 33: 37–49.

730 Huang, D.-Y., Xu, Y.-G., Peng, P.a., Zhang, H.-H. and Lan, J.-B. (2009). Chemical Composition
731 and Seasonal Variation of Acid Deposition in Guangzhou, South China: Comparison with
732 Precipitation in Other Major Chinese Cities. *Environ. Pollut.* 157: 35–41.

733 Huang, W.R., Hsu, H.H., Wang, S.-y. and Chen, J.-P. (2015). Impact of Atmospheric Changes on

734 the Low-Frequency Variations of Convective Afternoon Rainfall Activity over Taiwan. *J.*
735 *Geophys. Res. Atmos.* 120: 8743 – 8758.

736 Inomata, Y., Ohizumi, T., Saito, T., Morohashi, M., Yamashita, N., Takahashi, M., Sase, H.,
737 Takahashi, K., Kaneyasu, N., Fujihara, M., Iwasaki, A., Nakagomi, K., Shiroma, T. and
738 Yamaguchi, T. (2019). Estimating Transboundary Transported Anthropogenic Sulfate
739 Deposition in Japan Using the Sulfur Isotopic Ratio. *Sci. Total Environ.* 691: 779–788.

740 Itahashi, S., Uno, I., Hayami, H. and Fujita, S.-i. (2015). Variation of the Ratio of Nitrate to Non-
741 Seasalt Sulfate in Precipitation over East Asia with Emissions from China. *Atmos.*
742 *Environ.* 118: 87–97.

743 Itahashi, S., Yumimoto, K., Uno, I., Hayami, H., Fujita, S.I., Pan, Y. and Wang, Y. (2018). A 15-
744 Year Record (2001–2015) of the Ratio of Nitrate to Non-Sea-Salt Sulfate in Precipitation
745 over East Asia. *Atmos. Chem. Phys.* 18: 2835–2852.

746 John Kerns, B.W., Chen, Y.-L. and Chang, M.-Y. (2010). The Diurnal Cycle of Winds, Rain, and
747 Clouds over Taiwan during the Mei-Yu, Summer, and Autumn Rainfall Regimes. *Mon.*
748 *Weather Rev.* 138: 497–516.

749 Kalnay, E., Kanamitsu, M., Kistler, R., Collins, W., Deaven, D., Gandin, L., Iredell, M., Saha, S.,
750 White, G., Woollen, J., Zhu, Y., Chelliah, M., Ebisuzaki, W., Higgins, W., Janowiak, J.,
751 Mo, K.C., Ropelewski, C., Wang, J., Leetmaa, A., Reynolds, R., Jenne, R. and Joseph, D.

752 (1996). The Ncep/Ncar 40-Year Reanalysis Project. *Bull. Am. Meteorol. Soc.* 77: 437–471.
 753 Kaneyasu, N., Yamamoto, S., Sato, K., Takami, A., Hayashi, M., Hara, K., Kawamoto, K.,
 754 Okuda, T. and Hatakeyama, S. (2014). Impact of Long-Range Transport of Aerosols on
 755 the Pm2.5 Composition at a Major Metropolitan Area in the Northern Kyushu Area of
 756 Japan. *Atmos. Environ.* 97: 416–425.
 757 Kato, Y., Ohizumi, T., Nakata, M. and Sase, H. (2025). Temporal and Seasonal Variations in the
 758 Frequency of High-Concentration Precipitation Samples at Eanet Sites in Japan: 20 years
 759 of Daily Wet Deposition Data. *Asian J. Atmos. Environ.* 19: 15.
 760 Keene, W.C., Pszenny, A.A.P., Galloway, J.N. and Hawley, M.E. (1986). Sea-Salt Corrections and
 761 Interpretation of Constituent Ratios in Marine Precipitation. *J. Geophys. Res. Atmos.* 91:
 762 6647–6658.
 763 Kim, B.-U., Bae, C., Kim, H.C., Kim, E. and Kim, S. (2017a). Spatially and Chemically
 764 Resolved Source Apportionment Analysis: Case Study of High Particulate Matter Event.
 765 *Atmos. Environ.* 162: 55–70.
 766 Kim, H.-S., Kim, D.-S., Kim, H. and Yi, S.-M. (2012). Relationship between Mortality and Fine
 767 Particles during Asian Dust, Smog–Asian Dust, and Smog Days in Korea. *Int. J. Environ.*
 768 *Health Res.* 22: 518–530.
 769 Kim, H.C., Kim, E., Bae, C., Cho, J.H., Kim, B.U. and Kim, S. (2017b). Regional Contributions

770 to Particulate Matter Concentration in the Seoul Metropolitan Area, South Korea:
771 Seasonal Variation and Sensitivity to Meteorology and Emissions Inventory. *Atmos.*
772 *Chem. Phys.* 17: 10315–10332.

773 Klemm, O., Tseng, W.-T., Lin, C.-C., Klemm, K. and Lin, N.-H. (2015). Ph Control in Fog and
774 Rain in East Asia: Temporal Advection of Clean Air Masses to Mt. Bamboo, Taiwan.
775 *Atmos.* 6: 1785–1800.

776 Kong, S., Pani, S., Griffith, S., Ou-Yang, C.-F., Ravindra Babu, S., Chuang, M.-T., Ooi, M.C.G.,
777 Huang, W.-S., Sheu, G.-R. and Lin, N.-H. (2022). Distinct Transport Mechanisms of East
778 Asian Dust and the Impact on Downwind Marine and Atmospheric Environments. *Sci.*
779 *Total Environ.* 827: 154255.

780 Koo, Y.-S., Kim, S.-T., Cho, J.-S. and Jang, Y.-K. (2012). Performance Evaluation of the Updated
781 Air Quality Forecasting System for Seoul Predicting Pm10. *Atmos. Environ.* 58: 56–69.

782 Kurokawa, J. and Ohara, T. (2020). Long-Term Historical Trends in Air Pollutant Emissions in
783 Asia: Regional Emission Inventory in Asia (Reas) Version 3. *Atmos. Chem. Phys.* 20:
784 12761–12793.

785 Lee, S., Ho, C.-H., Lee, Y.G., Choi, H.-J. and Song, C.-K. (2013). Influence of Transboundary Air
786 Pollutants from China on the High-Pm10 Episode in Seoul, Korea for the Period October
787 16–20, 2008. *Atmos. Environ.* 77: 430–439.

