National Assessment Report for Japan

Chapter 1. Basic Information on National Monitoring Activities

1.1 Outline of activities on acid deposition and air pollution in National Monitoring Plan

In Japan, around 1975, during the rainy season, cases of eye and skin irritation caused by fog and drizzle, particularly in the northern Kanto region, together with adverse effects on agricultural crops, became evident. These incidents attracted considerable public attention and led to the first recognition of wet atmospheric pollution (the acid rain issue) as an environmental problem. Subsequently, with the rapid economic development of East Asia, emissions of air pollutants increased significantly. Concerns grew that acid rain precursors and other air pollutants emitted in the region would be transported to Japan, located downwind of the Asian continent. As a result, the issue evolved from a domestic environmental concern into a broader regional environmental problem, a situation that continues to the present day.

Based on the results of a five-year investigation on acid rain pollution conducted from 1975 under the title Wet Atmospheric Pollution Survey in Tokyo and six surrounding prefectures, as well as growing recognition that acid rain was causing serious impacts on ecosystems, including lakes and forests, in Europe and North America, the Ministry of the Environment (then the Environment Agency) launched the First Acid Deposition Survey in fiscal year (FY) 1983. The survey established acid rain monitoring programs covering atmospheric deposition, soils and vegetation, and inland waters throughout Japan. The results of the 20-year survey through FY2002 were compiled in June 2004 as the Comprehensive Report on the Acid Deposition Survey. The report revealed that acid rain with acidity comparable to that observed in Europe and North America was occurring nationwide, suggested that inflow of pollutants originating from the Asian continent affected areas along the Sea of Japan coast, and identified changes in the physical and chemical characteristics of inflowing rivers and surrounding soils in the watershed of Lake Ijira (Yamagata City, Gifu Prefecture) that were considered indicative of the effects of acid rain. In FY2003, the government established the Long-term Acid Deposition Monitoring Plan to ensure comprehensive and continuous nationwide monitoring of acid deposition, and monitoring has since been conducted in accordance with this plan. The results were summarized in the Long-term Acid Deposition Monitoring Report (FY2003–FY2007), which clarified acidification processes in the Lake Ijira watershed and highlighted the need for integrated monitoring of transboundary air pollution, including ozone and particulate matter.

In response to growing domestic and international concern over transboundary air pollution, the need arose to better understand the long-range transport and long-term trends of acid deposition precursors and air pollutants such as ozone (O_3) and fine particulate matter ($PM_{2.5}$) in Japan. Accordingly, the *Long-term Monitoring Plan for Transboundary Air Pollution and Acid Deposition* was established in March 2009, and monitoring based on this plan commenced in FY2009. The findings were summarized in the *Long-term Monitoring Report on Transboundary Air Pollution and Acid Deposition (FY2008–FY2012)*, published in March 2014. The report indicated that precipitation in Japan continued to exhibit relatively high acidity and that the regional and seasonal variations in

gaseous and particulate pollutant concentrations continued to suggest the influence of transboundary pollution transported from the Asian continent. In addition, long-term changes potentially associated with acid deposition, such as declining pH in soils and stream waters, were observed at some monitoring sites. Areas requiring continued surveillance of the impacts of O_3 on vegetation were also identified. Based on these findings, together with growing public concern regarding $PM_{2.5}$ following the establishment of environmental quality standards in Japan, the monitoring plan was partially revised in March 2014. The revisions included the optimization of the monitoring network, expansion of $PM_{2.5}$ monitoring, and the introduction of intensified monitoring in priority areas requiring close observation. The *Long-term Monitoring Report on Transboundary Air Pollution and Acid Deposition (FY2013–FY2017)*, published in March 2019, showed that although the acidity of precipitation in Japan remained higher than that observed in Europe and North America, signs of a gradual decline had begun to emerge. Atmospheric SO_2 concentrations also showed a decreasing trend, largely reflecting reductions in SO_2 emissions across the Asian continent. The report concluded that continued long-term monitoring is essential to track future changes in $PM_{2.5}$ and O_3 and to further assess their environmental impacts. Furthermore, in March 2019, the monitoring plan was revised again to ensure the continued, efficient, and reliable assessment of the status and impacts of domestic emission sources as well as transboundary air pollution and acid deposition in Japan. The revision included a review and optimization of the monitoring network, including the reassessment of monitoring sites. Table 1 shows the number of monitoring sites in each survey from Fiscal Year (FY: From April 1st to March 31st in Japan) 1983 to FY2022. This report focuses on the results of monitoring conducted in FY2018-FY2022.

Table 1 Number of monitoring sites in each survey

Survey	First	Second	Third	Fourth	—	Long-term			
Period (FY)	1983-1987	1988-1992	1993-1997	1998-2000	2001-2002	2003-2007	2008-2012	2013-2017	2018-2022
Wet/Dry Deposition	14～34	29	48	55	48	31	27	24 (27 in 2013)	20
Soil /Vegetation	12	43	88	20	18	25	25	25	19
Inland aquatic environment	133 (Screening survey)	5	33	17	12	11	11	11	6
Catchment -scale						1	1	1	1
Others		2 (Snow)							

1.2 Monitoring program from FY2018 to FY2022

1.2.1 Introduction

The Ministry of the Environment Government of Japan (MOEJ) * has been conducting acid deposition monitoring since 1983. From the viewpoint of clarifying that ozone and aerosols should be monitored as transboundary air pollutants as well as SO_2, NO_x, etc., wet deposition (precipitation), dry deposition including air pollutants, such as gases and aerosols, soil and vegetation, inland aquatic environment and catchment-scale monitoring are currently implemented according to the Long-Term Monitoring Plan (revised in March 2014). This report is compiled the monitoring results for the five years from FY2018 to FY2022.

*: The original organization was the Environment Agency of Japan until 2001.

1.2.2 Objectives of tranboundary air pollution and long-term acid deposition monitoring

Atmospheric and ecological impact monitoring are carried out for a long period of time in close collaboration with the Acid Deposition Monitoring Network in East Asia (EANET);

- To understand long-distance transboudary transportation and long-term trends of air pollutants, such as acid rain precursors, ozone and $PM_{2.5}$, etc.and
- To early grasp the effects of transboundary air pollution and acid deposition and predict their future effects.

1.2.3 Detailes of monitoring

Wet and dry deposition monitoring were carried out to understand the status of acid deposition. Soil and vegetation, inland aquatic environment, and catchment-scale monitoring were carried out for clarifying the impact by acid deposition. (Table 2)

Table 2 Transboundary air pollution and long-term acid deposition monitoring (From FY2018 to FY2022)

	Classification	Monitoring content	The number of sites
Acid Deposition Monitoring	Wet deposition monitoring	Monitoring precipitation pH (both rain and snow) and concentrations of dissolved ionic components in precipitation, which includes calculating deposition amounts on the ground surface by multiplying each ionic concentration with its precipitation amount.	20
	Dry deposition monitoring	Monitoring air concentrations of gaseous substances, gravimetric concentrations of particulate matters and their constituents. Calculating of deposition amounts on the ground surface based on meteorological conditions such as wind speed.	
Ecological Impact monitoring	Soil and vegetation monitoring	Monitoring soil condition, including pH and ionic concentrations, as well as the degree of tree decline and understory vegetation.	19
	Inland aquatic environment monitoring	Monitoring pH and ion concentrations in rivers, lakes, etc.	6
	Catchment-scale monitoring	Monitoring to evaluate mass balance between acidic substances through the atmosphere or in/out flow river and its effect on the ecosystem focusing on a certain watershed (catchment).	1

Chapter 2. Acid Deposition and Air Pollution in Japan and Their Environmental Impacts

2.1 State of acid deposition and air pollution

2.1.1 Precipitation acidity (pH)

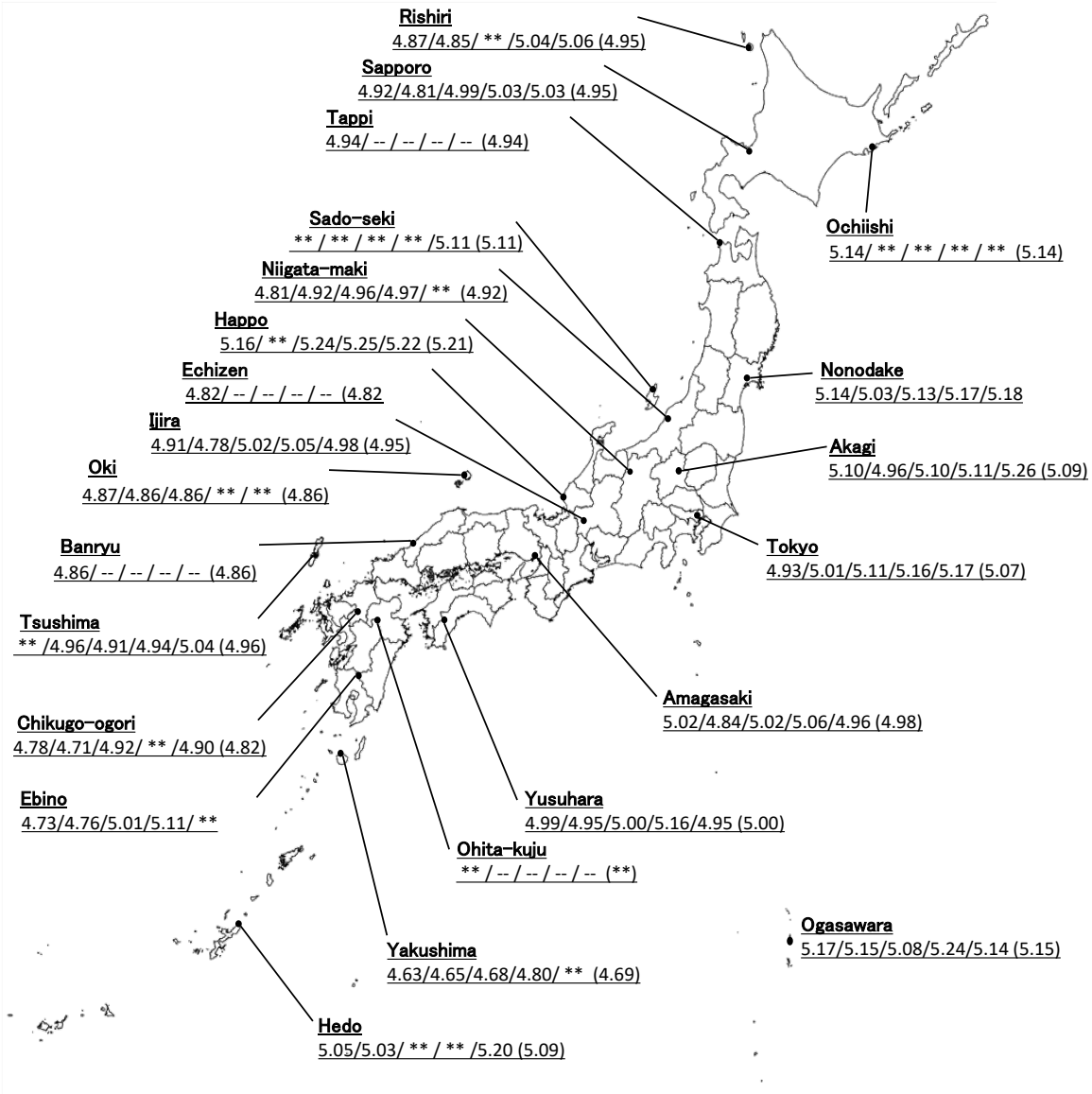
Precipitation in Japan remains acidic. Although the pH of precipitation in Japan is lower than that observed in Europe, North America, and other EANET participating countries, a gradual increase in precipitation pH (i.e., a decrease in acidity) has been observed in recent years. Emissions of acid deposition precursors in both Japan and China have shown declining trends. In particular, the contribution of sulfur oxides emitted from China to the acidity of precipitation in Japan is considered to have continued to decrease.

- The five-year weighted mean precipitation pH values, calculated for each monitoring site in Japan for FY2018–FY2022 with precipitation amount taken into account, ranged from pH 4.63 to 5.26. Relatively high values were observed at Ogasawara (5.15), Ochiishi (5.14), and Nonodake (5.12), while relatively low values were observed at Echizen-misaki (4.82), Chikugo-ogori (4.82), and Lake Banryu (4.86). The five-year weighted mean across all sites was 4.94, indicating that precipitation in Japan remains acidic (Figure 1).

- The pH of precipitation in Japan is lower than the average values observed in Europe, North America, and EANET participating countries (Cambodia, China, Indonesia, Japan, Lao PDR, Malaysia, Mongolia, Myanmar, the Philippines, the Republic of Korea, Russia, Thailand, and Viet Nam) (Figure 2-1). A comparison among international monitoring networks of the

contributions of acidic species that acidify precipitation—non-sea-salt sulfate (nss-SO_4^{2-}) and nitrate (NO_3^-)—and alkaline species that neutralize acidity—ammonium (NH_4^+) and non-sea-salt calcium (nss-Ca_2^+)—suggests that the relatively smaller contribution of alkaline species compared with acidic species is one of the reasons why precipitation pH in Japan is lower than that in many other countries (Figure 2-2).

