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FOURTH PERIODIC REPORT ON THE STATE OF ACID DEPOSITION IN EAST ASIA

PART III : EXECUTIVE SUMMARY

CAMBODIA
CHINA
INDONESIA
JAPAN
LAO PDR
MALAYSIA
MONGOLIA
MYANMAR
PHILIPPINES
REPUBLIC OF KOREA
RUSSIA
THAILAND
VIETNAM

ACID DEPOSITION MONITORING NETWORK
IN EAST ASIA (EANET)

The Fourth Periodic Report
on the State of Acid Deposition in East Asia

Part III: Executive Summary

December 2021

Preface

East Asian countries have been experiencing acid deposition for several decades, and therefore, there are various resulting risks in the region as consequences of fast-growing economies and fossil fuel consumption. The pollutants are causing adverse effects on the human body, plants, animals, materials, and cultural heritage.

The Acid Deposition Monitoring Network in East Asia (EANET) has gained international recognition for its success in promoting regional cooperation on acid deposition monitoring in East Asia and facilitating inter-regional exchange on data measurement and assessments of the state of acid deposition.

EANET published scientific assessment reports on the state of acid deposition in East Asia in 2006, 2011, and 2016 based on EANET monitoring data for the periods of 2000-2004, 2005-2009, and 2010-2014, respectively.

This is the *Fourth Periodic Report on the State of Acid Deposition in East Asia* (PRSAD4), describing the outcomes of 20 years of EANET activities (2000-2019). The PRSAD4 is comprised of three parts; Part I: Regional Assessments, Part II: National Assessments, and Part III: Executive Summary. PRSAD4 Part I presents the main activities of EANET and provides an assessment of the state of acid deposition focusing on the special distribution, trend analysis, and, possibly, impact assessment of acid deposition and air pollution in the region. The data from the 2000-2019 period was also utilized for evaluating the current status of the environment in the EANET region, to share the results of the past 20 years provided by EANET in its regular phase, and to scientifically evaluate the monitoring data generated. It includes quality assurance (QA) and quality control (QC), impacts on ecosystems in East Asia, cross-cutting studies on the atmospheric environment in the EANET region, a summary, conclusions, and recommendations.

To develop this report, a drafting committee comprising experts from EANET participating countries was established to lead the scientific evaluation of monitoring data and the achievements of EANET (2000-2019), mainly implemented based on the Medium-Term Plans for EANET with the goals of producing a comprehensive assessment of acid deposition and further developing the EANET network. Part II is a compilation of National Assessments describing monitoring activities, air quality assessments, and control measures implemented at the national level in EANET participating countries. However, it must be noted that the contents of Part II are not covered in this executive summary.

This fourth report in the series represents a significant scientific achievement by EANET. Its publication owes much to the ongoing efforts of EANET participating countries and related collaboration coordinated by the network. The report is expected to promote a better understanding of acid deposition issues in the region and marks an important step in the continuous development of EANET.

Summary for Policy Makers

Summary of Key Points

EANET is an intergovernmental regional network established for promoting cooperation among countries in East Asia to address acid deposition and other related atmospheric pollutants. It aims to create a common understanding of the state of acid deposition in East Asia, provide useful inputs for decision making at local, national, and regional levels, prevent or reduce adverse impacts on the environment caused by acid deposition, and contribute to cooperation on issues related to acid deposition among participating countries.

To assess the state of acid deposition in the East Asia region, the Periodic Report on the State of Acid Deposition in East Asia (PRSAD) was developed every five years. The PRSAD4, based on EANET monitoring data for the 2000-2019 period, is its fourth publication following PRSAD1 in 2006, PRSAD2 in 2011, and PRSAD3 in 2016.

For the PRSAD4, the Network Center (NC) for EANET established the Drafting Committee (DC) to prepare the implementation plan and other related works. The PRSAD4 consists of 3 parts, namely, Part I: Regional Assessment Report, Part II: National Assessment Report, and Part III: Executive Summary. The PRSAD4 provides useful information to EANET participating countries to understand the state of acid deposition in the East Asian region based on monitoring data.

Current status of EANET Monitoring activities

EANET monitoring covers five environmental items namely wet deposition, dry deposition, soil/vegetation, inland aquatic environment, and catchment scale monitoring. The monitoring activities have been conducted following a set of monitoring guidelines and technical manuals. Monitoring for wet and dry deposition is implemented to measure concentrations and fluxes of acidic and other substances deposited in the ground while monitoring soil/vegetation, inland aquatic environment, and catchment are being implemented to assess adverse impacts on terrestrial and aquatic ecosystems. Currently, wet and dry deposition monitoring is being carried out at dozens of sites in the EANET network (including remote, rural, and urban regions). Data and information for ecological impact studies are currently collected from more than dozens of inland aquatic monitoring sites and soil and forest vegetation monitoring sites. All the sites follow a standardized set of methodologies for site selection, sampling, and chemical analyses to ensure technical conformity within the network.

EANET monitoring sites are classified into two basic categories, namely, acid deposition monitoring sites and ecological survey sites. Acid deposition monitoring sites are sites for the collection of fundamental data on the temporal and spatial distribution of acid deposition and related chemical substances, and they are further classified into three sub-categories: remote sites, rural sites, and urban sites, for specific objectives of the monitoring. Ecological survey sites are those that provide basic data for assessing the effects of acidification on terrestrial ecosystems, and they are further classified into two sub-categories: basic survey sites and ecosystem analysis sites.

The number of wet/dry monitoring sites in EANET has been increasing in the past 20 years. The number of the wet deposition monitoring sites was 61 in 2019 (Cambodia: 1, China: 10, Indonesia: 7, Japan: 12, Lao P.D.R: 1, Malaysia: 4, Mongolia: 2, Myanmar: 1, Philippines: 3, the Republic of Korea: 3, Russia: 4, Thailand: 6, Vietnam: 7), representing a 52%-increase from 2000. There are 21 urban sites, 19 rural sites, and 21 remote sites in EANET. So far, there are 54 monitoring sites of dry deposition in EANET, including 41 sites using the filter pack method, 34 sites using the automatic method, and 6 sites using the passive sampler method. Soil monitoring has been conducted at 31 sites in 10 countries.

Since 2000, vegetation monitoring has been conducted at 24 sites in 8 countries. The basic survey was principally carried out for the initial objectives (namely, the establishment of baseline data and early detection of possible impact) in participating countries in accordance with the *Technical Documents for Soil and Vegetation Monitoring in East Asia* (EANET 2000). The EANET has been monitoring forest vegetation at 24 sites in 8 countries. The vegetation monitoring includes conducting comprehensive forest survey, tree decline survey, and understory vegetation survey, but the content and frequency of surveys vary widely. The data on the inland aquatic environment are collected in 20 sites from 11 countries.

Major results of EANET monitoring

Quality Assurance of monitoring data is a crucial condition to ensure reliable and proven results of measurements, which is a solid base to create a common understanding of the status of the acid deposition problems through EANET activities. Monitoring of wet and dry deposition was implemented to evaluate fluxes of acidic substances to the land surface through the measurement of the chemical composition of precipitation and atmospheric aerosols as well as gaseous pollutants, while monitoring for soil/vegetation and inland aquatic environment has been carried out to assess adverse impacts on terrestrial and aquatic ecosystems. QA/QC plays an important role in acid deposition monitoring to ensure that the measurement is accurate, comparable, and of quantifiable quality. The EANET produced four QA/QC programs (Wet Deposition, Air Concentration, Soil and Vegetation, and Inland Aquatic Environment). Several documents regarding QA/QC have been developed by EANET to guide receiving reliable data that is comparable among the participating countries as well as with other monitoring networks outside of the East Asian region.

Summary of Technical Evidence

Data quality

Throughout the implementation of Inter-laboratory Comparison project, the submitted data was evaluated by using the prepared values which were specified by the NC. Using this prepared value, a bias was observed in the statistical data. The possible likelihood value, which should be used for the evaluation, can be the median of all of the compiled data.

The National Monitoring Plan (NMP) of each participating country was prepared after starting formal phase. The NMP should be reviewed every year and revised if necessary. When there is a revision in the monitoring activity, the NMP should be revised immediately and accordingly submitted to the NC.

The Technical Mission has been conducted as one of the additional activities of EANET. It could be effective to build the acid deposition monitoring capacity in participating countries. The potential problems in the laboratories in charge of EANET monitoring in participating countries may not be able to be stimulated to maintain and improve their abilities and their data quality. Even in the laboratories, which have been already regarded as showing high potential, there might remain an unrevealed problem. It may be important to reconsider the modality of the Technical Mission.

The following items should be focused on for the improvement of the quality of monitoring data in EANET: 1) Selection of the monitoring sites focusing on the site category; 2) Establishment of a common point of view on the methodology including sampling and measurement; 3) Implementation of the ILC project; 4) Implementation of the technical mission; 5) Establishment of quality management, and; 6) Reporting of the monitoring data.

Wet and Dry Deposition of Acidic Substances in East Asia

Under the EANET framework, there are currently 61 wet deposition monitoring sites and 54 dry deposition monitoring sites, located in 13 EANET member countries. Following EANET criteria, the monitoring sites are categorized into three types: urban, rural, and remote. Sampling techniques, sample storage and analysis, data processing, and QA/QC procedures of all monitoring sites and laboratories participating in EANET are conducted according to EANET's Guidelines and Manuals.

Precipitation chemistry was evaluated mainly based on pH, pAi, the acid concentration sum (nss-SO_4^{2-} + NO_3^-), and the base concentration sum (NH_4^+ + nss-Ca^{2+}). The lowest pH values were all observed at sites where the acid concentration sum was larger than the base concentration sum. The acid concentration sums at the remote site tend to decrease in Northeast Asia, as at the rural and urban sites. In the 2010s, H^+ concentrations at monitoring sites in China, Korea, and Japan have been declining significantly. The acid concentration sums have increased in recent years in Vietnam and Malaysia.

From the comparison of precipitation chemistry among EANET, NADP, and EMEP, the pH values and nss-SO_4^{2-} concentrations showed significant increasing and decreasing trends, respectively, in all networks from 2000 to 2019. The NO_3^- concentrations showed significantly a decreasing trend from NADP and EMEP, and a slightly decreasing trend from EANET. The decreasing trend of nss-SO_4^{2-} concentration has been significant from the period 2004-2014 to that of 2009-2019 in Northeast Asia, especially, at monitoring sites in China, the Republic of Korea, and Japan. The nss-SO_4^{2-} concentration in Southeast Asia had exhibited an insignificant trend. The NO_3^- concentration did not show a significant trend in Northeast Asia and Southeast Asia. Since the nss-Ca^{2+} and NH_4^+ concentrations showed an increasing trend, the pH increasing trend in Southeast Asia was possibly caused by the increase of cations.

Total (dry and wet) depositions of S and N were evaluated at the sites carrying out the 4-stage filter pack monitoring in Russia, Mongolia, Japan, Vietnam, Thailand, Malaysia, and Indonesia. Dry depositions were estimated by simplifying the inferential method using available monthly meteorological data. Low total S depositions were found at the remote sites located in the northern inland and the Pacific Ocean, and high total S depositions were found in the Japanese remote sites near the Asian continent. In the high S deposition sites, a decreasing trend of S depositions was found in recent years. A high total S depositions of more than 30 kg S/ha/year were found at urban sites in Southeast Asia. The dry S depositions decreased from the 2000s at Hanoi and Bangkok. In remote sites, spatial and temporal trends of the total N depositions were similar to those of the total S depositions, and reduced N depositions contributed more than half the total N depositions at lower N deposition sites, and oxidized N depositions contributed more than half the total N depositions at higher N deposition sites. Regardless of the site categories, reduced N depositions contributed more than half the total N depositions for many years at many sites, except at Sado-seki, Oki, and Petaling Jaya. The trend analysis for the total S and N, as well as oxidized and reduced N depositions, has shown a significant increase in some sites in Southeast Asia and a significant decrease in some sites in Northeast Asia, especially over the last decade.

Gas and Aerosol Pollution in East Asia

EANET has conducted continuous monitoring in various countries in 20 years and the number of gas and aerosol monitoring sites has increased each year. There were 54 air concentration monitoring sites operating in the EANET network as of 2019. The measuring components, which are SO_2, NO, NO_2, NOx, $PM_{10}/_{2.5}$, O_3, HNO_3, HCl, NH_3, and Particulate Matter Components (PMCs) vary, depending on the methodology employed at each site.

Concentrations of SO_2, NO_2, NOx, O_3, PM_{10}, and $PM_{2.5}$ were monitored in each EANET member country from 2015 to 2019. The spatial distribution of SO_2 in Russia and Mongolia varied from 0.18 to 9.10 ppb. This significant difference in the spatial distribution of concentration shows the influence of population distribution, as well as the effect of heating and fuel composition such as coal burning. After continuous improvement, SO_2 concentrations at monitoring sites in China have been significantly reduced and the 5-year average range for the 5 stations in China is 0.1 ppb-3.54 ppb. The SO_2 concentrations at all sites in Japan have been very low for a long time, with a 5-year average range of 0.03 ppb-1.3 ppb, with the maximum value occurring in Tokyo. The SO_2 concentration range at the three land sites in the Republic of Korea is similar to that of China, with a 5-year average range of 0.66 ppb-2.44 ppb. It was observed that, except for in Indonesia, SO_2 concentrations in Southeast Asian countries are generally low.

Among the ozone observations in eight countries namely Cambodia, Indonesia, Japan, Mongolia, Philippines, the Republic of Korea, Russia, and Thailand, the Republic of Korea had the highest nationwide O_3 5-year average concentration of 40.47 ppb from 3 stations. Japan reported the next highest nationwide average O_3 concentration. In contrast, O_3 concentration in Southeast Asian countries was found to be low.

Among the NOx observations at monitoring sites of six countries of China, Japan, Lao PDR, Malaysia, Mongolia, Philippines, and Thailand, Mongolia had the highest NOx 5-year average concentration of 55.0 ppb at Ulaanbaatar. Among the NO_2 observations at monitoring sites of eight countries of China, Indonesia Japan, Lao PDR, Mongolia, Philippines, the Republic of Korea, and Thailand, there are large spatial differences in the distribution of NO_2 concentrations.

Among the $PM_{2.5}$ observations in ten countries namely Cambodia, Indonesia, Japan, Lao PDR, Mongolia, Myanmar, Philippines, the Republic of Korea, Thailand, and Vietnam, Mongolia had the highest $PM_{2.5}$ 5-year average concentration. Jakarta in Indonesia reported the next highest nationwide averaged $PM_{2.5}$ concentration and Hoa Binh in Vietnam is close to that of Jakarta.

Among the PM_{10} observations at monitoring sites of seven countries namely China, Japan, Lao PDR, Mongolia, Philippines, the Republic of Korea, and Thailand, there are large spatial differences in the distribution of PM_{10} concentrations. Ulaanbaatar in Mongolia and Vientiane in Lao PDR have the highest and the second-highest PM_{10} concentrations with a 5-year mean concentration.

As for HNO_3, NH_3, and HCl at monitoring sites of each EANET member country from 2015 to 2019, the concentration of HNO_3 at one site in China was relatively high during the period from 2015 to 2019. China exhibited a relatively high NH_3 concentration with a 5-year average concentration at the Hongwen site, followed by Indonesia. The highest HCl concentration was found in Russia with a 5-year mean concentration with large differences in its geospatial distribution.

As for SO_4^{2-}, NO_3^-, NH_4^+, and Ca^{2+} components in particulate matter in each EANET member country from 2015 to 2019, relatively high SO_4^{2-}, NO_3^-, and NH_4^+ concentrations were found in Hongwen in China. The highest 5-year averaged Ca^{2+} concentration was found in Vietnam.

Impacts on Ecosystems in East Asia

Soil chemical properties and forest growth of EANET sites were not necessarily parallel to the input of atmospheric deposition because of their diversity of characteristics such as anthropogenic disturbance, and pest infestation (bark beetle). However, in some sites, changing patterns of ion concentrations in lakes/streams were associated with those of atmospheric deposition at the nearest deposition monitoring sites. Monitoring sites in China, Japan, and Indonesia showed symptoms suggesting their recovery from acidification. Moreover, biogeochemical cycles in two Japanese forest catchments have sensitively responded to the recent decline of atmospheric deposition (in particular, SO_4^{2-}) resulting in phenomena indicating recovery from acidification. However, various factors, such as forest types, maturation of trees, and climatic conditions, appear to have influenced N leaching to stream water.

Relevant Studies on Atmospheric Environment Assessment in the EANET Region

Several studies have been carried out in the last few years to elucidate the cause and response of regional acid deposition and air pollution and their human health and ecological impacts in East Asia based on observational and modeling studies. Additionally, some original and/or summarized analyses are conducted on the satellite observation, acidification in forest catchments, impact studies on human health and ecological system, and trends of air quality, acid deposition, and their environmental impacts. Most of those studies are strongly linked to EANET in that the entire EANET monitoring data have been used or the results were obtained in the EANET “additional activities” by the collaboration of scientists of participating countries. These kinds of scientific investigations are crucial for EANET for conveying the implications of these activities to policymakers and the public. More active integration of emission inventory, model simulation, monitoring including satellite observation, and human health and ecological impact studies would be key for the future successful operation of EANET.

Table of Contents

1. Introduction

The Acid Deposition Monitoring Network in East Asia (EANET) is an intergovernmental regional network that was established to promote cooperation among countries in East Asia to address the issue of acid deposition. EANET has gained international recognition for its success in promoting regional cooperation for acid deposition monitoring in East Asia and facilitating inter-regional exchange on data measurement and assessments of the state of acid deposition. Other than conducting technical acid deposition monitoring, EANET also implements public awareness and research activities to promote a common understanding of the state of acid deposition and prevailing air pollution problems.

The objective of issuing the Fourth Periodic Report on the State of Acid Deposition in East Asia (PRSAD4) is to provide comprehensive information to the international community on long-term trends and the regional distribution of acid deposition and air pollution in East Asia. EANET published scientific assessment reports on the state of acid deposition in East Asia in 2006, 2011, and 2016 based on the EANET monitoring data for the periods 2000-2004, 2005-2009, and 2010-2014, respectively.

Preparation of the fourth period report

EANET published scientific assessment reports on the state of acid deposition in East Asia in 2006, 2011, and 2016 based on the EANET monitoring data accumulated from 2000 to 2004, 2005 to 2009, and 2010 to 2014, respectively. This report is the fourth periodic report on the state of acid deposition in East Asia based on the monitoring data from 2000 to 2019 (i.e., twenty years), focusing on the spatial distribution, trend analysis, and, possibly, impact assessment of acid deposition and air pollution in the region. The 2015-2019 data was also utilized for evaluating the current status of the environment in the EANET region, following the first, second, and third periodic reports, to share the results of the previous five years provided by EANET in its regular phase, as well as to scientifically evaluate the monitoring data generated.

The Nineteenth Session of the Scientific Advisory Committee (SAC19) in October 2019 approved the establishment of the Drafting Committee (DC) for the PRSAD4 to prepare the implementation plan and other related works. DC was endorsed by the Twenty-First Session of the Intergovernmental Meeting (IG21) in November 2019, in Beijing, China. The first DC meeting for the PRSAD4 was held virtually on May 12, 2020, due to the COVID-19 pandemic. It was organized by the Network Center (NC) for EANET in Niigata, Japan. The DC members from 13 participating countries of EANET participated in the meeting. Prof. Fan Meng, Institute of Chinese Research Academy of Environmental Sciences, China, was elected as the Chairperson of the DC.

The DC developed the format and contents of the PRSAD4. The PRSAD4 consists of three parts: Part I: Regional Assessment, Part II: National Assessment, and Part III: Executive Summary. The DC also nominated the lead authors and contributors to draft the report. The contents of the PRSAD4 are as follows:

- Chapter 1: Introduction
- Chapter 2: Data Quality
- Chapter 3: Wet and Dry Deposition of Acidic Substances in East Asia
- Chapter 4: Gas and Aerosol Pollution in East Asia
- Chapter 5: Impacts on Ecosystems in East Asia
- Chapter 6: Relevant Studies on Atmospheric Environment Assessment in the EANET region
- Chapter 7: Summary and Recommendations for Future Activities

The report was prepared as a scientific assessment of the state of acid deposition in East Asia based on the data accumulated from the network. A trend analysis was also undertaken to investigate the long-term variation in acid deposition using data dating prior to 2015 in addition to the data from the 2015-2019 period for this report.

EANET activities in 2000-2019

EANET was established as an important initiative for regional cooperation among participating countries to create a common understanding of the state of acid deposition problems and provide useful inputs to policymakers at various levels.

Monitoring activities

EANET monitoring covers five environmental media namely, wet deposition, dry deposition, soil/vegetation, inland aquatic environment, and catchment-scale monitoring. Monitoring activities were conducted following a set of monitoring guidelines and technical manuals. Monitoring for wet and dry deposition is implemented to measure concentrations and fluxes of acidic and other substances to the ground. In addition, monitoring soil/vegetation, inland aquatic environment, and catchment are being implemented to assess adverse impacts on terrestrial and aquatic ecosystems. Currently, wet and dry deposition monitoring is being carried out at dozens of sites in the EANET network (including remote, rural, and urban regions). Data and information for ecological impact studies are currently collected from more than dozens of inland aquatic monitoring sites and soil and forest vegetation monitoring sites. All the sites follow a standardized set of methodologies for site selection, sampling, and chemical analyses to ensure technical conformity within the network.

EANET monitoring sites are classified into two basic categories: acid deposition monitoring sites and ecological survey sites. Acid deposition monitoring sites are sites for collecting fundamental data on the temporal and spatial distribution of acid deposition. They are further classified into three sub-categories: remote sites, rural sites, and urban sites for specific objectives of the monitoring. Ecological survey sites provide basic data for assessing the effects of acidification on terrestrial ecosystems. They are further classified into two sub-categories: basic survey sites and ecosystem analysis sites.

Thirteen EANET countries, namely, Cambodia, China, Indonesia, Japan, Lao PDR, Malaysia, Mongolia, Myanmar, Philippines, the Republic of Korea, Russia, Thailand, and Vietnam have participated in acid deposition monitoring. A total of sixty-six sites were nominated for the monitoring, including 26 urban, 19 rural, and 21 remote sites.

Research activities

The promotion of research activity is specified in the Medium Term Plan of EANET. Scientific findings from the research activities are shared with the scientists from participating countries and their scientific and technical research results are published in relevant international and national journals, including in the EANET Science Bulletin.

Research activities are implemented to improve acid deposition monitoring methodologies, deposition estimations, and building capacity in the development of emission inventories, and will also promote efforts to develop and use appropriate models to assess and analyze the trend of acid deposition and other relevant air pollutants at all scales.

Research activities related to acid deposition problems include the promotion of the following:

- Research studies, particularly on the applicability of various methodologies for the measurement of air concentrations in East Asia;
- Studies on the effects of acid deposition and other priority chemical species on the ecosystem, human health, and other social aspects from the viewpoint of the socio-economics; and
- Studies on proposed models to assess and analyze the trend of national and regional acid deposition and other air pollutants in East Asia by conducting an evaluation of existing models and providing a suitable model, and the promotion of the atmospheric simulation model through activities such as workshops, training courses; and

- Emission inventories through initiatives such as workshops, training courses, pilot studies, and the preparation of reference materials. Joint research projects are also implemented as well. Several joint scientific research projects on acid deposition and its effects were conducted by the EANET participating countries.

The fellowship research program is also introduced annually. It has been promoted since 2005 and is an effective mechanism for encouraging young researchers from across the EANET region to participate in air pollution research activities.

Capacity building activities

The EANET individual training course at the NC has been implemented once or twice a year from the commencement of the EANET activities. The training courses consist of wet deposition, dry deposition, soil/vegetation and inland aquatic environment monitoring, and data management. The NC carries out the annual questionnaire survey on training activities conducted by the participating countries to gather both information on training requirements and suggestions on new training areas.

The technical capabilities and skills of the participating countries for acid deposition monitoring and assessment were significantly enhanced through many EANET activities. The NC dispatched technical missions annually to participating countries to assist them in monitoring performance, laboratory operations, data management, and other procedures. Other activities to enhance the skills and knowledge of personnel included national workshops, annual expert meetings, and scientific workshops on ecological impacts and other topics related to acid deposition. Numerous EANET publications (e.g., technical manuals and guidelines, data reports, reports on QA/QC projects, training materials, etc.) have been produced for use by the technical staff, specialists, and researchers involved in EANET monitoring, data quality, and data management. All these materials are available on the EANET website (https://www.eanet.asia).

Public awareness activities

The promotion of public awareness of acid deposition and sharing a common understanding of atmospheric environmental issues is an important component of EANET activities. Public awareness activities on acid deposition and other priority chemical species, including their effects, control, and mitigation measures, are being regularly conducted by EANET.

Reports for policymakers titled "Goals, Achievements and Way Forward," "Clean Air for Sustainable Future," "EANET and Clean Air for Sustainable Development," and "Toward Clean Air for Sustainable Future in East Asia through Collaborative Activities" were published. EANET has undertaken joint public awareness projects with participating countries to develop brochures on acid deposition and conducted the "Workshop on Public Awareness on Acid Deposition Problems" in participating countries. The capacity-building workshops for government officials, academicians, non-government organizations, and private sectors were held by EANET to raise awareness of the adverse impacts of acid deposition on the environment. The factsheet titled "Countries' efforts and achievements in combating acid deposition" was developed by all participating countries of EANET through collaboration and coordination with the NC and Secretariat. The published public awareness materials were also issued on the EANET website.

The Science Bulletin Vol.1-5 was published to share the research findings from the EANET Research Fellowship Program, Joint Research Project, and other scientific papers using EANET monitoring data.

2. Data Quality

QA/QC

Quality Assurance (QA) and Quality Control (QC) play important roles in acid deposition monitoring to ensure that the measurements are accurate and of comparable and quantifiable quality. EANET produced four QA/QC programs (Wet Deposition, Air Concentration, Soil and Vegetation, and Inland Aquatic Environment) in 2000. Those are composed of several procedure types, as follows: i) siting, ii) sampling procedures and sample handling, iii) chemical analysis, and iv) QA/QC for the measurement data at each site. Several documents regarding QA/QC have been developed by EANET to guide receiving reliable data that can be compared among the participating countries as well as with other monitoring networks outside the East Asian region.

Preparation of National Monitoring Plan (NMP)

Every participating country of EANET has been required to prepare its NMP as per the "Data Reporting Procedures and Formats for Acid Deposition Monitoring in East Asia." The NMPs were prepared by participating countries after starting formal phase and have been reviewed every year with possible revisions, if necessary.

Siting

EANET monitoring sites are classified into two basic categories: acid deposition monitoring sites and ecological survey sites. Acid deposition monitoring sites are operating bodies collecting fundamental data on air concentrations and wet acidified deposition to investigate its temporal and spatial distribution, and they are classified into three sub-categories: remote sites, rural sites, and urban sites, with some differences in the objectives of the monitoring. Ecological survey sites are those that provide basic data for assessing the effects of acidification on terrestrial ecosystems, and they are principally classified into two sub-categories: survey sites and ecosystem analysis sites.

Sampling and sample handling

The sampling was carried out according to the monitoring manuals for each monitoring category. Especially for the collected wet deposition samples, dry monitoring samples using the filter pack method, and inland water samples, samples were stored in a refrigerator or a biocide such as thymol was added to them to prevent deterioration or at least minimize possible conversion of the chemical species in the collected sample before chemical analysis. The collected soil samples were air-dried, sieved, and then stored in a cool and dark place before the analysis.

Chemical analysis

The major ions in the collected water samples namely, hydronium ion (H^+, as pH), ammonium ion (NH_4^+), calcium ion (Ca^{2+}), potassium ion (K^+), magnesium ion (Mg^{2+}), sodium ion (Na^+), sulfate ion (SO_4^{2-}), nitrate ion (NO_3^-), and chloride ion (Cl^-), are analyzed. Alkalinity, as an indicator for acid neutralizing capacity, is also analyzed in the inland water samples, while exchangeable cations, including Ca^{2+}, Mg^{2+}, K^+, Na^+, H^+, and Al^{3+}, are analyzed in the soil samples. The operation manuals for each monitoring category provide several acceptable analytical techniques to be applied in the laboratories of participating countries.

QA/QC procedure prior to the data submission to the national and network center

The acceptable ranges for ion balance, R1, and conductivity agreement, R2, are defined as a function of the concentration sums of the analytical suites. When the value of either R1 or R2 does not meet the criteria, the samples are supposed to be a subject of reanalysis as well as a more detailed examination of all analytical procedures is involved. Then, it is also recommended that some ionic species, including hydrogen carbonate, HCO_3^-, or organic acids, be added to the analytical suites or the criteria should be modified as a function of the potential of hydrogen (pH). The acceptable ranges are encouraged to be revised considering the ionic composition and concentration levels as well as the current status of analytical apparatus in the laboratory.

Evaluation of Inter Laboratory Comparison (ILC) Projects

The ILC project of EANET is a well-designed worldwide approach of the round-robin test performed with a common set of simulated water (artificial rainwater or surface water) or unified common samples in all analytical laboratories for EANET monitoring. The purpose of this project is to evaluate the analytical systems through the evaluation of analytical accomplishments, analytical instruments, their operating condition, and other relevant and appropriate practices.