788 Lei, W., Li, X., Yang, X., Zhang, L., Li, X., Zhao, W. and Pan, Y. (2026). Seasonal
 789 Characteristics, Sources, and Regional Transport Patterns of Precipitation Components at
 790 High-Elevation Mountain in South China. *Atmos.* 17: 87.

791 Li, X., Zhao, B., Zhou, W., Shi, H., Yin, R., Cai, R., Yang, D., Dällenbach, K., Deng, C., Fu, Y.,
 792 Qiao, X., Wang, L., Liu, Y., Yan, C., Kulmala, M., Zheng, J., Hao, J., Wang, S. and Jiang,
 793 J. (2020). Responses of Gaseous Sulfuric Acid and Particulate Sulfate to Reduced SO_2
 794 Concentration: A Perspective from Long-Term Measurements in Beijing. *Sci. Total*
 795 *Environ.* 721: 137700.

796 Likens, G.E., Butler, T.J., Claybrooke, R., Vermeylen, F. and Larson, R. (2021). Long-Term
 797 Monitoring of Precipitation Chemistry in the U.S.: Insights into Changes and Condition.
 798 *Atmos. Environ.* 245: 118031.

799 Lin, C., Huo, T., Yang, F., Wang, B., Chen, Y. and Wang, H. (2020). Characteristics of Water-
 800 Soluble Inorganic Ions in Aerosol and Precipitation and Their Scavenging Ratios in an
 801 Urban Environment in Southwest China. *Aerosol Air Qual. Res.* 21: 200513.

802 Lin, C.Y., Wang, Z., Chen, W.N., Chang, S.Y., Chou, C.C.K., Sugimoto, N. and Zhao, X. (2007).
 803 Long-Range Transport of Asian Dust and Air Pollutants to Taiwan: Observed Evidence
 804 and Model Simulation. *Atmos. Chem. Phys.* 7: 423–434.

805 Lin, N.-H. and Peng, C.-M. (1999). Estimates of the Contribution of Below-Cloud Scavenging to

806 the Pollutant Loadings of Rain in Taipei, Taiwan. *Terrestrial, Atmospheric and Oceanic*
807 *Sciences* 10: 693–704.

808 Lu, Z., Zhang, Q. and Streets, D.G. (2011). Sulfur Dioxide and Primary Carbonaceous Aerosol
809 Emissions in China and India, 1996–2010. *Atmos. Chem. Phys.* 11: 9839–9864.

810 Marchetto, A., Rogora, M. and Arisci, S. (2013). Trend Analysis of Atmospheric Deposition Data:
811 A Comparison of Statistical Approaches. *Atmos. Environ.* 64: 95–102.

812 Martins, E.H., Nogarotto, D.C., Mortatti, J. and Pozza, S.A. (2019). Chemical Composition of
813 Rainwater in an Urban Area of the Southeast of Brazil. *Atmos. Pollut. Res.* 10: 520–530.

814 Nguyen, L.S.P., Sheu, G.-R., Lin, D.-W. and Lin, N.-H. (2019). Temporal Changes in
815 Atmospheric Mercury Concentrations at a Background Mountain Site Downwind of the
816 East Asia Continent in 2006–2016. *Sci. Total Environ.* 686: 1049–1056.

817 Oh, H.-R., Ho, C.-H., Kim, J., Chen, D., Lee, S., Choi, Y.-S., Chang, L.-S. and Song, C.-K.
818 (2015). Long-Range Transport of Air Pollutants Originating in China: A Possible Major
819 Cause of Multi-Day High-Pm10 Episodes during Cold Season in Seoul, Korea. *Atmos.*
820 *Environ.* 109: 23–30.

821 Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X. and Hayasaka, T. (2007). An
822 Asian Emission Inventory of Anthropogenic Emission Sources for the Period
823 1980–2020. *Atmos. Chem. Phys.* 7: 4419–4444.

824 Ohizumi, T., Take, N., Inomata, Y., Yagoh, H., Endo, T., Takahashi, M., Yanahara, K. and
825 Kusakabe, M. (2016). Long-Term Variation of the Source of Sulfate Deposition in a
826 Leeward Area of Asian Continent in View of Sulfur Isotopic Composition. *Atmos.*
827 *Environ.* 140: 42–51.

828 Park, S.-M., Seo, B.-K., Lee, G., Kahng, S.-H. and Jang, Y.W. (2015). Chemical Composition of
829 Water Soluble Inorganic Species in Precipitation at Shihwa Basin, Korea. *Atmos.* 6: 732–
830 750.

831 Pascaud, A., Sauvage, S., Coddeville, P., Nicolas, M., Croisé, L., Mezdoor, A. and Probst, A.
832 (2016). Contrasted Spatial and Long-Term Trends in Precipitation Chemistry and
833 Deposition Fluxes at Rural Stations in France. *Atmos. Environ.* 146: 28–43.

834 Prakash, J., Agrawal, S.B. and Agrawal, M. (2023). Global Trends of Acidity in Rainfall and Its
835 Impact on Plants and Soil. *J. Soil Sci. Plant Nutr.* 23: 398–419.

836 Pu, W., Quan, W., Ma, Z., Shi, X., Zhao, X., Zhang, L., Wang, Z. and Wang, W. (2017). Long-
837 Term Trend of Chemical Composition of Atmospheric Precipitation at a Regional
838 Background Station in Northern China. *Sci. Total Environ.* 580: 1340–1350.

839 Rattigan, O.V., Civerolo, K.L. and Felton, H.D. (2017). Trends in Wet Precipitation, Particulate,
840 and Gas-Phase Species in New York State. *Atmos. Pollut. Res.* 8: 1090–1102.

841 Roy, A., Chatterjee, A., Tiwari, S., Sarkar, C., Das, S.K., Ghosh, S.K. and Raha, S. (2016).

842 Precipitation Chemistry over Urban, Rural and High Altitude Himalayan Stations in
843 Eastern India. *Atmos. Res.* 181: 44–53.

844 Schindler, D.W. (1988). Effects of Acid Rain on Freshwater Ecosystems. *Science* 239: 149–157.

845 Seinfeld, J.H. and Pandis, S.N. (2016). *Atmospheric Chemistry and Physics: From Air Pollution*
846 *to Climate Change*. John Wiley & Sons.

847 Sen, P.K. (1968). Estimates of the Regression Coefficient Based on Kendall's Tau. *J. Am. Stat.*
848 *Assoc.* 63: 1379–1389.

849 Shah, V., Jacob, D.J., Moch, J.M., Wang, X. and Zhai, S. (2020). Global Modeling of Cloud
850 Water Acidity, Precipitation Acidity, and Acid Inputs to Ecosystems. *Atmos. Chem. Phys.*
851 20: 12223–12245.