- The pH of precipitation in Japan has continued to show signs of an upward trend in recent years (Figure 3). This is considered to reflect decreases in emissions of acid deposition precursors, including sulfur and nitrogen oxides, in both Japan and China as a result of emission control measures. In particular, sulfur oxide emissions in China have shown a marked downward trend, suggesting that the acid contribution to precipitation in Japan from transboundary air pollution has continued to decline.



FY2018/FY2019/FY2020/FY2021/FY2022 (5-year average)

-- : No measurement

**:The annual average did not meet the validity criteria and was rejected.

Note 1: The annual average was calculated as precipitation amount weighted mean. But each value in parentheses shows their 5-year arithmetic average, as well as the all-sites' average.

Note 2: Hokkaido area: Rishiri, Sapporo, and Ochiishi, Pacific Ocean side: Nonodake, Akagi, Ijira, Tokyo, Ogasawara, and Yusuhara, mid-North Honshu, Sea of Japan side: Tappi, Sado-seki, Niigata-maki, Happo, and Echizen, Seto Island Sea Coast: Amagasaki and Ohita-kuju, San'in: Oki and Banryu, East China Sea Coast: Chikugo-ogori, Tsushima, and Ebino, Nansei Islands: Yakushima and Hedo.

Note 3: Tappi, Echizenmisaki, Banryu, and Ohita-kuju sites stopped monitoring in FY2018.

Note 4: Wet deposition monitoring was not conducted at Goto.

Figure 1. Distribution of precipitation pH in Japan (FY2018-FY2022).

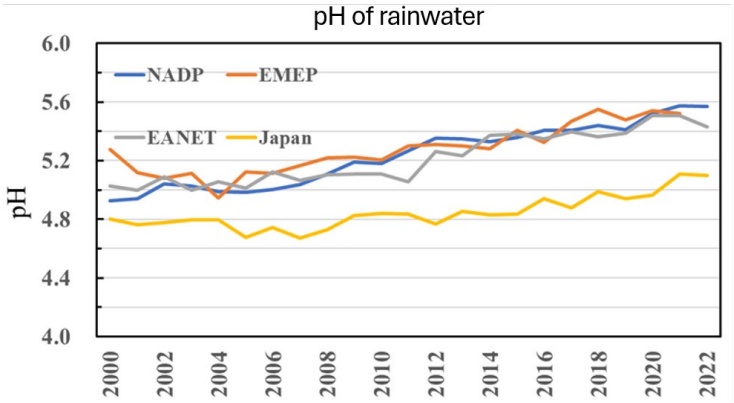


Figure 2-1. Comparison of precipitation pH trends between Japan and other international networks (EMEP[1]: Europe, NADP[2]: North America, EANET: East Asia including Japan).

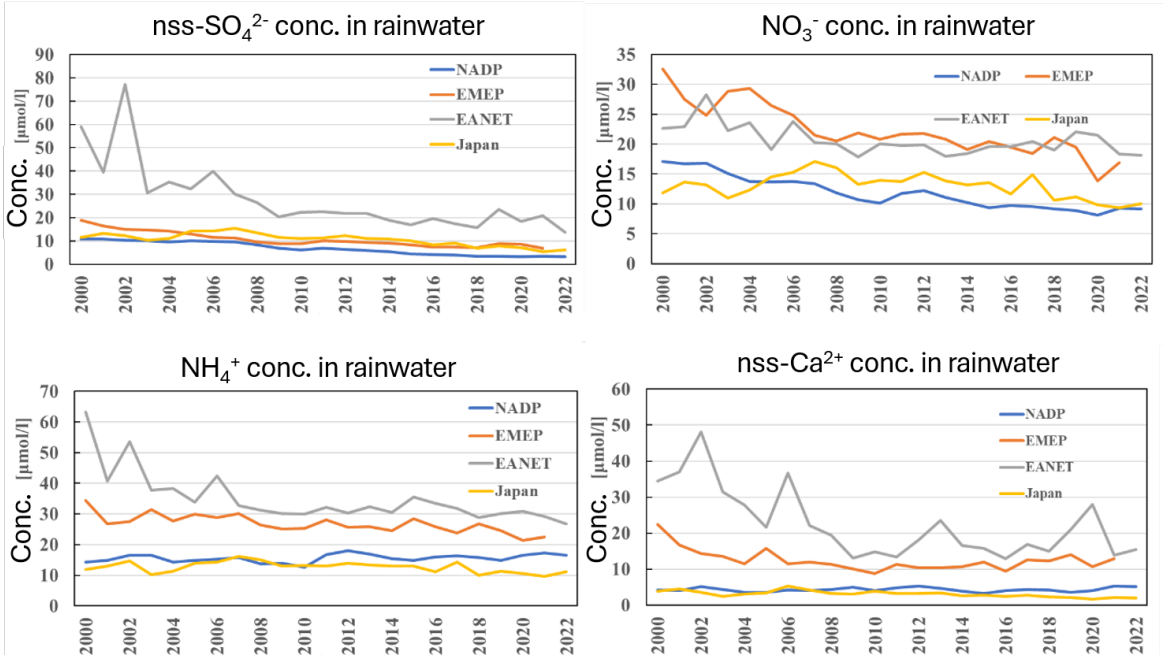


Figure 2-2. Comparison of trends in concentrations of chemical constituents in precipitation between Japan and other international monitoring networks (nss-SO_4^{2-}: Non-sea salt sulfate ion, NO_3^-: Nitrate ion, NH_4^+: Ammonium ion, and nss-Ca^{2+}: Non-sea salt calcium ion).

[1] EMEP: Co-operative Program for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe

[2] NADP: National Atmospheric Deposition Program

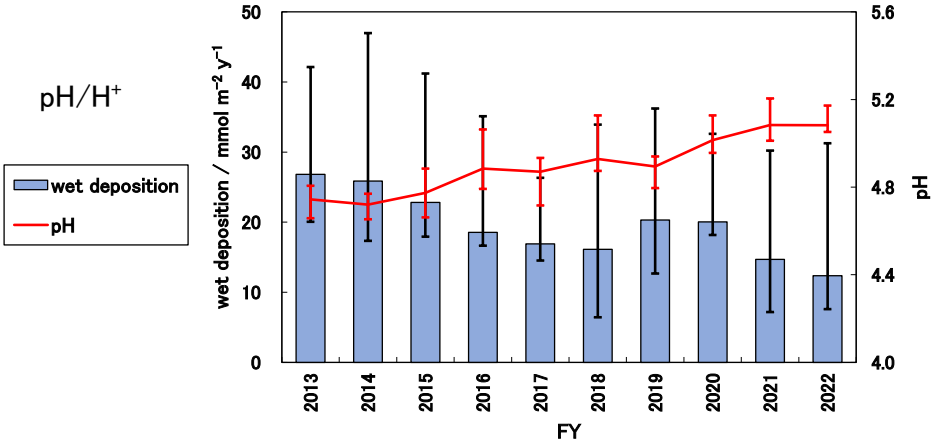


Figure 3. Annual variation of precipitation pH and hydrogen ion (H^+) deposition (median) in the last ten years.

Error bars mean the range of 25th to 75th percentile values[3] for each year in pH and hydrogen ion deposition. The annual data which did not meet the criteria of completeness[4] are not included in calculation.

2.1.2 Seasonal variations of substance in precipitation

Concentrations of non-sea-salt sulfate ions and other chemical constituents in acidified precipitation showed marked increases in winter, particularly in areas such as the San'in region, which is close to the Asian continent. This suggests the influence of transboundary transport from the continent, in addition to domestic emission sources.

- The concentrations of nss-SO_4^{2-}, NO_3^-, and NH_4^+ ions in precipitation were examined by region across Japan (Figure 4). All three species exhibited a common seasonal pattern, with relatively low concentrations in summer and higher concentrations from winter to spring. Although precipitation chemistry in Japan is generally influenced by widespread transboundary air pollution originating from the Asian continent, wet deposition is also considered to vary depending on the distance from and geographical relationship to the continent, domestic emission sources, and the amount and frequency of precipitation.

[3] The percentile value is a value located at a specified percentage of the total when the target data groups are arranged from the smallest.
[4] Completeness means the ratio of valid data for a certain monitoring period.

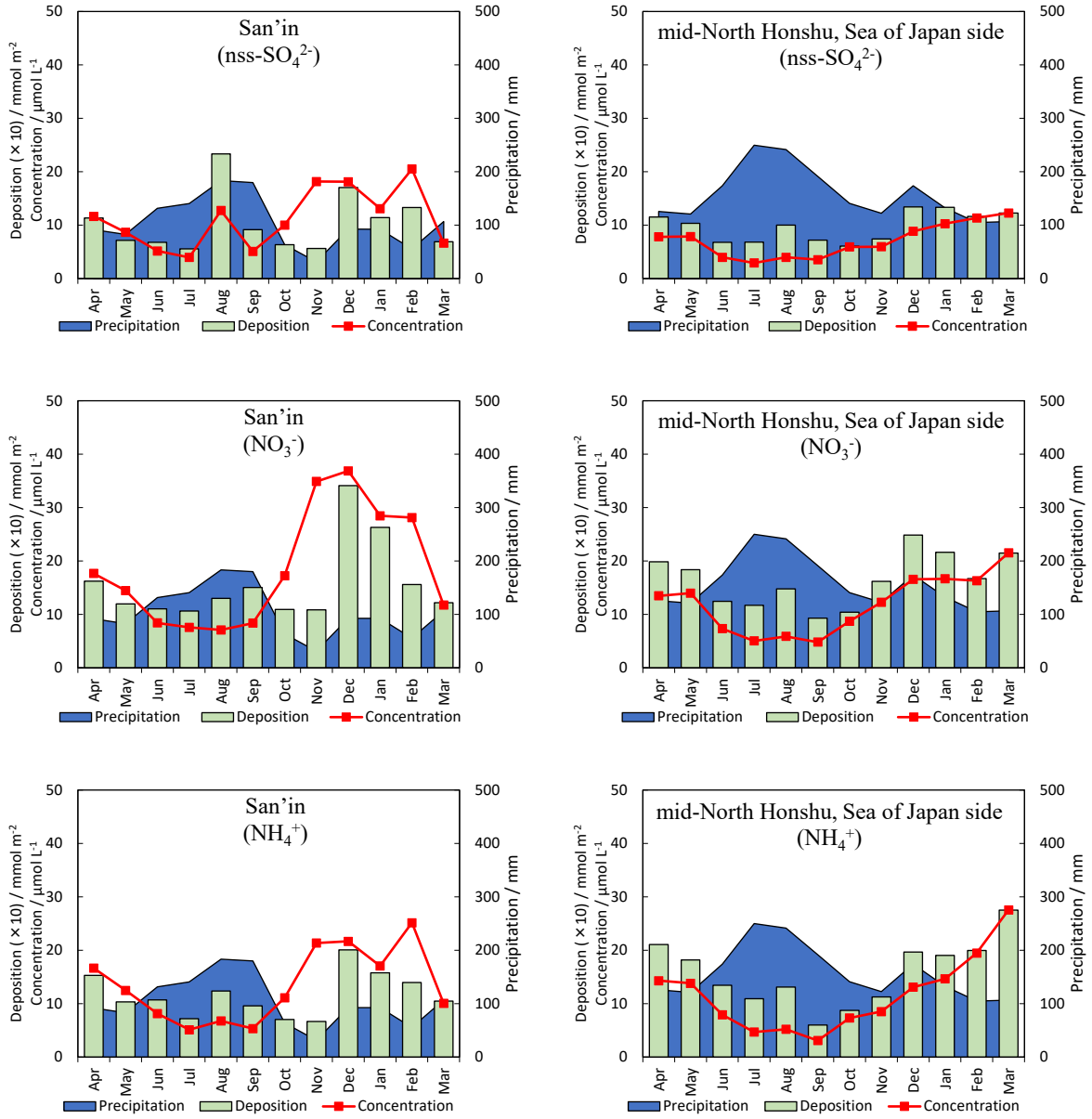


Figure 4. Seasonal variations in nss-SO_4^{2-}, NO_3^- and NH_4^+ concentration, deposition and precipitation at San' in area and mid-North Honshu, Sea of Japan side area (FY2018 – FY2022).

2.1.3 Current status and long-term trends of acid deposition

For total deposition, including both dry and wet deposition, although some sites showed a marked decrease in total sulfur deposition compared with five years earlier, the overall trends in total sulfur and nitrogen deposition in recent years were not clear.

- The annual total deposition of sulfur and nitrogen compounds at the EANET sites in Japan ranged from 2.1 to 8.2 kg-S/ha/year and from 1.7 to 12.7 kg-N/ha/year, respectively. As shown in Figures 5 and 6, an examination of the interannual variation in total deposition (wet deposition + dry deposition) over the five-year period from 2018 to 2022 indicated that, overall, no clear increasing or decreasing trends were observed.
- At Oki, although sufficient wet deposition data were not available for 2021 and 2022, the sulfur deposition decreased by approximately 4 kg-S/ha during the period 2018–2020 (Figure 5). While no substantial change was observed in wet deposition, dry deposition showed a marked decrease.
- At Happo, Yusuhara, and Hedo, although there were year-to-year variations, sulfur deposition also showed a decreasing trend over the period from 2018 to 2022.