Wet deposition

The ILC surveys on wet deposition were carried out 22 times. The results of the analysis in the project were evaluated in terms of the exceedance of the values of the Data Quality Objectives (DQOs) which had been prescribed in the EANET QA/QC program as a deviation of ±15% from the prepared value for each analytical suite. The flag "E" was placed on the data exceeding DQOs by a factor of 2 (±15%~±30%), and the flag "X" on the data exceeding DQOs by a factor of over 2 (<-30% or >30%). A set of the data for each sample was evaluated with the data checking procedures. The overall percentages of flagged data, "E" and "X," respectively, are shown in Figure 2.1. The percentage of data within the DQOs increased from 75.4-78.3% (1998) to 86.1-94.1% (2019). The proportion of flag data that depends on the component concentration of artificial precipitation sample has been declining recently, despite the low component concentration.

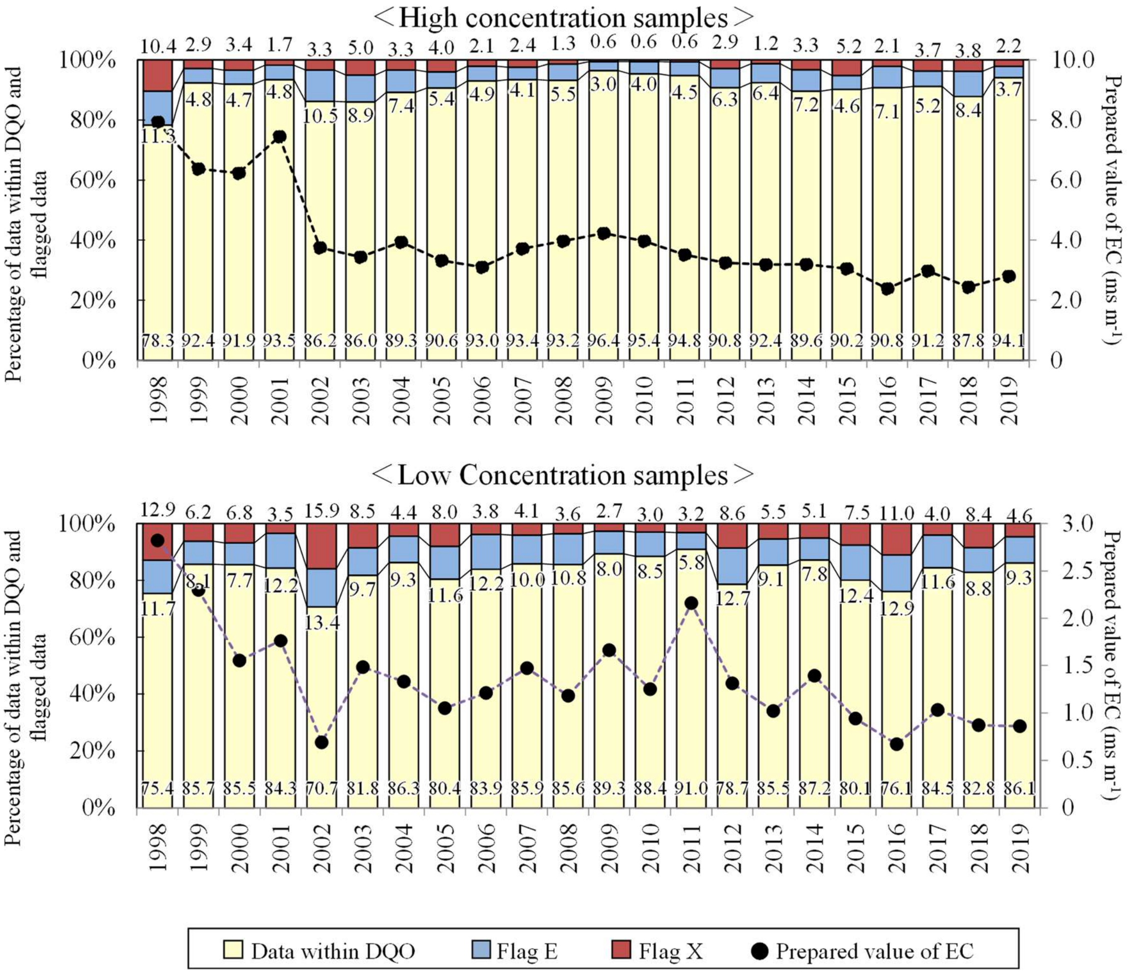
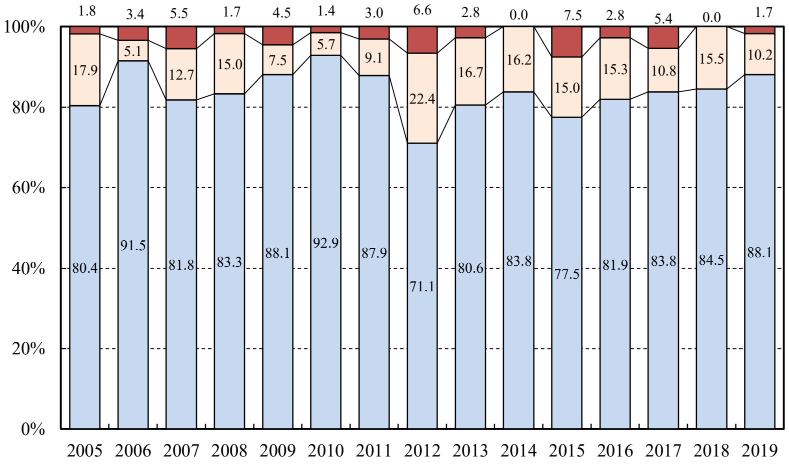


Figure 2.1. Overall comparisons of all Inter-Laboratory Comparison projects for 1998-2019.

Dry deposition

The ILC surveys were carried out 15 times. According to *Technical Manual for Air Concentration Monitoring in East Asia* (2013), the Data Quality Objectives of EANET require that the determined values be within ±15% of the setting value. Each laboratory analyzed each sample three times, and these average values were evaluated based on the deviation from the corresponding prepared values. The flag "E" indicates that its deviation exceeds ±15% but not ±30%, and the flag "X" indicates that its deviation exceeds ±30%. Figure 2.2. shows the overall percentages of flagged data. The percentage of data within the DQOs decreased from 73.2% (2005) to 62.7% (2019) for small quantity samples and increased from 80.4% (2005) to 88.1% (2019) for large quantity samples.

<Large quantity samples >



< Small quantity samples >

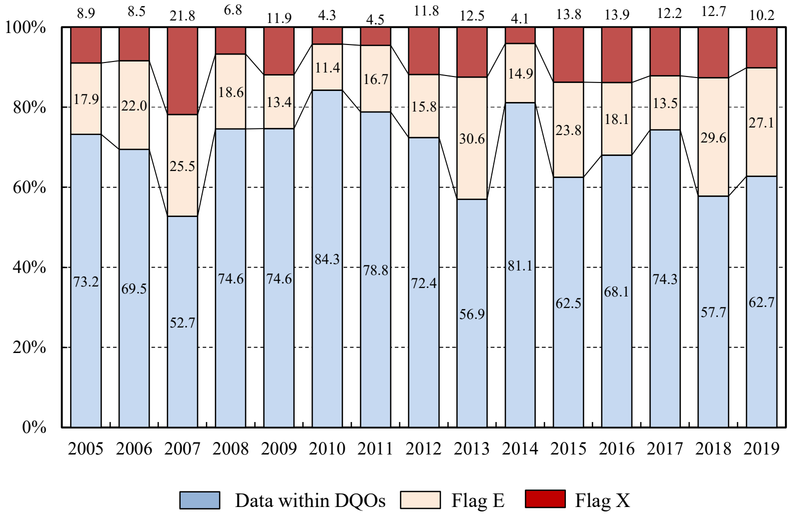


Figure 2.2. Overall comparisons of the Inter-Laboratory Comparison projects.

Soil

The project on soil sample analysis commenced in 1999 as one of the activities within the QA/QC programs. Air-dried soil samples, which were prepared by the NC, were distributed to the laboratories every year except in 2001. The laboratories carried out the complete procedures of the soil analysis including extraction, instrumental analysis or titration, and reporting. The laboratories analyzed each sample twice under the within-laboratory reproducibility condition (wherein time, analysts, and/or instruments were different in the same laboratory). Moreover, triplicate analysis under the repeatability condition was carried out each time. Each parameter was statistically evaluated according to several procedures. One of the procedures is the detection of outliers using the Cochran and Grubbs method. The evenness of within-laboratory precision (variation in each laboratory) and inter-laboratory precision (variation between participant laboratories) are verified using this method. Figure 2.3. shows the change of outlier ratio in all properties and laboratories from 2002 to 2019 [the ratio is calculated by {(N of entire dataset) - (N of verified dataset)} / (N of entire dataset)]. Although the ratio decreased from the first experiment in 2002, it is still high (10-

25% from 2003 to 2019). The outlier ratio in 2019 was lower than the average outlier ratio from 2003 to 2018, which was approximately 14%. Outliers may disturb the evaluation and understanding of actual monitoring data. For an ILC project on soil, a decrease in the outliers is most important task in the near future. The appropriate standard solution, extraction solution, dilution rate, and calculation should be checked to reduce extremely different values which would be considered outliers.

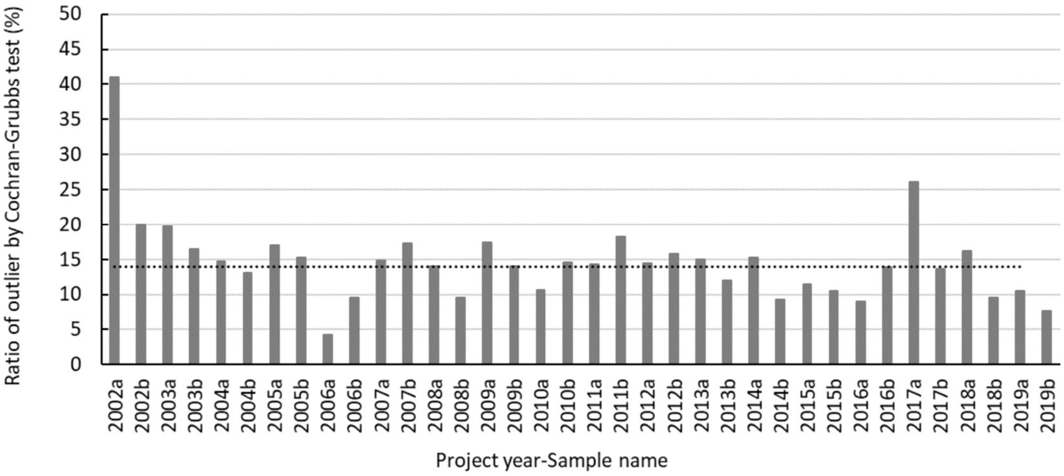


Figure 2.3. Change of the outlier ratio in all properties and laboratories from 2002 to 2019 calculated by [(*N* of entire dataset) – (*N* of verified dataset)] / (*N* of entire dataset). 'a' and 'b' show the two kinds of samples in each year (e.g., 191s and 192s). The dotted line represents the average outlier ratio from 2003 to 2018.

Inland aquatic environment

The project on the inland aquatic environment sample analysis commenced in 2000 as one of the activities within the QA/QC programs. One artificial inland water sample, which contained all analyzed ions and was prepared by the NC, was distributed to the laboratories every year. The laboratories carried out the complete procedures of the inland water analysis including extraction, instrumental analysis or titration, and reporting. The performance at each step was responsible for any inter-laboratory variations. The ILC projects of EANET were carried out 20 times, and the results showing the percentage of flagged data and that of data that satisfied the DQOs are shown in Figure 2.4.

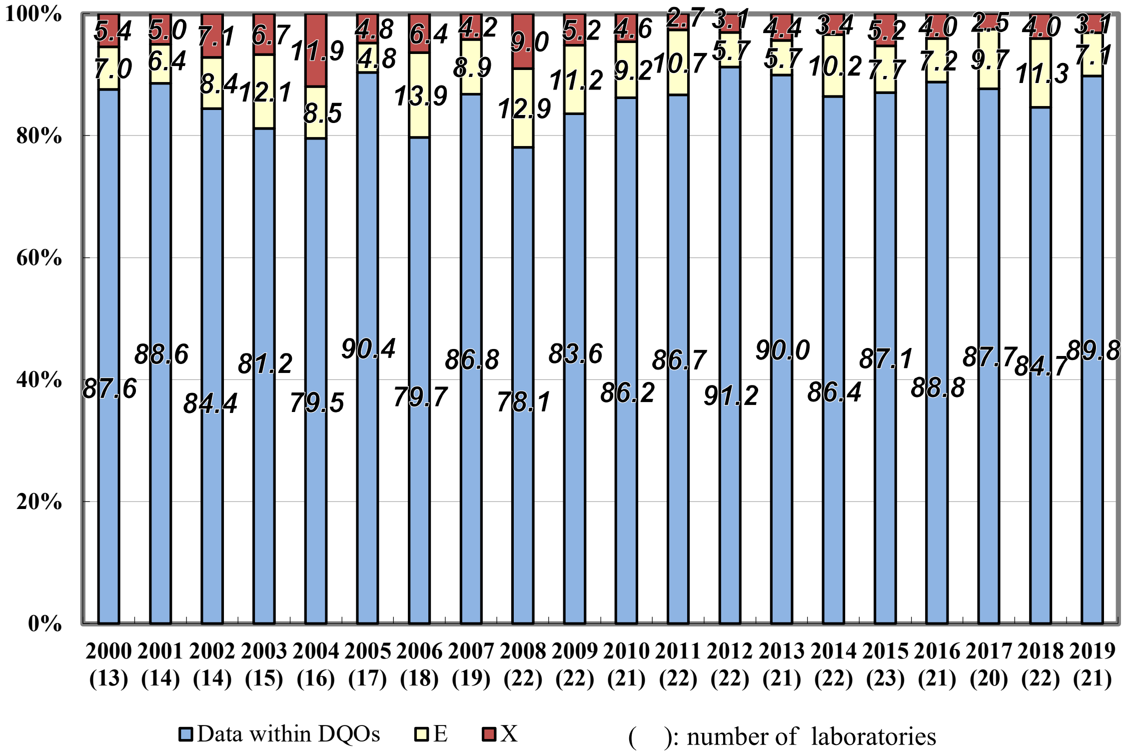


Figure 2.4. Comparison of the results from the Inter-Laboratory Comparison projects.

Evaluation of the measurements

The objectives of the QA/QC program are to obtain reliable data that can be comparable among the countries of the East Asian region, as well as with other networks by ensuring data accuracy, precision, representativeness, and completeness in acid deposition monitoring. All activities of data management are directed to ensure acceptable quality assurance levels. This section describes data evaluation processes for QA/QC of EANET monitoring.

Flow for the data reporting and verification

All datasets are eventually collected at the NC where the datasets are further examined and qualified through communications with national centers. The datasets qualified during that period are submitted to the international data verification group for the final checking process. The international experts in the monitoring activities qualify each of the datasets in a careful and detailed manner. After a discussion between the NC and national centers, the verified datasets are submitted to the Scientific Advisory Committee (SAC) to be scientifically and technically approved. The annual data report is completed after SAC consideration, which is officially and finally approved in the Intergovernmental Meeting.

Validation of the data for wet deposition

The tables with annual sets of wet deposition data include certain flags to reflect values from the criteria or questions to the individual samples or their analysis. The most frequent problems include an absence of value due to “insufficient sample volume”(for analysis) or “low precision”, therefore, the correspondent flags are attached to those data, both for a whole sample or individual datum. The list of flags is included in the Technical Manual for Wet Deposition Monitoring in East Asia (2010). Most of them are completely in compliance with similar flags recommended by the European Monitoring and Evaluation Program (EMEP) or World Meteorological Organization (WMO) in their manuals on precipitation chemistry monitoring (EMEP, 2001; WMO, 2004).

Data completeness

Data Completeness is one of the data quality indicators used when measurement data are summarized statistically in monthly, seasonal, or annual periods. If large amounts of data are missing or regarded as invalid during the summary period, the summarized statistics (e.g., the mean, median, maximum, and minimum values) can be highly misleading. Therefore, periodic reporting of summarized data should be based on the full operation measurements without any missing data, which is inevitable in the long-term regional monitoring network activities.

Site representativeness

The characteristics of the individual site were reported by the NMP of each EANET member country. It is important to consider the nature of the sites (e.g., urban, rural, remote, or ecological) for the evaluation of monitoring data.

3. Wet and Dry Deposition of Acidic Substances in East Asia

Under the EANET framework, there are currently 61 wet deposition monitoring sites and 54 dry deposition monitoring sites located in 13 EANET member countries. In accordance with EANET criteria, the monitoring sites are categorized into three types: urban, rural, and remote. Sampling techniques, sample storage and analysis, data processing, and QA/QC procedures of all monitoring sites and laboratories participating in EANET are conducted according to EANET's guidelines and manuals. The details of each process can be found in the main text.

Precipitation chemistry in East Asia

In the average precipitation composition for 20 years at each observation site, cations and anions are balanced, and it can be confirmed that the nine components are the main ions at those sites. In contrast, at other sites, the cation sum tends to be higher than the anion sum. This suggests the existence of unmeasured anions, most of which are thought to be due to bicarbonate ions, fluorine ions, and organic acids, and actual measurements of those ions have been reported from many sites. Therefore, the determination of dissolved components has been performed without defects in EANET and the uncertainty of the measured value is considered small (Figure 3.1). In addition, when unmeasured anions are not dissolved in precipitation, the sum of non-sea-salt anions (nss-SO_4^{2-}, NO_3^-) and the sum of non-sea-salt cations (H^+, NH_4^+, nss-Ca^{2+}) are balanced. These relationships between anions and cations can be understood as a result of the neutralization of sulfuric and nitric acids produced by the gas-phase and liquid-phase reactions by basic ammonia gas and calcium particles. In other words, it can be confirmed from actual observation data that the acidity (H^+ concentration) of precipitation is determined by the balance of acids and bases dissolved in precipitation.

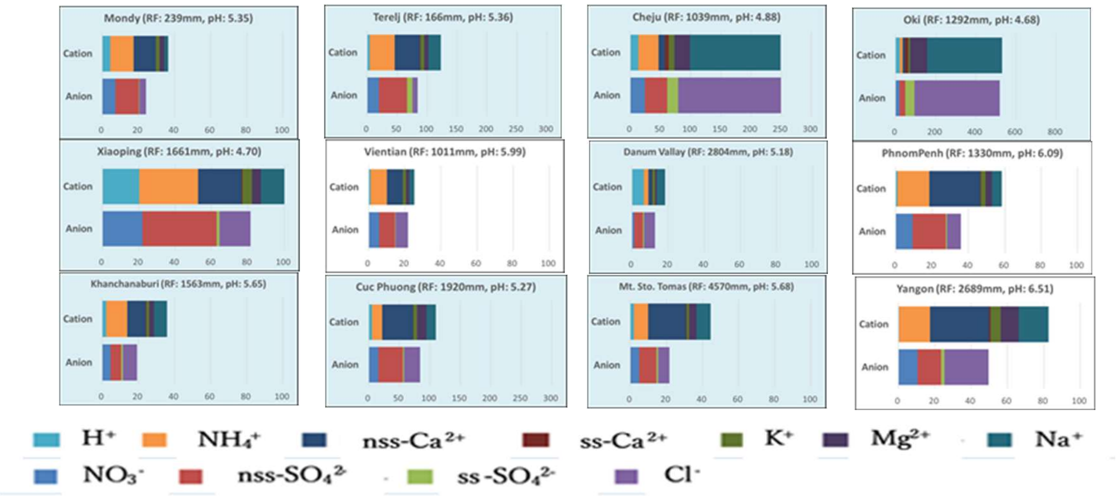


Figure 3.1. Ion balance of total precipitation collected in selected EANET sites between 2000 and 2019, 1) Figures of remote and rural sites are hatched, 2) RF: Averaged annual precipitation, 3) Concentration unit: μeq L^{-1}.

As described above, at the EANET sites where the concentration of acidic component tends to be higher than that of the basic component in an annual basis, high acidity precipitation may be observed on a monthly or daily basis. Table 3.1 summarizes the lowest pH value for each year from the observation data for 20 years. The lowest values were observed at sites where the acid concentration sum was larger than the base concentration sum. Additionally, the lowest pH values were observed not only in urban sites but also in rural and remote sites from the latter half of the 2000s to the former half of the 2010s. This factor is attributed to the increase in the sum of acid concentrations during the period. In addition, it is believed that such data were acquired because EANET's wet deposition monitoring is performed with a high time resolution (daily) using a cooling storage device.

Table 3.1 Low precipitation pHs in the twenty-year observation by EANET (2000-2019)

Year	Number of sites	Daily or event			Annual mean		
		pH	Site name	Precipitation (mm) and Date	pH	Site name	Precipitation (mm)
2000	40	3.54	Guanyinqiao	1.6 (10/21)	4.22	Nanshan	1259
2001	41	3.25	Jinyunshan	2.1 (10/18)	4.18	Jinyunshan	710
2002	41	3.16	Bandung	14.5 (12/28)	4.23	Petaling Jaya	2660
2003	44	3.41	Jinyunshan	4.5 (12/2)	4.28	Petaling Jaya	3041
2004	46	3.52	Xioping	14.8 (9/2)	4.33	Petaling Jaya	2996
2005	47	3.40	Kanghwa	1.0 (11/21)	4.25	Kanghwa	811
2006	49	3.14	Listvyanka	3.6 (8/17)	4.34	Cheju	1127
2007	49	3.35	Hedo	0.5 (11/3)	4.49	Banryu	1273
2008	52	2.84	Haifu	0.8 (9/13)	4.20	Haifu	1010
2009	54	3.06	Haifu	1.3 (11/27)	4.22	Haifu	1066
2010	53	2.97	Jinyunshan	1.1 (12/29)	3.94	Jinyunshan	979
2011	53	3.08	Jinyunshan	2.3 (4/3)	4.04	Jinyunshan	848
2012	53	2.79	Haifu	0.4 (5/27)	4.09	Jinyunshan	1005
2013	52	3.17	Jinyunshan	1.1 (1/16)	4.20	Jinyunshan	1127
2014	53	3.11	Jinyunshan	0.9 (1/26)	4.39	Jinyunshan	1490
2015	57	3.37	Banryu	2.0 (5/25-6/1)	4.39	Petaling Jaya	3751
2016	57	3.55	Yusuhara	1.5 (8/16)	4.26	Petaling Jaya	3432
2017	57	3.25	Kanghwa	3.5 (2/19)	4.44	Kanghwa	588
2018	57	3.57	Hedo	0.5 (7/27)	4.50	Petaling Jaya	3848
2019	61	3.54	Jinyunshan	0.5 (1/26)	4.68	Kanghwa	502

Figure 3.2 shows the 20-year secular changes in acid concentration sums (nss-SO_4^{2-} + NO_3^-), base concentration sums (NH_4^+ + nss-Ca^{2+}), and H^+ concentrations for representative EANET sites. The higher acid concentration sums were observed in the 2000s and 2010s at the rural and urban EANET sites in China, the Republic of Korea, and Japan in Northeast Asia, in the comparison with base concentration sums. As a result, higher H^+ concentrations were observed at EANET sites mainly in the 2000s in Korea, in the 2010s in China, and the 2000s and 2010s in Japan. After that period, it was observed that H^+ concentrations in these countries have been declining significantly. In Mongolia, the acid concentration sum in Ulaanbaatar has tended to be high in recent years. In addition, at rural and urban sites in Southeast Asian countries, like Vietnam and Malaysia, the acid concentration sums and the H^+ concentrations have increased in recent years. Thus, whether following the tendency of the base concentration sum or evaluating the situation completely independently, the acid concentration sums are maintained at a high level at some sites while there are sites where the acid concentration sum tends to increase. The acid concentration sums at the remote sites tend to decrease in Northeast Asia, as at the rural and urban sites. Besides, in Southeast Asia, there is no upward trend in acid concentration in remote sites. As mentioned herein, precipitation acidity is the result emerging from the balance between acids and bases dissolved in precipitation. Since the situation may change periodically due to air pollution in the surrounding region in the northeastern and southeastern Asian countries as compared with North America and European countries, it is important to continue wet deposition monitoring focusing on not only acids but also bases in precipitation in EANET countries.

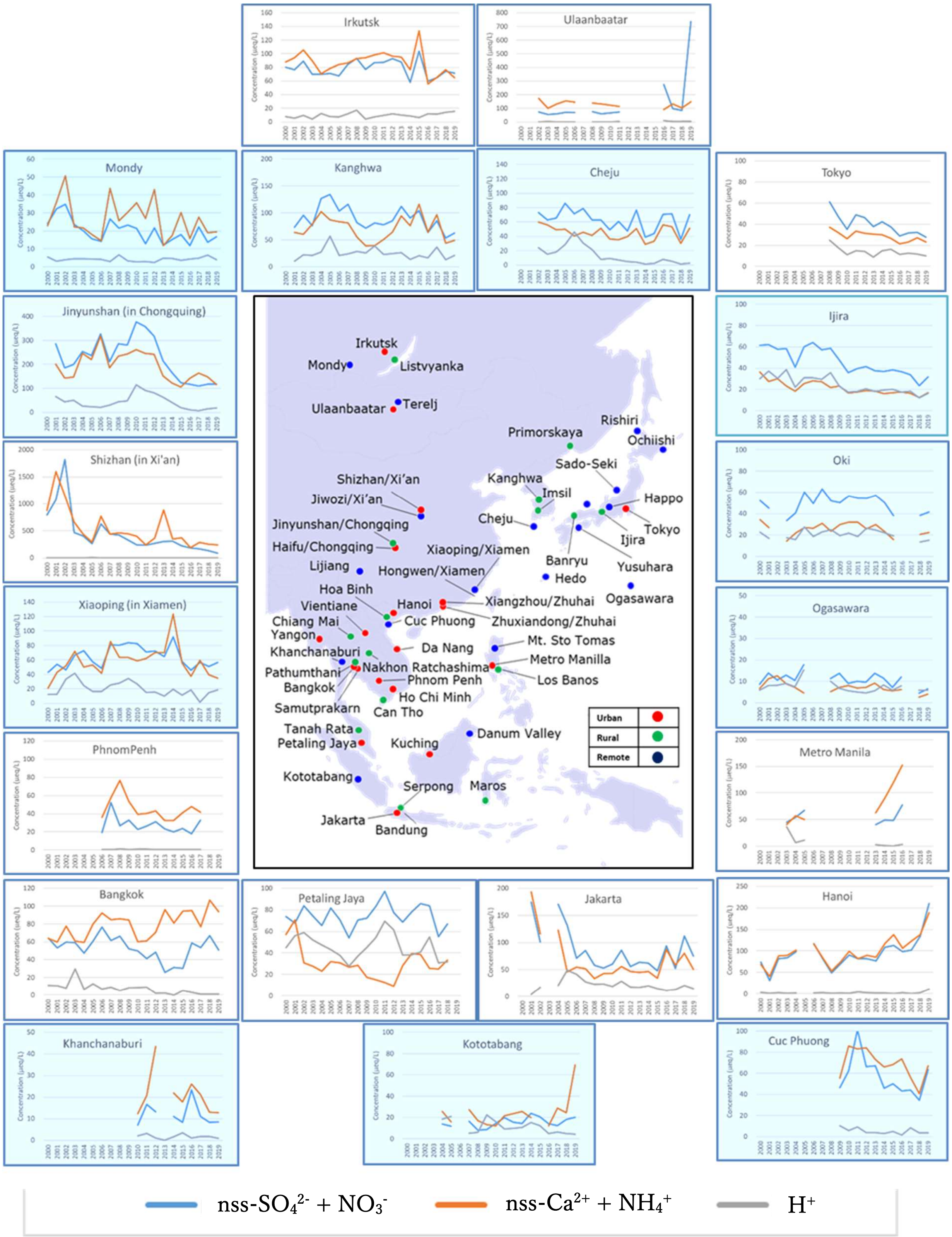


Figure 3.2. Long term trends of precipitation acidity (H^+) and concentrations of acids (nss-SO_4^{2-}+NO_3^-) and bases (nss-Ca^{2+}+NH_4^+) that acidified or neutralized precipitation in selected EANET sites (2000-2019). * Figures of remote and rural sites are hatched.

Spatial and temporal variation of wet deposition in East Asia

The trend of the annual average of precipitation chemistry and annual wet deposition amount was compared among EANET, National Trends Network (NTN) of National Atmospheric Deposition Program (NADP) in the US, and EMEP in Europe from 2000 to 2019 (the EMEP data is available until 2017). The NTN network currently has 263 sites. As for EMEP, precipitation data from 89 sites are presented in the report from 2018.

In the comparison among three regions, it should be kept in mind that in the following discussion, the annual average concentration and annual deposition amount in EANET were obtained using the data from all the urban, rural, and remote sites, but the urban sites were excluded in EMEP and NADP network. Therefore, it is important to pay attention to the comparison of increasing and decreasing trends in each region.

The pH value of precipitation exhibits a significantly increasing trend from all NADP, EMEP, and EANET from 2000 to 2019 (Figure 3.3). In 2000, the pH values of precipitation from EMEP, NADP, and EANET were 5.3, 4.9, and 5.0, respectively. In 2017, the pH values of precipitation from EMEP, NADP, and EANET were 5.5, 5.4, and 5.4, respectively. It suggests that the acid rain pollution in the whole of Europe, North America, and East Asia has gradually reduced in the past 20 years. It can primarily be concluded that the increasing trend of pH value in precipitation from NADP, EMEP, and EANET is probably due to a more significant reduction of acid components (nss-SO_4^{2-} and NO_3^-) than alkali components (NH_4^+, and nss-Ca^{2+}) in the past 20 years.