852 Sheu, G.-R. and Lin, N.-H. (2013). Characterizations of Wet Mercury Deposition to a Remote
853 Islet (Pengjiayu) in the Subtropical Northwest Pacific Ocean. *Atmos. Environ.* 77: 474–
854 481.

855 Sheu, G.-R., Lin, N.-H., Wang, J.-L., Lee, C.-T., Ou Yang, C.-F. and Wang, S.-H. (2010).
856 Temporal Distribution and Potential Sources of Atmospheric Mercury Measured at a
857 High-Elevation Background Station in Taiwan. *Atmos. Environ.* 44: 2393–2400.

858 Sheu, G.-R., Phu Nguyen, L.S., Truong, M.T. and Lin, D.-W. (2019). Characteristics of
859 Atmospheric Mercury at a Suburban Site in Northern Taiwan and Influence of Trans-

860 Boundary Haze Events. *Atmos. Environ.* 214: 116827.

861 Shi, Z., Zhang, J., Xiao, Z., Lu, T., Ren, X. and Wei, H. (2021). Effects of Acid Rain on Plant
862 Growth: A Meta-Analysis. *J. Environ. Manag.* 297: 113213.

863 Stein, A.F., Draxler, R.R., Rolph, G.D., Stunder, B.J.B., Cohen, M.D. and Ngan, F. (2015). Noaa's
864 Hysplit Atmospheric Transport and Dispersion Modeling System. *Bull. Am. Meteorol.*
865 *Soc.* 96: 2059–2077.

866 Stohl, A. (1998). Computation, Accuracy and Applications of Trajectories—a Review and
867 Bibliography. *Atmos. Environ.* 32: 947–966.

868 Su, Z., Wang, Y., Zheng, Y., Wang, Y., Li, P. and Zhang, X. (2024). Acid Deposition and
869 Meteorological Factors Together Drive Changes in Vegetation Cover in Acid Rain Areas.
870 *Ecol. Indic.* 167: 112720.

871 Sun, M., Wang, Y., Wang, T., Fan, S., Wang, W., Li, P., Guo, J. and Li, Y. (2010). Cloud and the
872 Corresponding Precipitation Chemistry in South China: Water-Soluble Components and
873 Pollution Transport. *J. Geophys. Res. Atmos.* 115.

874 Tang, A., Zhuang, G., Wang, Y., Yuan, H. and Sun, Y. (2005). The Chemistry of Precipitation and
875 Its Relation to Aerosol in Beijing. *Atmos. Environ.* 39: 3397–3406.

876 Vellingiri, K., Kim, K.-H., Lim, J.-M., Lee, J.-H., Ma, C.-J., Jeon, B.-H., Sohn, J.-R., Kumar, P.
877 and Kang, C.-H. (2016). Identification of Nitrogen Dioxide and Ozone Source Regions

878 for an Urban Area in Korea Using Back Trajectory Analysis. *Atmos. Res.* 176-177: 212–
879 221.

880 Vet, R., Artz, R.S., Carou, S., Shaw, M., Ro, C.-U., Aas, W., Baker, A., Bowersox, V.C., Dentener,
881 F., Galy-Lacaux, C., Hou, A., Pienaar, J.J., Gillett, R., Forti, M.C., Gromov, S., Hara, H.,
882 Khodzher, T., Mahowald, N.M., Nickovic, S., Rao, P.S.P. and Reid, N.W. (2014). A
883 Global Assessment of Precipitation Chemistry and Deposition of Sulfur, Nitrogen, Sea
884 Salt, Base Cations, Organic Acids, Acidity and Ph, and Phosphorus. *Atmos. Environ.* 93:
885 3–100.

886 Wang, S., Zhang, X., Ma, Y., Li, X., Cheng, M., Zhang, X. and Liu, L. (2018). Detecting Sulfuric
887 and Nitric Acid Rain Stresses on *Quercus Glauca* through Hyperspectral Responses.
888 *Sensors* 18: 830.

889 Wang, S.X., Zhao, B., Cai, S.Y., Klimont, Z., Nielsen, C.P., Morikawa, T., Woo, J.H., Kim, Y., Fu,
890 X., Xu, J.Y., Hao, J.M. and He, K.B. (2014). Emission Trends and Mitigation Options for
891 Air Pollutants in East Asia. *Atmos. Chem. Phys.* 14: 6571–6603.

892 Wang, X., Liu, Z., Niu, L. and Fu, B. (2013). Long-Term Effects of Simulated Acid Rain Stress
893 on a Staple Forest Plant, *Pinus Massoniana* Lamb: A Proteomic Analysis. *Trees* 27: 297–
894 309.

895 Wu, Q., Wang, S., Li, G., Liang, S., Lin, C.-J., Wang, Y., Cai, S., Liu, K. and Hao, J. (2016).

896 Temporal Trend and Spatial Distribution of Speciated Atmospheric Mercury Emissions in
897 China during 1978–2014. *Environ. Sci. Technol.* 50: 13428–13435.

898 Yim, S.H.L., Gu, Y., Shapiro, M.A. and Stephens, B. (2019). Air Quality and Acid Deposition
899 Impacts of Local Emissions and Transboundary Air Pollution in Japan and South Korea.
900 *Atmos. Chem. Phys.* 19: 13309–13323.

901 Zeng, J., Han, G., Zhang, S., Xiao, X., Li, Y., Gao, X., Wang, D. and Qu, R. (2022). Rainwater
902 Chemical Evolution Driven by Extreme Rainfall in Megacity: Implication for the Urban
903 Air Pollution Source Identification. *J. Clean. Prod.* 372: 133732.

904 Zhang, L., Wang, J., Wang, S., Wang, C., Yang, F. and Li, T. (2024). Chemical Characteristics of
905 Long-Term Acid Rain and Its Impact on Lake Water Chemistry: A Case Study in
906 Southwest China. *J. Environ. Sci.* 138: 121–131.

907 Zhang, X.Y., Chuai, X.W., Liu, L., Zhang, W.T., Lu, X.H., Zhao, L.M. and Chen, D.M. (2018).
908 Decadal Trends in Wet Sulfur Deposition in China Estimated from OMI SO₂ Columns. *J.*
909 *Geophys. Res. Atmos.* 123.