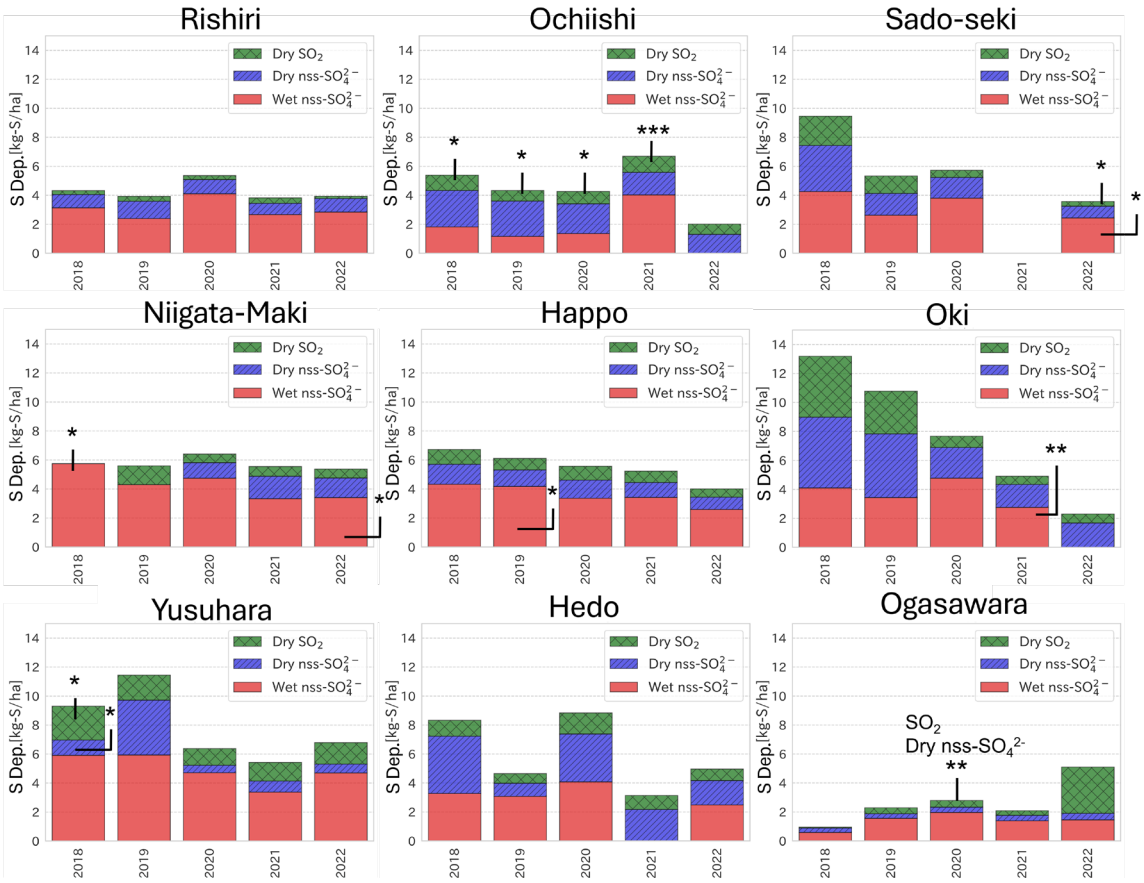


Figure 5. Annual total sulfur deposition at each site (FY2018–FY2022)

*, Dry : > 50 % and < 70 %, Wet : > 50 % and < 80 %,

**, Dry and Wet : > 30% and < 50 % <=

***, Dry and Wet : <= 30%

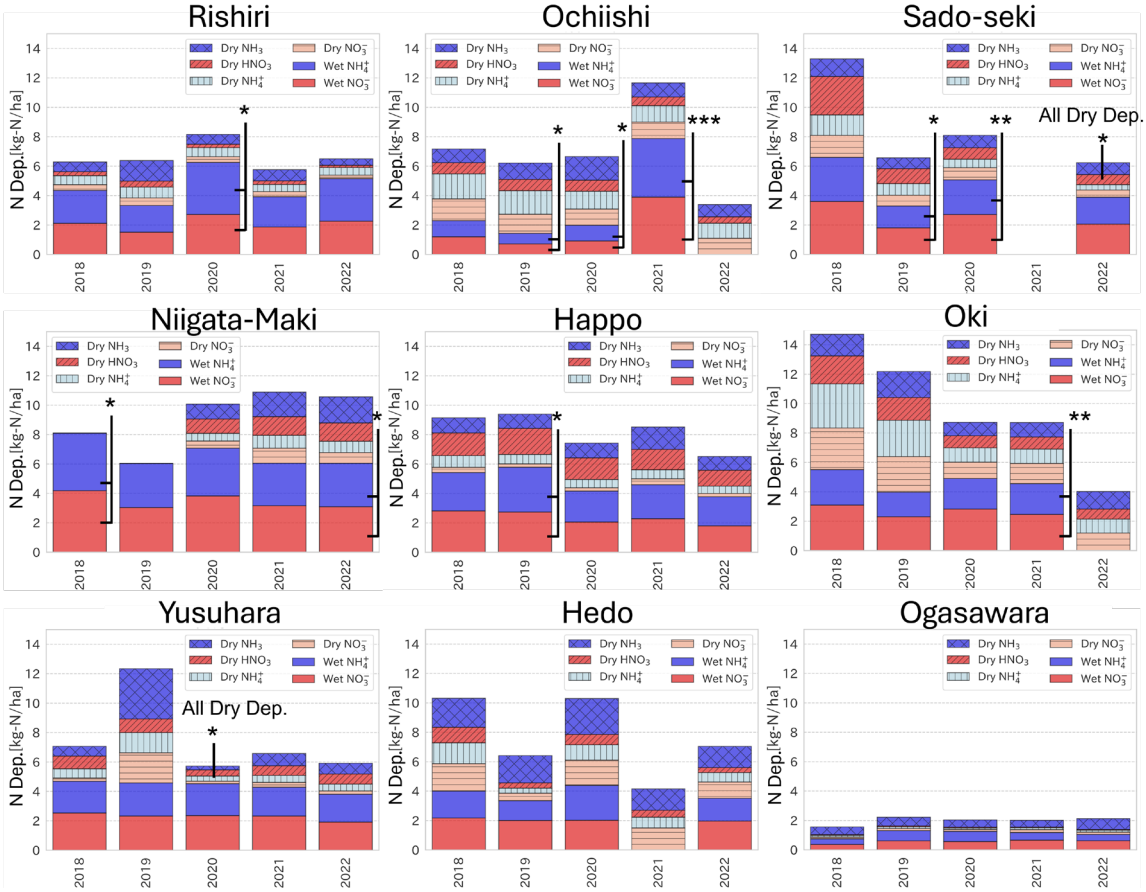


Figure 6. Annual total nitrogen deposition at each site (FY2018–FY2022)

*, Dry : > 50 % and < 70 %, Wet : > 50 % and < 80 %,

**, Dry and Wet : > 30% and < 50 % <=

***, Dry and Wet : <= 30%

2.1.4 Long term trends of O_3 concentrations

The annual mean ozone concentration at remote sites showed an increasing trend until FY2007, followed by a decreasing trend after FY2015. In contrast, the frequency distribution of the daily maximum 8-hour average ozone concentration showed no long-term trend indicative of reductions resulting from emission control measures at either remote or rural sites. Note that trend analysis in this report was performed using the Mann–Kendall test.

- The annual mean ozone concentration at remote sites showed an increasing trend until FY2007, followed by a decreasing trend after FY2015 (Figure 7). Analysis of the 20-year trends in annual mean ozone concentrations at individual sites during FY2003–FY2022 (Figure 8) showed statistically significant decreasing trends ($p < 0.05$) at Rishiri (−0.51 ppb yr^{-1}), Tappi (−0.65 ppb yr^{-1}), Sado-seki (−0.49 ppb yr^{-1}), Happo (−0.61 ppb yr^{-1}), Hedo (−0.42 ppb yr^{-1}), Banryu Lake (−0.23 ppb yr^{-1}), and Akagi (−0.90 ppb yr^{-1}). In contrast, statistically significant increasing trends ($p < 0.05$) were observed at Amagasaki (+0.60 ppb yr^{-1}) and Chikugo-Ogori (+0.36 ppb yr^{-1}).
- The frequency distribution of the daily maximum 8-hour average ozone concentration showed no long-term trend indicative of reductions resulting from emission control measures at the remote and rural sites, which account for the majority of the monitoring sites. Instead, substantial interannual variability was observed. A similar pattern was found in analyses of the annual 99th percentile of the daily maximum 8-hour average ozone concentration* derived from simulation model results. In the Kanto region, reductions in NO_x and VOCs emissions were suggested to have contributed to lower summertime peak ozone concentrations. In contrast, no clear decreasing trends were observed in the other regions, where considerable interannual variability remained.

  *The annual 99th percentile of the daily maximum 8-hour average ozone concentration is calculated from hourly ozone measurements obtained by continuous automatic monitoring. It is defined as the value corresponding to the 99th percentile (the 362nd highest-ranked value) after all daily maximum 8-hour average concentrations for a given year are arranged in ascending order. This metric reflects high-concentration ozone events while being less sensitive to meteorological conditions and interannual variability. It is also consistent with findings from studies on the ecological impacts of ozone and is widely used as an indicator for evaluating long-term trends.
- The observed interannual variability in ozone concentrations is considered to be influenced by year-to-year variations in meteorological conditions, which affect the transport of ozone from the Asian continent.

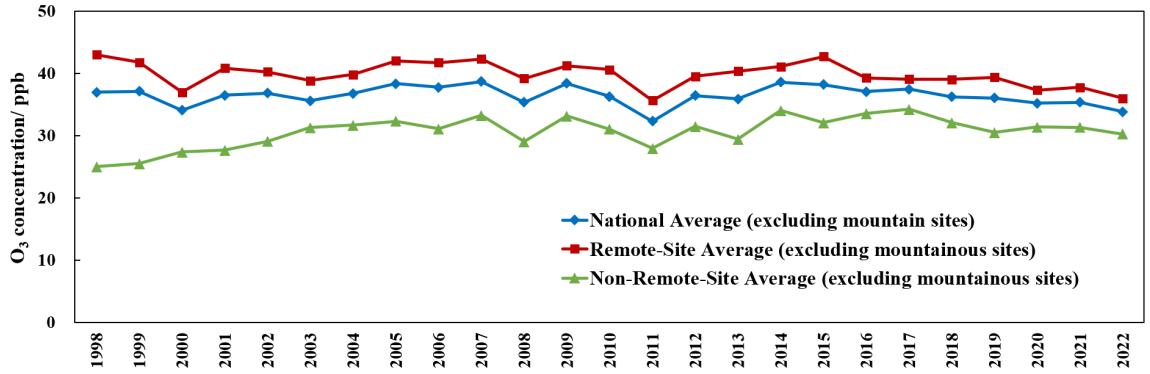


Figure 7. Long-term trends in annual mean ozone concentrations at remote and non-remote sites.

Annual values with a data completeness of less than 70% were excluded from the calculation.

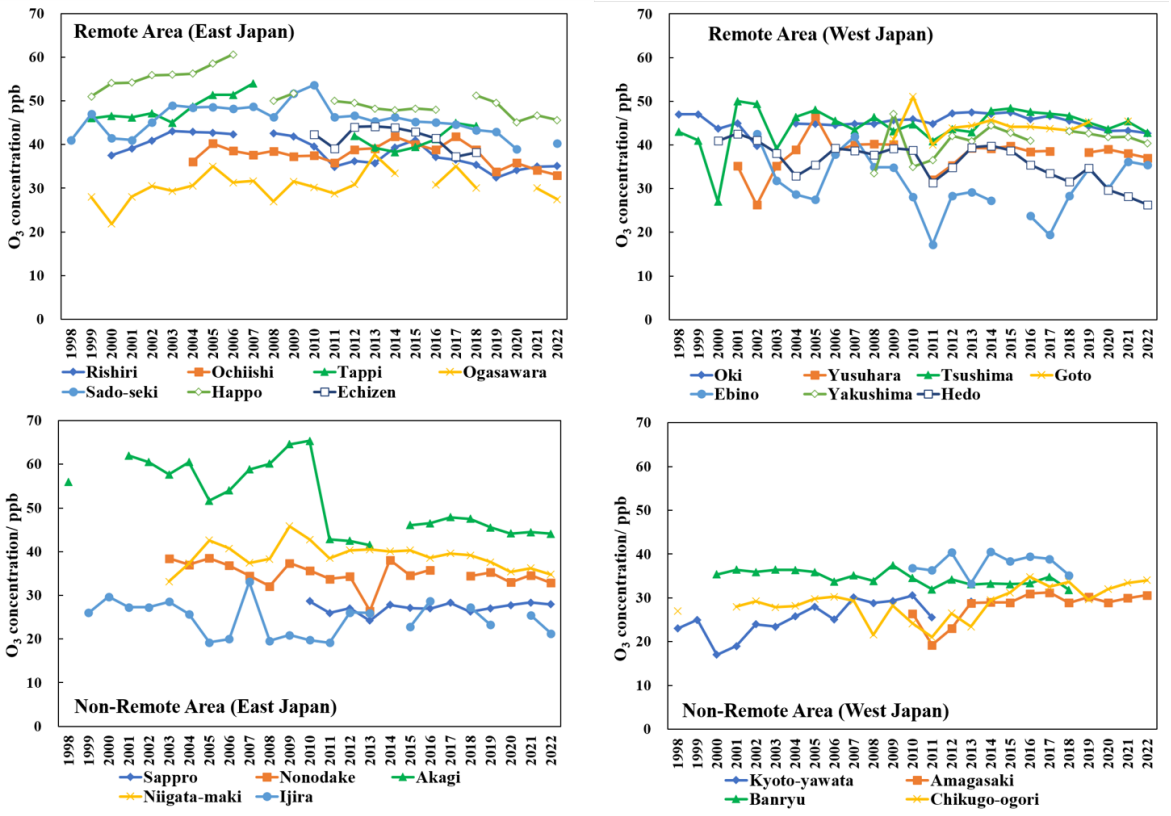


Figure 8. Long-term trends in ozone concentrations at individual sites.