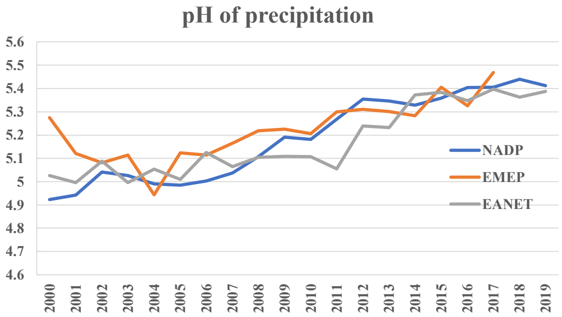


Figure 3.3. The trend of annual average pH value in precipitation from EANET, EMEP, and NADP since 2000 to 2019.

The wet deposition of nss-SO_4^{2-} exhibits a significantly declining trend from NADP, EMEP, and EANET in the past 20 years (Figure 3.4). The wet deposition amount of NO_3^- does not exhibit a clear change from EANET, but a significantly declining trend from both EMEP and NADP. The wet deposition amounts of NH_4^+ from NADP exhibit a slightly increasing trend and slightly decreasing trend from EANET, but no significant trend was observed from EMEP. The wet deposition amounts of nss-Ca^{2+} from EMEP exhibit a slightly declining trend, but no significant trend was observed from NADP and EANET. The precipitation amount from NADP exhibits a significantly increasing trend in the past 20 years, but in the other networks, it does not exhibit any obvious trend.

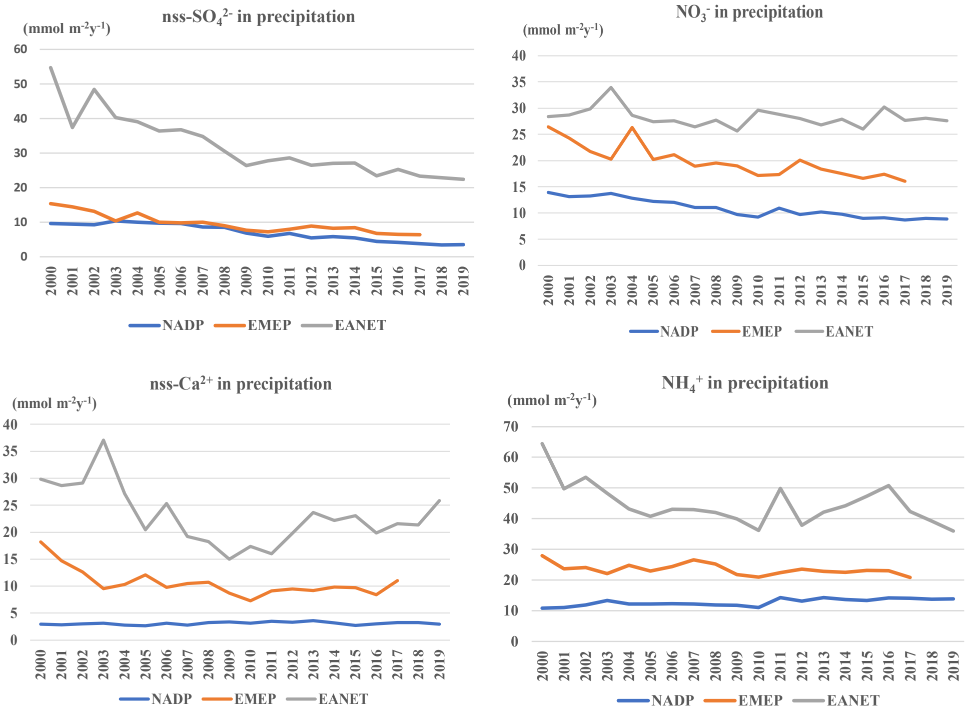


Figure 3.4. The trend of annual wet deposition amount of nss-SO_4^{2-}, NO_3^-, NH_4^+, and nss-Ca^{2+} and precipitation amount from EANET, EMEP, and NADP from 2000 to 2019.

Trends of wet deposition of EANET in Northeast Asia and Southeast Asia

The annual trend of the ion concentrations of precipitation and the wet deposition fluxes were estimated using the Mann-Kendall method. The trend analysis was focused on the wet deposition of H^+, nss-SO_4^{2-}, NO_3^-, NH_4^+, and nss-Ca^{2+}, and the ion concentration of nss-SO_4^{2-}, NO_3^-, NH_4^+, nss-Ca^{2+}, and pH, and the precipitation amount. The wet deposition fluxes and concentrations were obtained from 2000 in EANET participating countries. The annual trends which satisfy the criteria of EANET were estimated by the 11-year moving analysis within the 20 years (%PCL > 80% and %TP > 80%).

The percentage of EANET monitoring sites with a significant positive trend, insignificant positive trend, insignificant negative trend, and significant negative trend in EANET participating countries are shown in Figure 3.5, including Northeast Asian countries (Russia, Mongolia, China, the Republic of Korea, and Japan) and Southeast Asian countries (Cambodia, Indonesia, Lao P.D.R, Malaysia, Myanmar, Philippines, Thailand, and Vietnam). Based on the data analysis, some maps were prepared to show the geographical distribution of acids and bases wet deposition trends (in the periods 2000-2010, 2005-2015, and 2009-2019). The pH shows an increasing trend in Northeast Asian and Southeast Asian countries (Figure 3.6). The concentration of nss-SO_4^{2-} shows ta decreasing trend in Northeast Asian countries (Figure 3.7). There were no significant trends in the other components such as NO_3^-. The decline of the nss-SO_4^{2-} in North Asian countries contributes to the increase in pH. A the concentration of nss-SO_4^{2-} in Southeast Asian countries exhibited an insignificant trend. The concentration of nss-Ca^{2+} and NH_4^+ exhibited the increasing trend in Southeast Asia. Therefore, the increasing trend of pH in Southeast Asian countries was possibly caused by increasing cation contribution (nss-Ca^{2+} and NH_4^+).

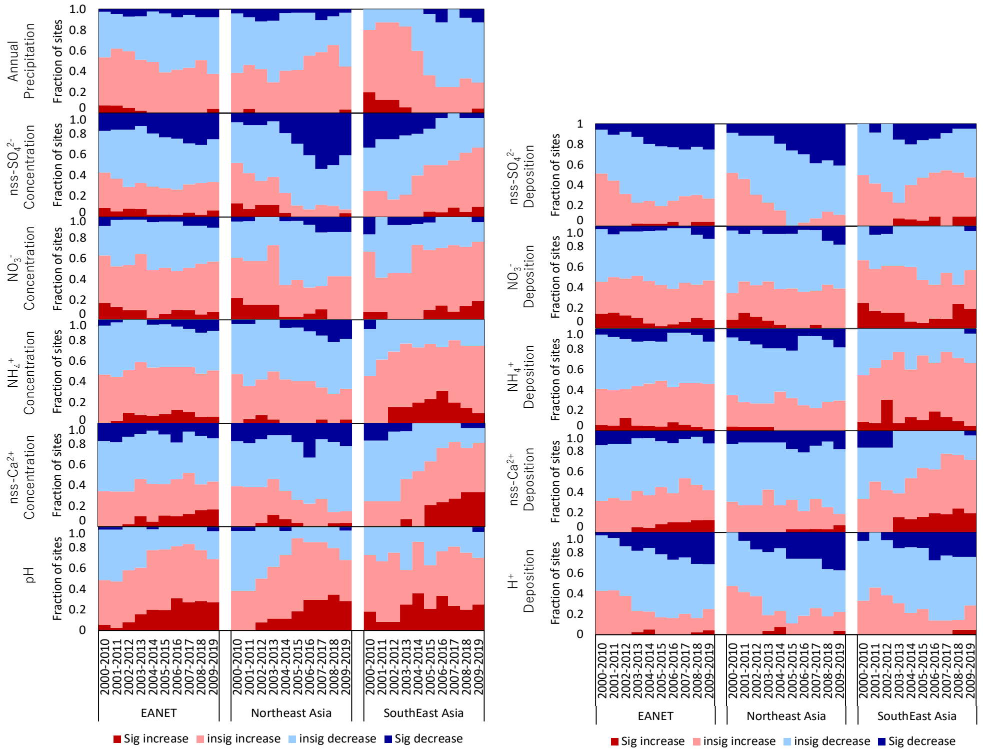


Figure 3.5. Percentage of significant positive trend (red), insignificant positive trend (light red), insignificant negative trend (light blue), and significant negative trend (blue) in EANET, northeast Asian sites, and southeast Asian sites, which are obtained by the Mann-Kendall test using the annual averaged concentrations of nss-SO$_4^{2-}$, NO$_3^-$, NH$_4^+$, nss-Ca^{2+}, pH, and precipitation amount, and the annual averaged wet deposition flux of nss-SO$_4^{2-}$, NO$_3^-$, NH$_4^+$, nss-Ca^{2+}, and H$^+$.

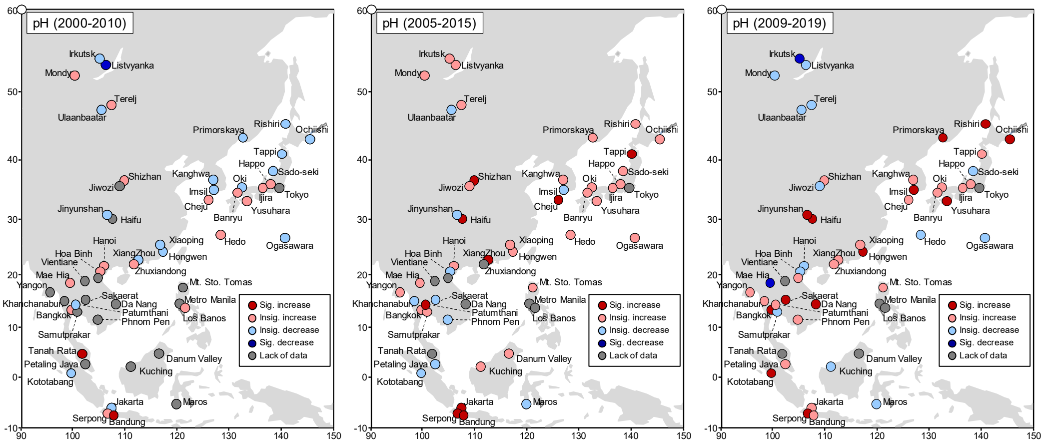


Figure 3.6. Spatial distribution of pH trend at EANET sites in the period of 2000-2010, 2005-2015, and 2009-2019.

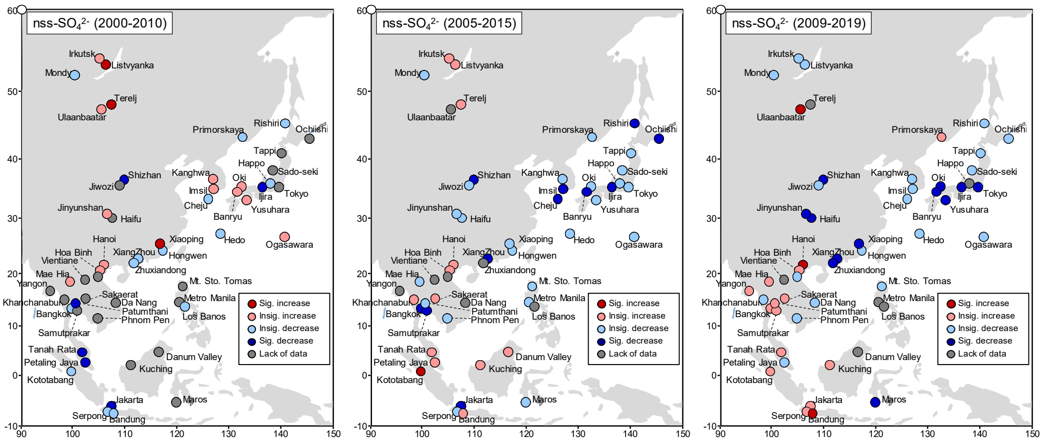


Figure 3.7. Spatial distribution of the trend for nss-SO$_4^{2-}$ concentration at EANET sites in the period of 2000-2010, 2005-2015, and 2009-2019.

Dry deposition

The dry deposition was estimated by the inferential method based on the Technical Manual for Dry Deposition Flux Estimation in East Asia (EANET, 2010). A detailed description of the methodology is shown in the Technical Manual for Air Concentration Monitoring in East Asia (https://www.eanet.asia/wp-content/uploads/2019/04/techacm.pdf). For the evaluation of dry deposition, the sampling period of air concentrations ranges from being longer than one day to one or two weeks. This monitoring might employ either real-time monitors or integrate manual samplers (filter packs, denuders, or passive samplers, as may be determined appropriate). So far, there are 54 monitoring sites of dry deposition in EANET, including 41 sites using the filter pack method (chemical species in particulate matter and gaseous compounds), 34 sites using the automatic method (SO_2, NO, NOx, etc.), and 6 sites using the passive sampler method (SO_2, NO_2, and NH_3) in 2019. The number of wet/dry monitoring sites in ENAET is increasing over the past 20 years.

Trends of annual amounts of total sulfur (S) deposition by dry and wet depositions at remote, rural, and urban sites are shown in Figure 3.8. Low total S depositions were found at the remote sites located in the northern inland (Mondy and Terelj) and Pacific Ocean (Ogasawara), and high total S depositions were found at the Japanese remote sites near the Asian continent (Sado-seki and Oki). Dry deposition largely contributed to the high total S at Sado-seki and Oki. In Japanese remote sites, except Rishiri and Ogasawara, more than 10 kg S/ha/year of total S deposition were found over many years. In these sites, the total S depositions had decreased from approximately 2013. Especially in Yusuhara and Happo, the decreases started from approximately the late 2000s. A clear decrease in wet S deposition was also found in Ijira (a rural site) from the early 2000s. More than 10 kg S/ha/year of total S deposition was found at all rural and urban sites. Especially high total S depositions of more than 30 kg S/ha/year were found at urban sites in Southeast Asia (Hanoi, Bangkok, and Petaling Jaya). At Hanoi and Bangkok, dry S depositions have decreased from the 2000s.

Trends of annual amounts of total nitrogen (N) deposition by dry and wet depositions at remote, rural, and urban sites are shown in Figure 3.9. Ban et al. (2016) estimated total N deposition at Japanese remote sites from 2003 to 2012 following the inferential method of EANET (2010), using hourly input data observed at each site. The amounts and trends of annual total N deposition estimated in this report using monthly data were almost the same as those of Ban et al. (2016). In remote sites, spatial and temporal trends of the total N depositions were similar to those of the total S depositions [low in northern inland (Mondy and Terelj) and Pacific Ocean (Ogasawara); high in Japanese sites near the Asian continent (Sado-seki and Oki)]. In Japanese remote sites, except Rishiri and Ogasawara, more than 10 kg N/ha/year of total N deposition was found over many years.

At the sites with high N deposition (Sado-seki and Oki), the total N depositions had decreased around 2013. There were no clear trends indicating decreases or increases in the total N depositions at other

Japanese remote sites. Regarding rural sites, a clear decrease in wet N deposition was found at Ijira, similar to that of wet S deposition. At Serpong, there were quite a high total N depositions of more than 30 kg N/ha/year, which were frequently found in the 2010s. In urban sites, in Hanoi and Bangkok, dry N depositions clearly did not decrease from the 2000s, unlike the decreases in dry S depositions.

Trends of annual amounts of total nitrogen (N) deposition by oxidized N and reduced N at remote, rural, and urban sites are shown in Figure 3.10. Here, oxidized N deposition is defined as the sum of wet depositions of NO_3^- and dry depositions of HNO_3 and particulate NO_3^-; reduced N deposition is defined as the sum of wet depositions of NH_4^+ and dry depositions of NH_3 and particulate NH_4^+. In remote sites, reduced N deposition contributed more than half of the total N depositions at lower N deposition sites (Mondy, Terelj, Rishiri, and Ogasawara), and oxidized N deposition contributed more than half of the total N depositions at higher N deposition sites (Sado-seki and Oki). At Sado-seki and Oki, the decreases in the total N depositions were largely contributed by the decreases in the oxidized N depositions. Regardless of the site categories, reduced N depositions contributed more than half the total N depositions in many years at all the sites in Russia, Mongolia, and Southeast Asia, except Petaling Jaya.

The trend analyses (2000-2010, 2005-2015, and 2009-2019, respectively, using the Mann-Kendall method) were conducted for the total depositions of sulfur, nitrogen, oxidized nitrogen, and reduced nitrogen, respectively (Figure 3.11). Generally, significant trends were observed from some sites in EANET during the period 2000-2019.

For the total deposition of sulfur, a significant decreasing trend was observed from Hanoi (2000-2010), Ijira (2000-2010, 2005-2015, and 2009-2019), Primorskaya (2005-2015), Yusuhara (2005-2015) and Rishiri (2009-2019); on the contrary, the significant increasing trend was observed from Irkutsk from 2000 to 2010, Hanoi (2009-2019) and Serpong (2009-2019). For the total deposition of nitrogen, a significant decreasing trend was observed from Ijira (2005-2015 and 2009-2019), and a significant increasing trend was observed from Oki from 2005 to 2015. For the total deposition of oxidized nitrogen, a significant decreasing trend was observed from Ijira and Ogasawara from 2005 to 2015 and 2009 to 2019, and Yusuhara from 2005 to 2015; a significant increasing trend was observed from Hanoi (2005-2015 and 2009-2019) and Hoa Binh (2009-2019). For the total depositions of reduced nitrogen, a significant decreasing trend was observed from Terelj (2000-2010) and Ijira (2005-2015 and 2009-2019); a significant increasing trend was found from Oki (2000-2010 and 2005-2015).

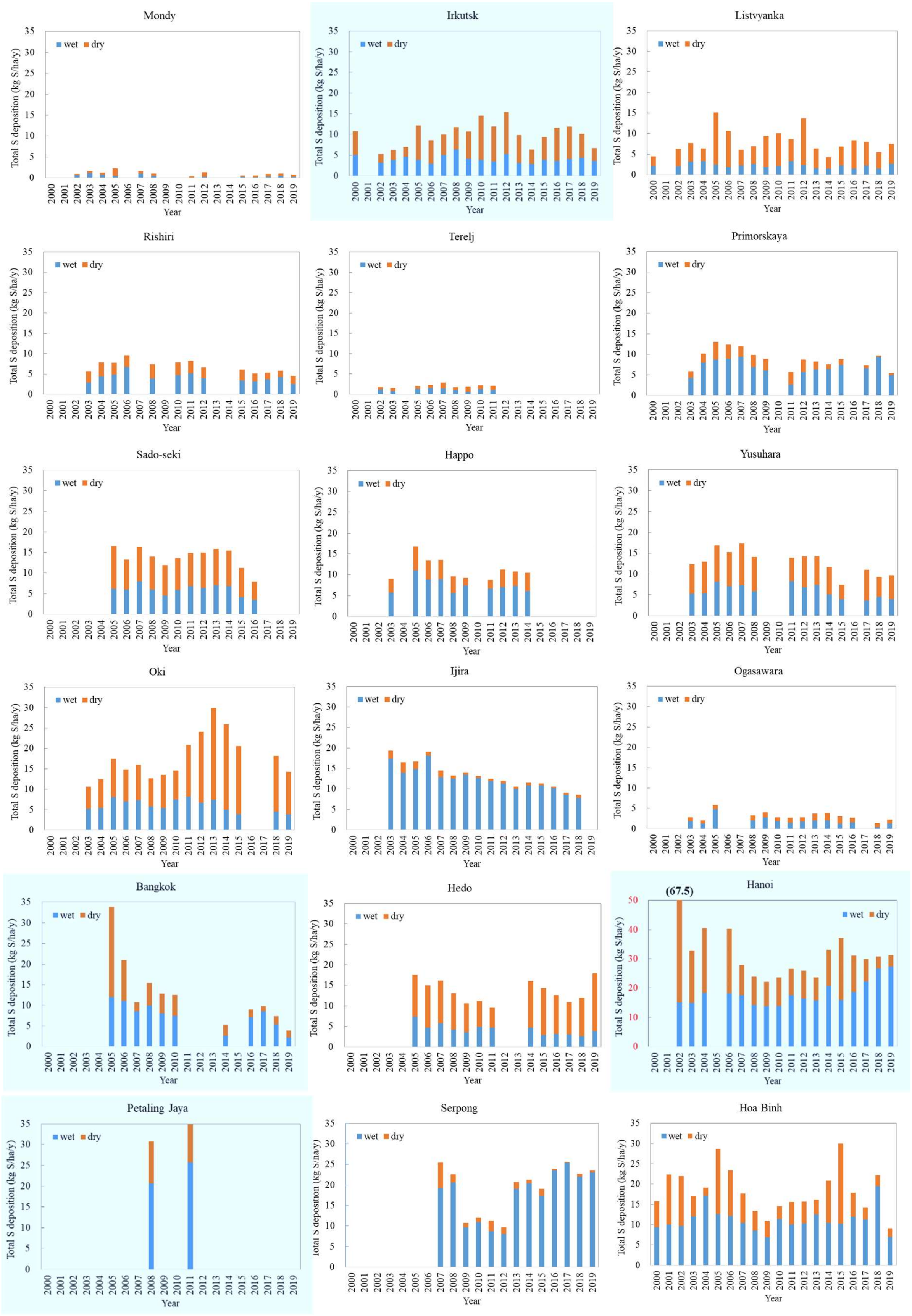


Figure 3.8. Trends of annual amounts of total sulfur depositions in EANET sites. The blue and orange columns indicate wet and dry depositions, respectively. * The figures of urban sites are hatched.

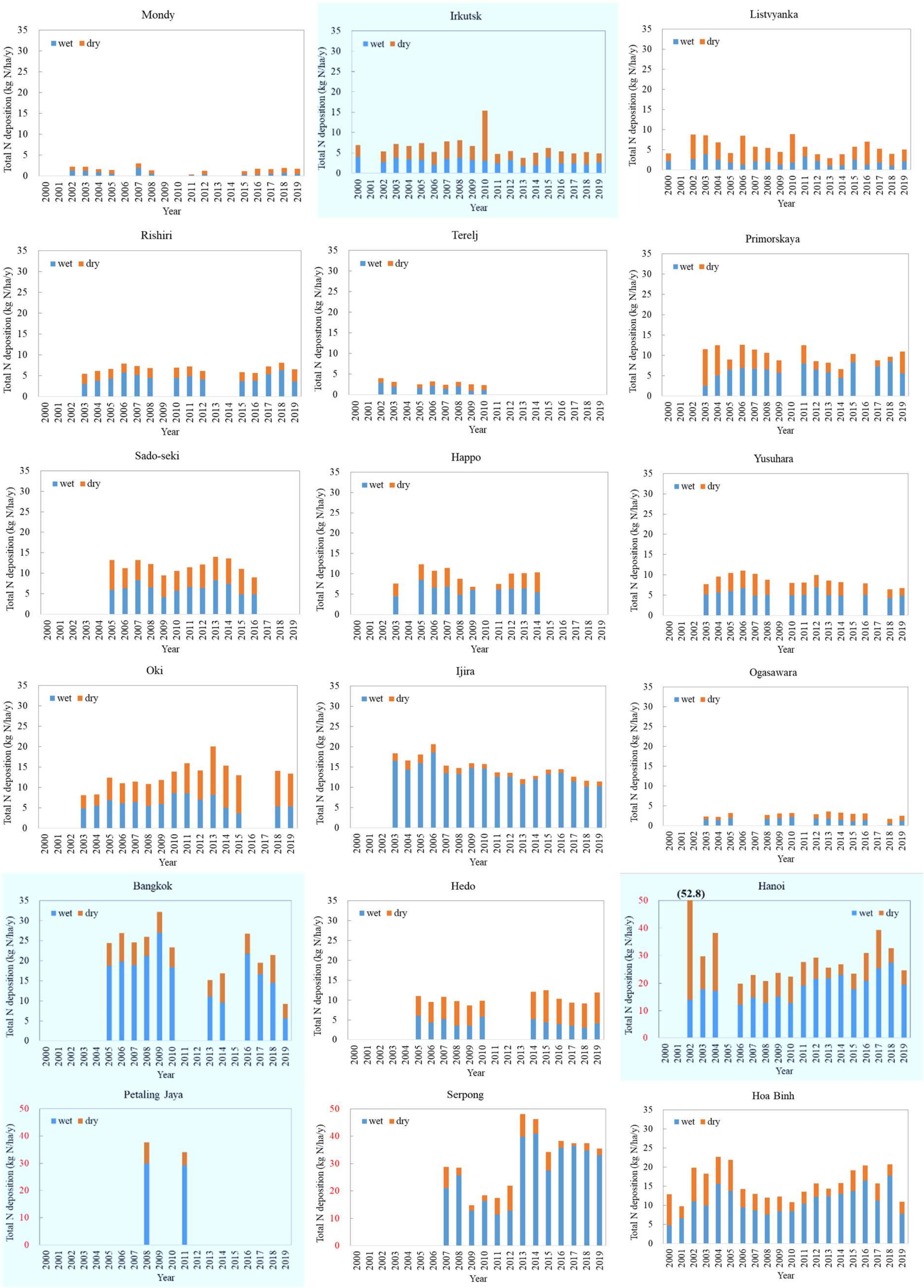


Figure 3.9. Trends of annual amounts of total nitrogen depositions in EANET sites. The blue and orange columns indicate wet and dry deposition, respectively. * The figures of urban sites are hatched.

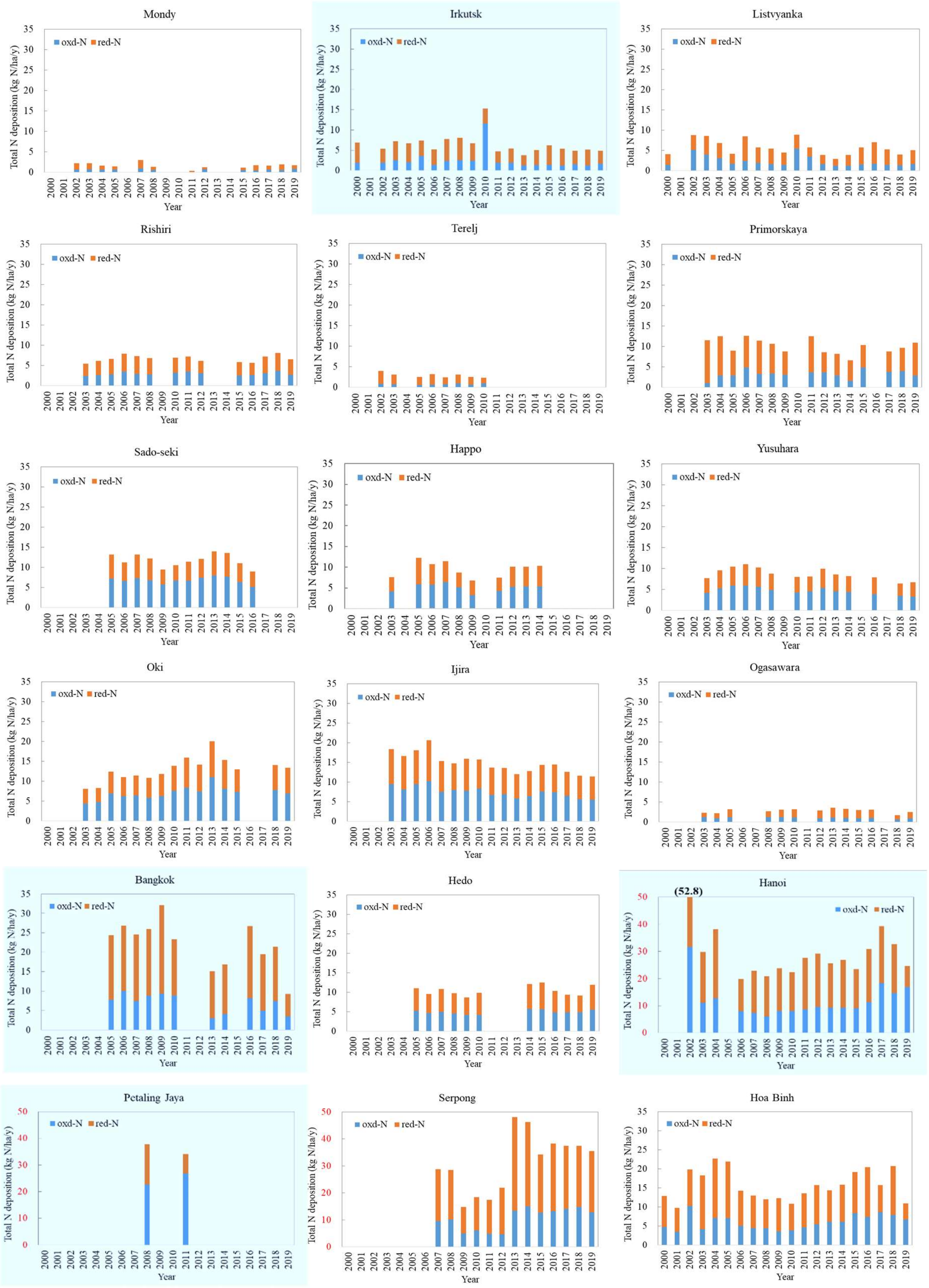


Figure 3.10. Trends of annual amounts of total nitrogen depositions in EANET sites. The blue and orange columns indicate oxidized and reduced nitrogen depositions, respectively. * The figures of remote and rural sites are hatched.