910 Zheng, B., Tong, D., Li, M., Liu, F., Hong, C., Geng, G., Li, H., Li, X., Peng, L., Qi, J., Yan, L.,
911 Zhang, Y., Zhao, H., Zheng, Y., He, K. and Zhang, Q. (2018). Trends in China's
912 Anthropogenic Emissions since 2010 as the Consequence of Clean Air Actions. *Atmos.*
913 *Chem. Phys.* 18: 14095–14111.

914 Zhou, K., Xu, W., Liu, X. and Zhao, Y. (2025). China Played a Greater Role in Reducing the
915 Deposition of Japan and Korea Than Themselves. *Environ. Sci. Technol.* 59: 24888–
916 24898.

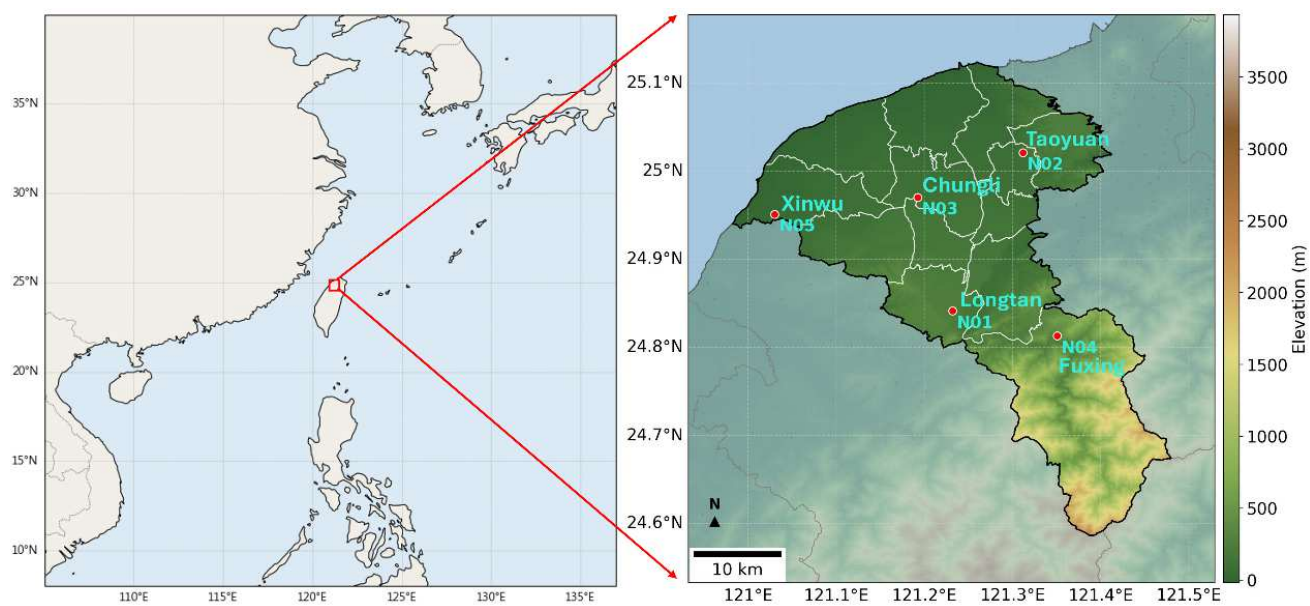


Fig. 1. Location of Taoyuan City and the sampling sites.

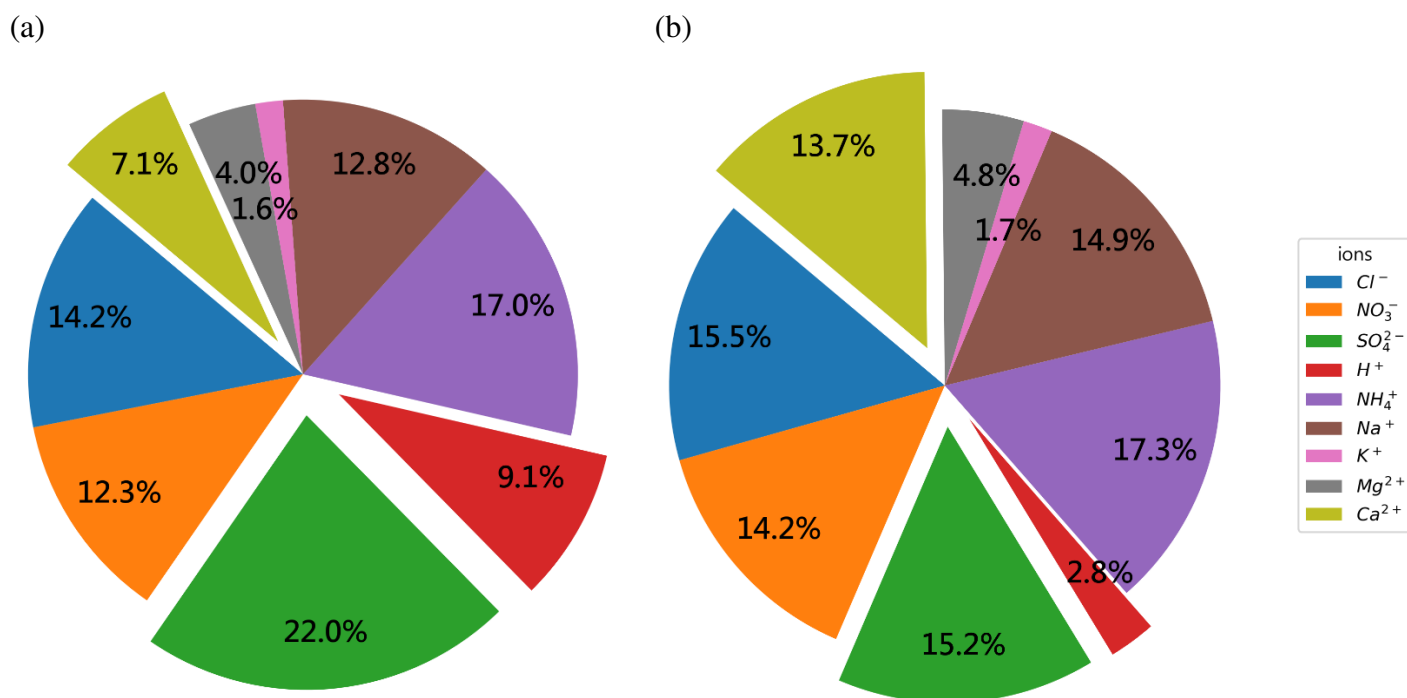
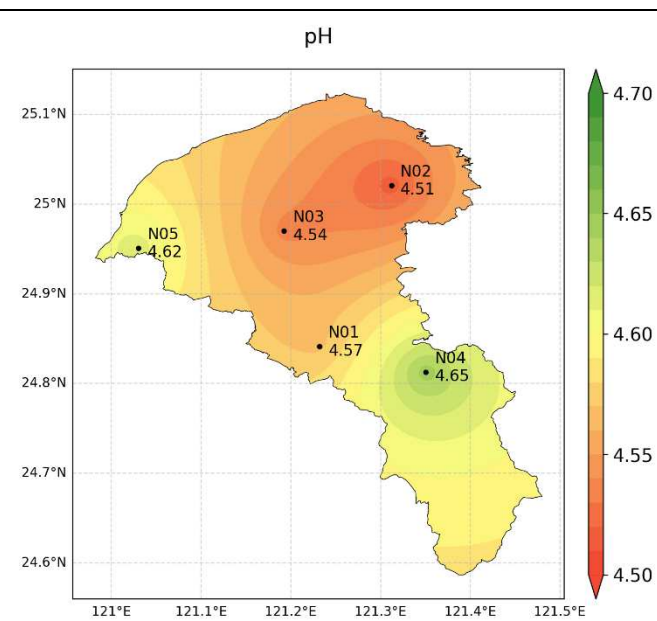