Annual values with a data completeness of less than 70% are not shown.

2.1.5 Long term trends of PM concentrations

The annual mean concentrations of particulate matter (PM_{10}) and fine particulate matter ($PM_{2.5}$) showed long-term decreasing trends in many regions. In addition, the frequency of high $PM_{2.5}$ concentration events also decreased. Data from the National Air Monitoring Stations and EANET sites further indicated a downward shift in the mode of the annual mean $PM_{2.5}$ concentrations.

- Analysis of the long-term trends in annual mean PM_{10} and $PM_{2.5}$ concentrations over the 20-year period from FY2003 to FY2022 (Figure 9) showed statistically significant decreasing trends ($p < 0.05$) at 13 of the 17 sites for $PM_{2.5}$ and at 7 of the 13 sites for PM_{10}. In contrast, statistically significant increasing trends ($p < 0.05$) were observed only for PM_{10} at Rishiri and Ogasawara.
- Since 2015, the occurrence of high-concentration $PM_{2.5}$ events has shown a decreasing trend. In particular, the proportion of events lasting only a single day, as shown in Figure 10, has decreased since 2015. In contrast, the annual occurrence of events lasting two or more consecutive days varied considerably from year to year.
- Frequency distribution analysis of $PM_{2.5}$ concentrations based on annual mean concentrations and hourly mean concentrations for FY2010–FY2021 indicated a decreasing frequency of high $PM_{2.5}$ concentrations. In addition, the mode of the annual mean $PM_{2.5}$ concentrations showed a downward trend (Figure 11).

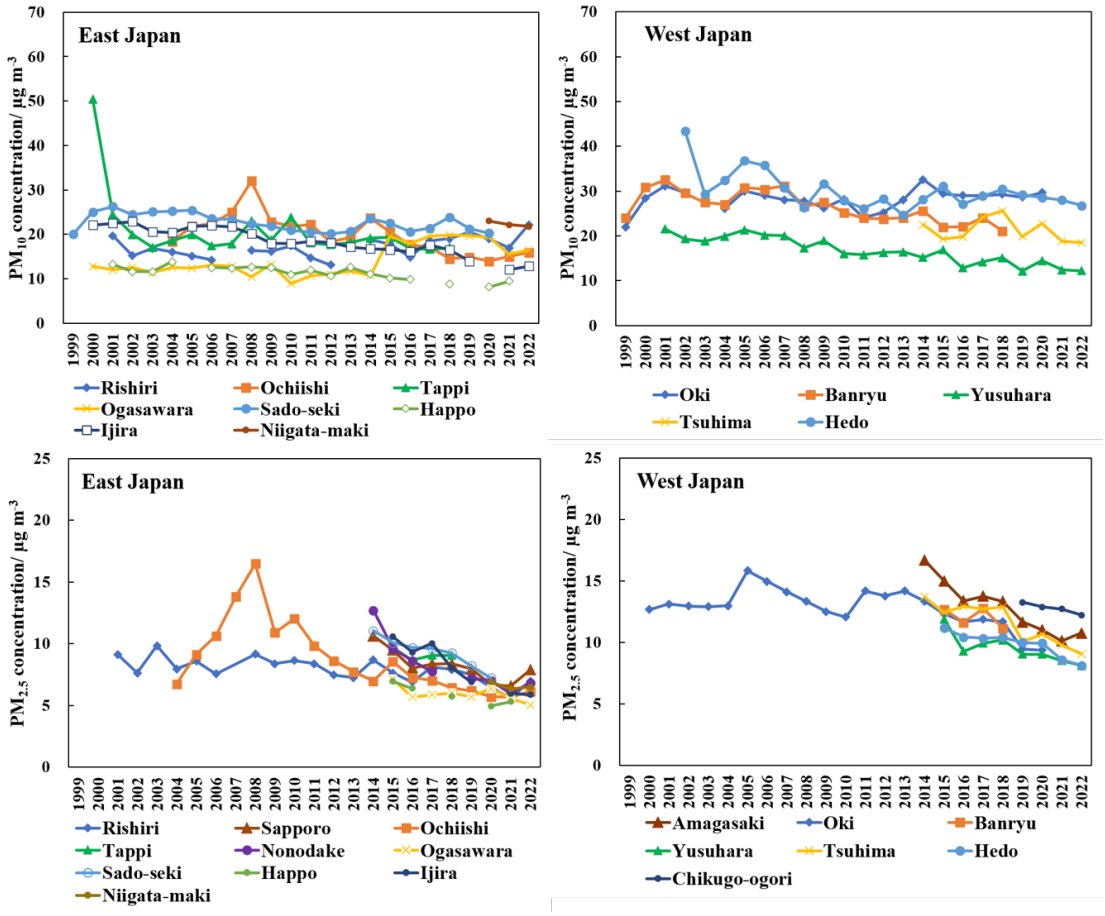


Figure 9. Long-term trends in PM_{10} and $PM_{2.5}$ concentrations.

Annual values with a data completeness of less than 70% are not shown.

For the $PM_{2.5}$ concentrations at Ochiishi, Rishiri, and Oki, a change in the measurement instrument should be taken into account when interpreting the data. The concentrations reported for FY2012 and earlier include the contribution of moisture, whereas the concentrations reported for FY2013 onward exclude the contribution of moisture.

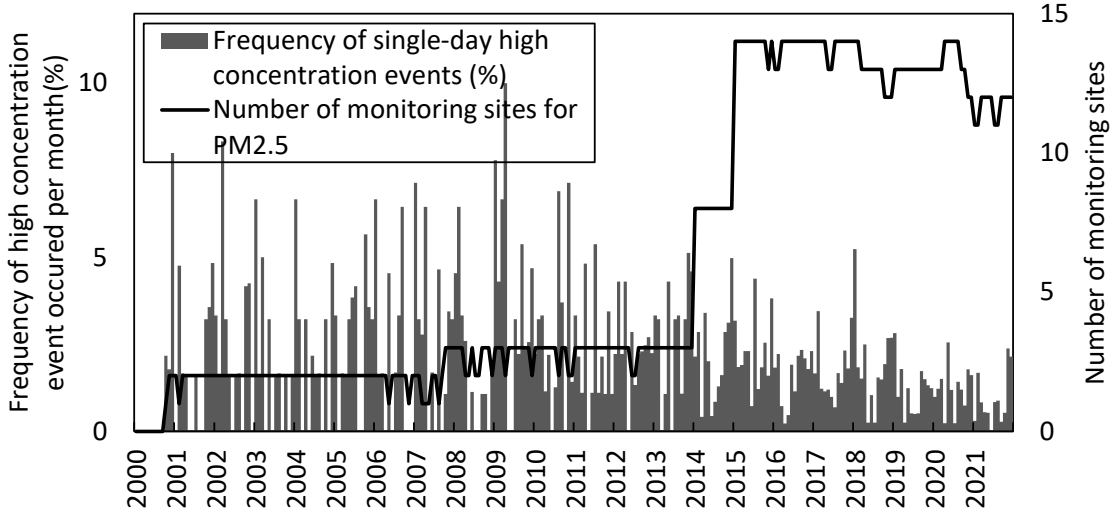


Figure 10. Long-term trends in the monthly frequency of high-concentration $PM_{2.5}$ events and the number of $PM_{2.5}$ monitoring sites.

The monthly frequency of high-concentration events is based on the proportion of high-concentration events determined from daily mean concentrations. The number of monitoring sites represents the sites with no missing data for each month.

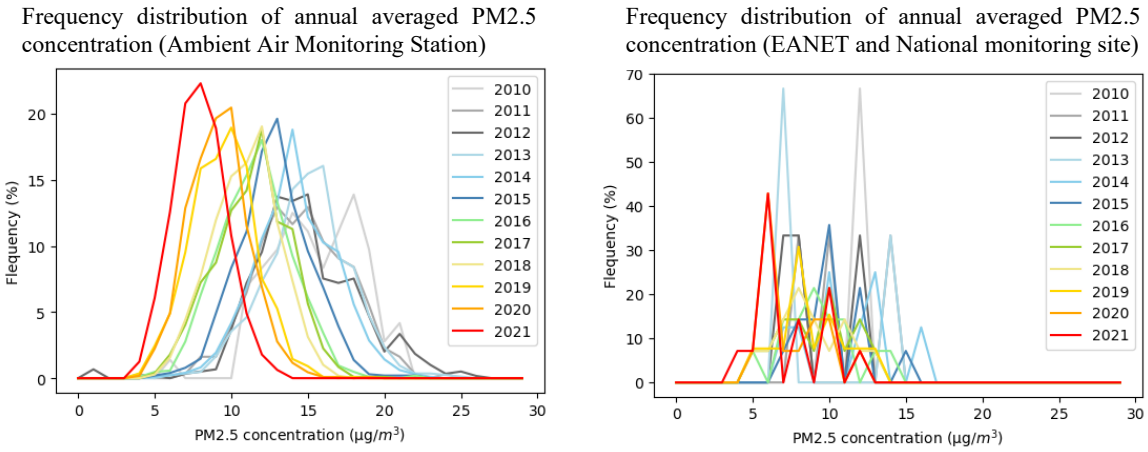


Figure 11. Relative frequency distribution of annual mean $PM_{2.5}$ concentrations (FY2010–FY2021). (Left) Ambient air monitoring stations; (Right) EANET and national monitoring stations.

2.1.6 Long term trends of air pollutant concentrations (Except ozone and PM)

Ambient SO_2 concentrations were generally lower in summer and higher in winter. At many sites, concentrations of SO_2 and particulate nss-SO_4^{2-} showed decreasing trends, which were considered to reflect the decline in SO_2 emissions from the Asian continent. In contrast, for nitrogen compounds, fewer sites exhibited decreasing trends than for sulfur compounds.

- Ambient SO_2 concentrations were generally lower in summer and higher in winter (Figure 12), suggesting the influence of transport from the Asian continent by the northwesterly winter monsoon. At the remote sites on the Sea of Japan side, SO_2 concentrations at Oki, which is located closer to the Asian continent, tended to be higher than those at Sado-seki, suggesting a greater contribution of continental transport in western Japan
- Concentrations of particulate nss-SO_4^{2-}, particulate NO_3^-, and particulate NH_4^+ were generally higher from spring to summer and lower from autumn to winter. Particulate nss-SO_4^{2-} concentrations tended to be higher at monitoring sites in western Japan than at those in eastern Japan, suggesting, as with SO_2, a greater contribution of continental transport in western Japan.
- Concentrations of SO_2 and particulate nss-SO_4^{2-} showed decreasing trends at many sites (Figure 13), which are considered to reflect the declining trend in SO_2 emissions from the Asian continent.
- At the remote sites of Yakushima, Ebino, and Yusuhara, elevated concentrations thought to originate from volcanic emissions were observed, indicating that volcanic activity is an important source in Japan in addition to anthropogenic emissions.
- For nitrogen compounds, statistically significant decreasing trends were observed in gaseous nitric acid (HNO_3) concentrations at 6 of the 14 sites and in particulate nitrate (NO_3^-) concentrations at one site. However, fewer sites exhibited decreasing trends for nitrogen compounds than for sulfur compounds.