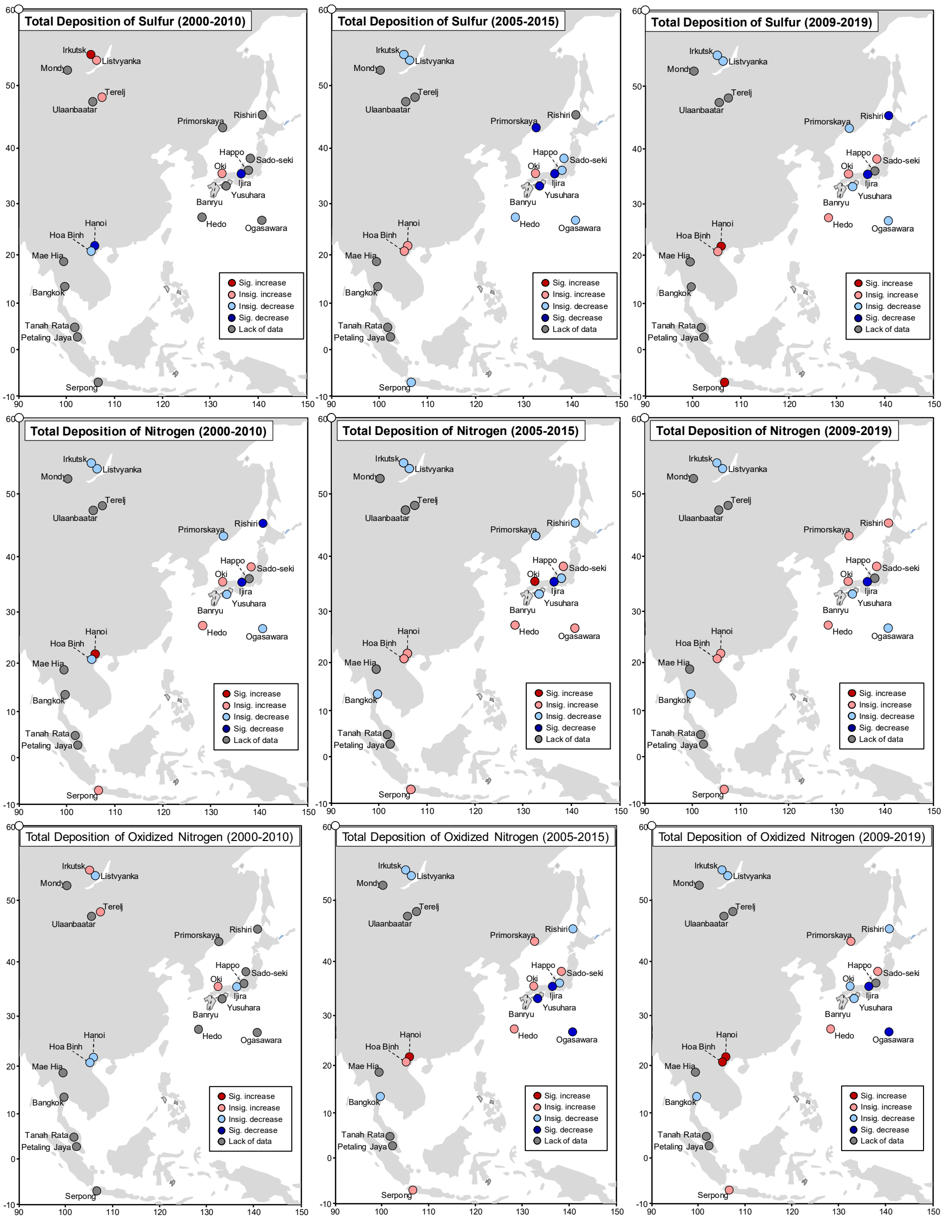


Figure 3.11 (1). Spatial distribution of the trend for the total (wet and dry) depositions of sulfur, nitrogen, and oxidized nitrogen at EANET sites in the periods 2000-2010, 2005-2015, and 2009-2019.

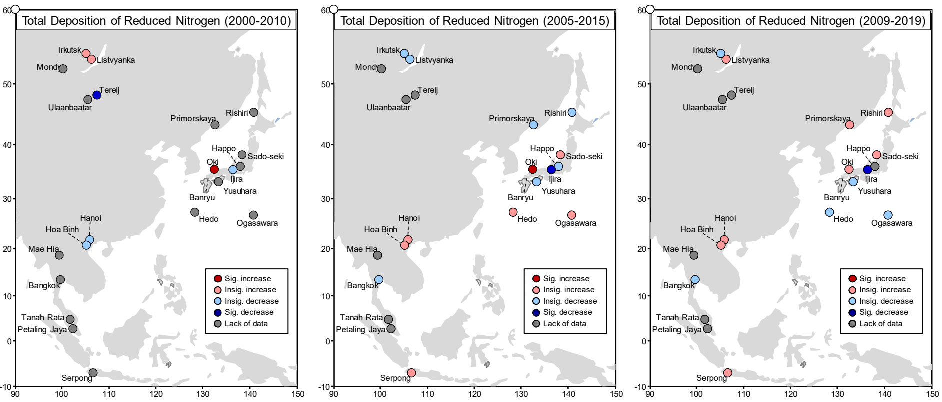


Figure 3.11 (2). Spatial distribution of the trend for the total (wet and dry) depositions of reduced nitrogen at EANET sites in the periods 2000-2010, 2005-2015, and 2009-2019.

The total (dry and wet) depositions of S and N were evaluated at the sites carrying out the wet-only sampling and the 4-stage filter pack monitoring in Russia, Mongolia, Japan, Vietnam, Thailand, Malaysia, and Indonesia. Dry depositions were estimated by simplifying the inferential method using available monthly meteorological data. Low total S depositions were found at the remote sites located in the northern inland and the Pacific Ocean, and high total S depositions were found in the Japanese remote sites near the Asian continent. In the high S deposition sites, a decreasing trend of S depositions was found in recent years. High total S depositions of more than 30 kg S/ha/year were found at urban sites in Southeast Asia. It was observed that the dry S depositions have decreased from the 2000s at Hanoi and Bangkok. In remote sites, spatial and temporal trends of the total N depositions were similar to those of the total S depositions and reduced N depositions contributed more than half of the total N depositions at lower N deposition sites and oxidized N depositions contributed more than half the total N deposition at higher N deposition sites. Regardless of the site categories, reduced N depositions contributed more than half the total N deposition in many years at many sites, except at Sado-seki, Oki, and Petaling Jaya. The trend analysis for total S and N, as well as oxidized and reduced N depositions, has shown a significant increase in some sites in Southeast Asia and a significant decrease in some sites in Northeast Asia, especially in the last decade.

4. Gas and Aerosol Pollution in East Asia

Monitoring status by each country

Once the pollutants are emitted and mixed in the air, they are deposited locally and across the border. The feature of long-range transportation shows that it is necessary to establish a continuous monitoring network and deal with this environmental phenomenon not only from a domestic perspective but also as a regional or hemispherical problem. Therefore, EANET has conducted continuous monitoring in various countries for over 20 years and the number of gas and aerosol monitoring sites has increased each year (Fig 4.1). This will allow for the establishment of a high-resolution monitoring network in East Asia and provide the scientific evidence necessary for policymakers to confront this environmental issue as well. There were 54 air concentration monitoring sites operated in the EANET network as of 2019. The measuring components, which are SO_2, NO, NO_2, NOx, $PM_{10/2.5}$, O_3, HNO_3, HCl, NH_3, and Particulate Matter Components (PMCs), vary depending on the methodology at each site.

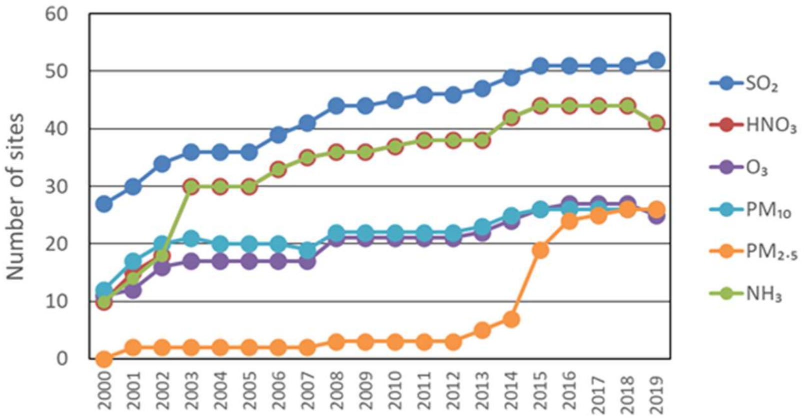


Figure 4.1. Annual variations of the number of gas and aerosol monitoring sites in EANET.

Country based characteristics of SO_2, NO_2, NOx, O_3, PM_{10}, and $PM_{2.5}$ concentrations

Figure 4.2 shows the average concentrations of SO_2, NO_2, NOx, O_3, PM_{10}, and $PM_{2.5}$ in each EANET member country from 2015 to 2019. The spatial distribution of SO_2 at EANET sites in Russia and Mongolia varied from 0.18 to 9.10 ppb. This significant difference in the spatial distribution of concentration shows the influence of population distribution, as well as the effect of heating and fuel composition such as coal burning. After continuous improvement, SO_2 concentrations at EANET sites in China have been significantly reduced and the 5-year average range for the five EANET stations in China is 0.1 ppb-3.54 ppb. The SO_2 concentrations at all EANET sites in Japan were very low for a long time, with a 5-year average range of 0.03 ppb-1.3 ppb, along with the maximum value occurring in Tokyo. The SO_2 concentration range at the three land EANET sites in the Republic of Korea is similar to that of China, with a 5-year average range of 0.66 ppb-2.44 ppb. Except for Indonesia, SO_2 concentrations in Southeast Asian countries are generally low for the 5-year-average concentration.

Among the ozone observations in eight EANET member countries including Cambodia, Indonesia, Japan, Mongolia, Philippines, the Republic of Korea, Russia, and Thailand, the Republic of Korea had the highest nationwide O_3 5-year average concentration of 40.5 ppb from 3 stations. Japan reported the next highest nationwide average O_3 concentration. Alternately, O_3 concentration in Southeast Asian countries is lower.

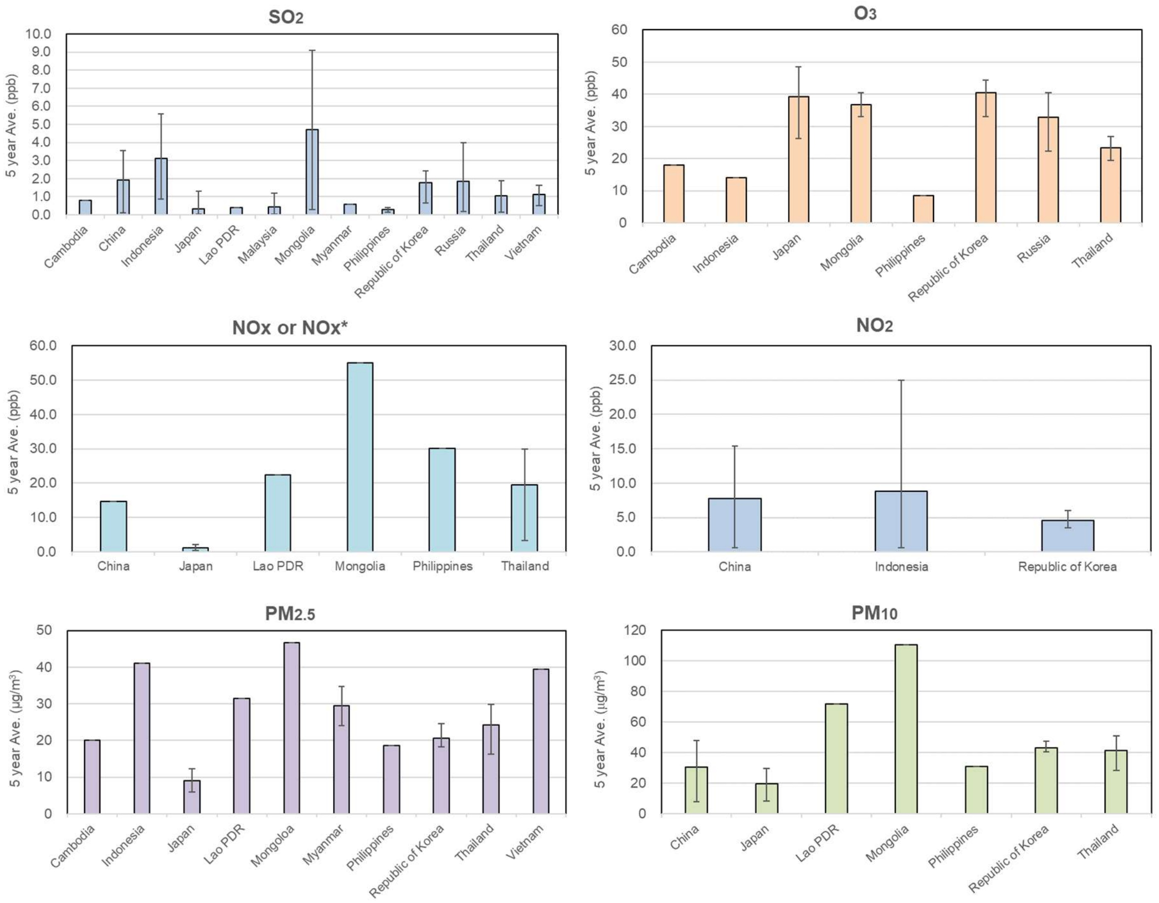


Figure 4.2. Average concentrations of SO_2, NO_2, NOx, O_3, PM_{10}, and $PM_{2.5}$ at the EANET monitoring sites in each country (annual average from 2015 to 2019).

Among the NOx observations in six EANET member countries including China, Japan, Lao PDR, Malaysia, Mongolia, Philippines, and Thailand, Mongolia had the highest NOx 5-year average concentration of 55.0 ppb at Ulaanbaatar. Metro Manila in the Philippines reported the next highest 5-year averaged NOx concentration of 30.1 ppb. Among the NO_2 observations in eight EANET member countries including China, Indonesia, Japan, Lao PDR, Mongolia, Philippines, the Republic of Korea, and Thailand, there were large spatial differences in the distribution of NO_2 concentrations. Indonesia had the highest NO_2 concentration level with a 5-year mean concentration of 8.82 ppb.

Among the $PM_{2.5}$ observations in ten EANET member countries including Cambodia, Indonesia, Japan, Lao PDR, Mongolia, Myanmar, Philippines, the Republic of Korea, Thailand, and Vietnam, Mongolia had the highest $PM_{2.5}$ 5-year average concentration of 40.5 µg/m^3 at Ulaanbaatar. Jakarta of Indonesia reported the next highest nationwide average $PM_{2.5}$ concentration of 41.0 µg/m^3. The $PM_{2.5}$ concentration in Hoa Binh of Vietnam was close to that of Jakarta of Indonesia, with a 5-year average concentration of 39.4 µg/m^3. The 5-year average $PM_{2.5}$ concentrations of the remaining countries were less than 30 µg/m^3.

Among the PM_{10} observations in seven EANET member countries including China, Japan, Lao PDR, Mongolia, Philippines, the Republic of Korea, and Thailand, there were large spatial differences in the distribution of PM_{10} concentrations. Ulaanbaatar of Mongolia had the highest PM_{10} concentration level with a 5-year mean concentration of 110 µg/m^3. Vientiane of Lao PDR had the second-highest PM_{10} concentration with a 5-year mean of 72 µg/m^3. The PM_{10} concentration levels in China, Korea, the Philippines, and Thailand were similar. The 5-year nationwide average PM_{10} concentrations of 12 stations in Japan had a range of 8 to 30 µg/m^3, with a mean concentration of 19.9 µg/m^3, which is generally low and has a relatively large variation among sites.

Country-based characteristics of gaseous HNO_3, NH_3, and HCl concentrations

Figure 4.3 shows the average concentrations of HNO_3, NH_3, and HCl in the monitoring sites of each EANET member country from 2015 to 2019. The concentration of HNO_3 at the site in China was relatively high during the period from 2015 to 2019, with Hongwen concentrations ranging from 1.1 ppb to 1.2 ppb, with a 5-year average value of 1.15 ppb. The concentration at the sites in Indonesia was the second-highest in the EANET region with a more consistent concentration distribution, with the 5-year averaged HNO_3 concentration of 3 stations ranging from 0.62 ppb to 1.14 ppb from 2015 to 2019. The spatial distribution of HNO_3 concentrations in Japan and Malaysia showed large fluctuations, with the 5-year mean HNO_3 concentrations at 12 stations in Japan ranging from 0.03 ppb to 0.62 ppb. The 5-year mean HNO_3 concentration at Petaling Jaya in Malaysia was 0.63 ppb, while those at Tanah Rata and Danum Valley was only 0.63 ppb. The 5-year mean concentration at Tanah Rata and Danum Valley was only 0.06 ppb, These variations would be caused by the spatial difference in population distribution and emissions, and atmospheric chemistry in Japan and Malaysia. The HNO_3 concentrations in the rest of the countries were generally low, with a 5-year mean concentration of less than 0.5 ppb.

Ammonia gas affects particulate formation in the atmosphere due to the ammonium component and acidity of deposition. Most EANET countries are agricultural countries where the use of fertilizers and livestock breeding are common practices. In Mongolia, as well as in China, there are also large pastoral areas. At the same time, population hygiene conditions in the city and countryside, as well as ammonia escape during the denitrification of industrial and mobile sources, can also affect ammonia concentrations. Generally, the atmosphere of most EANET countries can be classified as an ammonia-rich condition. There are various NH_3 concentration distributions in East Asia. First, the spatial distribution of NH_3 in the Philippines, Malaysia, and Mongolia has shown a large variation. The 5-year mean concentration at the Metro Manila station in the Philippines was 13.2 ppb, while the mean concentration at the Mt. Sto. Tomas station was only 1.91 ppb. The Petaling Jaya station had the highest 5-year mean concentration of 9.61 ppb among the three stations in Malaysia, while the Tanah Rata and Danum Valley stations had lower 5-year mean concentrations of 1.23 ppb and 2.38 ppb, respectively. The Ulaanbaatar station in Mongolia had a 5-year mean concentration of 8.0 ppb and the Terelj station had a 5-year mean concentration of 1.70 ppb. China had a relatively high NH_3 concentration among the countries with a 5-year average concentration of 9.99 ppb at the Hongwen site, followed by Indonesia with a 5-year average NH_3 concentration range of 8.76 ppb-9.18 ppb at three stations and an overall average of 9.23 ppb. The 5-year mean concentration at the Cambodia station was 7.20 ppb, the average NH_3 concentration at four stations in Thailand ranged from 4.48 to 8.85 ppb, and the mean 5-year concentration at three stations in the Republic of Korea ranged from 1.96 to 7.70 ppb. The other countries reported lower average NH_3 concentrations for the 5 years. The 5-year mean NH_3 concentration at the Yangon station in Myanmar was 4.93 ppb, the 5 stations in Vietnam had mean NH_3 concentrations ranging from 1.42 to 4.28 ppb with an overall mean of 3.16 ppb, the Vientiane station in Lao PDR had a mean NH_3 concentration of 2.64 ppb, with four stations in Russia having mean concentrations ranging from 1.18 to 2.84 ppb. Ammonia concentrations in Japan were generally low, with a 5-year average of only 0.97 ppb at 12 stations.

Among the EANET countries, the highest HCl concentration was found in Russia with a 5-year mean concentration of 3.43 ppb and large differences in the geospatial distribution. The mean concentrations at the Listvyanka and Irkutsk stations were 6.94 ppb and 4.40 ppb, respectively, while the 5-year mean concentrations at the Mondy and Primorskaya stations were lower, being 1.96 ppb and 0.42 ppb. The second-highest concentration of HCl in the EANET region was found in Mongolia, where the 5-year mean concentrations ranged from 1.30 ppb to 3.55 ppb.

The remaining EANET countries had relatively similar lower HCl concentration levels. The 5-year average concentration levels were 0.66 ppb in Malaysia, 0.53 ppb in Japan, 0.52 ppb in Thailand, 0.50 ppb in the Philippines, 0.48 ppb in Myanmar, and 0.25 ppb in Lao PDR. The differences among the three stations in the Republic of Korea were not significant, with the mean concentration being 0.33 ppb.

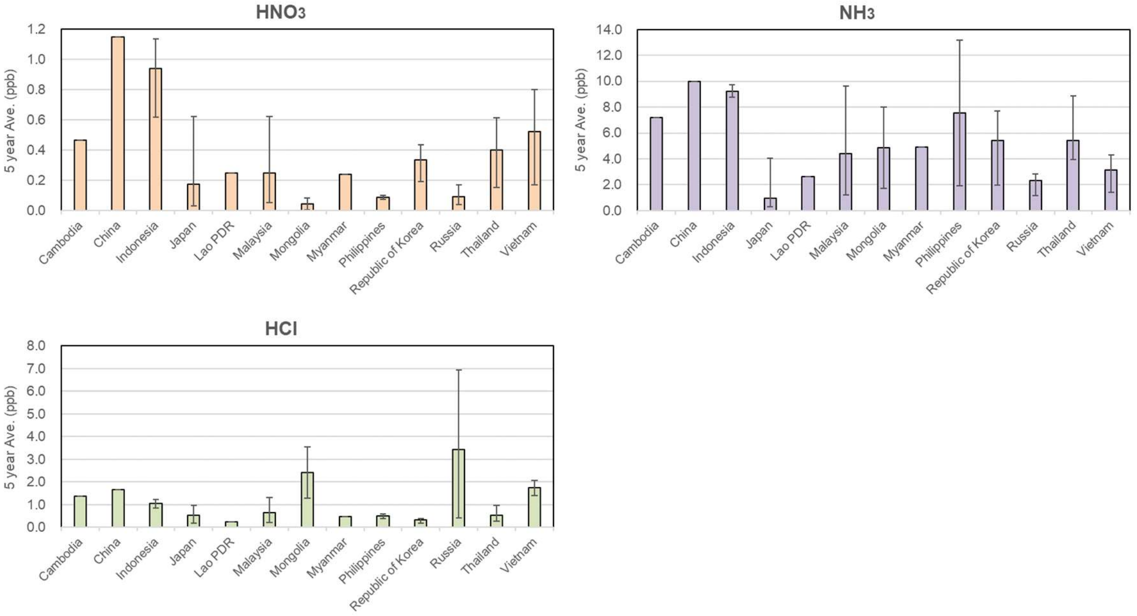


Figure 4.3. Average concentrations of HNO_3, NH_3, and HCl at the EANET monitoring sites in each country (annual average from 2015 to 2019).

Country-based characteristics of SO_4^{2-}, NO_3^-, NH_4^+, and Ca^{2+} component concentrations in particulate matter

Figure 4.4 shows the average concentrations of SO_4^{2-}, NO_3^-, NH_4^+, and Ca^{2+} components in particulate matter in each EANET member country from 2015 to 2019. The highest SO_4^{2-} concentration was found at Hongwen of China, the only station reporting data, with a 5-year average SO_4^{2-} concentration of 10.0 µg/m^3. The second highest 5-year averaged SO_4^{2-} concentration was found in Vietnam with a mean of 5.59 µg/m^3, showing some differences in spatial distribution. The 5-year mean SO_4^{2-} concentrations of the remaining EANET countries were relatively low. The 5-year mean SO_4^{2-} concentrations at Phnom Penh of Cambodia was 1.95 µg/m^3. The 5-year mean SO_4^{2-} concentrations at three EANET stations in Indonesia ranged from 4.66 to 5.34 µg/m^3. The 5-year mean SO_4^{2-} concentration at Vientiane of Lao PDR was 4.26 µg/m^3. The 5-year mean SO_4^{2-} concentrations at three EANET stations in Malaysia ranged from 1.18 to 3.66 µg/m^3. The 5-year mean SO_4^{2-} concentration at Yangon of Myanmar was 1.23 µg/m^3. The 5-year mean SO_4^{2-} concentrations at two EANET stations in the Philippines ranged from 0.29 to 2.35 µg/m^3. The 5-year mean SO_4^{2-} concentrations at three EANET stations in the Republic of Korea ranged from 4.04 to 4.63 µg/m^3. The 5-year mean SO_4^{2-} concentration at five EANET stations in Thailand ranged from 0.76 to 5.27 µg/m^3. The SO_4^{2-} concentration levels of Japan, Mongolia, and Russia were low and more consistent with a 5-year mean range of 1.67 to 4.45 µg/m^3 for 12 EANET stations in Japan, 0.51 to 1.68 µg/m^3 for two EANET stations in Mongolia, and 0.38 to 1.90 for three EANET stations in Russia, respectively.

The highest NO_3^- concentration was found at Hongwen of China, the only EANET station reporting data, with a 5-year average NO_3^- concentration of 7.94 µg/m^3. The second highest 5-year averaged NO_3^- concentration was found in Vietnam with a mean of 4.07 µg/m^3 and a range of 1.54 µg/m^3-6.73 µg/m^{3}a for five EANET stations, showing some differences in spatial distribution. The 5-year mean NO_3^- concentrations of the remaining countries were relatively low. The 5-year mean NO_3^- concentration at Phnom Penh of Cambodia was 1.66 µg/m^3. The 5-year mean NO_3^- concentrations at three EANET stations in Indonesia ranged from 1.15 to 2.06 µg/m^3. The 5-year mean NO_3^- concentrations at 12 EANET stations in Japan ranged from 0.34 to 3.23 µg/m^3. The 5-year mean NO_3^- concentration at Vientiane of Lao PDR was 0.44 µg/m^3. The 5-year mean NO_3^- concentrations at three EANET stations in Malaysia ranged from 0.09 to 1.56 µg/m^3. The 5-year mean NO_3^- concentration at Yangon of Myanmar was 0.80 µg/m^3. The 5-year mean NO_3^- concentrations at two

EANET stations in the Philippines ranged from 0.21 to 1.37 µg/m^3. The 5-year mean NO_3^- concentrations at three EANET stations in the Republic of Korea ranged from 1.20 to 3.06 µg/m^3. The 5-year mean NO_3^- concentrations at four EANET stations in Thailand ranged from 0.21 to 2.09 µg/m^3. The NO_3^- concentration levels in Mongolia and Russia were low and more consistent with a 5-year mean range of 0.01 to 0.16 µg/m^3 for two EANET stations in Mongolia and 0.03 to 0.61 µg/m^3 for three EANET stations in Russia, respectively.

The highest NH_4^+ concentration was found at Hongwen of China, the only station reporting data, with a 5-year average NH_4^+concentration of 3.26 µg/m^3. The second highest 5-year averaged NH_4^+ concentration was in the Republic of Korea with a mean of 2.12 µg/m^3 and a range of 1.66 µg/m^3 to 2.66 µg/m^3. The third highest 5-year averaged NH_4^+ concentration was found in Vietnam with a mean of 1.65 µg/m^3 and a range of 1.11to 1.97 µg/m^{3}a for five EANET stations. The 5-year mean NH_4^+ concentrations of the remaining countries were relatively low. The 5-year mean NH_4^+ concentration at Phnom Penh of Cambodia was 0.49 µg/m^3. The 5-year mean NH_4^+ concentrations at three EANET stations in Indonesia ranged from 0.71 to 1.34 µg/m^3. The 5-year mean NH_4^+ concentration at 12 EANET stations in Japan ranged from 0.20 to 1.17 µg/m^3, with the highest concentration occurring in Tokyo. The 5-year mean NH_4^+ concentration at Vientiane of Lao PDR was 1.17 µg/m^3. The 5-year mean NH_4^+ concentrations at three EANET stations in Malaysia ranged from 0.15 to 1.03 µg/m^3. The 5-year mean NH_4^+ concentrations at three EANET stations in Mongolia ranged from 0.05 to 1.48 µg/m^3. The 5-year mean NH_4^+ concentrations at Yangon of Myanmar was 0.47 µg/m^3. The 5-year mean NH_4^+ concentrations at two EANET stations in the Philippines ranged from 0.05 to 0.57 µg/m^3. The 5-year mean NH_4^+ concentrations at four EANET stations in Russia ranged from 0.06 to 0.66 µg/m^3. The 5-year mean NH_4^+ concentrations at four EANET stations in Thailand ranged from 0.23 to 1.09 µg/m^3.

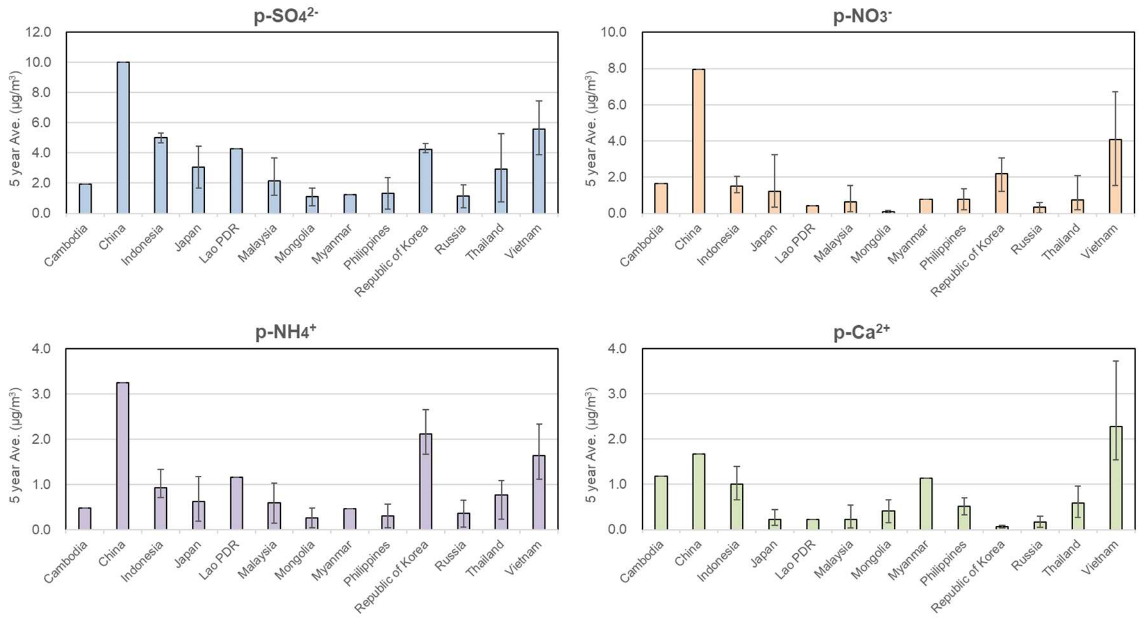


Figure 4.4. Average concentrations of particulate SO_4^{2-}, NO_3^-, NH_4^+, and Ca^{2+} at the EANET monitoring sites in each country (annual average from 2015 to 2019).