Fig. 2. Percentage composition of major ionic species in precipitation during the periods of 1994–2005 and 2014–2024.

(a) Phase I (1994-2005)



(b) Phase II (2014-2024)

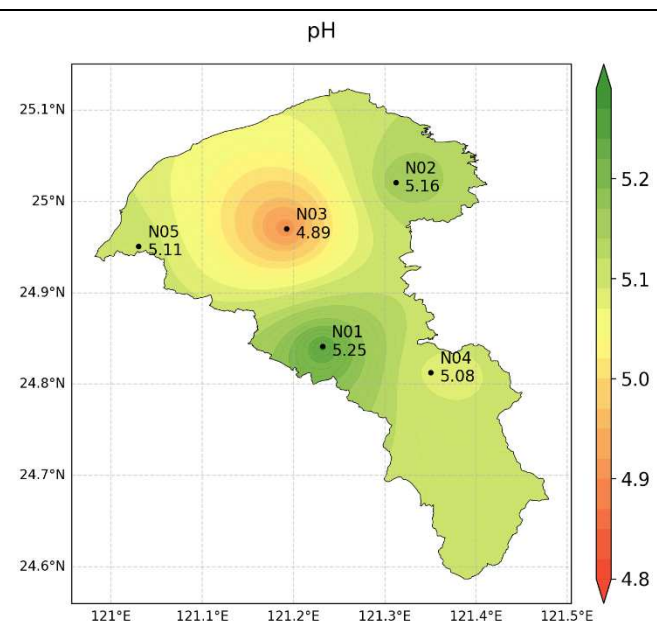


Fig. 3. Spatial distributions of volume-weighted mean (VWM) pH during Phase I and Phase II.

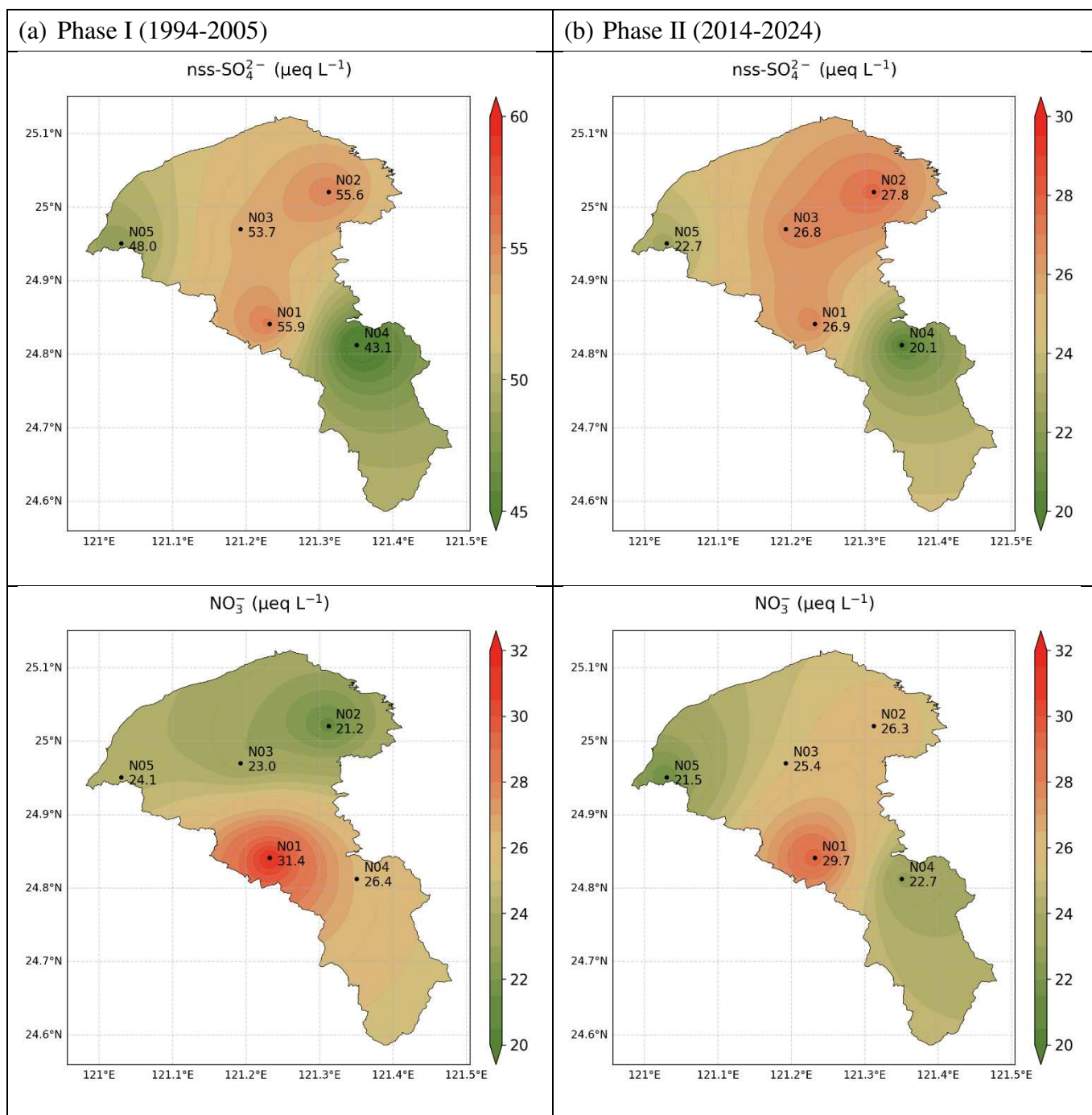


Fig. 4. Spatial distributions of VWM nss-SO_4^{2-} and NO_3^- concentrations during Phase I and Phase II.