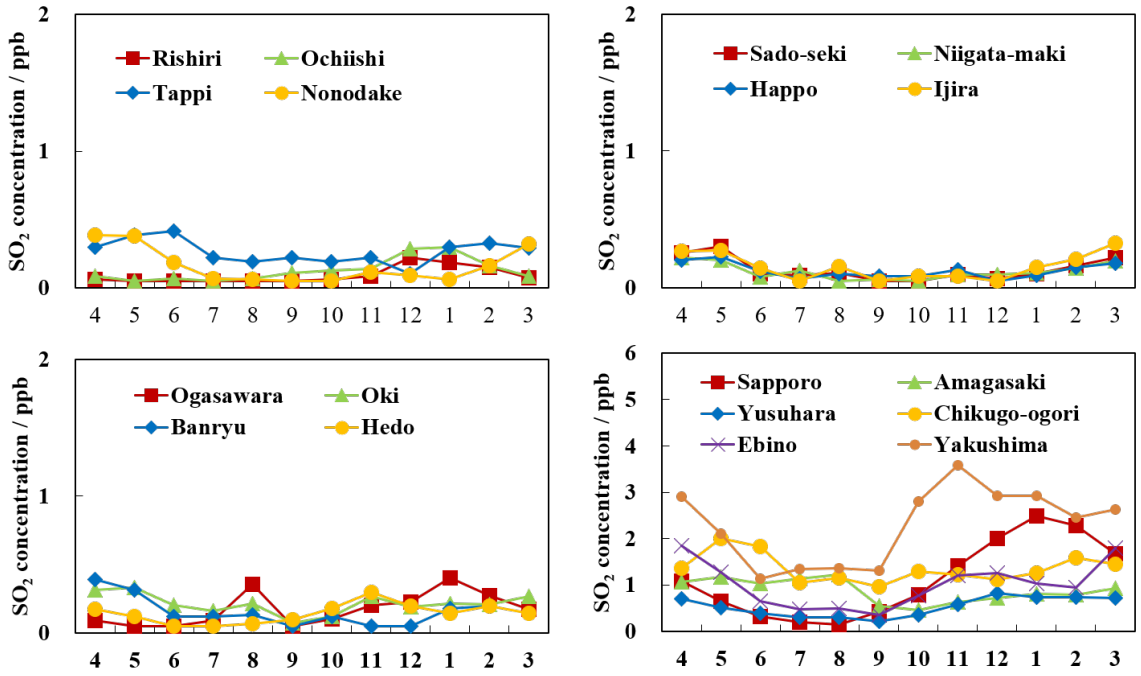


Figure 12. Monthly variation in SO_2 concentration (average for FY2018–FY2022).
Data for Tappi and Banryu Lake were compiled using only FY2018 data.

Monthly values with a data completeness of less than 70% were excluded from the calculation.

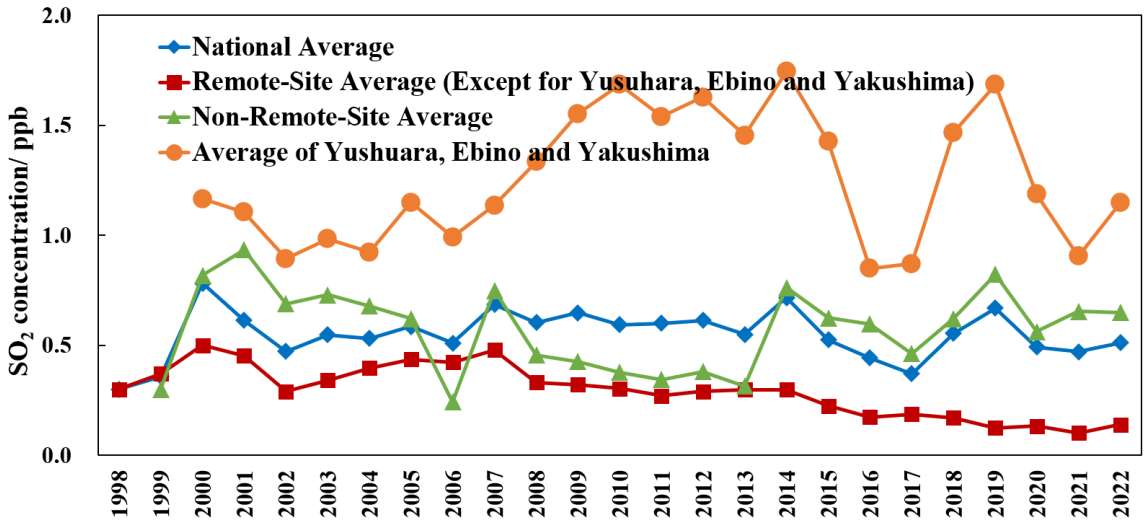


Figure 13. Long-term trends in annual mean SO_2 concentrations for the national average, remote sites, non-remote sites, and high-concentration sites (Yusuhara, Ebino, and Yakushima) (FY2003–FY2022).

Annual values with a data completeness of less than 70% were excluded from the calculation. The average for non-remote sites was calculated using data from Ijira Lake and Banryu Lake through FY2013, with data from Sapporo, Nonodake, Niigata-Maki, and Amagasaki included from FY2014 onward.

2.2 State of soil and vegetation

2.2.1 Long-term trends in soil chemical properties in forest ecosystems

The five-year interval soil monitoring showed no evidence of soil acidification based on the long-term changes in soil chemical properties.

- Soil pH (H_2O) generally ranged from 4.0 to 5.5, except at a few sites (Figure 14). No consistent increasing or decreasing trends were observed at most sites.
- Because soil chemical properties are influenced by factors such as soil type, vegetation, topography, and climate, it is necessary to monitor soils under a wide range of environmental conditions.
- Although no sites have shown clear evidence of progressive soil acidification to date, continued soil monitoring is necessary to assess long-term trends.

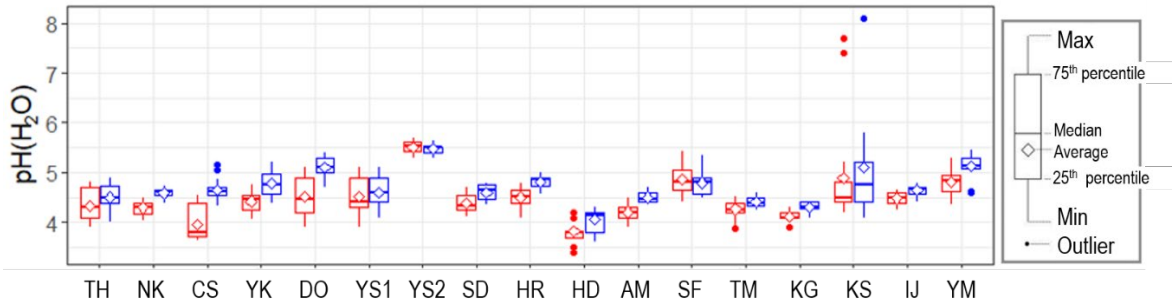


Figure 14. Range of soil pH (H_2O) values (FY2018–FY2022 survey).
Red, Surface layer (0–10 cm); Blue, Subsurface layer (10–20 cm); TH, Towada-Hachimantai; NK, Nikko; CS, Chubu-sangaku; YK, Yoshino-Kumano; DO, Daisen-Oki; YS1, Yakushima-1; YS2, Yakushima-2; SD, Sekidosan; HR, Horyuzan; HD, Hodoji; AM, Amanosan; SF, Shimofuridake; TM, Tokusagamine; KG, Kashigu; KS, Koshosan; IJ, Ijira; YM, Yamato

2.2.2 Current status of forest ecosystems

No tree decline was observed at most sites. Although tree decline was observed at a few sites, it was considered to be attributable to natural factors such as adverse weather conditions, wildlife damage, and pests and diseases. No forest decline attributable to anthropogenic influences, including air pollution, has been identified.

- Soil and vegetation monitoring has been conducted in forest ecosystems at a total of 15 sites (19 survey plots), including 11 sites primarily focused on the impacts on trees and 4 sites focused on the impacts on soils. The monitoring is carried out on a five-year rotational basis.
- The total basal area, based on the tree-by-tree survey for description of trees, showed an increasing trend at most monitoring sites, except for a few sites. From the perspective of tree growth, no evidence of forest decline was observed (Figure 15).
- The tree decline survey indicated relatively high levels of tree decline at Towada-Hachimantai, Daisen-Oki, Yoshino-Kumano, and Hodoji, and some sites showed a continuing increase in tree decline. In most cases, the observed decline was considered to be attributable to natural factors, such as adverse weather conditions, pests and diseases, and wildlife damage. However, the extent to which anthropogenic influences, including air pollution, have contributed remains unclear.

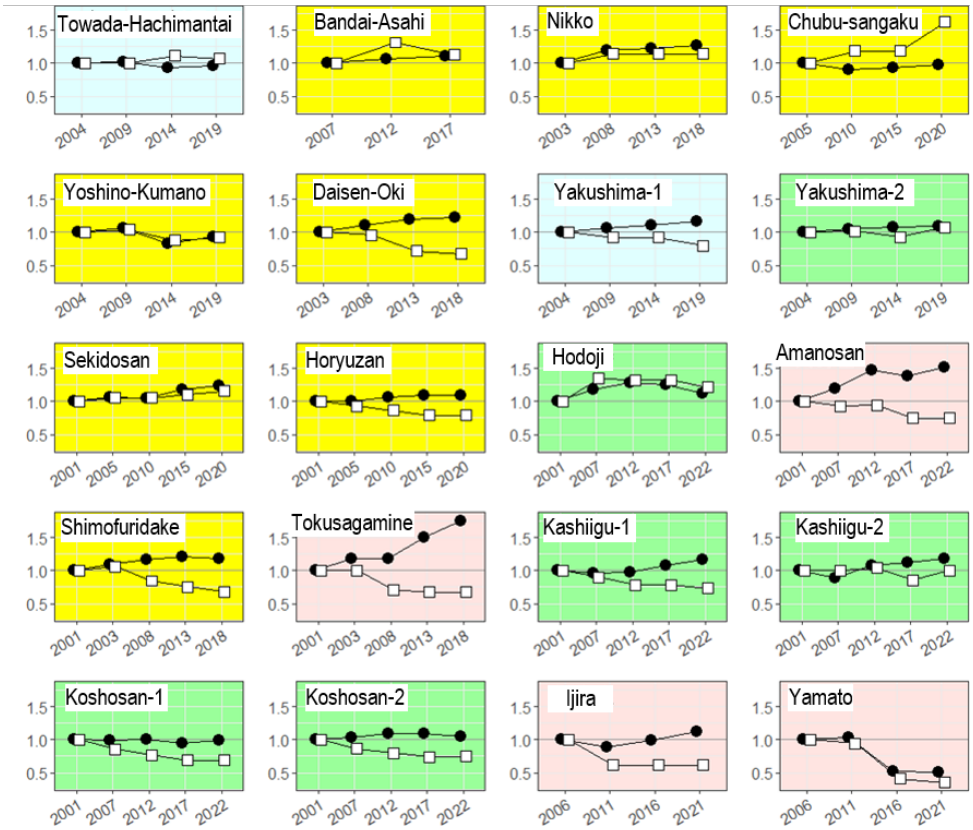


Figure 15. Long-term trends in total basal area and tree density.
●Total basal area, □Tree density. Values shown in the figure are expressed as the ratio relative to the first survey (set to 1.0). Background colors indicate vegetation type; Light blue, Natural coniferous forest; Yellow, Natural deciduous broad-leaved forest; Green, Natural evergreen broad-leaved forest; Pink, Hinoki plantation.

2.3 State of Inland aquatic environment

2.3.1 Trends in water quality at inland water monitoring sites

Decreasing trends in sulfate and nitrate concentrations were observed in most of the monitored lakes.

- Analysis of the data collected since 2006 showed an increasing number of sites with statistically significant increases in pH. Although long-term changes in alkalinity and cation concentrations varied among sites, SO_4^{2-} and NO_3^- concentrations showed decreasing trends in most of the monitored lakes (Table 3).
- For SO_4^{2-}, lower concentrations than those observed at the beginning of the monitoring period were recorded in the most recent survey at Imagamiike (Yamagata Prefecture) and Futagoike (Nagano Prefecture). In addition, at Ijira Lake (Gifu Prefecture), where no significant change in SO_4^{2-} concentration had been detected over the period 1989–2017 in the previous report (FY2013–FY2017), a statistically significant decrease was observed in the updated analysis covering the period from 2006 to 2022. These changes are considered to represent the response of inland waters to reduced atmospheric deposition associated with the recent decline in emissions of acidic substances in East Asia.

Table 3 Variation of water quality at inland water monitoring sites.