The highest 5-year averaged Ca^{2+} concentration was found in Vietnam with a mean of 2.28 µg/m^3 and a range of 1.61 to 3.73 µg/m^3 for five EANET stations. The second highest Ca^{2+} concentration was found in Hongwen of China and Phnom Penh, with the same 5-year average Ca^{2+} concentration of 1.68 µg/m^3. The 5-year mean Ca^{2+} concentrations of the remaining countries were relatively low. The 5-year mean Ca^{2+} concentrations at three EANET stations in Indonesia ranged from 0.96 to 1.40 µg/m^3. The 5-year mean Ca^{2+} concentrations at 12 EANET stations in Japan ranged from 0.09 to 44 µg/m^3, with the highest concentration occurring in Tokyo. The 5-year mean Ca^{2+} concentration at Vientiane of Lao PDR was 0.22 µg/m^3. The 5-year mean Ca^{2+} concentrations at three EANET stations in Malaysia ranged from 0.03 to 0.54 µg/m^3. The 5-year mean Ca^{2+} concentrations at two EANET stations in Mongolia ranged from 0.15 to 0.67 µg/m^3. The 5-year mean Ca^{2+} concentration at Yangon of Myanmar was 1.14 µg/m^3. The 5-year mean Ca^{2+} concentrations at two EANET stations in the Philippines ranged from 0.33 to 0.70 µg/m^3. The 5-year mean Ca^{2+} concentrations at three EANET stations in the Republic of Korea ranged from 0.03 to 0.09 µg/m^3. The 5-year mean Ca^{2+} concentrations at four EANET stations in Russia ranged from 0.05 to 0.30 µg/m^3. The 5-year mean Ca^{2+} concentrations at four EANET stations in Thailand ranged from 0.26 to 0.97 µg/m^3.

The annual trends of gas and aerosol concentration levels were estimated using the Mann-Kendall method. The trend analysis focused on the annual average of the atmospheric concentration of SO_2, NO_2, NOx, NH_3, O_3, PM_{10}, and $PM_{2.5}$. Table 4.1. shows the annual trend of SO_2 concentration at each EANET site from 2000 to 2019. Among the 51 sites, 27 sites showed a significant decreasing trend ($p<0.1$) and 8 sites showed a strongly significant decreasing trend ($p<0.001$). The decreasing trend of SO_2 at many sites in East Asia is associated with the decrease in anthropogenic emissions of SO_2. SO_2 emissions in EANET participating countries significantly increased in the early 2000s mainly due to contributions from coal-fired power plants, which increased rapidly along with large economic growth. Then, the SO_2 emissions exhibited decreasing trends after the middle of the 2000s, reflecting the effects of control measures.

Table 4. 1. Annual trend of SO_2 concentration at each EANET site from 2000 to 2019

Unit: ppb yr^{-1}

Country	Site	Signific.	Slope
Cambodia	Phnom Penh	*	-0.20
China	Jinyunshan	***	-0.91
	Hongwen	***	-0.37
	Haibin-Park	*	-0.26
Indonesia	Jakarta	+	-0.23
	Serpong	*	-0.03
	Kototabang	*	-0.28
	Bandung		-0.08
Japan	Rishiri		0.00
	Ochiishi		0.00
	Tappi		0.00
	Sado-seki	***	-0.02
	Happo	**	-0.03
	Ijira	**	-0.02
	Oki	+	-0.02
	Banryu	***	-0.05
	Yusuhara		-0.01
	Hedo	*	-0.01
	Ogasawara		0.00
	Tokyo	***	-0.08
Lao PDR	Vientiane		0.00
Malaysia	Petaling Jaya	***	-0.10
	Tanah Rata	+	-0.04
	Danum Valley	*	-0.01
Mongolia	Ulaanbaatar		-0.40
	Terelj		0.00
Myanmar	Yangon	+	-0.13
Philippines	Metro Manila	**	-0.27
	Los Banos	+	-0.08
	Mt. Sto. Tomas		-0.04
Republic of Korea	Kanghwa	+	-0.06
	Cheju	*	-0.13
	Imsil		0.01
Russia	Mondy		-0.01
	Listvyanka		0.06
	Irkutsk		0.10
	Primorskaya		-0.03
Thailand	Bangkok		0.05
	Samutprakarn	***	-0.25
	Pathumthani		-0.19
	Khanchanaburi		0.01
	Mae Hia	***	-0.02
	Chang Phueak		-0.01
	Si Phum		-0.05
	Sakaerat	+	0.02
	Nai Mueang		-0.08
Vietnam	Hanoi	+	-0.09
	Hoa Binh	+	-0.07
	Can Tho		-0.13
	Ho Chi Minh		-0.20
	Yen Bai	+	-0.53

Siginificance
*** if trend at α = 0.001 level of significance
** if trend at α = 0.01 level of significance
* if trend at α = 0.05 level of significance
+ if trend at α = 0.1 level of significance

Many EANET sites observed a significant increasing trend in the annual O_3 concentration, and the other surface ozone databases in East Asia also exhibited an increasing trend of summertime mean of daytime average and the daily maximum 8-hour average over East Asia. It is important to keep track of the trend of surface ozone in East Asia combined with EANET data and other monitoring databases. Since many sites installed a $PM_{2.5}$ monitor after 2015, a clear trend of annual $PM_{2.5}$ concentration has not been observed to date. Continuous monitoring is important to elucidate long-term trends in East Asia.

5. Impacts on Ecosystems in East Asia

Long-term monitoring data on soil, forest vegetation, inland water, and forest catchment

The EANET has been monitoring various ecosystem components, such as soil, forest vegetation, and inland water, to assess the impacts of atmospheric deposition on ecosystems since 2000. Important datasets over 10 years have been accumulated in many monitoring sites. Additionally, EANET has been promoting a catchment-scale analysis since 2010 considering biogeochemical cycles in forest ecosystems. The results should be discussed not only from the perspective of atmospheric deposition but also from other relevant factors, such as climate change.

Soil chemical properties and trends

Soil monitoring has been conducted at 31 sites in 10 countries, including the preliminary survey in 1999. Official survey records have been made since 2000 and 6 surveys were conducted at the most intensive sites during the last 20 years. As previous chapters have shown, the atmospheric environment has been improving over the 10 years, and attention should be paid to whether this reflects on soil chemical properties. This section focused mainly on soil pH (H_2O).

The geographical locations of the EANET soil monitoring sites cover a very widespread area. Latitude directly relates to climatic conditions and reflects diverse precipitation and air temperature. These climatic conditions also regulate vegetation and soil formation, both of which comprise soil type. The correlation between precipitation and soil pH (H_2O) was significantly negative ($p < 0.001$, t-test), whereas that between latitude and pH (H_2O) was significantly positive ($p < 0.001$, t-test), which means that soil pH (H_2O) was low in the site where precipitation was high and latitude was low, that is, near the equatorial region. There was a significant correlation found between latitude and mean annual precipitation ($p < 0.001$, t-test), and therefore, the effect of the climatic condition was significant on soil chemical properties. Generally, high precipitation and air temperature induce chemical weathering of soil minerals and leaching of base cations, resulting in soil acidification and low soil pH (H_2O) (Breemen et al., 1984).

To elucidate which factor was the most important to control soil pH (H_2O) distribution among these four factors (i.e., soil type, forest type, mean annual precipitation, and latitude), a partial least squares regression (PLS-R) analysis was performed. This method is similar to multiple regression analysis but suitable to deal with explanation variables having multi-collinearity. The result showed that soil type and mean annual precipitation were the dominant factors for variation of pH (H_2O) (Table 5.1). As mentioned above, it was suggested that the soil formation process and climatic conditions, as represented by the mean annual temperature, affected soil chemical properties.

Table 5.1 Results of PLS-R

Explanatory variable for pH(H_2O)	VIP *
- Soil type	1.286
- Mean annual precipitation	1.254
- Forest type	0.834
- Latitude	0.277

*, VIP (variable importance for projection) means the strength of each explaining variable.

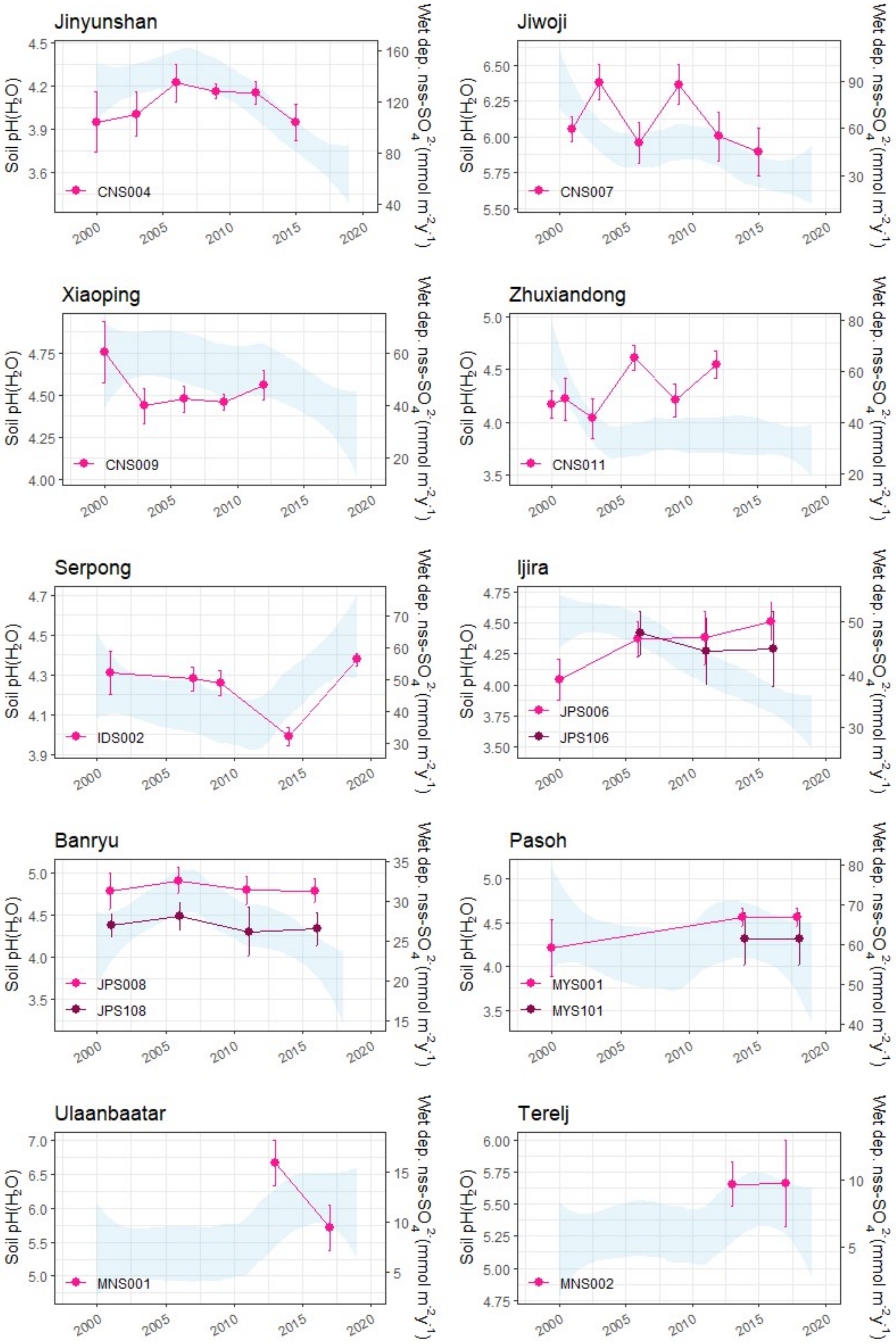


Figure 5.1 a. Temporal changes in the soil pH (H_2O) with the wet deposition nss-SO_4^{2-}.

The line chart represents the soil pH (H_2O) in each survey year and the band represents the variation of wet deposition nss-SO_4^{2-} derived from locally weighted scatter plot smooth (loess) with a 95% confidence interval.

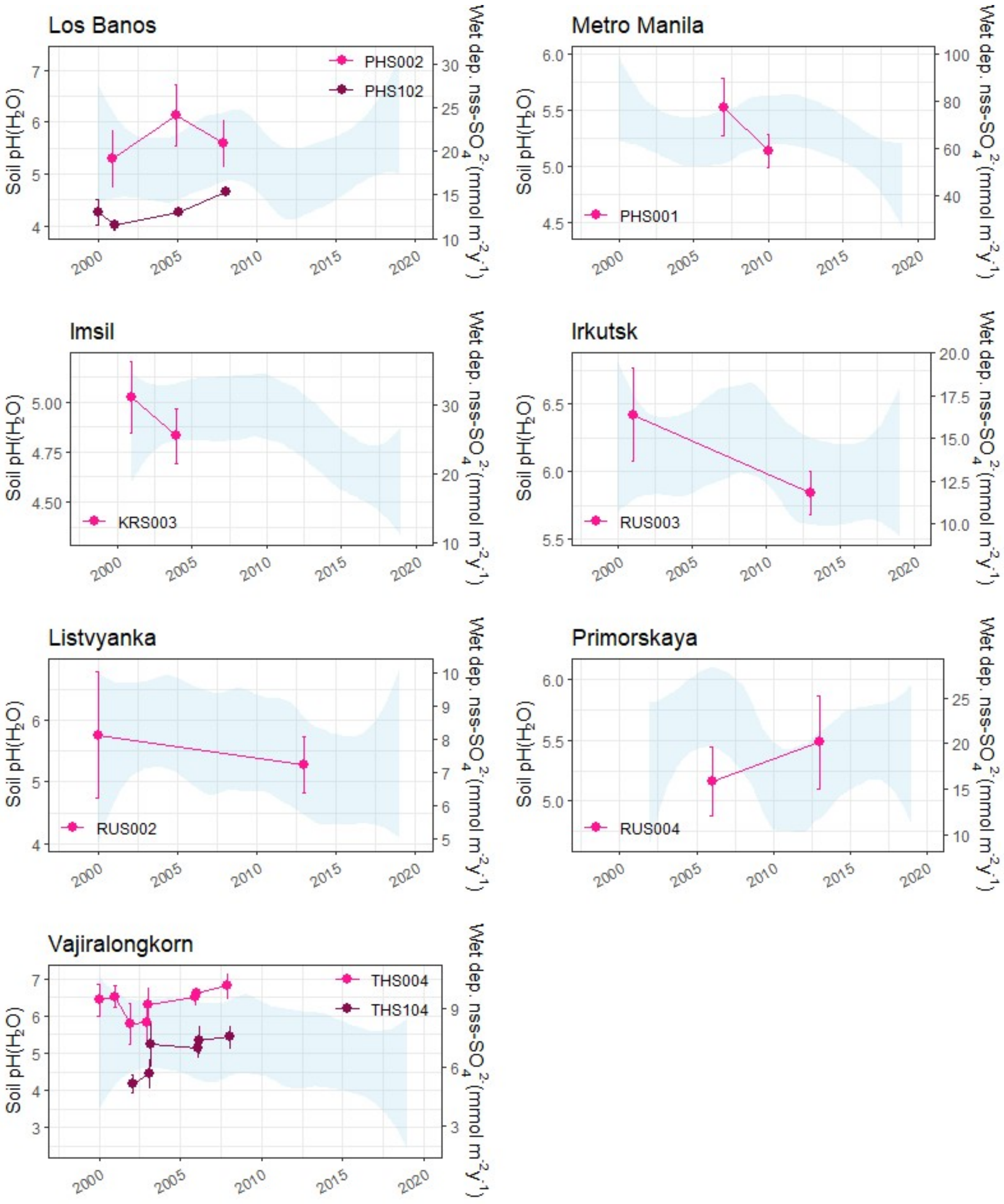


Figure 5.1 b. Temporal changes in the soil pH (H_2O) with the wet deposition nss-SO_4^{2-}.
The line chart represents the soil pH (H_2O) in each survey year and the band represents the variation of wet deposition nss-SO_4^{2-} derived from locally weighted scatter plot smooth (loess) with a 95% confidence interval.

Figures 5.1a and 5.1b illustrate the temporal changes of pH (H_2O) at each EANET site and the background band represents a trend of annual wet deposition of nss-SO_4^{2-} observed at the corresponding atmospheric monitoring station to each soil monitoring site. At most of the sites, the temporal change in soil pH (H_2O) appeared to fluctuate following the trend of acid deposition. The annual wet deposition of nss-SO_4^{2-} tended to decrease at sites in China, Japan, Metro Manila in the Philippines, and the Republic of Korea. Following this decrease, the pH (H_2O) in CNS009, CNS001, and JPS006 showed an increasing trend (Figure 5.1a). Although soil pH (H_2O) should increase in response to this atmospheric trend theoretically, the temporal change of soil pH (H_2O) did not necessarily comply with this theory in some sites (e.g., CNS004, CNS007, JPS106, JPS008, JPS108, and KRS003). It was suggested that the effect of acid deposition from the atmosphere on soil chemical properties was complicated because it reflected not only the acid neutralization reaction of soil minerals but also other many processes including interactions between atmospheric deposition and the forest trees and litter layer.

It was suggested that the soil reaction was not necessarily parallel to the input of atmospheric deposition. Considering the diverse distribution of soil characteristics among EANET sites, it was also suggested the reaction process would be different for each site. However, there are a considerable number of sites where soil monitoring has not been conducted enough times for the last 20 years. As this monitoring system will also provide valuable data for understanding the effects of climate change on ecosystems, soil monitoring should be continued more intensively.

Stand dynamics of forest vegetation

Vegetation monitoring has been conducted at 24 sites in eight countries since 2000. There is a large variation in the content and frequency of surveys among sites. Regarding vegetation monitoring, it was assumed that a comprehensive forest survey (General description of the forest; GDF), tree decline survey (Observation of tree decline; OTD), and understory vegetation survey would be conducted. Typically, trees subject to GDF are individually identified and recorded every three to five years. OTD is conducted every year for some GDF target trees, but the number of trees surveyed is small. The sites that have been surveyed more than once since 2015 are only those in China and Japan; four monitoring sites can track the diameter at breast height (DBH) growth of standing trees in China; there also are four monitoring sites in Japan.

Tree growth is influenced by various environmental factors such as meteorological conditions (i.e., temperature, precipitation, strong winds, and snowfall), CO_2 concentration, atmospheric depositions, intra- and inter-specific competition, diseases, pests, and anthropogenic physical disturbance (Forzieri et al. 2021; Hisano et al. 2020). The load of acid deposition (air pollution) is one of them. As a result of air pollution control measures in China, the amount of wet deposition of nss-SO_4^{2-}, which is an indicator of the intensity of regional air pollution, has been declining at each survey site since 2007 (Figure 5.1).

The stand dynamics of monitoring trees in the EANET plots did not exhibit clear responses to changes in regional air pollution. However, it was suggested that thinning as a method of forest management and/or damage caused by bark beetles had also influenced DBH and the total trunk cross-sectional area at breast height (BA) in some of the sites.

Even considering the effects of thinning and bark beetles, it is not clear whether the changes in the atmospheric environment had a positive effect on tree growth.

The amount of wet deposition of nss-SO_4^{2-}, which is an indicator of the intensity of regional air pollution, has been declining at each survey site since 2007. However, no corresponding increase in forest growth was observed. This may be due to anthropogenic disturbance, pest infestation (i.e., bark beetles), or intensified competition among individual trees as the age of the forest increases. Assessing the effects of air pollution through field monitoring requires appropriate monitoring plans to distinguish the effects of air pollution from other factors. To quantitatively assess the impact of air pollution on a wide range of forests, it is necessary to identify individual trees and conduct long-term growth measurements accompanied by air quality monitoring with little or no anthropogenic disturbance.

Inland water chemistry and trend

The data on the inland aquatic environment have been collected in 20 sites from 11 countries, while Banryu Lake in Japan was replaced by Futago-ike Lake (consists of two neighboring lakes, Oike and

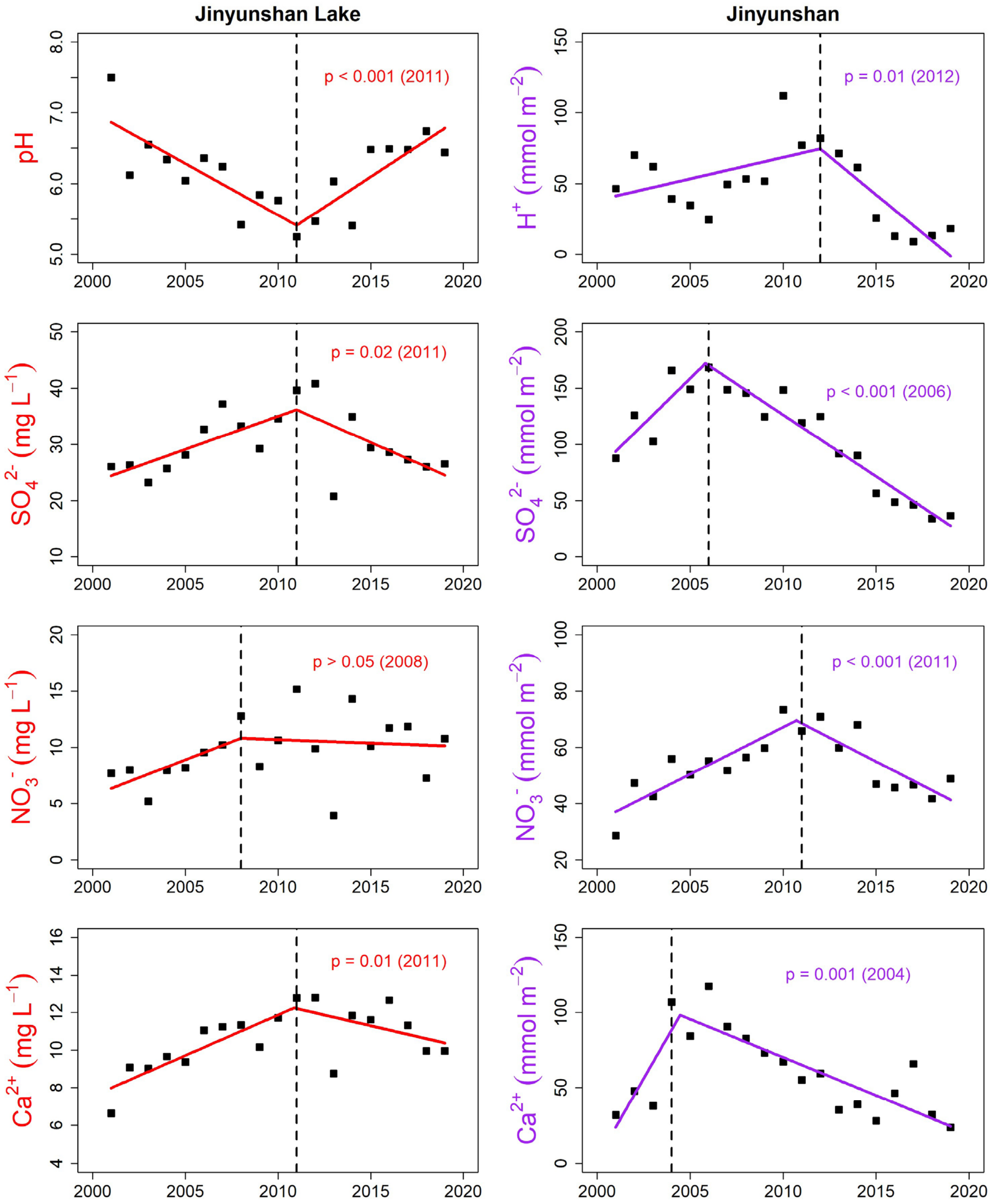
Meike) in 2019.

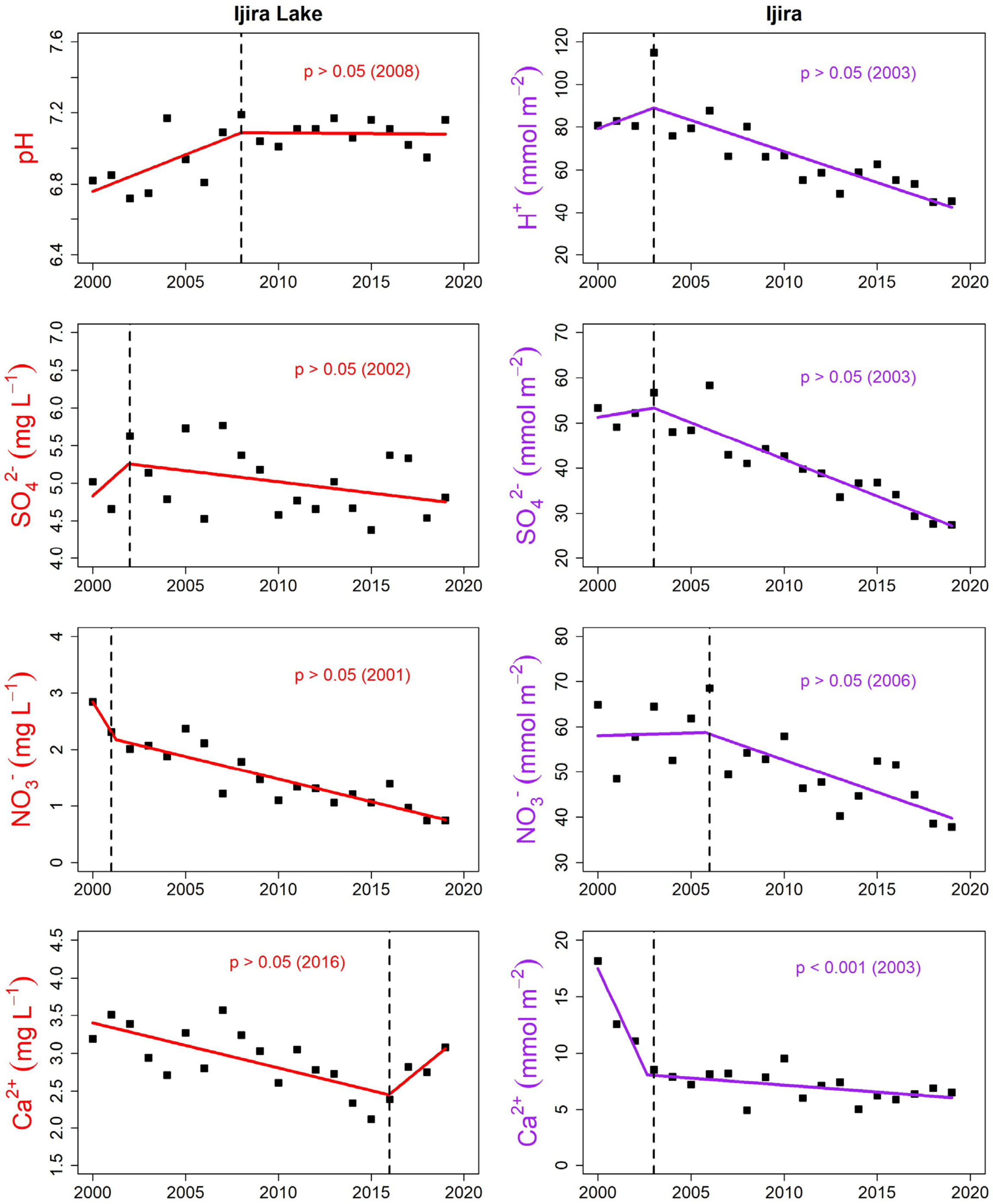
The Jinyunshan Lake, Xiaoping Dam, Jiwozi River, and Zhuxiandong Stream in China showed overall decreasing trends in pH values and increasing trends in the concentrations of anions (especially SO_4^{2-}) from the beginning of the monitoring period to either the end of its first ten years or the beginning of its last ten years, where overall increasing trends in pH values and decreasing trends in the concentrations of anions occurred. The discovered patterns of water chemistry across four sites suggest that these sites are currently recovering from their acidified states. Similar phenomena of acidification and recent recovery were observed in Ijira Lake in Japan and Patenggang Lake in Indonesia.

To understand the influence of atmospheric deposition on the observed patterns of water chemistry, the piecewise linear regression model in the segmented R package (Muggeo, 2008; R Development Core Team, 2020) was applied to the Jinyunshan Lake, Ijira Lake, and Patenggang Lake with wet deposition data in their nearest sites (Fig. 5.2). The results in Jinyunshan indicated the occurrence of inflection points (breakpoints). Before these points, the trends were decreasing for pH while increasing for anions and cations, and after these points, the trends were increasing for pH but generally decreasing for anions and cations (Fig. 5.2 top). Atmospheric wet H^+ depositions at the Jinyunshan site were found to follow trends different from those of pH values in the Jinyunshan Lake, with the year 2012 as the breakpoint between increasing and decreasing trends. The values of SO_4^{2-} and NO_3^- at the lake and its nearest deposition site showed similar patterns.