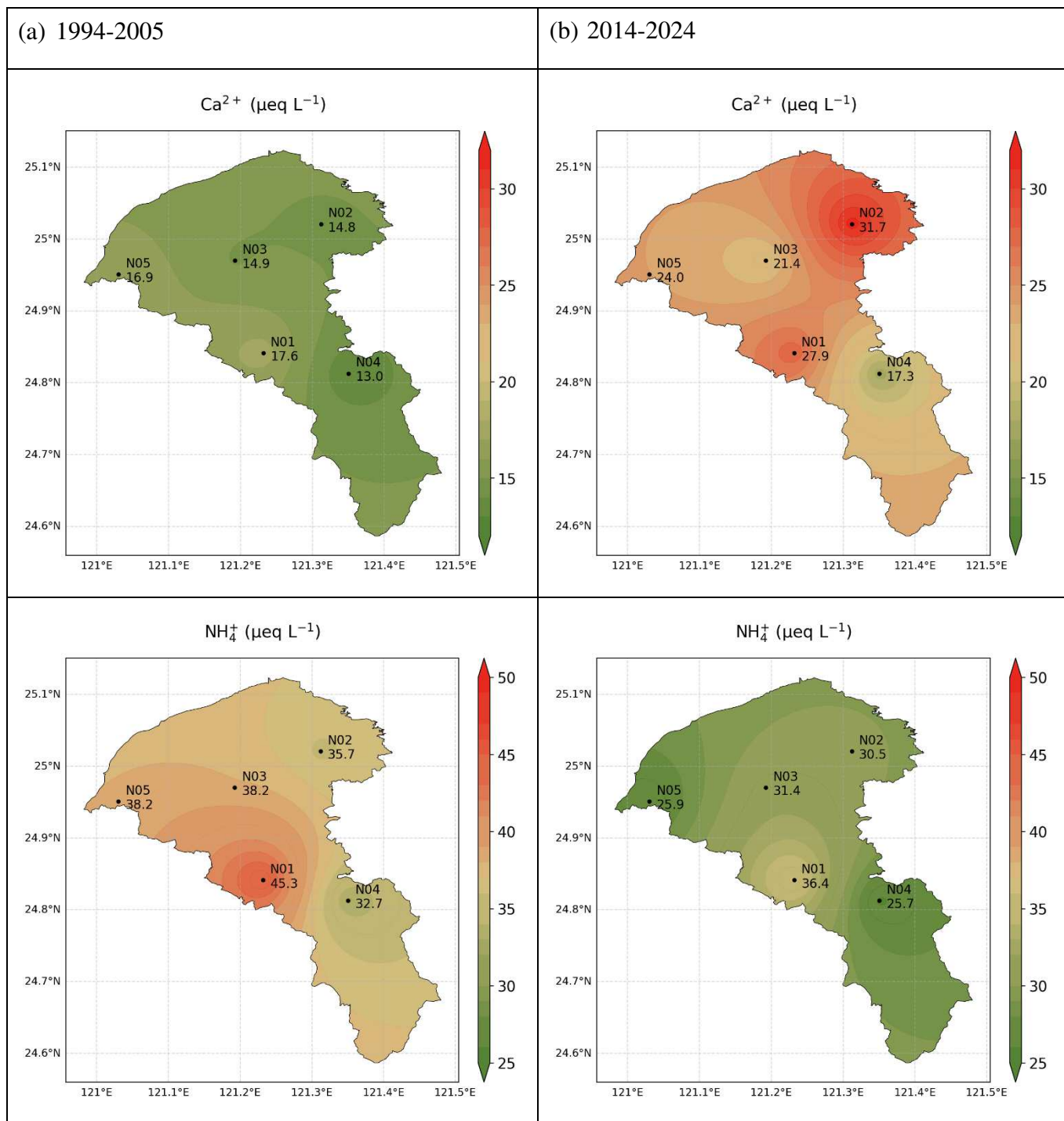


Fig. 5. Spatial distributions of VWM Ca^{2+} and NH_4^+ concentrations during Phase I and Phase II.