Lake (Liver) Name	Start Year (FY)	n	Z-score										
			pH	EC	Alkali-nity	SO_4^{2-}	NO_3^-	Cl^-	NH_4^+	Na^+	K^+	Ca^{2+}	Mg^{2+}
Imagami-	2001	84	2.38	-1.65	0.97	-3.42	-1.32	0.14	-1.22	1.64	0.55	-0.29	0.96
oike Lake	2006	66	2.32	-2.15	0.28	-5.55	-1.91	-0.34	-3.21	-1.14	-0.85	-0.04	-0.13
Karikomi	2003	80	3.36	4.47	6.20	1.09	-1.63	-0.49	-0.95	4.09	4.12	4.58	4.28
Lake	2006	68	3.30	3.34	4.68	1.81	-1.83	-0.71	-1.78	3.90	3.27	3.81	3.27
Ohata	2003	63	4.24	1.47	2.44	-4.76	-0.38	-2.05	-0.13	-0.07	3.67	1.75	1.89
Lake*2	2006	51	2.79	-0.82	2.32	-5.64	-2.44	-1.85	-0.62	-0.13	2.24	1.82	0.25
Yashaga-	2000	77	-0.82	-4.36	-1.45	-5.58	0.43	-3.59	-3.06	-3.73	-2.21	-6.91	-4.84
ike Lake*3	2006	54	2.45	-3.38	2.70	-4.84	0.32	-1.65	-1.79	-2.02	-1.86	-6.09	-2.89
Futago-ike	1998	75	1.61	0.43	5.50	-4.02	-5.12	-5.70	-2.10	3.28	0.80	1.16	-0.21
Lake (Oike)	2006	50	-0.49	-0.39	4.27	-7.98	-5.56	-3.95	-0.29	-0.34	0.15	-1.69	-2.51
Futago-ike	1998	75	2.68	-7.22	3.42	-6.17	-6.09	-5.25	-3.55	0.81	-1.58	-4.69	-6.08
Lake (Meike)	2006	50	2.94	-5.95	4.16	-7.94	-4.41	-3.89	-0.66	-1.97	-0.42	-4.28	-3.98
Ijira Lake	1989	132	-0.28	-1.32	5.01	-3.37	-2.99	-7.34	-5.93	-4.67	-6.73	-2.00	-4.53
	2006	64	0.69	-5.03	-2.01	-3.87	-5.10	-6.62	-3.31	-3.92	-3.42	-3.87	-4.21
Kamagatani	1989	136	-0.90	-2.83	1.09	-2.64	-2.62	-6.38	-5.50	-3.05	-4.62	-5.03	-4.77
River	2006	68	2.50	-5.01	-2.91	-2.53	-6.68	-6.94	0.00	-3.71	-2.52	-4.37	-4.03
Kobora	1989	107	5.94	0.33	2.31	1.24	-2.84	-6.67	-5.05	-1.35	-0.38	-3.53	-2.90
River	2006	56	4.33	-3.28	-1.43	-1.58	-4.66	-6.42	-*1	-2.82	0.90	-2.90	-2.67
Sawano-ike	2003	80	4.82	-6.82	3.41	-5.65	-2.37	-7.57	0.66	-0.07	-0.62	-5.50	-3.98
Lake	2006	68	3.88	-6.27	1.91	-5.53	-0.74	-7.14	0.24	-0.45	-1.89	-8.20	-4.88
Banryu	1989	120	0.84	4.66	5.86	-3.02	-1.61	6.41	3.99	5.72	0.52	3.74	5.97
Lake*2	2006	52	-2.02	-2.66	-0.09	-2.54	-3.72	0.00	0.48	-2.29	-2.91	1.10	-0.18

$p<0.05$	-	+
$p<0.01$	-	+
$p<0.001$	-	+

Z-score represents the test statistic used to evaluate temporal trends. Positive values indicate an increasing trend, whereas negative values indicate a decreasing trend. Values highlighted in the table indicate that the trend test detected statistically significant trends at 5%, 1%, and 0.1% significance levels, respectively.
*1 The Z-score could not be calculated because all measurements were below the detection limit.
*2 Analysis was conducted using data up to FY2018.
*3 Analysis was conducted using data up to FY2019.

2.4 State of catchment-scale monitoring

2.4.1 Lake-Ijira catchment-scale monitoring

In the Lake Ijira catchment, where the impacts of atmospheric deposition have been continuously monitored, decreasing trends in dissolved inorganic nitrogen (DIN) concentrations and increasing trends in pH have been observed in precipitation and stream water, suggesting recovery from nitrogen saturation and acidification.

- In the Lake Ijira catchment (Gifu Prefecture), where acidification and nitrogen saturation were first reported in the mid-1990s, inputs (total deposition) of SO_4^{2-} and DIN have shown

decreasing trends since catchment monitoring began in 2006, indicating a reduction in the atmospheric loading of these pollutants.

- In response to these changes in atmospheric deposition, increasing trends in pH and Gran's acid neutralizing capacity (Gran's ANC), together with decreasing trends in SO_4^{2-} and NO_3^- concentrations, were observed at the three stream sampling sites. In particular, the annual volume-weighted mean NO_3^- concentration in stream water showed a clear decreasing trend, consistent with that observed in precipitation, suggesting recovery from nitrogen saturation in response to the decline in atmospheric deposition (Figure 16).
- Phenomena associated with recovery from acidification that have also been reported in Europe and North America, such as decreasing electrical conductivity (EC) and the accompanying increase in total organic carbon (TOC) concentrations in stream water, were also observed. Continued monitoring will therefore be necessary to determine whether recovery from nitrogen saturation and acidification in the Ijira Lake catchment will continue to progress.

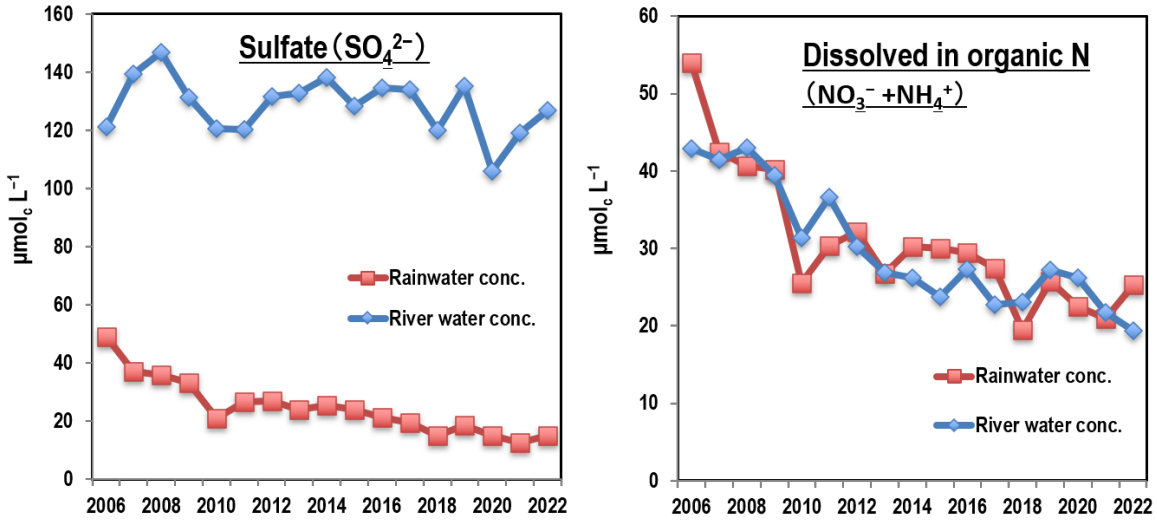


Figure 16. Long-term trends in volume-weighted mean concentrations of SO_4^{2-} and DIN in the Ijira Lake catchment.

Stream water concentrations are from the sampling site (RW1) where stream discharge is measured.

2.5 Integrated analysis

2.5.1 Isotope-based analysis

Sulfur isotope ratio analysis suggested that sulfur oxides originating from the Asian continent were transported to Japan by the northwesterly winter monsoon, although their contribution has declined in recent years. Analyses of hydrogen and oxygen isotope ratios, lead isotope ratios, and strontium isotope ratios suggested changes in precipitation patterns, variations in pollution sources, and the seasonal transport of Asian dust.

- Application of various isotope ratio analyses to monitoring samples that had previously been analyzed mainly for ionic components provided new insights into changes in the influence of transboundary air pollution and the biogeochemical processes within forest ecosystems.
- The sulfur isotope ratio ($\delta^{34}S$) in precipitation exhibited a clear seasonal pattern, with higher values in winter and lower values in summer, reflecting the stronger influence of sea salt transported by the northwesterly winter monsoon. Hydrogen and oxygen isotope ratios in precipitation also suggested the influence of water vapor originating from the Sea of Japan during the winter monsoon season.
- In the past, transboundary air pollution derived from coal combustion in China, which is characterized by relatively high $\delta^{34}S$ values, resulted in a pronounced seasonal increase in the non-sea-salt $\delta^{34}S$ values during winter. In recent years, however, this seasonal pattern has become less distinct. These findings suggest that the decline in transboundary air pollution, changes in economic activity associated with the COVID-19 pandemic, and climate change, including rising winter temperatures and shifts in precipitation patterns, may all have influenced the input of substances to forest ecosystems.
- Estimation of the contributions of individual sources based on $\delta^{34}S$ suggested that the decrease in deposition from the first half to the second half of the isotope monitoring period was attributable to a decline in domestic sources (DOM) in the Lake Ijira catchment (Gifu Prefecture), and to declines in both domestic sources and transboundary air pollution (TRB) in the Kajikawa catchment (Niigata Prefecture) (Figure 17).
- The $\delta^{34}S$ values from precipitation (rainfall, RF), through soil solution (SS), to river water (RW) were sufficiently homogenized within the ecosystem (Figure 18). The markedly high SO_4^{2-} deposition observed in winter in the Kajikawa catchment on the Sea of Japan side was suggested to be buffered by soils, thereby preventing rapid acidification of stream water. In the Lake Ijira catchment, sulfur of geological origin with low $\delta^{34}S$ values was suggested to be the main source of sulfur discharged to stream water, while a large amount of atmospherically derived sulfur was accumulated in the soil. Continued attention is needed to understand how these sulfur cycling and accumulation processes will behave under climate change and during the recovery process of the atmospheric environment.
- Lead isotope ratios also suggested that the sources of pollutants and their contributions may vary depending on time and location between the inland Lake Ijira catchment on the Pacific side and the Kajikawa catchment on the Sea of Japan side. Strontium isotope ratios indicated the occurrence of Asian dust transport at Kajikawa on the Sea of Japan side.

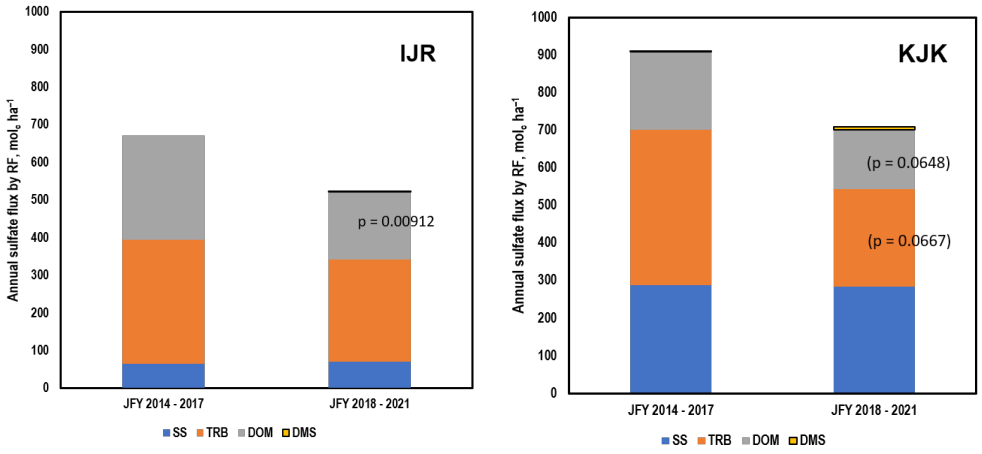


Figure 17. Contributions of individual sources to the mean annual SO_4^{2-} deposition during the first (FY2014–FY2017) and second (FY2018–FY2022) halves of the monitoring period in the Ijira Lake catchment (IJR; left) and the Kajikawa catchment (KJK; right).

Analysis was based on data collected from April 2014 to September 2022.

SS, TRB, DOM, and DMS denote sea salt, transboundary air pollution, domestic sources, and dimethyl sulfide emitted by marine phytoplankton, respectively, as the assumed sulfur sources.

The p-values shown in the figure are the results of Student's t-tests comparing the mean values.

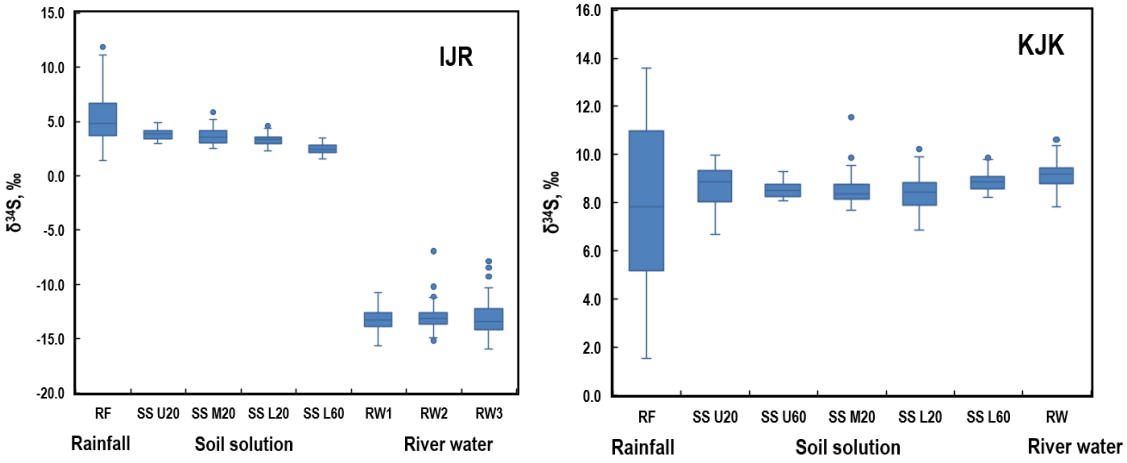


Figure 18. Vertical changes in sulfur isotope ratios (δ^{34}S) from precipitation (RF), through soil solution (SS), to stream water (RW) in the Ijira Lake catchment (IJR; left) and the Kajikawa catchment (KJK; right).