Ijira Lake in Japan has been generally experiencing an increasing trend in pH values and decreasing trends in SO_4^{2-} and NO_3^- concentrations throughout the monitoring period (Figure 5.2; middle). The long-term patterns of water chemistry observed in the Ijira Lake indicate continuous recovery of this lake from acidification and that decrease in deposition of acid constituents at Ijira site may have accelerated the recovery process.

Moreover, both a decreasing trend (2000-2008) and an increasing trend (2008-2019) in the values of pH were detected in the Patenggang Lake (Indonesia). In addition to the decrease in H^+ depositions throughout the monitoring period at Bandung, which is the nearest deposition site of Patenggang Lake, the ongoing recovery of this lake from its acidified state might also be attributed to the significant increase in Ca^{2+} depositions (Figure 5.2; bottom).


Jinyunshan Lake
Jinyunshan
pH
p < 0.001 (2011)
H+ (mmol m−2)
p = 0.01 (2012)
SO4 2- (mg L−1)
p = 0.02 (2011)
SO4 2- (mmol m−2)
p < 0.001 (2006)
NO3 - (mg L−1)
p > 0.05 (2008)
NO3 - (mmol m−2)
p < 0.001 (2011)
Ca2+ (mg L−1)
p = 0.01 (2011)
Ca2+ (mmol m−2)
p = 0.001 (2004)
2000
2005
2010
2015
2020


Ijira Lake
Ijira
pH
p > 0.05 (2008)
H⁺ (mmol m⁻²)
p > 0.05 (2003)
SO₄²⁻ (mg L⁻¹)
p > 0.05 (2002)
SO₄²⁻ (mmol m⁻²)
p > 0.05 (2003)
NO₃⁻ (mg L⁻¹)
p > 0.05 (2001)
NO₃⁻ (mmol m⁻²)
p > 0.05 (2006)
Ca²⁺ (mg L⁻¹)
p > 0.05 (2016)
Ca²⁺ (mmol m⁻²)
p < 0.001 (2003)
2000
2005
2010
2015
2020

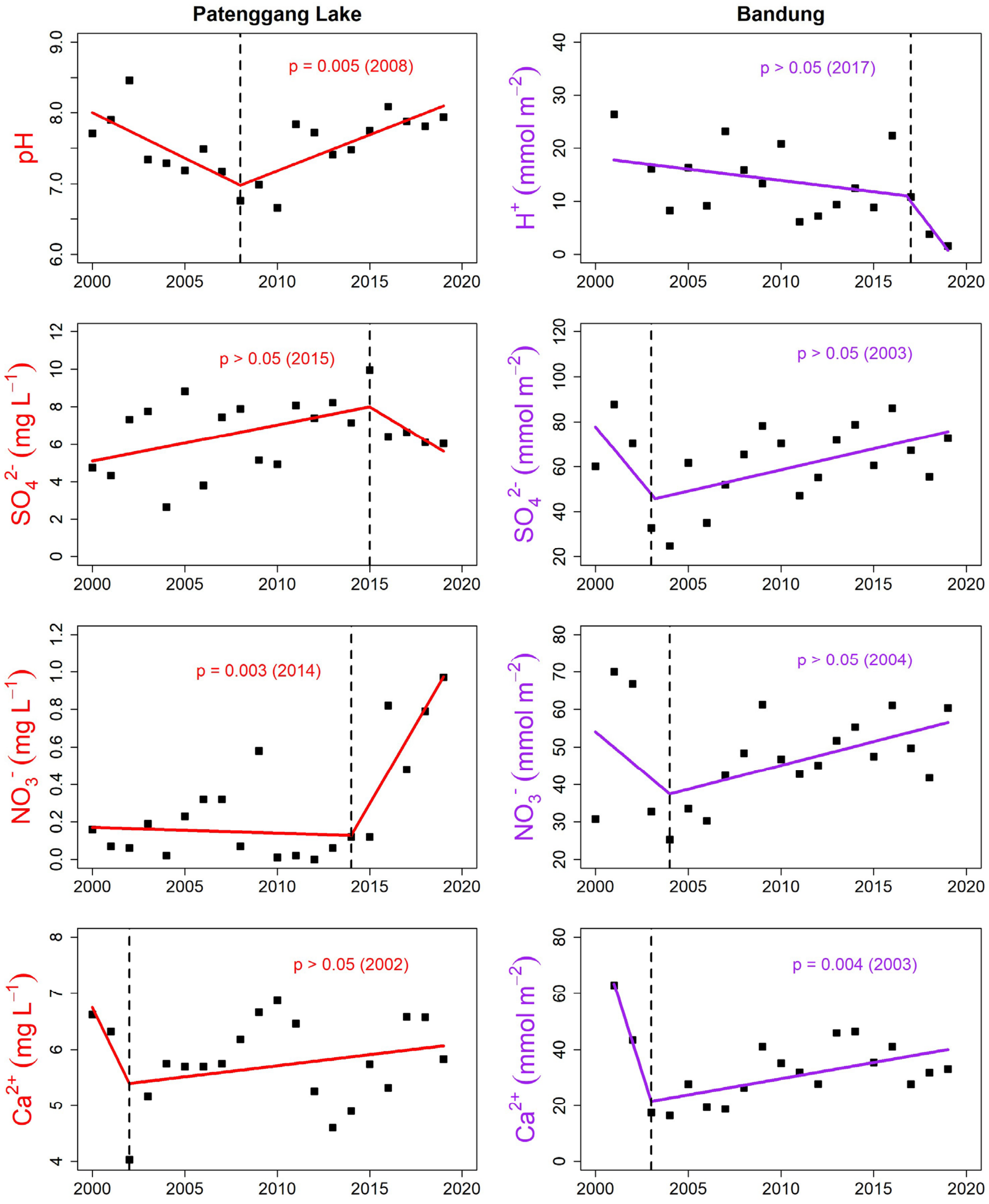


Figure 5.2. Long-term patterns of five selected chemical variables at three typical inland sites (Top, Jinyunshan Lake; Middle, Ijira Lake; Bottom, Patenggang Lake) and their nearest deposition sites. The vertical dashed lines indicate the breakpoint locations as estimated by piecewise linear regression model.

Chemical properties, such as the balance of ions, concentration levels, and seasonal changes, varied among the monitoring sites. Depending on the chemical properties, the response of inland waters to atmospheric deposition also differed. However, several monitoring sites, such as the Jinyunshan Lake, Xiaoping Dam, Jiwozi River, and Zhuxiandong Stream in China, Patenggang Lake in Indonesia, and Ijira Lake in Japan, showed symptoms suggesting their recovery from acidification. In some sites, changing patterns of atmospheric deposition at the nearest monitoring sites were associated with those of ion concentrations in lakes/streams, respectively. It is suggested that further reduction in atmospheric acid deposition and its monitoring are necessary to ensure their continued recovery from acidification.

Forest catchments

Following the recommendations described in the EANET strategy papers for ecological monitoring (e.g., EANET 2020), the research activities on catchment analysis were conducted from 2002 onwards at several reference forest sites, including the Kajikawa site (KJK) and the Lake Ijira catchment site (IJR) in Japan, in cooperation with scientists from EANET countries (Sase et al. 2019, 2021). Based on the Guideline for Catchment-scale Monitoring in East Asia (EANET 2010) developed through the research experience above, the regular catchment-scale monitoring has been conducted at two sites, IJR since 2007 and the La Mesa Watershed (LMW) in the Philippines since 2019. Moreover, the data analysis on a catchment scale has been implemented for the Komarovka River catchment (KMR) in Russia.

Field observations on a catchment scale at IJR and KJK have been implemented as EANET activities since 2007 and 2002, respectively. In the case of IJR, the EANET data on wet deposition and stream water chemistry from 2000 onwards are also available, while the stream water data from 2000 onwards have just been collected every quarter. Based on long-term national monitoring data since 1988, it was reported that the stream water at IJR was acidified with the increase of NO_3^- concentration in the mid-1990s, suggesting the N saturation of the ecosystems (Nakahara et al. 2010). However, recently, Sase et al. (2019) pointed out that the stream water has been recovering from acidification and N saturation is showing a recovery of the pH to its original level (approximately 7.0) and a decline in NO_3^- concentration in the early 2000s (see the inland water section). The recovery phenomena were accompanied by decreases in atmospheric S and N depositions. The stream water at KJK also shows an increase in pH with decreases in SO_4^{2-} concentrations, suggesting the recovery process, in particular since 2006. This was accompanied by decreases in atmospheric S and N depositions, too. However, the NO_3^- concentration in stream water at KJK still showed an increasing trend.

The input-output budgets of S at IJR and KJK are shown in Figure 5.3. In the case of IJR, the S outputs were generally 2 - 3 times larger than the inputs, suggesting additional S sources other than atmospheric depositions, even though the input has been declining smoothly. Based on the S isotopic analysis, Sase et al. (2019) suggested that geological S derived from bedrock largely contributed as an S source of the stream water and that S derived from atmospheric depositions has been accumulated in forest soil. Since changes in precipitation amounts/patterns and/or temperatures may affect the retention – release process of S in forest ecosystems, a recent increase in the net export at IJR should carefully be monitored. In the case of KJK, the input and output were relatively well-balanced and both gradually declined. It was suggested that atmospheric S is the main source of the stream water S, which was also supported by the S isotopic analysis (Sase et al. 2021). However, because the response of the output to the input was delayed and slightly weaker than the input, the net export has been increased and the output exceeded the input in certain instances. Similar phenomena were observed in Europe (Vuorenmaa et al. 2017). The S isotopic analysis suggested that S derived from atmospheric depositions was retained or cycled in the ecosystems and well homogenized before leaching the stream water (Sase et al. 2021). In this process as well, the impact of climatic conditions should carefully be monitored.

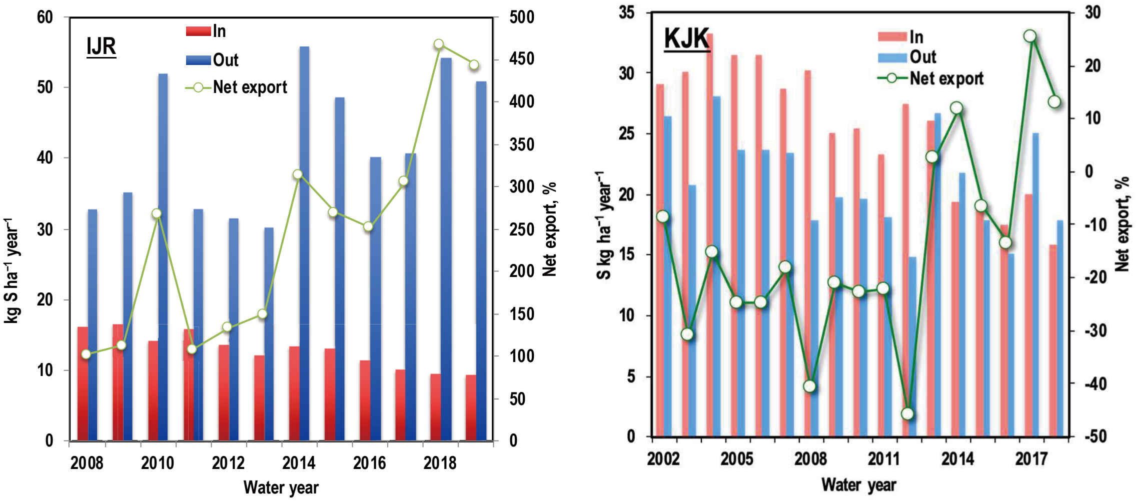


Figure 5.3. Input-output budgets of S at IJR (left) and KJK (right). The input of IJR, estimated as the flux by wet and dry depositions, input of KJK, estimated as the flux by TF+SF. Net export (%) = (output – input) / input × 100. The figure on the right was updated with the latest data after Sase et al. (2021). Water years (WYs) were defined as November of one year to October of the following year and June of one year to May of the following year at IJR and KJK, respectively.

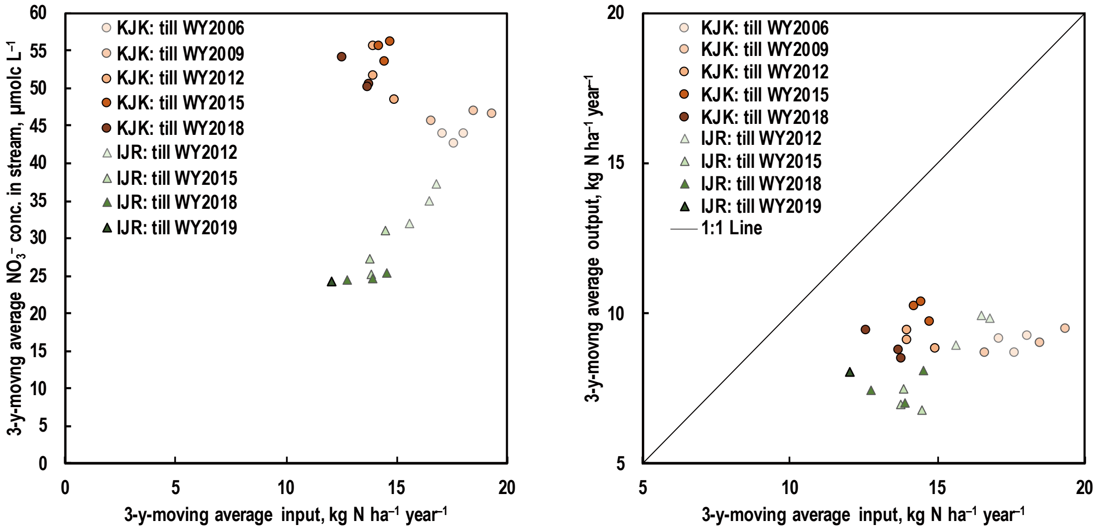


Figure 5.4. Changes in 3-year-moving averages of NO_3^- concentrations in stream water (left) and stream water N outputs (right) with those of atmospheric N inputs at IJR and KJK. The data since 2007 to 2019 and the data since 2002 to 2019 were applied to calculate the 3-year-moving averages at IJR and KJK, respectively. WYs were defined as from November of one year to October of the following year and June of one year to May of the following year at IJR and KJK, respectively. WYs were referred to by the following year and starting year for IJR and KJK, respectively; e.g., WY2019 at IJR, from November 2018 to October 2019; WY2018 at KJK, from June 2018 to May 2019. The N inputs were based on the total depositions by wet and dry depositions and depositions by TF+SF at IJR and KJK, respectively.

To enable an understanding of the changes in NO_3^- concentrations in stream water and stream water N outputs, their 3-year-moving averages are plotted with those of the atmospheric N input in Figure 5.4. On the one hand, the NO_3^- concentration at IJR clearly declined with the atmospheric N input, while those at KJK increased. On the other hand, the stream water N output at IJR did not decline recently and rather slightly increased and those at KJK slightly increased as well. The stream water N outputs were still lower than the 1:1 line of N balance but have been approaching the line (the right side of Figure 5.4). Large portions of the inputs leached the stream waters at IJR and KJK. The recent high precipitation amounts at IJR and the continuous decline of precipitation at KJK appeared to have influenced the stream water N outputs. The monsoon climate with warm and wet summer appeared to have contributed to N leaching from forest ecosystems (Fang et al. 2011). Thus, N leaching should carefully be monitored taking into account climate and forest conditions. Moreover, the current atmospheric N inputs are still enough large, over 10 kg N ha^{-1} $year^{-1}$.

The catchment biogeochemical cycles have sensitively responded to the recent decline of atmospheric depositions (in particular, SO_4^{2-}), resulting in phenomena indicating recovery from acidification. However, NO_3^- concentration in stream water at KJK has been increasing and the decline rates of NO_3^- concentration in stream water at IJR have become lower in recent years. Forest types, maturation of trees, and meteorological variability appeared to affect N leaching to stream water. It is suggested that interactions with meteorological variations be taken into consideration in recovery processes from acidification and N saturation. New catchment analysis sites, KMR and LMW, may also contribute to further understanding of ecological response to atmospheric deposition and climate change.

Summary of ecological impacts

The aforementioned results suggest that the ecological monitoring data should be discussed not only with atmospheric deposition but also with other relevant factors, such as natural characteristics of soil and vegetation, disturbance of various environmental factors, and climate (including year-to-year meteorological variability). Although changes in soil chemical properties and forest growth did not necessarily correspond to those of atmospheric deposition, inland water chemistry, including streams of forest catchments, have sensitively responded to the atmospheric deposition. In order to detect the effects of atmospheric deposition on forest ecosystems, data with low noise factors such as site disturbances (including anthropogenic disturbances) are required. As shown in the cases in Europe and the United States, it is important to continue monitoring ecosystems and model analyses using the monitoring results, even after the atmospheric deposition has decreased sufficiently.

6. Relevant Studies on Atmospheric Environment Assessment in the EANET region

Introduction

Due to its continued rapid economic growth after 2000, Asia continues to attract much more concern than before from the view of global, hemispherical, and regional air pollution. In addition to EANET's efforts, there have been many research activities and organizational initiatives focusing on regional air pollution in East Asia as well as from global and hemispherical perspectives. In this chapter, such research outputs and initiatives in the last few years are reviewed to provide useful information on the present status of acid deposition and regional air pollution. The covered topics are atmospheric observation, including satellite observation, emission inventory, chemical transport models (CTMs), and ecological and human health impact studies, which are not necessarily directly related to EANET but would be useful for understanding air pollution and acid deposition in East Asia.

In the previous part, the observation studies, emission inventory studies, and CTM are systematically reviewed. In the following part, ecological impact assessment studies and human health impact studies are reviewed, respectively. In the final part, other international activities in the EANET region are introduced and a cross-chapter analysis of acid deposition based on measurements in EANET sites, emissions inventory, satellite measurement, and CTM is carried out.

This part does not intend to provide a comprehensive review, but the atmospheric observational studies focusing on field campaigns and satellite measurement, as well as studies on acidification and nitrogen leaching in forest catchments, global and regional emision inventories, the Model Inter-Comparison Studies for Asia (MICS-Asia) utilizing EANET data for validation and analysis, and ecological impact assessment studies on the national and/or regional scales and impacts of PM2.5 and ozone on human health, are introduced with higher priority. Additionally, some original analyses are conducted in the satellite observation, emission trends, health impacts, and acid deposition.

Observational studies for the EANET region

Field campaigns for regional air quality

One of the objectives of acid deposition monitoring in EANET is to grasp the effect of acidification and the long-term trends of acidifying species and related chemical substances and the other relevant air pollutants. Long-term monitoring datasets are useful for the evaluation of the impact on the ecosystem and human health. However, to elucidate the impacts of specific atmospheric events and chemical and physical processes relevant to assessing the effects of air pollutants, intensive observational studies based on field campaigns will be helpful in addition to long-term monitoring studies. In this section, field campaigns for acid deposition and regional air quality in East Asia are reviewed.

The Seven South East Asian Studies (7-SEAS) project was established to characterize aerosol-meteorological interactions in Southeast Asia. The 7-SEAS program was organized through a collaborative effort with the United States and governments in Southeast Asia. There are several published research papers and project reports. These sections introduce some important research results.

The Sonla campaign aimed to investigate aerosol chemical characteristics and obtain the chemical profile of near-source biomass burning aerosols at a site in Sonla, Northern Vietnam (Lee et al., 2016). A 24-hour sampling of $PM_{2.5}$ was conducted during the 7-SEAS campaign in the spring of both 2012 and 2013. The collected particles were measured for mass concentrations, carbonaceous fractions, and water-soluble components (Fig 6.1).

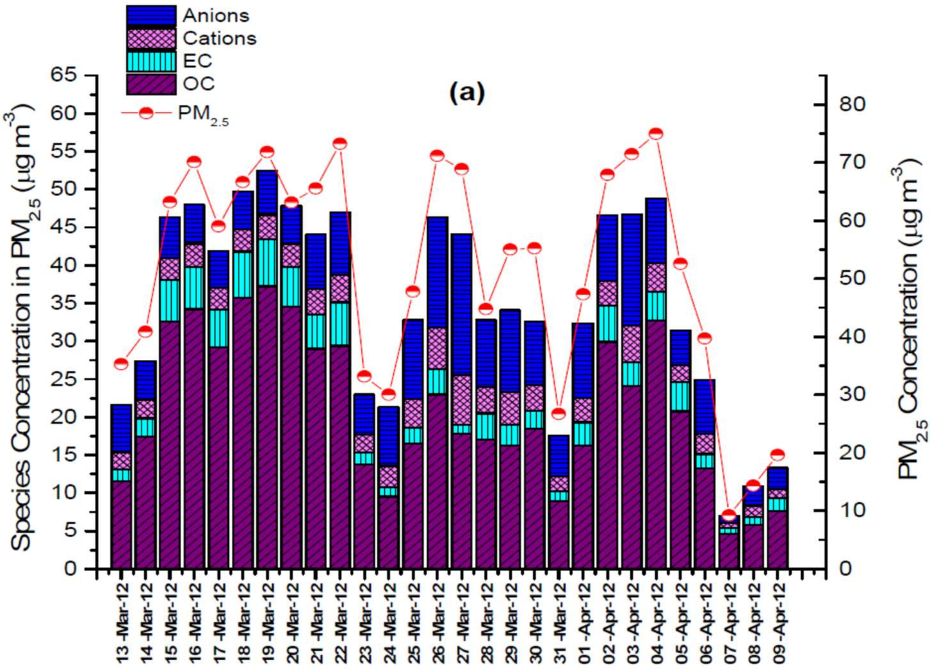


Figure 6.1. Temporal variations in $PM_{2.5}$ aerosol mass concentrations and inorganic ions and carbonaceous fractions in 2012 in Sonla, Northern Vietnam (Lee *et al.*, 2016).

The Korea–United States Air Quality (KORUS-AQ) field study was conducted between May and June 2016 and was jointly sponsored by the National Institute of Environmental Research, the Republic of Korea, and the National Aeronautics and Space Administration of the United States (Crawford et al., 2021). KORUS-AQ consists of three aircraft observations, an extensive ground-based observation network, and three ship-based observations and air quality forecast models. Information gathered during this study is contributing to an improved understanding of the factors controlling air quality in the Republic of Korea. KORUS-AQ also provided valuable scientific knowledge for future air quality-observing strategies involving geostationary satellite instruments being launched by both countries to examine air quality throughout the day over Asia and North America. Figure 6.2 shows the diurnal statistics of ozone with altitude by aircraft observation over the Seoul Metropolitan Area.

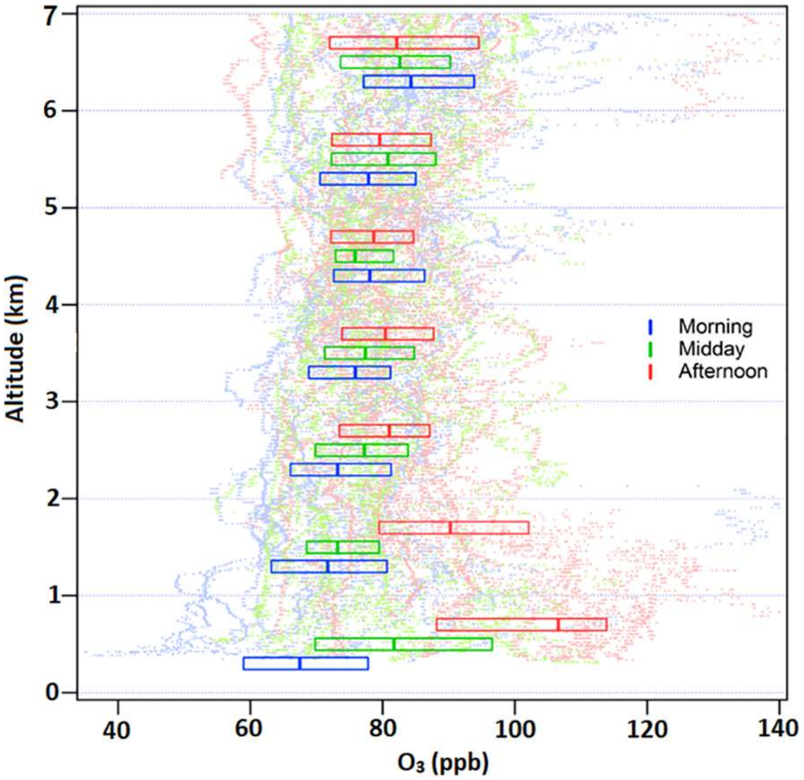


Figure 6.2. Vertical distribution of ozone observed by aircraft observation over the Seoul Metropolitan Area. The boxes show the median and inner quartile values for 1 km increments of altitude and are plotted over the individual measurements separated into the morning, midday, and afternoon observations (Crawford et al., 2021).

In early autumn 2012 and summer 2013, an intensive atmospheric observations campaign was carried out at the Field Museum TAMA of Tokyo University of Agriculture and Technology located in the suburbs of Tokyo. The observations focused on plant-derived VOCs and total OH reactivity in a typical area where anthropogenic and plant-derived substances are mixed (Ramasamy et al., 2018). Total OH reactivity, which gives the instantaneous loss rate of OH radicals due to reactive species, is an invaluable technique to understand regional air quality, as it gives the overall reactivity of the air mass including the fraction of each trace species reactive to OH, the fraction of missing sinks, O_3 formation potential. The average measured OH reactivities during that autumn and summer were 7.4 s^{-1} and 11.4 s^{-1}, respectively. In summer, isoprene was the major contributor, accounting for 28.1% of the OH reactivity, as a result of enhanced light-dependent biogenic emission, whereas NO_2 was a major contributor in autumn, accounting for 19.6%, due to the diminished contribution from isoprene as a result of lower solar strength (Fig 6.3).

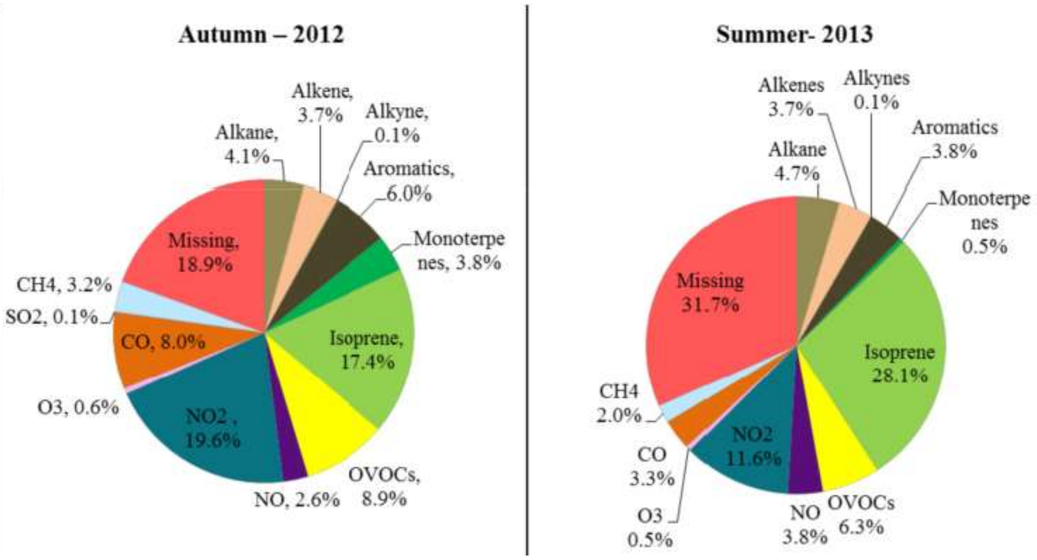


Figure 6.3. The average contribution of trace species to the total OH reactivity during the autumn 2012 and summer 2013 campaign (Ramasamy et al., 2018).

Satellite observations

Satellite observations have been useful for monitoring the air quality from space in near real-time, and are also widely used as a proxy for emissions. Representative satellite data is the NO_2 column used to capture NOx emissions. This space-based view of the NO_2 column was first reported by Richter et al. (2005) from 1996 to 2004. The NO_2 and SO_2 column dataset, which was observed by Ozone Monitoring Instruments (OMI) onboard National Aeronautics and Space Administration (NASA) Earth Observing System (EOS) Aura satellite, was used and analyzed. Aura satellite was launched on 15 July 2004 in a sun-synchronous ascending polar orbit with a local equator crossing time of 13:45±0:15. The data from 2005 to 2019 are analyzed for NO_2 and SO_2 columns. Retrieval algorithms were based on the products provided by NASA. The level-3 daily product (OMNO2d) of the latest version 3.0 (Krotkov et al., 2019) for the NO_2 column was used. This product contains the total and tropospheric column for all atmospheric conditions and for clear sky conditions (cloud fraction is less than 30%). The tropospheric column with screened for clear sky conditions was analyzed. The level-3 daily product (OMSO2e) of the latest version 3.0 (Krotkov et al., 2016) for the SO_2 column was used. This product contains SO_2 in the planetary boundary layer. Both data of NO_2 and SO_2 columns were gridded at a resolution of 0.25° × 0.25°. For the SO_2 column, the smoothed method to average out the noise contained in the SO_2 column was additionally adopted (Koukouli et al., 2016). This method smoothed the SO_2 column at each grid by weighting the surrounding eight cells, and if the negative values were found at the grid, it was treated as a zero value. The explosive volcanic eruptive events outside Asia had impacts on the Asian SO_2 column and these periods were discarded when averaging annual data in 2009 and 2011 based on the same approach in the previous report (Itahashi et al., 2018). In addition to NOx and SO_2 columns, the Aerosol Optical Depth (AOD) represents the attenuation of sunlight by aerosols and is an important measure of the aerosol column concentration (Kaufman et al., 2002) is analyzed. To obtain the long-term trends of AOD over the Asian region, AOD measured by Moderate resolution Imaging Spectroradiometer (MODIS) onboard Terra satellite was analyzed. The MODIS product in the latest Collection 6.1 (Levy et al., 2013) was provided in the period starting from 24 February 2000 to the present. Level 3 of the MOD08_3D dataset gridded into 1.0°×1.0° (NASA, 2021) was used, and the product of AOD with Dark Targett (DT)/Dark Blue (DB) algorithms were analyzed from 2000-to 2019. The analyzed areas are Northeast Asia (China, the Republic of Korea, Japan, Mongolia, and Russia) and Southeast Asia (Vietnam, Cambodia, Lao PDR, Myanmar, Thailand, Malaysia, Indonesia, and the Philippines). For Russia, to cover four EANET sites (see Fig. 1.5.1), the region of far east Russia from 90°E and up to 60°N is analyzed. The long-term trends of the NO_2 column, SO_2 column, and AOD is summarized as follows.