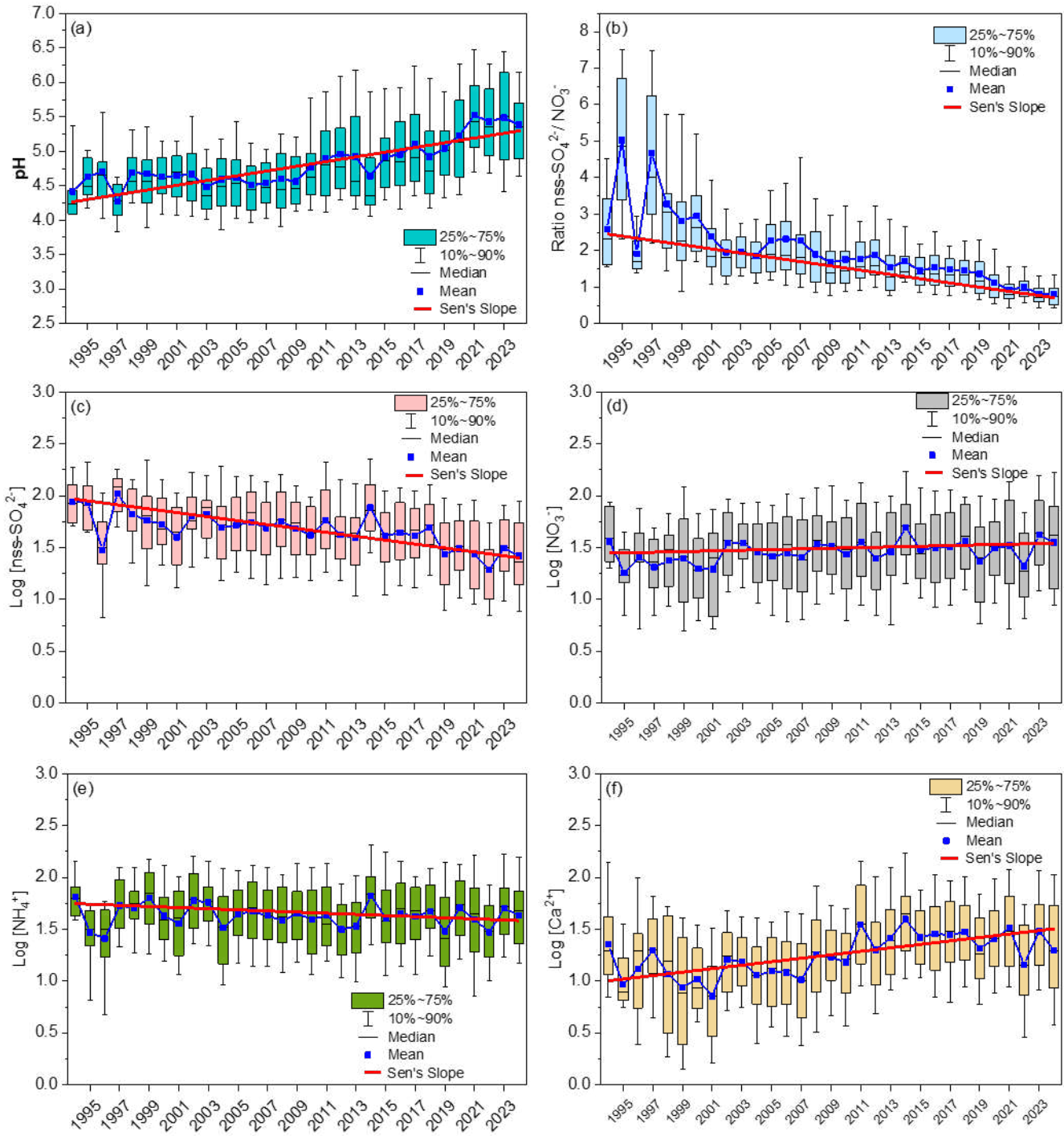


Fig. 6. Box plots (10th–90th percentile whiskers, 25th–75th percentiles, and median) of Chungli station daily concentrations for (a) pH, (b) ratio of $\text{nss-SO}_4^{2-}/\text{NO}_3^-$, (c) $\text{Log}[\text{nss-SO}_4^{2-}]$, (d) $\text{Log}[\text{NO}_3^-]$, (e) $\text{Log}[\text{NH}_4^+]$, and (f) $\text{Log}[\text{Ca}^{2+}]$ during 1994–2024. The red line indicates Sen's slope trend

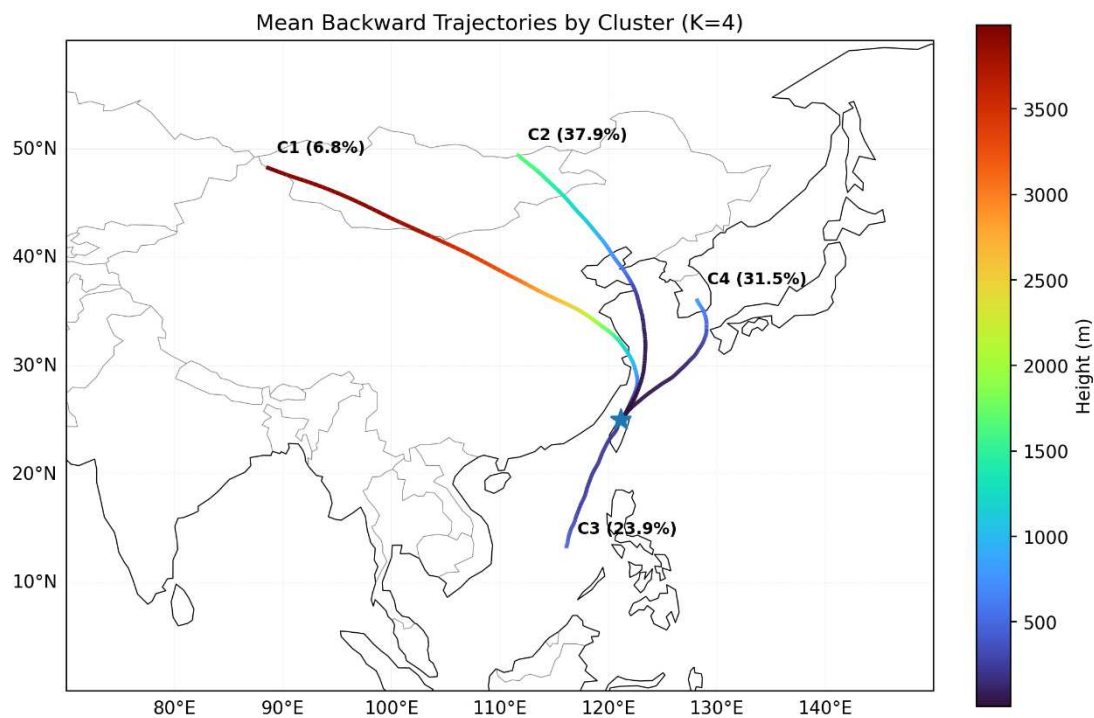
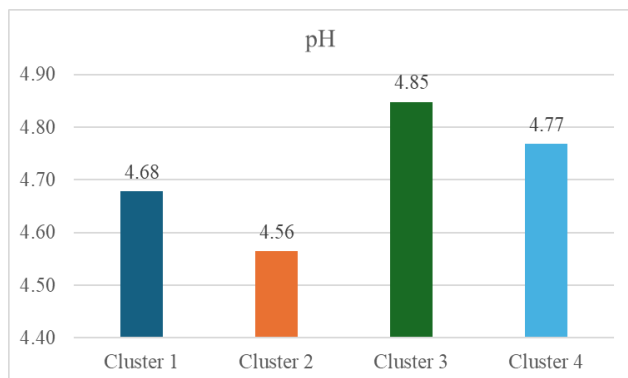
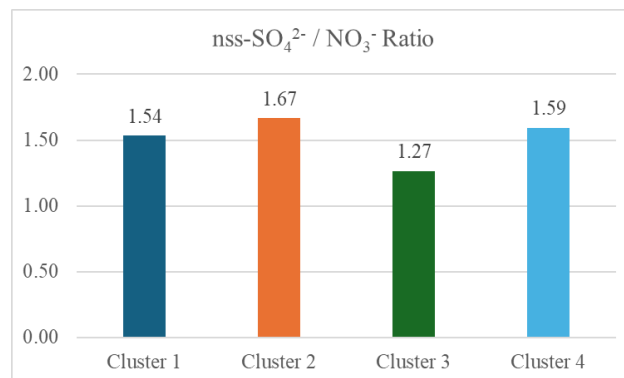


Fig. 7. Mean backward air-mass trajectories for the four identified clusters arriving at Chungli station during 2006-2024.