For soil solution (SS), U, M, and L denote the upper, middle, and lower parts of the slope, respectively, and the numbers indicate the sampling depth (cm).

2.5.2 Ozone in forest areas

Measurements of ozone concentrations during the growing season in forest and mountainous areas, where such observations had previously been unavailable, suggested that current ozone levels at Mt. Hakkaisan and Mt. Hikosan are high enough to potentially reduce tree growth. Furthermore, analyses using an atmospheric chemistry model estimated that the ozone concentrations during the period 2015–2019 reduced carbon dioxide uptake by trees by more than 10% at maximum.

- The AOT40 values (accumulated ozone exposure over a threshold of 40 ppb) at Mt. Hakkaisan (Niigata Prefecture) and Mt. Hikosan (Fukuoka Prefecture) showed decreasing trends over the past decade, but remained at relatively high levels (Figure 19). Japanese beech (*Fagus crenata*), the dominant tree species at these sites, is relatively sensitive to ozone. Exposure experiments using seedlings over a period of approximately two years have reported that an AOT40 value of 8–15 ppm·h during a single growing season (six months) reduces tree growth by 10%. These results suggest that the current ozone levels at Mt. Hakkaisan and Mt. Hikosan are sufficiently high to potentially reduce tree growth. In contrast, ozone levels at the outer rim of Lake Mashu were substantially lower than those at Mt. Hakkaisan and Mt. Hikosan. Ozone concentrations in these forest and mountainous areas also exhibited smaller diurnal variations than those at nearby low-elevation monitoring sites, resulting in much higher values of cumulative exposure indices such as AOT40.
- Based on the nationwide distribution of ozone concentrations simulated by an atmospheric chemistry model, reductions in carbon dioxide uptake were estimated using the phytotoxic ozone dose index (POD_0), which represents cumulative stomatal ozone uptake by vegetation. Estimated reductions in carbon dioxide uptake were approximately 4–5% for Japanese beech (*Fagus crenata*) around the borders of Fukui, Shiga, and Kyoto prefectures and from Kanagawa Prefecture to the Izu Peninsula; approximately 14% for konara oak (*Quercus serrata*), with particularly high reductions estimated from Kanagawa Prefecture to the Izu Peninsula; approximately 1–2% for Japanese oak (*Quercus crispula*) throughout its distribution range; and approximately 8% in northern Hokkaido and 10–12% in Honshu for Japanese white birch (*Betula platyphylla* var. *japonica*) (Figure 20).
- These findings suggest that continued attention should be paid to ozone concentrations in forest areas, together with the accumulation of fundamental data on ozone uptake by different tree species.

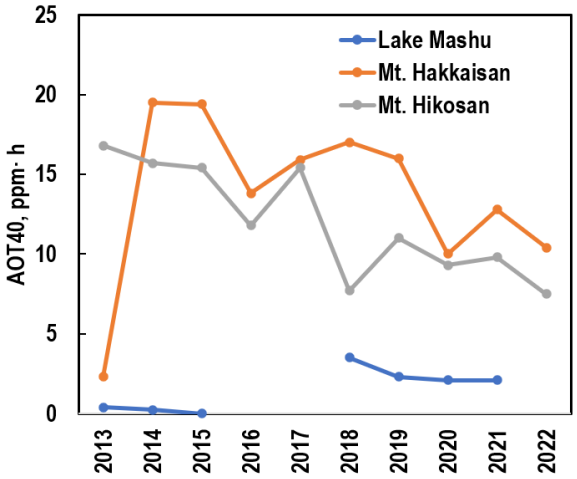


Figure 19. Changes in AOT40 at the outer rim of Lake Mashu, Mt. Hakkaisan, and Mt. Hikosan.

Only data with a data completeness of 80% or higher are shown. The relatively low AOT40 values at Mt. Hakkaisan in 2013 and Mt. Hikosan in 2018 were due to missing data during early spring, when ozone concentrations are relatively high.

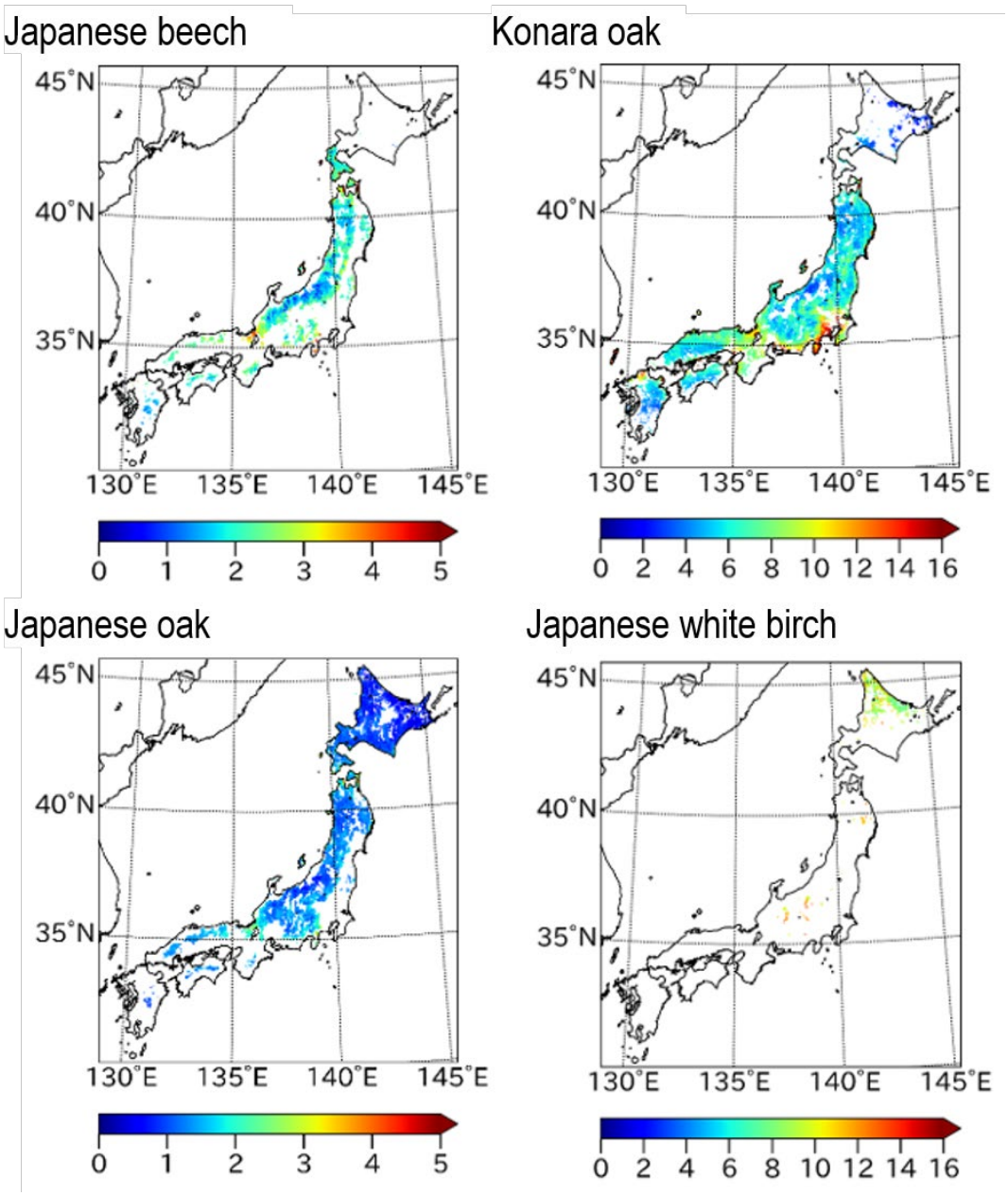


Figure 20. Mean distribution of the estimated reduction in carbon dioxide uptake due to ozone during 2015–2019 (based on simulation model results).
The maps are shown according to the distribution of each tree species. Note that the color scale differs among species.

2.5.3 Impacts of changes in the atmospheric environment on soils, surface waters, and catchments

The decreases in wet deposition of SO_4^{2-} and NO_3^- were accompanied by increases in soil pH (H_2O) and decreases in SO_4^{2-} and NO_3^- concentrations in inland waters at multiple monitoring sites, suggesting mitigation of soil acidification and responses of surface waters associated with improvements in the atmospheric environment. Furthermore, integrated analyses of atmospheric and ecosystem monitoring data made it possible to investigate the processes linking atmospheric pollution to ecosystem responses, from upstream atmospheric inputs to downstream ecological effects. These analyses also demonstrated their value in evaluating the effectiveness of emission control measures and environmental regulations.

- The pH of precipitation in Japan has been increasing. Together with trends in SO_2 and NO_x emissions in Japan and China, this suggests that, in addition to the effects of domestic emission control measures, the contribution of transboundary air pollution has also been declining.
- Accompanying the long-term decrease in atmospheric deposition, including non-sea-salt sulfate (nss-SO_4^{2-}) in wet deposition, increases in soil pH (H_2O) were observed at several monitoring sites (Figure 21). Increased inputs of non-sea-salt sulfate (nss-SO_4^{2-}), ammonium (NH_4^+), and nitrate (NO_3^-), all of which contribute to soil acidification, lead to decreases in soil pH (H_2O). At Bandai-Asahi, Yoshino-Kumano, Daisen-Oki, Amanosan, and Ijira, soil pH (H_2O) showed significant negative correlations with the wet deposition of these ions. These findings suggest that improvements in the atmospheric environment have contributed to the mitigation of soil acidification.
- Since the mid-2000s, decreasing trends in wet deposition of SO_4^{2-} and NO_3^-, as well as decreasing trends in SO_4^{2-} and NO_3^- concentrations in inland waters, have been observed at many sites (Figure 22). Wet deposition of SO_4^{2-} at many atmospheric monitoring sites peaked around 2005 and then showed a decreasing trend (blue band in the figure). Inland water concentrations also peaked in the late 2000s and subsequently declined, except at Lake Karikomi and Ijira Lake, where geological influences are substantial (red band in the figure). Although the trend in NO_3^- concentration was not clear at Imakamiike, where concentration levels were low, decreasing trends were observed in lake water at Ijira Lake and in Futagoike (Oike), together with decreases in wet deposition (Figure 22, right). These findings suggest that changes in the atmospheric environment may have affected inland water concentrations.
- Long-term data on the atmosphere, soils, and stream water accumulated in the Lake Ijira catchment since the 1980s showed interrelated changes, and the mechanisms of acidification and nitrogen saturation triggered by extreme weather events in the 1990s were also clarified. Since 2006, decreases in wet deposition of H^+, SO_4^{2-}, and DIN have been accompanied by increases in soil pH, decreases in stream water NO_3^- concentrations, and increases in stream water pH, suggesting recovery of the forest ecosystem from acidification and nitrogen saturation.
- Integrated analyses of atmospheric and ecosystem monitoring data, including catchment monitoring, enable discussion of processes linking atmospheric pollution to ecosystem responses from upstream atmospheric inputs to downstream effects. These analyses are therefore suggested to be useful for evaluating the effects and effectiveness of emission control measures and environmental regulations. They are also considered effective for understanding nitrogen deposition and its cycling processes, which remain important current issues.