- The annual-averaged NO_2 column during the period 2005-2019 overall showed constant trends in 15 years with 0.10 D.U. and 0.03 D.U. over Northeast Asia and Southeast Asia, respectively. In detail, Northeast Asia posed a slight declining trend in the late 2010s, whereas Southeast Asia showed a slight increase. This trend is generally matched to the estimation of bottom-up emission inventories.
- The annual-averaged SO_2 column over Northeast Asia showed a peak in 2006-2007 and then declined. This trend is partly corresponded to the estimation of bottom-up emission inventories, and partly due to the effect of volcanic SO_2 emissions around Japan. Southeast Asia showed a peak in 2006 and 2008, and gradual increasing trends for the SO_2 column. These increasing trends found in the SO_2 column are consistent with the growth of SO_2 emissions estimated by the bottom-up emission inventory.
- The long-term trends of aerosol pollution status inferred from AOD overall posed large year-to-year variations; suggesting that the aerosol pollution did not straightforwardly relate to precursor emissions or satellite observations for NOx and SO_2 columns. This would be because AOD contains both fine- and coarse-mode aerosols, and hence is attributed to both anthropogenic and natural sources. Over Northeast Asia, it was noticed that there was a declining trend after 2013. Under the reduction of anthropogenic emissions from China, a decline in PM2.5 concentration has been reported since 2013 (e.g., Zhang et al., 2019), and it has also influenced downwind regions (e.g., Uno et al., 2020). Over Southeast Asia, for example, the highest peak was seen in 2015-2016, and then the drop was found in 2017. The broad impact caused by the biomass burning over this region will be large and will cause large year-to-year variations.

Acidification and nitrogen leaching in forest catchments

Integrated monitoring which takes into account hydrological/biogeochemical processes in forest catchments is effective to assess the effects of atmospheric deposition on terrestrial ecosystems. Therefore, as shown in Chapter 5.4 in the main text, EANET has been promoting catchment-scale monitoring and relevant studies continuously. Of course, studies on acidification and nitrogen leaching in forest catchments have been conducted by many scientists outside the EANET community. Some EANET-relevant studies have been terminated. This section reviews other relevant studies in the East Asian region. A similar review on acidification and nitrogen leaching in forest catchments has already been carried out for the Review of the State of Air Pollution (RSAP) (EANET, 2015). At that time, the Executive Summary of RSAP pointed out the following situations:

- Inland water acidification, probably due to high acid loads, occurred over the last several decades in China, Japan, and Russia, and both SO_4^{2-} and NO_3^- contributed to the acidification in the region. ANC of the bedrock geology is one of the most important factors for the manifestation of inland water acidification.
- Based on input–output measurements conducted in Japan and China, the majority of ecosystems exhibit a high retention ability for nitrogen, whereas nitrogen saturation occurred in some particular forested ecosystems. The spatial distribution of nitrate concentrations in stream water in Japan showed that there were some areas with a high concentration in the vicinity of large cities and intensive agricultural areas.

Although the scientific papers published until 2014 were reviewed in the RSAP, the atmospheric environment in the region has been dynamically changing for the last decade. The situations on forest catchments may have slightly been changed as responses to the changing atmosphere. Therefore, in this section, some updates will be highlighted, mainly based on relatively new works of literature after 2010.

The aforementioned responses of forest catchments to changing emission/deposition of air pollutants are summarized in Table 6.1. Recovery from acidification is still not clear in many cases, probably because the datasets only until 2013/2014 could be used for these assessments. Despite the decrease of S depositions, the response of ecosystems may be slightly delayed due to the internal S cycle (Sase et al., 2021). In some cases, acidification on depositions and/or stream water seems to be still ongoing. The changing climate may affect both deposition processes and ecosystem responses. The reduction of NO_X emissions after 2011/2012 has not been enough reflected in observational studies in forest areas. Moreover, not only N depositions but also other factors, such as climatic conditions and forest maturation and management, may influence N leaching processes. Monitoring and assessment for the next 5-year term will be more important to evaluate recovery from acidification and N leaching.

Table 6.1 Response of forest catchments to changing emission/deposition of pollutants in the EANET countries

Country	Monitored area (period)	Emission/ deposition	Stream water/ lake water	Source
Japan	Ijira FC (1988 – 2014)	SO_4^{2-} D decrease DIN D decrease (after 2006)	pH increase NO_3^- C decrease (after 2005/2006)	Sase et al. 2019
	Kajikawa FC (2002 – 2018)	SO_4^{2-} D decrease (after 2006) DIN D decrease (after 2007)	pH increase SO_4^{2-} C decrease (after 2006) NO_3^- C increase	Sase et al. 2021
	Tatara R basin	DIN D increase (1991 – 2007)	NO_3^- C increase (1977 – 2007)	Chiwa et al. 2012
	608 FC in the suburbs of Tokyo (spatial surveys)	Partly high DIN D (2007 – 2010)	Partly high NO_3^- C (2007 – 2010)	Nishina et al. 2017
Russia	Komarovka R (2005 – 2019)	No trend	pH decrease NO_3^- C increase	Zhigacheva in preparation
	Pereemnaya R (2001 – 2014)	SO_2 increase NO_X increase	pH decrease SO_4^{2-} C increase NO_3^- C increase	Obolkin et al. 2016
Malaysia	Baru FC (2008-2011)	No trend	No trend (well neutralized)	Yamashita et al. 2014
Thailand	Sakaerat FC	SO_4^{2-} D decrease	pH decrease SO_4^{2-} C increase	Sase et al. 2017

Note: R, river; FC, forest catchment; E, emission; D, deposition, C, concentration; L, leaching rate; DIN, dissolved inorganic nitrogen. Blue and yellow markers represent phenomena showing recovery from acidification and progress of acidification (or pollution), respectively.

Emission inventories

Air pollution, including acid deposition, is a crucial problem of atmospheric environment that we have been facing. The fundamental reason why atmospheric environmental problems occur is clear: concentrations of related species increased too much mainly due to emissions from anthropogenic activities. Therefore, to mitigate atmospheric environmental problems, it is important to find an effective way to reduce anthropogenic emissions and for this purpose, understanding details about the current status and trends of emissions is fundamentally important. It is a basic role of emission inventory, which is defined as a dataset of air pollutants emissions estimated for specified source categories and related activities in certain administrative or geographical areas and in certain time periods.

In this section, global emission inventories, regional emission inventories in Asia, and national emission inventories in EANET member countries are reviewed. For methodologies to develop emission inventories, see emission inventory guidelines and manuals such as EMEP/EEA air pollutant emission inventory guidebook (https://www.eea.europa.eu/themes/air/air-pollution-sources-1/emep-eea-air-pollutant-emission-inventory-guidebook/emep) and ATMOSPHERIC BROWN CLOUDS EMISSION INVENTORY MANUAL (Shrestha et al., 2013).

Overview of global emission inventories

Global emission inventories provide emissions data from all countries in the world, which are necessary for comparing and analyzing emissions among different countries and regions. For performing global model simulations, global gridded emission datasets are essential. Furthermore, for countries without national emissions data, we often have to rely on estimates from global emission inventories. Table 6.2 summarizes general information of major global anthropogenic emission inventories. Emission Database for Global Atmospheric Research (EDGAR) is a global database of anthropogenic emissions of greenhouse gases and air pollutants for sub-sector levels generally based on international statistics (Crippa et al., 2018). EDGAR has been providing both datasets for countries emissions and gridmaps. Evaluating the Climate and Air Quality Impacts of Short-Lived Pollutants (ECLIPSE) global emission fields provide gridded data not only for past years, but also for future projections estimated using the Greenhouse Gas and Air Pollution Interaction and Synergies (GAINS) integrated assessment model (Amman et al., 2011; Klimont et al., 2017). The EDGAR and ECLIPSE emissions inventories are developed based on consistent methodologies for each database. In addition, recently, “mosaic approaches” using independent emission inventories have also been used for the development of global emission inventories. The Task Force Hemispheric Transfer of Air Pollution (TF HTAP) developed HTAPv2 emission database by compiling different national and regional inventories based on official and latest available regional information. In HTAPv2, EDGARv4.3 was used as default data and the MIX inventory, official emission inventories for the Model Intercomparison for Asia Phase III (MICS Asia III) (see Section 6.3.2 in the main text) was used for the Asian region (Janssens-Maenhout et al., 2015). Recently, the Community Emission Data System (CEDS) inventory was developed for the Coupled Model Intercomparison Project Phase 6 (CMIP6). The CEDS calibrated default global emission estimates using independent national and regional emission inventories (Hoesly et al., 2018; McDuffe et al., 2020).

For atmospheric environmental problems, biomass burning is also an important source of related species especially in Southeast Asia. Global Fire Emissions Database (GFED) version 4 provides monthly, daily, and 3-hour emissions with burned area information (https://www.globalfiredata.org). The Fire Inventory from NCAR (FINN) version 1.0 provides daily, 1 km resolution, global estimates of air pollutants emissions from open biomass burning including wildfire, agricultural fires, and prescribed burning (https://www2.acom.ucar.edu/modeling/finn-fire-inventory-ncar).

For the development of accurate emission inventories and utilizing the data for improving atmospheric environment, international cooperation is essential among both scientific researchers and policy makers. The Global Emission InitiAtive (GEIA), which was created in 1990, is a community of emission experts for building emissions data access and analysis platforms and communicating with the emissions community through online resources and in-person meetings (http://www.geiacenter.org). GEIA is also collecting emission datasets which are provided from Emissions of atmospheric Compounds and Compilation of Ancillary Data (ECCAD), the GEIA data portal (https://eccad.aeris-data.fr).

Table 6.2 General information of major global anthropogenic emission inventories

Names	Descriptions
EDGARv5.0	Species: SO_2, NO_x, CO, NMVOCS, NH_3, CO_2, CH_4, N_2O, PM_{10}, $PM_{2.5}$, BC, OC, and Mercury; Year: 1970-2015; Resolution; 0.1°×0.1°; https://edgar.jrc.ec.europa.eu
ECLIPSEv5 (v6: Baseline scenario)	Species: SO_2, NO_x, CO, NMVOCS, NH_3, CO_2, CH_4, PM_{10}, $PM_{2.5}$, BC, OC, and OM; Year: 1990-2050; Resolution; 0.1°×0.1°; https://iiasa.ac.at/web/home/research/researchPrograms/air/ECLIPSEv5.html https://iiasa.ac.at/web/home/research/researchPrograms/air/ECLIPSEv6.html
HTAPv2	Species: SO_2, NO_x, CO, NMVOCS, NH_3, PM_{10}, $PM_{2.5}$, BC, and OC; Year: 2008 and 2010; Resolution; 0.1°×0.1°; https://edgar.jrc.ec.europa.eu/dataset_htap_v2
CEDS	Species: SO_2, NO_x, CO, NMVOCS, NH_3, CO_2, CH_4, BC, and OC; Year: 1750-2014/1970-2017; Resolution; 0.5°×0.5°; https://gmd.copernicus.org/articles/11/369/2018 https://zenodo.org/record/3754964#.YHgs3ujN1hE

Overview of regional emission inventories in East Asia

With an increase in demand for energy, motorization, and industrial and agricultural products, all the anthropogenic air pollutant emissions in East Asia increased drastically during these six decades. However, situations of the trends were different among countries, sources, and species and thus, spatial and temporal variation of emissions in East Asia are becoming complicated (Kurokawa & Ohara, 2020). Therefore, the development of emission inventories, focusing on the Asian region, has been conducted and continuous efforts for their updates and new development are required.

The earliest Asian emission inventory developed by Kato and Akimoto (1992), estimated SO_2 and NO_x emissions in East, Southeast, and South Asia in 1975, 1980, and 1985-1987. Akimoto and Narita (1994) provided gridded data with a resolution of 1°×1°. There are two major project-based emission inventories in Asia. Streets et al. (2003a, b, 2006) developed Asian emission inventories for the Transport and Chemical Evolution over the Pacific (TRACE-P) field campaign and Zhang et al. (2009) created its successor data for the Intercontinental Chemical Transport Experiment-Phase B (INTEX-B) project. Woo et al. (2020) developed the Comprehensive Regional Emissions inventory for Atmospheric Transport Experiment (CREATE) based on GAINS-Asia model. The Regional Emission inventory in ASia (REAS) series provided historical emission datasets in Asia: REASv1.1 for actual emissions during 1980-2003 and projected ones in 2010 and 2020 (Ohara et al., 2007), REASv2.1 focusing on the period between 2000 and 2008 (Kurokawa et al., 2013), and REASv3.2 for a long historical period from 1950 to 2015 (Kurokawa & Ohara, 2020). For East Asian countries, a lot of research-based national emission inventories have been developed. MEIC (Multi-resolution Emission Inventory for China) developed by Tsinghua University is a widely used emission inventory data base for China (Zheng et al., 2018). Zhao et al. (2013, 2014) developed recent and projected emission inventories of air pollutants in China. For Japan, several project-based emission data sets were developed such as the Japan Auto-Oil Program (JATOP) Emission Inventory-Data Base (JEI-DB) (JPEC, 2014) and emission data for Japan's Study for Reference Air Quality Modeling (J-STREAM) (Chatani et al., 2018). Pham et al. (2008) and Permadi et al. (2017)

developed emission inventories in Thailand and Indonesia, respectively. Furthermore, similar to global data, a mosaic-based emission inventory was developed in Asia. The MIX inventory, a component of the HTAPv2 inventory as described in Sect. 6.3.1 (in the main text), is a mosaic of up-do-date regional emission inventories (Li et al., 2017): MEIC for China, JEI-DB for Japan, Clean Air Policy Support System (CAPSS) inventory for the Republic of Korea (Lee et al., 2011), and REASv3.2 for default. Table 6.3 summarizes general information of major regional emission inventories in Asia. Figure 6.4 presents time series of the total emissions from EANET participating countries except for Russia during 1995-2015. Note that emissions from Russia are not included in Fig. 6.4 because majority of emissions of Russia were not from the East Asia region.

Table 6.3 General information of major regional emission inventories in Asia

Names	Descriptions
REASv3.2	Species: SO_2, NO_x, CO, NMVOCS, NH_3, CO_2, PM_{10}, $PM_{2.5}$, BC, and OC; Year: 1950-2015; Resolution; 0.25°×0.25°; https://doi.org/10.5194/acp-20-12761-2020
INTEX-B	Species: SO_2, NO_x, CO, NMVOCS, PM_{10}, $PM_{2.5}$, BC, and OC; Year: 2006; Resolution; 0.5°×0.5°; https://doi.org/10.5194/acp-9-5131-2009
TRACE-P	Species: SO_2, NO_x, CO, NMVOCS, NH_3, CO_2, CH_4, BC, and OC; Year: 2000; Resolution; 0.5°×0.5°; https://doi.org/10.1029/2002JD003093
CREATE	Species: SO_2, NO_x, CO, NMVOCS, NH_3, CO_2, CH_4, N_2O, PM_{10}, and $PM_{2.5}$; Year: 2010; Resolution; 0.1°×0.1°; https://doi.org/10.3390/su12197930
MIX	Species: SO_2, NO_x, CO, NMVOCS, NH_3, CO_2, PM_{10}, $PM_{2.5}$, BC, and OC; Year: 2008 and 2010; Resolution; 0.25°×0.25°; https://doi.org/10.5194/acp-17-935-2017

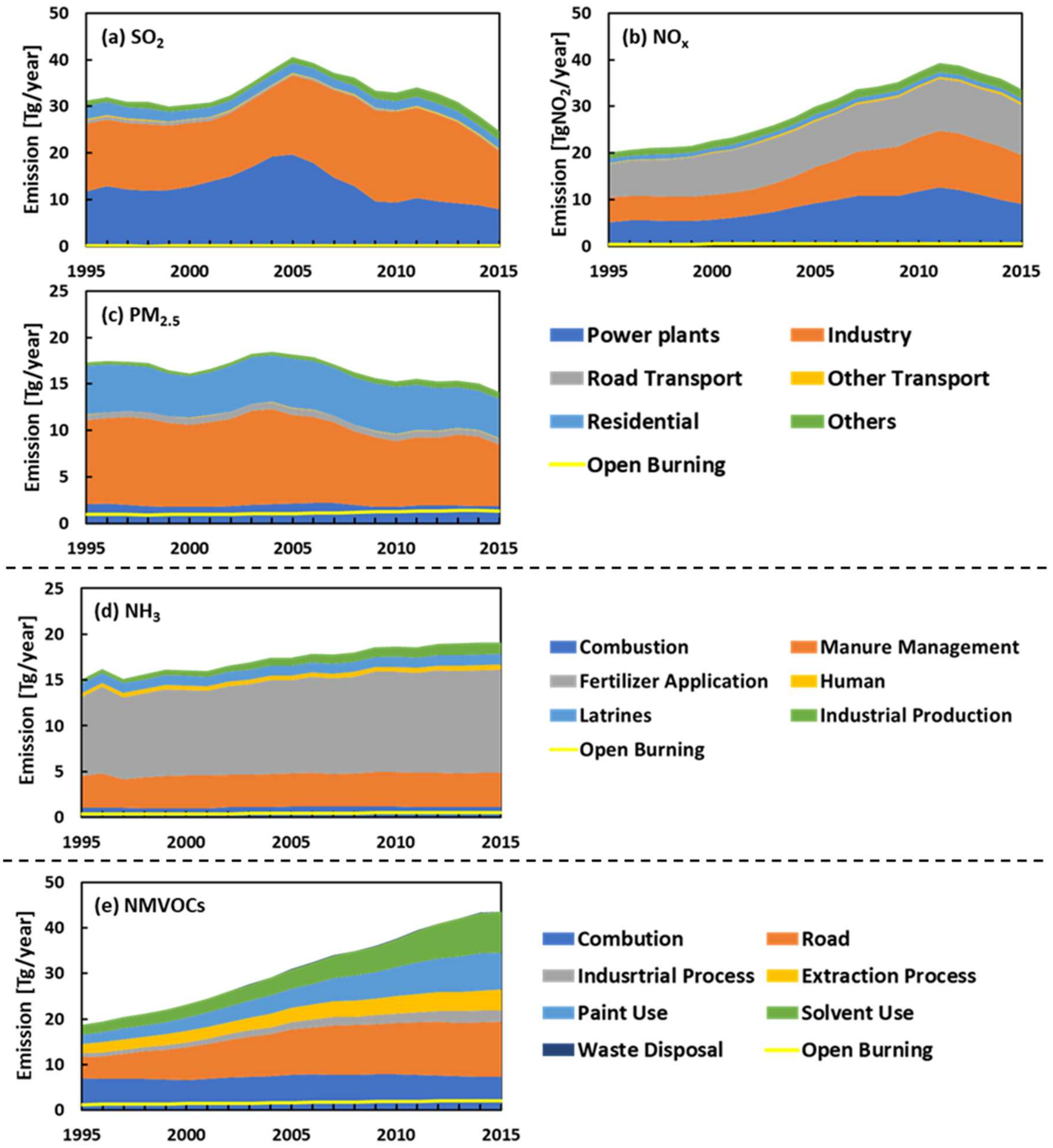


Figure 6.4. Time series of anthropogenic emissions from major sectors of (a) SO_2, (b) NO_x, (c) $PM_{2.5}$, (d) NH_3, and (e) NMVOCs in East Asia (total of EANET participating countries except for Russia) during 1995-2015. Data on emissions were obtained from REASv3.2 except for those from open biomass and waste burning, which were taken from EDGARv5.0.

Chemical transport modeling studies

In this section, studies on the CTM are reviewed with the related studies for atmospheric composition (mainly focused on $PM_{2.5}$ and O_3) and acid deposition.

Introduction of CTM studies

Air pollutants and depositions are affected by not only local sources but also the long-range transport. Observation networks such as EANET are vitally important to measure phenomena in the atmosphere. However, interpreting such phenomena from observation itself is sometimes difficult due to the complex impacts of both nearby and distant sources. CTMs, which numerically representing the processes of emissions, transport, chemical reactions, and depositions have been recognized as an important approach. The emission data is one of the important input datasets for

CTMs prepared using emission inventories (see, Section 6.3 in the main text). Another important input data is the meteorological field generated by the meteorological model. For example, the meteorological data of wind fields are used for the transport (advection and diffusion) process. The chemical reactions and depositions are internally calculated by CTMs, but the required temperature and relative humidity in the chemical reaction processes are also taken from meteorological fields. The application of CTMs varies the target phenomena. With regard to the horizontal grid resolution, CTMs can be divided into two types of frameworks: the regional CTMs and the global CTMs. Generally, the regional CTMs targeted local-to-regional scales with a few to tens kilometers and the global CTMs targeted whole over the world with coarse-resolution greater than a hundred kilometers. Because the targeted domain by the regional CTMs also affected by the global-scale ambient pollution, the lateral boundary condition for the regional CTMs is taken from global CTMs. CTMs have been developed by each researcher or group; however, the open-source models are also available currently. For example, the Community Multiscale Air Quality (CMAQ) modeling system (Byun and Schere, 2006) released by the U.S. Environmental Protection Agency (EPA) is one of the typical regional CTMs. Apart from understanding the spatiotemporal behavior of air pollutants by CTMs, CTMs can be also applied to investigate the important processes. For example, by reducing the specific emission sources and then conduct the additional simulation, the impact caused by the specific emission sources can be estimated. These applications are components of the so-called the source-receptor (S/R) analysis, and the simplest way is changing the input emission data and subsequently conducting the additional simulation using different emissions (i.e., brute-force method). Other ways are the tagged tracer method and the direct method (Clappier et al., 2017). The former uses tags for S/R analysis. In this case, the tags can be applied for the emission source itself or the source region where air pollutants formed. The latter is a method to directly calculate sensitivities for S/R analysis on along with simulation (i.e., decoupled direct method; DDM). Today, CTMs research are one of the most important approaches to foster our understanding on the air quality. The example of a feedback loop between observation and CTMs is illustrated in Figure 6.5.

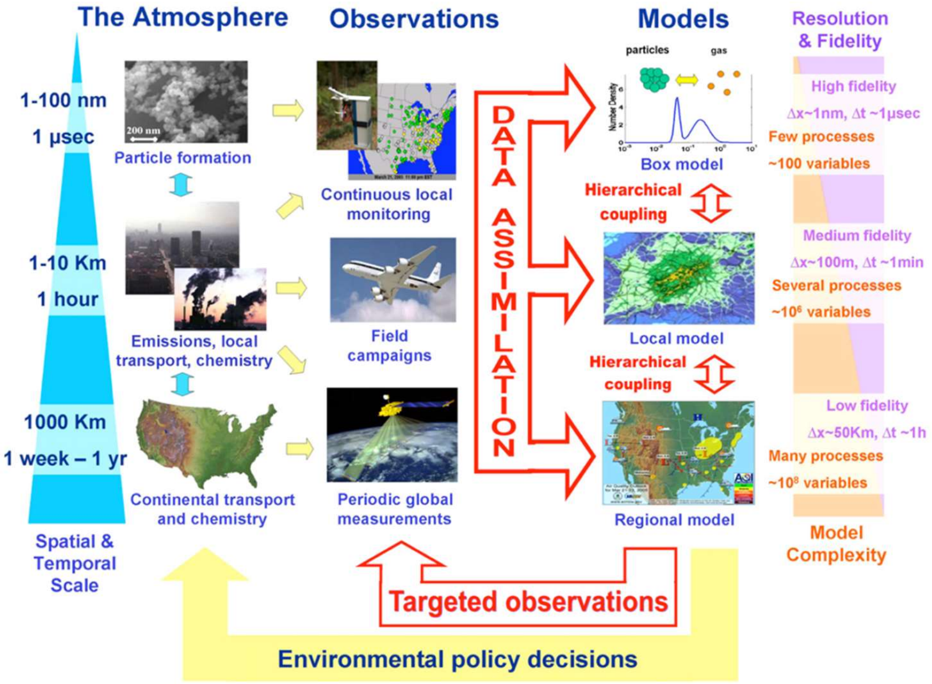


Figure 6.5. Information feedback loops between models and observations as they relate to predicting air quality. Complex CTMs incorporate chemical, aerosol, radiation modules, and use information from meteorological simulations (e.g., wind and temperature fields, turbulent diffusion parameterizations) and from emission inventories to produce chemical weather forecasts. Yellow arrows represent the data flow for predictions using the first principles. Another source of information for concentrations of pollutants in the atmosphere is the observations. Data assimilation combines these two sources of information to produce an optimal analysis state of the atmosphere, consistent with both the physical/chemical laws of evolution through the model (first principles) and with reality through measurement information. Pink arrows illustrate the data flow for dynamic feedback and control loop from measurements/data assimilation to simulation. Targeted observations locate the observations in space and time such that the uncertainty in predictions is minimized (Carmichael et al., 2008a).

Although CTMs are based on state-of-the-art science, it should be taken into account that uncertainties exist in CTMs (Carmichael et al., 2008a). An interpretation based on one CTM can cause misunderstanding of phenomena due to its uncertainty. To further our understanding of CTMs over Asia, the Model Inter-Comparison Study for Asia (MICS-Asia) has been established. Phase I was conducted in 1998–2000 (Carmichael et al., 2002), Phase II was in 2003–2008 (Carmichael et al., 2008b), and Phase III was in 2010–2020. MICS-Asia Phase I focused on sulfur (SO_2 and SO_4^{2-}) concentrations and deposition in January and May 1993. Observation datasets of sulfur were prepared by a cooperative monitoring network in East Asia (Fujita et al., 2000). A total of 18 sites located in China, the Republic of Korea, and Japan were compared with models. MICS-Asia Phase II extended its focus on O_3 and PM (Carmichael et al., 2008b). To compare the seasonality, the four representative months of March, July, and December 2001 and March 2002 were analyzed. Historically, a consistent and reliable observation dataset over the Asian region has been a bottleneck for the analysis of ambient concentration and deposition, and hence the evaluation of CTM. To address this issue, EANET has been established in 2001 and providing observation data up to now. In MICS-Asia Phase II, the EANET observation data at 43 sites were compared with models. As the lesson learned in Phase II, emission data were made to be uniform; however, participant models used different modeling domains with different horizontal and vertical structures, also using different meteorological models. In MCS-Asia Phase III, the targeted year was 2010 to fully cover the entire year. The modeling domain was coordinated, and the modeling inputs data such as meteorological field and emission data are also unified. For emission data, the topic 2 of MICS-Asia Phase III established the MIX emission inventory (Li et al., 2017). In the following subsections, scientific results from MICS-Asia activities are presented. In addition, relevant results from CTMs studies conducted over East Asia are also reviewed.

Ecological Impact Assessment Studies

The EANET has been monitoring atmospheric deposition and its effects on ecosystems since 2001. Many of sites have already accumulated the data for more than 10 years. However, the number of monitoring sites is still limited for ecological monitoring on soil, vegetation, inland water, and forest catchments. Therefore, only by the EANET data, it is difficult to assess impacts of air pollution/total atmospheric (wet + dry) deposition on ecosystems on the East Asian scale. Fortunately, impacts of atmospheric deposition on ecosystems have also been assessed by scientists in the region on the national and/or regional scales, although the number of studies were not many recently. In this section, the recent studies on ecological impact assessment in the region are briefly summarized and a future view on forest environmental risk assessment is presented.

Risk of acidification and eutrophication

In Asia, a similar integrated assessment utilizing the CL (Critical Load) concept, RAINS-ASIA, was promoted by IIASA in cooperation with the World Bank and the Asian Development Bank in the 1990s (Shah et al., 2000; IIASA, 2008). According to the IIASA website, this was the “first” integrated assessment of air pollution in Asia, which was conducted with a large international network of scientific collaborators from 20 countries in Asia. Unfortunately, the second assessment has not been conducted on the regional scale, at least by international initiatives like the EANET. Several studies were conducted by scientists on the national or regional scale, although they did not cover the entire EANET region.

Zhao et al. (2011) calculated CL exceedances and assessed soil acidification risks on the national scale in China taking account of the future scenario of PM emission control. Since base cation (BC) deposition derived from anthropogenic emissions may have an important role in neutralizing soil acidification (Larssen and Carmichael, 2000), it was suggested that acidification risks increased in some parts of the country due to a decrease in BC deposition with PM emission control. Duan et al. (2016) also emphasized the importance of BC (in particular Ca) deposition even for the risk of regional soil acidification.

Xie et al. (2020) produced an empirical-N CL map for the Northeast Asian region, including China, Korea and Japan. The empirical CLs for each vegetation type were estimated based on the literatures conducting field N fertilization experiments. According to their assessment, the area of CL exceedance in East Asia decreased by at least 4.6% during 2010 – 2015, and they concluded that NO_x emission reduction (by 15% for the same period) in China had great benefits to natural ecosystems (Xie et al., 2020). They also suggested the importance of NH_3 emission reduction.