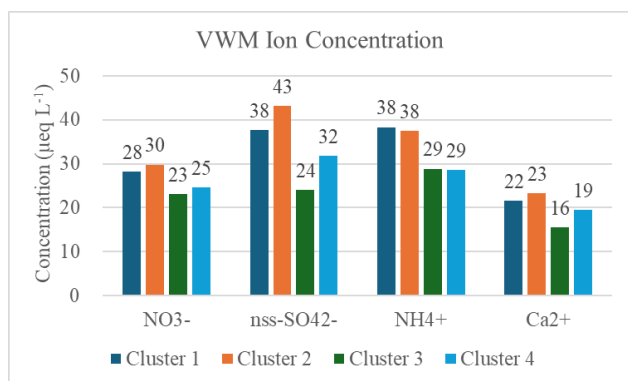
(a)



(b)



(c)



(d)

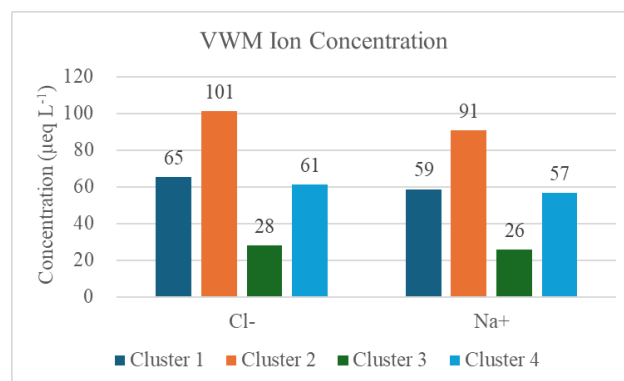


Fig. 8. Comparison of (a) VWM pH, (b) nss-SO₄²⁻/NO₃⁻ ratio, (c) VWM concentrations of major ions, (d) sea salt ions among the four air mass trajectory clusters.

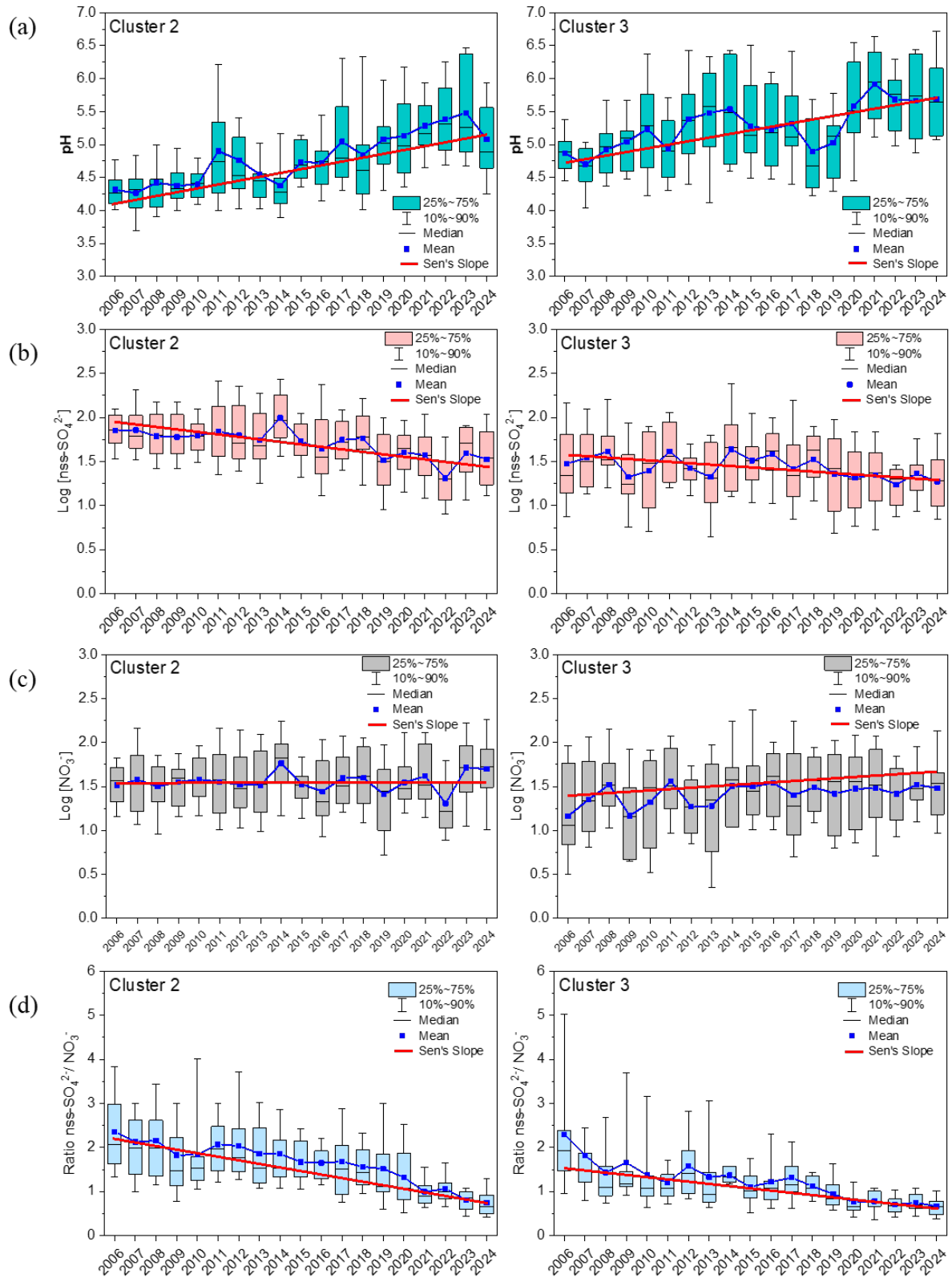


Fig. 9. Box plots (10th–90th percentile whiskers, 25th–75th percentiles, and median) of Chungli station daily concentrations under C2 (left panel) and C3 (right panel) trajectory clusters for (a) pH, (b) Log[nss-SO₄²⁻], (c) Log[NO₃⁻], (d) ratio of nss-SO₄²⁻/NO₃⁻ during 2006–2024. The red line indicates Sen's slope trend.

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- [2.Tables.docx](#)
- [4.SupFigure.docx](#)