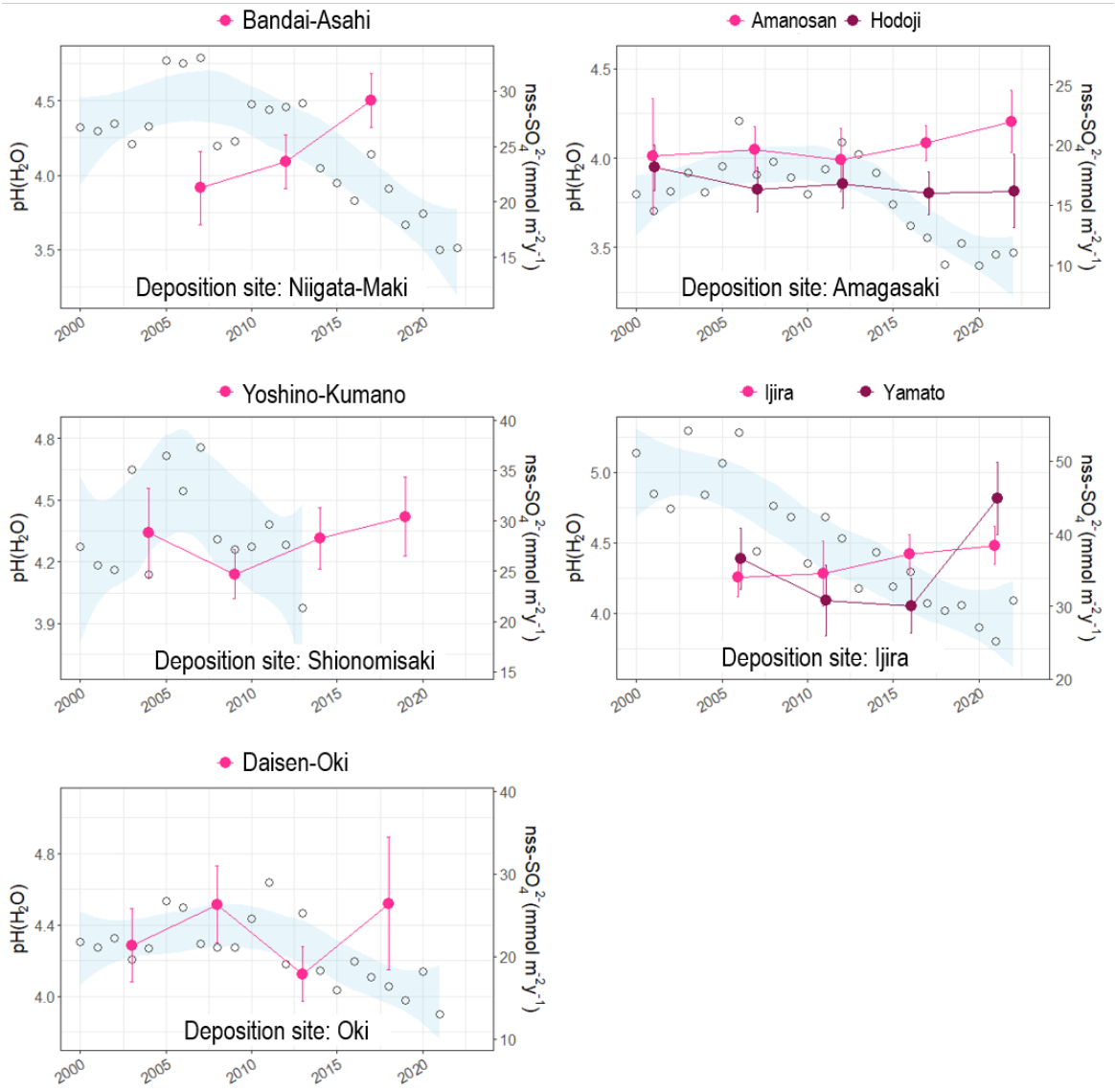


Figure 21. Long-term trends in soil pH (H_2O) and wet deposition of nss-SO_4^{2-}.
Open circles indicate annual wet deposition of nss-SO_4^{2-}. Light blue shaded area, Trend in nss-SO_4^{2-} wet deposition (95% confidence interval estimated by locally weighted smoothing); Solid line, Soil pH (H_2O)

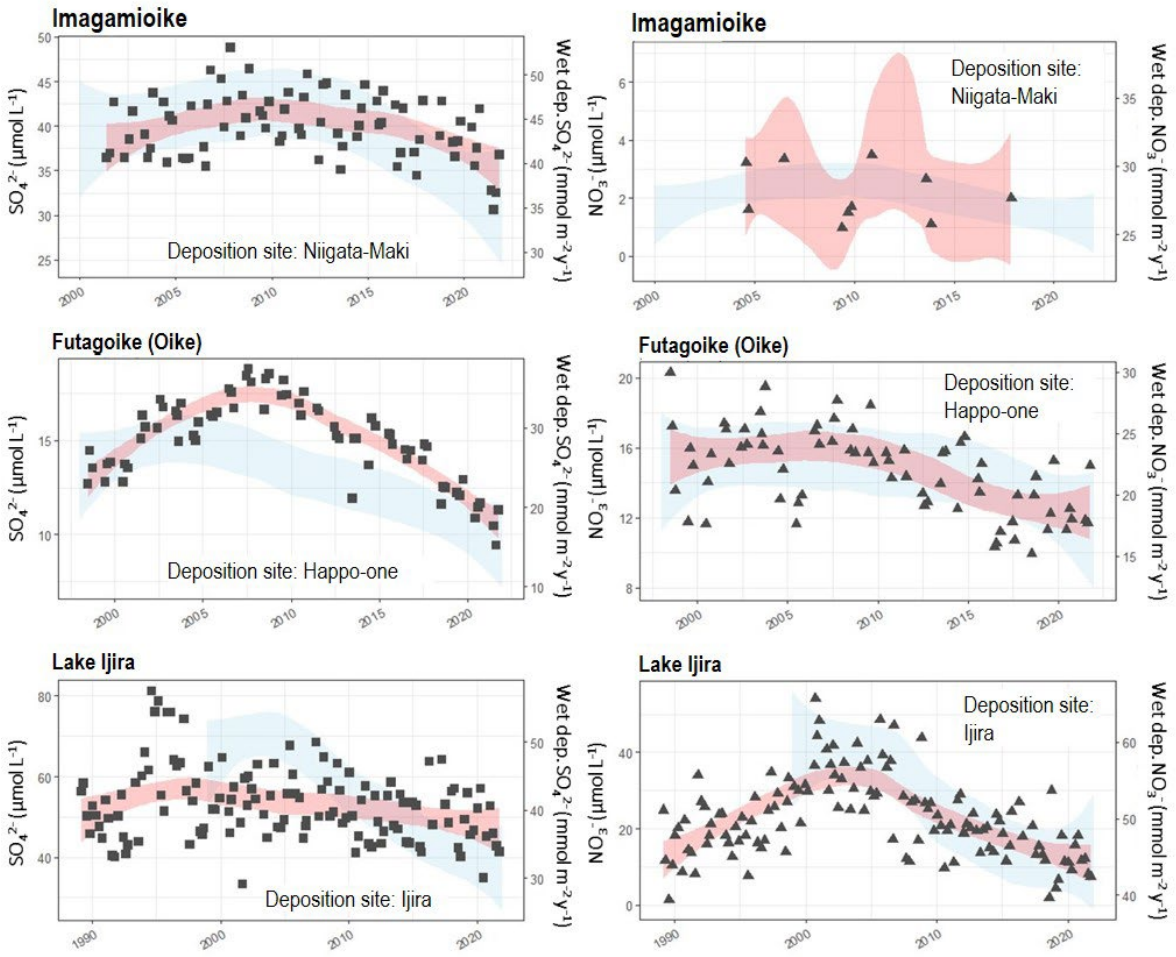


Figure 22. Changes in SO_4^{2-} concentrations (left) and NO_3^- concentrations (right) in inland water associated with long-term changes in wet deposition.

Black squares and triangles indicate SO_4^{2-} concentration (left) and NO_3^- concentration (right) in inland water, respectively. Red shaded area, Trend in inland water NO_3^- concentration (95% confidence interval of the LOESS smoothing curve); Blue shaded area, Trend in wet deposition of NO_3^- (95% confidence interval of the LOESS smoothing curve).

Chapter 3. Future challenges for transboundary air pollution and acid deposition control

3.1 Outcomes of long-term monitoring

- This monitoring program originated from acid rain monitoring in the fields of the atmosphere, soils and vegetation, and surface waters. It has since been expanded into a long-term, nationwide monitoring program for acid deposition and, with particular attention to transboundary transport from the Asian continent, has continuously monitored not only acid deposition but also ozone and particulate matter over an extended period.

- Although the acidity of precipitation in Japan remains relatively high compared with that in Europe and North America, it has shown a continuing decline in recent years, and concentrations of acidifying pollutants have remained at low levels. Furthermore, reductions in acidifying pollutants have been accompanied by signs of recovery from acidification in soils and surface waters at several monitoring sites, reflecting the effectiveness of domestic and international emission control measures implemented to date. In this way, the monitoring program, originally established to assess the acid rain problem, has made significant contributions to the surveillance of transboundary transport of acidifying substances and other air pollutants, as well as to the assessment of long-term trends and the detection of early ecosystem responses.

- The results of this monitoring program, together with its coordination with EANET, are considered to have contributed not only to domestic emission control measures in Japan but also to efforts to promote reductions in emissions of acidifying substances and other air pollutants across East Asia.

3.2 Future challenges

- Building on the achievements of this national monitoring program, it will be important to continue quantitatively assessing transboundary air pollution while strengthening coordination with Japan's national ambient air quality monitoring network. The combination of this national monitoring program, which is suitable for characterizing background concentrations, including the influence of transboundary air pollution, and national ambient air quality monitoring, which is suitable for monitoring domestic influences, provides a basis for distinguishing and assessing the respective contributions of transboundary and domestic sources. Such coordination will contribute to a more comprehensive understanding of air quality in Japan and provide fundamental information for addressing a wide range of air pollution issues.

- For ozone, although high-concentration indicators—particularly in urban areas—have shown decreasing trends as a result of domestic emission control measures, the attainment rate of the environmental quality standard remains extremely low. Ozone also remains a major environmental issue in neighboring upwind countries, including China and the Republic of Korea. Continued assessment of the impacts of transboundary air pollution is therefore expected to remain essential.

- For $PM_{2.5}$, attainment of the environmental quality standard has remained at a high level in recent years. This improvement is attributable not only to progress in domestic emission control measures but also to substantial reductions in $PM_{2.5}$ concentrations across East Asia, particularly in China, resulting from emission control efforts that have reduced the influence of transboundary air pollution. Nevertheless, continued monitoring of transboundary air pollution associated with increasing industrial activity in Northeast Asia remains necessary and will continue to be one of the key roles of this monitoring program.
- New challenges of international importance have also emerged, including nitrogen cycling and nitrogen management, the interactions between air pollution and climate change, and initiatives related to short-lived climate forcers (SLCFs).

Chapter 4. Summary

This monitoring program originated from acid deposition monitoring of the atmosphere, soil and vegetation, and inland aquatic environments, initiated with the First Acid Rain Survey in FY1983. Since then, long-term monitoring has been conducted over a wide geographical area, initially focusing on acid deposition and later expanding to ozone and particulate matter, with particular attention to transboundary transport from the Asian continent.

Although precipitation in Japan remains more acidic than that in Europe and North America, its acidity has decreased in recent years, while concentrations of acidifying substances have remained low. Signs of recovery from acidification have also been observed in soils and inland waters at some sites. These changes are considered to reflect the effects of emission control measures implemented both in Japan and abroad.

Annual mean ozone concentrations at remote sites increased until FY2007 but have generally decreased since FY2015. During FY2003–FY2022, significant decreasing trends were observed at many sites, although increases occurred at some sites. In contrast, daily maximum 8-hour average ozone concentrations showed no clear long-term decreasing trend at most remote and rural sites, with considerable interannual variability. In the Kanto region, reductions in NO_x and VOC emissions were suggested to have reduced summertime peak ozone concentrations. Meteorological variability affecting transboundary transport may partly explain year-to-year variations. Current ozone levels at Mt. Hakkaisan, Niigata Prefecture, and Mt. Hikosan, Fukuoka Prefecture, are sufficiently high to potentially reduce tree growth. A model simulation estimated that, during 2015–2019, ozone concentrations reduced carbon dioxide uptake by trees by up to more than 10% in some areas.

PM_{10} and $PM_{2.5}$ concentrations generally decreased during FY2003–FY2022. Significant decreases were observed at most monitoring sites, particularly for $PM_{2.5}$. Monthly $PM_{2.5}$ concentrations were significantly lower in FY2018–FY2022 than in FY2013–FY2017 at all sites, and high-$PM_{2.5}$ events have decreased since 2015. These long-term declines are considered to reflect reductions in anthropogenic $PM_{2.5}$ emissions, although open biomass burning may contribute to interannual variability.

For soil and vegetation monitoring, no clear soil acidification or tree decline symptoms attributable to acid deposition were reported during the monitoring period. In most cases, the observed decline symptoms were instead attributable to natural factors, such as adverse weather conditions, pests and diseases, and wildlife damage.

For inland aquatic environmental monitoring, SO_4^{2-} and NO_3^- concentrations showed decreasing trends in most of the monitored lakes, which were interpreted as a response of inland waters to reduced atmospheric deposition associated with the recent decline in emissions of acidic substances in East Asia.

In addition, decreasing trends in dissolved inorganic nitrogen (DIN) concentrations, together with increasing trends in pH in both precipitation and stream water at the Lake Ijira catchment, suggested recovery from nitrogen saturation and acidification. Sulfur isotope ratio analysis suggested that sulfur

oxides originating from the Asian continent were transported to Japan by the northwesterly winter monsoon, although their contribution has declined in recent years. Other isotope ratios in rainwater also indicated changes in precipitation patterns, variations in pollution sources, and the seasonal transport of Asian dust.

Integrated analyses of atmospheric and ecosystem monitoring data made it possible to investigate the processes linking atmospheric pollution to ecosystem responses, from upstream atmospheric inputs to downstream ecological effects. These analyses also demonstrated their value for evaluating the effectiveness of emission control measures and environmental regulations.