Environmental risk assessment of ozone

Risk assessments of ozone and PM have also been conducted on the national and regional scales, although no study covered the whole EANET region. Scientists from China and Europe have recently assessed the ozone risk on Asian forests using different indices (Figure 6.6; De Marco et al., 2020). So far, concentration-based indices, such as AOT40 or W126, were widely used for the assessment of ozone impacts in Europe and the United States, respectively, although a flux-based index, PODY, which is considered a stomatal ozone uptake, has been recommended to be used. As shown in Figure 6.6, the spatial distributions of risks using the respective indices are slightly different. It is suggested that further elaboration on the indices is necessary. Also, a regional assessment for the whole EANET region is expected to be conducted in the near future.

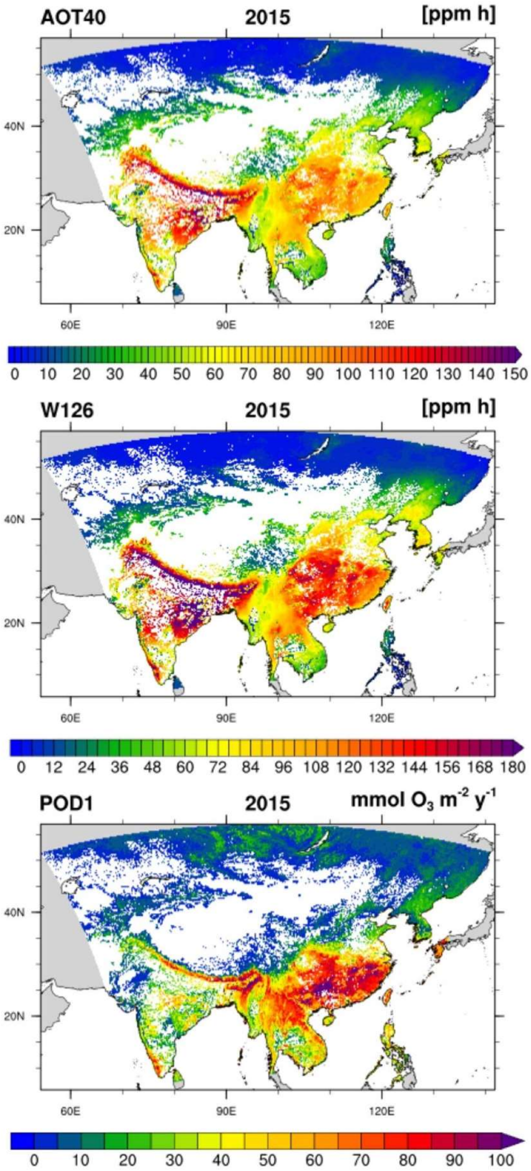


Figure 6.6. Ozone risk assessment for Asian forests estimated by three metrics (AOT40, W126 and POD1) in 2015. White color represents grid points without forest cover and grey color is used outside the domain (De Marco et al., 2020).

Health Impact studies

The health impact study on air pollution is mainly conducted by biological and epidemiological studies. Epidemiological studies are effective against public health problems such as air pollution. The epidemiology was defined as “the study of the distribution and determinants of health-related states or events in specified populations, and the application of this study to the prevention and control of health problems” (Last, 2001). The effects of air pollution in the London Smog episode and epidemiological surveys on the effects of air pollution in Yokkaichi, Japan are well known as the example of application of epidemiology to air pollution problems.

Indoor air pollution affects people indoors with gases and particulate matter that are emitted from burning fuels in stoves and kitchen cooking stoves, while outdoor air pollution is the air pollution contained in the air, which affects the person breathing it, indoors or outdoors. This chapter examines outdoor air pollution for broader considerations. The number of premature mortalities is often used as an index as an endpoint (event for which occurrence is predicted) for the health impact assessment.

The exposure-response function estimated by epidemiological studies calculates the probability of premature mortality (death shorter than life expectancy at that age) in people who have been exposed to a certain concentration of air pollutants for a certain period of time.

$PM_{2.5}$ and ozone have been focused for not only significant health effects but also characteristics as transboundary air pollutants in recent years. This chapter focuses on recent epidemiological findings of $PM_{2.5}$ and ozone in East Asia.

Impacts of $PM_{2.5}$ on human health

Previous epidemiological and toxicological studies have accumulated evidence on the health effects of particulate air pollution. Especially, fine particles with an aerodynamic diameter of less than 2.5 µm ($PM_{2.5}$) can easily penetrate deeply into lungs and cause various biological responses, such as oxidative stress and inflammation. The evidence established the causal link of exposure to $PM_{2.5}$ with respiratory and cardiovascular diseases, and lung cancer. Recently, there has also been increasing interest in the health effects of $PM_{2.5}$ on diabetes, the cenetral nervous system, and maternal outcomes.

Based on the health risk function underpinned by epidemiological evidence, the health burden attributable to $PM_{2.5}$ has been assessed. Here, research progress about the health impacts of $PM_{2.5}$ in the East Asian region are summarized.

Burden of diseases attributable to $PM_{2.5}$ and its projection in East Asia

The Global Burden of Disease study (GBD 2019 Risk Factors Collaborators, 2020) used the population-weighted annual average concentration of $PM_{2.5}$ which was estimated using the measurements of satellite-derived aerosol optical depth, ground-based monitoring stations, chemical transport models and land-use modeling. The annual mortality attributable to ambient $PM_{2.5}$ in East Asia (13 participating countries of EANET) was 1.805 million in 2019, which is slightly larger than that in 2010 (1.564 million). The annual DALYs attributable $PM_{2.5}$ also increased from 38.6 to 43.4 million years. The change in age distribution partialy contributed to in the increases in attributable mortality and DALYs from 2010 to 2019. Indeed, the percentage change in age-standardized rate for mortality and DALYs show a decreasing trend except for Japan and Mongolia (GBD 2019 Risk Factors Collaborators, 2020).

There has been increasing number of literatures estimating the country- or region- specific mortality attributable to $PM_{2.5}$. For example, in a study in Ulaanbaatar, Mongolia estimated the annual average concentration of $PM_{2.5}$ measured at the city center exceeded 75µg/m^3 during 2009-2010, which shows a clear seasonal variation. It is considered that coal and wood combustion in the low-income traditional housing area are the main sources. The estimated attributable mortality to air pollution was not low (Allen et al., 2013). These countries- and region- specific studies are useful to evaluate the impacts of specific air pollution measures although the estimates have uncertainty and further studies are warranted to estimate more accurate health impacts. According to a study (Dasadhikari et al., 2019) which estimated the mortality attributable due to $PM_{2.5}$ by sector from 2010 to 2015, in the Asian-Pacific region, agricultural, industrial, and residential sectors are the largest emission sources in 2015.

It is of interest how the burden of disease due to $PM_{2.5}$ will change in the future in which when climate change has impacts on air quality including $PM_{2.5}$ and ozone. Silva et al. (2016) estimated the attributable mortality due to $PM_{2.5}$ under various scenarios by atmospheric modeling. Because of reduced emissions, $PM_{2.5}$ concentration of 2100 is expected to decrease in all scenarios. The mortality attributable to $PM_{2.5}$ in 2100 is expected decrease in East Asia as well as other regions. However, some scenarios show increase in mortality during 2030 and 2050.

Impacts of ozone on human health

Epidemiological evidence

Health risks associated with ozone exposure can be simplified into two types: short-term and long-term. Short-term exposures usually last within days and weeks, whereas long-term take several months to years. Short-term exposures are usually paired with similar short-term health outcomes in the same temporal scale such as daily respiratory-related mortality or outpatient asthma visits. Whereas, health outcome measures from cohort studies such as lung cancer and chronic obstructive pulmonary disorder (COPD) among others, have been usually used alongside long-term exposure.

Ozone-related health burden and its future projection

In contrast to the wide spectrum of candidate health outcomes that were examined in $PM_{2.5}$-related cohort studies, ozone-related cohort studies have mostly observed a consistent finding whereby COPD mortality is associated with ozone pollution. In the recent GBD 2019 study, the authors utilized an inverse-variance weighted meta-analysis of the five cohorts, which estimated the relative risk of dying from COPD due to ozone exposure is at 1.06 (95% CI: 1.03, 1.10) (GBD 2019 Risk Factors Collaborators, 2020); visualized in Figure 6.7. Health risks below a theoretical minimum risk exposure level (TMREL) are assumed to be approaching the null association. Previous studies used a uniform distribution sourced out from the cohort studies (GBD 2019 Risk Factors Collaborators, 2020), with studies utilizing TMREL values of 32.4 ppb or 33.3 ppb (Anenberg et al., 2010; Anenberg et al., 2019).

Utilizing a global chemical transport model, Silva et al. (2016) estimated that 35% of the 493 (95% CI: 122 – 989) thousand global burdens of ozone-related respiratory mortality occurred in East Asia. Energy, residential and commercial, industry, land transportation, and shipping and aviation sectors were observed to contribute to the anthropogenic ozone emission in the region. In a similar global health burden estimation, Anenberg et al. (2018) noted that a substantial proportion of the ozone-related asthma emergency room visits occurred in Asia.

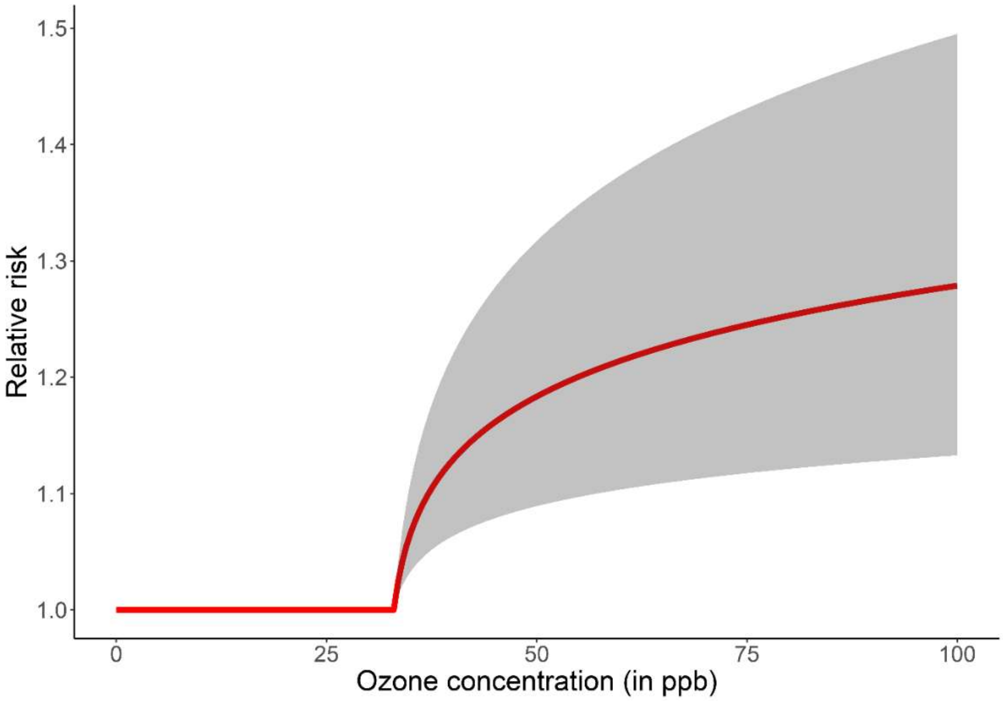


Figure 6.7. Visualization* of the concentration-response health risk function of ozone and COPD.

* Utilizing the ozone-COPD risk estimate from GBD 2019. Central estimate (in red) with 95% Confidence Interval in grey-shaded area.

A systematic analysis of the 2017 Global Burden of Disease (GBD) study revealed that ozone exposure resulted to an overall chronic respiratory disease-related, age-standardized DALY of 120.21 (45.70 – 193.34) per 100,000 population in Southeast Asia, East Asia, and Oceania (Soriano et al., 2020). In GBD 2019, East Asia (13 participating countries of EANET) has a total of 108,205 ambient ozone pollution-attributable deaths, which was 31% lower than the 2010 GBD estimate of 156,844.

Near- and far-term projections in ozone concentration have been projected to result to excesses in premature respiratory-related death. Silva et al. (2016) noted that future excess ozone-related mortality is discernable particularly in East Asia with 127,000 deaths per year under the representative concentration pathway (RCP) 8.5, which approximately constitutes 47.5% of global excess mortality in 2030 relative to 2000 concentrations. In brief, RCPs are scenarios which contain information regarding emission, concentration, and land-use trajectories (Moss et al., 2010). Further down in 2050, East Asia is projected to have the highest ozone-related mortality at 518,000 deaths per year under the RCP 6.0 pathway.

Other international initiatives on air pollution

Impacts on public and individual health and climate change are the well known scourges of air pollution. According to State of Global Air 2020, which is issued by the Health Effects Institute and the Institute for health Metrics and Evalution’s GBD project, air pollution is 4th leading risk factor for death globally, continuing to exceed the impacts of other widely recognized risk factors for chronic disease like obesity (high body-mass index), high cholesterol, and malnutrition (Figure 6.8). Furthermore, air pollution contributed to 6.7 million deaths in 2019. It was also pointed out that the findings in 2019 show that little or no progress has been made in many parts of the world. Major disparities continue to exist; air quality has improved in many high-income countries over the past several decades, while dangerous levels of air pollution persist in low- and middle-income countries.

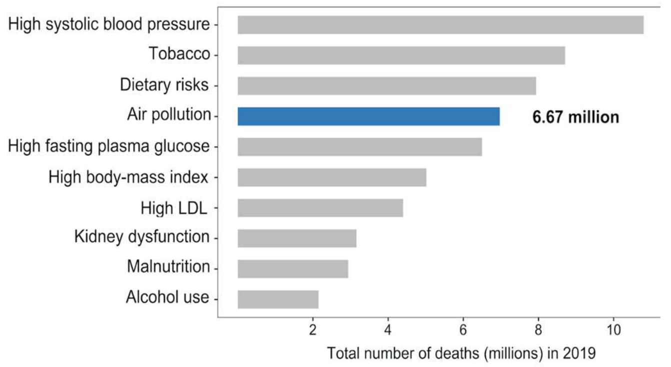


Figure 6.8. Global Ranking of Risk Factors by Total Deaths from all Causes in 2019.
Data: State of Global Air 2020

Analyzing the East Asia region, air pollution is a severe health crisis as well, and perhaps even more serious situation than the world. There are over 4 million lives jeopardizing due to exposure from outdoor and household air pollution in the Asia Pacific. The more significant numbers of the victims are the vulnerable ones: women, the elderly, and the poor.

In order to mitigate the issue of air pollution and improve air quality, not only the EANET but also lots of international organizations in the world have been conducting serial activities including monitoring air pollutants, evaluating and analyzing the monitoring data, providing efficient solutions how to improve, and so on.

In this part, the efforts of several international initiatives, Concretely, the Global Atmosphere Watch (GAW) Programme of WMO, the Task Force on Hemispheric Transport of Air Pollution (TF HTAP), Joint Research Project for Long-range Transboundary Air Pollutants in Northeast Asia (LTP) and Asia Pacific Clean Air Partnership (APCAP) will be introduced. By reviewing these initiatives’ activities, EANET can learn their advantages on how to address air pollution. Overmore, EANET is also able to find out the possibility to build or strengthen the cooperative relationship with other international initiatives.

Cross-chapter analysis of acid deposition

This part demonstrates three examples of cross-chapter analysis based on observed wet depositions, observed column density from satellite measurements, emission inventory and the results from CTM study. This part compares the long-term trends of observed wet depositions with emission inventories in EANET, NADP and EMEP for 20 years (2000-2019). The analyses in this part will provide useful information on trend factors of acid depositions in East Asia.

Trends of wet depositions and emissions among EANET, NADP and EMEP

Long-term trends of wet depositions and emissions are compared among EANET, NADP, and EMEP regions for 20 years (2000-2019). EANET region is separated to Northeast Asia (China, Korea, Japan, and Mongolia; NE Asia) and Southeast Asia (other countries; SE Asia). Figure 6.9 shows the multiple trends of concentrations in precipitation, wet depositions and emissions for sulfur and oxidized nitrogen in each region. Additionally, the relative ratios of emission and column density (satellite measurements in section 6. 2 in the main text) to the average during 2005-2015 is added for covering the emission trends in EANET region in the whole period during 2000-2019. The trends of NOx emissions are well consistent with those of column densities of NO_2 for the period during 2005-2015. For SO_2, the emission trends were almost similar to the column densities, though the latter shows a large fluctuation.

The SO_2 and NO_x emissions in SE Asia increased since 2000, while the emissions in NE Asia decreased from 2005 and 2011 for SO_2 and NO_x, respectively. Total amount of emissions for SO_2 as well as NO_x in EANET was around two times of those in NADP and UNEP, which decreased gradually since 2000. After 2015, the column density of NO_2 was almost stable in SE Asia, but slightly decreased in NE Asia. However, the column density of SO_2 after 2015 increased in SE Asia, but it was not a significant trend in the same period in NE Asia.

Wet depositions of SO_4^{2-} and NO_3^- in EANET region were higher than those in NADP and EMEP. Wet depositions of SO_4^{2-} in NE Asia decreased to one quarter for the period during 20 years in spite of the increase of SO_2 emissions before 2006. Inversely, the wet depositions of SO_4^{2-} in SE Asia increased after the last half of the 2000's in response to the increasing SO_2 emissions and became higher than that in NE Asia after 2013. The crossing point of increasing trend in SE Asia and decreasing trend in NE Asia is found around 2012. For NO_3^-, the wet depositions in NE Asia slightly decreased after 2011 according to the decrease of NO_x emissions. Wet depositions for NO_3^- in SE Asia were higher than other regions since 2001 and increased after the last half of the 2000's in response to the increase of NO_x emissions. The trends of the concentration in precipitation of SO_4^{2-} and NO_3^- are similar to those of their wet depositions in NE Asia as well as SE Asia after the middle of the 2000s.

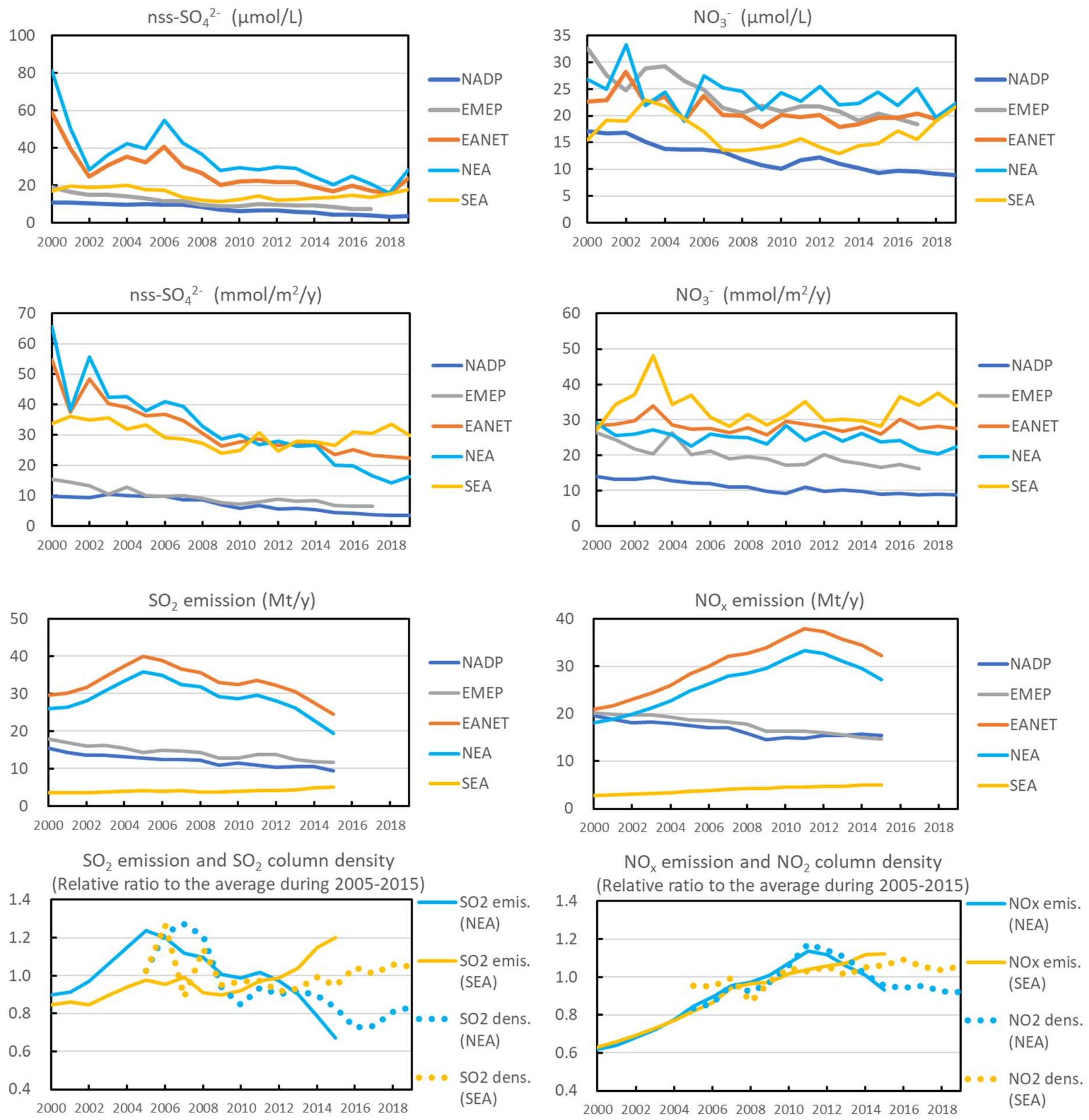


Figure 6.9. 20-years trends of observed concentrations in precipitation, observed wet depositions emissions and emissions of sulfur (left) and oxidized nitrogen (right) in EANET, NADP and EMEP regions during 2000-2019. The bottom shows the relative ratios of emission and column density (satellite measurements) to the average during 2005-2015 in EANET region. The emissions are approximately calculated as NADP = North America and EMEP = OECD Europe + Other Europe + Russia. The regional average column densities of SO_2 and NO_2 in EANET region are obtained by the area-weighted average of the column densities of each country.

7. Summary and Recommendations for Future Activities

EANET has been regularly monitoring acid deposition in East Asia since 2000. Thanks to the sincere efforts of the participating countries of Acid Deposition Monitoring Network in East Asia, the amount of sulfur dioxide and nitrogen oxides released in East Asia has been decreasing since around 2010 and it seems that the environmental impact of acid rain itself is being mitigated. However, there remain serious problems of air pollution including substances related to acid deposition such as ozone and $PM_{2.5}$ in this area. Impacts of air pollutants on human health and ecosystems are now considerable concern not only in the East Asian region but also in all over the world.

It goes without saying that acid rain is closely associated with air pollution. Therefore, overcoming air pollution will lead to the reduction of acid rain.

Data quality

QA/QC plays an important role in acid deposition monitoring to ensure that the measurement to be accurate, comparable and quantifiable quality. EANET produced four QA/QC programs (Wet Deposition, Air Concentration, Soil and Vegetation, Inland Aquatic Environment) in 2000. Those are composed of several procedure types: i) siting, ii) sampling procedures and sample handling, iii) chemical analysis, and iv) QA/QC for the measurement data at each site. Several documents regarding QA/QC have been developed by EANET to guide receiving reliable data that can be comparable among the participating countries as well as with other monitoring networks outside of the East Asian region.

The ILC project is also used for all analytical laboratories for the EANET monitoring involved. The purposes of this project are to evaluate the analytical systems through the evaluation of analytical results, analytical instruments and their operating condition and other relevant and appropriate practices. The ILC surveys were carried out 22 times, 15 times, 21 times, 20 times for wet deposition, dry deposition, soil and inland aquatic environment, respectively.

The objectives of QA/QC programs are to obtain reliable and comparable monitoring data among the participating countries in East Asian region, as well as with other networks by ensuring data accuracy, precision, representativeness, and completeness in the acid deposition monitoring. For that purpose, each participating country should continue to improve their data quality according to some reliable guidelines for QA/QC activities by using QA/QC Guidebook for Acid Deposition Monitoring Network in East Asia -2016 (https://www.eanet.asia/wp-content/uploads/2019/04/QAQC_Guidebook2016.pdf). Additionally, the National Monitoring Plan of each participating country should be reviewed every year and also should be revised, if necessary, according to the QA/QC Guidebook.

It is noteworthy that there was a remarkable improvement of the quality of analysis after collaborative research between a participating country and the NC. It must be important to continue ILC to confirm and improve the ability of the laboratories implementing EANET monitoring. The validity of this evaluation needs to be checked in an objective way, and the results should be feedbacked to each laboratory.

In order to eliminate the missing of data for a long period or to attain the DQOs for maintaining the completeness of the data, it is necessary to avoid power failure at the site due to lightning damage or unstable power supply, failure of the sampling inlet, malfunction of monitoring equipment due to deterioration over time or severe meteorological conditions, lack of financial and technical resources to repair the instruments, and so on. Timely inspection and renewal of monitoring instruments are very important. In order to improve data completeness, the alternative instruments for replacing the failed instruments should be kept in a laboratory, and lightning conductor, surge protector and/or uninterruptible power system (UPS) should be equipped in the station. Moreover, if the scope of EANET is expanded according to the decision made in IG22, addition of instruments is required.

Wet and Dry Deposition of Acidic Substances in East Asia

Under the EANET framework, there are currently 61 wet deposition monitoring sites and 54 dry deposition monitoring sites located in 2019 in 13 EANET member countries. In accordance with EANET criteria, the wet and dry deposition monitoring sites are categorized into three types: urban, rural, and remote. Sampling techniques, sample storage and analysis, data processing, QA/QC procedures of all monitoring sites and laboratories participating in EANET are conducted according to EANET's Guidelines and Manuals.

The 20-year secular changes in acid concentration sums (nss-SO_4^{2-} + NO_3^-), base concentration sums (NH_4^+ + nss-Ca^{2+}) and H^+ concentrations for representative sites are monitored in the EANET region. At the rural and urban sites, higher acid concentration sums were observed in 2000s and 2010s in China, the Republic of Korea, and Japan in Northeast Asia, in the comparison with base concentration sums. At rural and urban sites in Southeast Asian countries, the acid concentration sums and the H^+ concentrations have increased in recent years in Vietnam and Malaysia. The acid concentration sums at the remote site tend to decrease in Northeast Asia, as at the rural and urban sites. Besides, in Southeast Asia, there is no upward trend of acid concentration in remote sites. Since the situation may change periodically due to air pollution in surrounding region in Northeast and Southeast Asian countries, it is important to continue wet deposition monitoring focusing on not only acids but also bases in precipitation in EANET countries.

Based on the EANET data, geographical distribution of acids and bases wet deposition trends (in the periods of 2000-2010, 2005-2015, and 2009-2019) were analyzed. The pH shows the increasing trend in Northeast Asian and Southeast Asian countries. The concentration of nss-SO_4^{2-} had the decreasing trend in Northeast Asian countries. The increasing trend of pH in Southeast Asian countries was possibly caused by increasing cation contribution (nss-Ca^{2+} and NH_4^+).

From the comparison of precipitation chemistry among EANET, NADP, and EMEP, the pH values and nss-SO_4^{2-} concentrations showed significantly increasing and decreasing trend, respectively, in all networks from 2000 to 2019. The NO_3^- concentrations showed significantly decreasing trend from NADP, EMEP and slightly decreasing trend from EANET.

Total (dry and wet) deposition of S and N was evaluated at the sites carrying out the wet-only and the 4-stage filter pack monitoring in Russia, Mongolia, Japan, Vietnam, Thailand, Malaysia, and Indonesia. Low total S depositions were found at the remote sites located in northern inland and Pacific Ocean, and high total S depositions were found in the Japanese remote sites near Asian continent. In the high S deposition sites, the decreasing trend of S deposition was found in recent years. In remote sites, spatial and temporal trends of the total N depositions were similar to those of the total S depositions, and oxidized N depositions contributed more than half of total N deposition at higher N deposition sites.

Although the number of monitoring sites are increasing, it is still insufficient to describe the wet and dry deposition status of the whole area of each country. It is desirable to arrange denser sites according to the capabilities of each country. In addition, it becomes important to evaluate the obtained data by utilization of modeling analysis as well as comparison with the data reported by other networks such as NADP, EMEP. There are wet deposition monitoring data in East Asia for more than 20 years. Thus, such long-term variation of regional deposition should be analyzed in comparison with the change of emission amount of air pollutants in the region for getting insight into the effect of domestic and transported air pollution and their control measures.

Dry deposition was estimated by the inferential method based on Technical Manual for Dry Deposition Flux Estimation in East Asia (EANET, 2010). Namely, current method for estimating dry deposition is based on observations of gaseous and particulate S and N components and meteorological elements. As a way to compliment limited number of data, chemical transport model is effective to estimate the deposition velocity. NADP has developed a hybrid approach for estimating dry deposition using monitoring data and modeled data from chemical transport model

(Schwede and Lear, 2014). These days model analyses are indispensable to evaluate the field data and to analyze the regional effects of air pollution and acidic deposition. Utilization of model analyses is strongly recommended for estimation of dry deposition.

Gas and Aerosol Pollution in East Asia

EANET has conducted continuous monitoring in various countries in 20 years and the number of gas and aerosol monitoring sites have been increased each year. There were 54 air concentration monitoring sites operated in the EANET network as of 2019. The measuring components, which are SO_2, NO, NO_2, NOx, $PM_{10}/_{2.5}$, O_3, HNO_3, HCl, NH_3, and Particulate Matter Components (PMC), varies depending on the methodology at each site.

The spatial distribution of SO_2 in Russia and Mongolia varied by the influence of population distribution, as well as the effect of heating and fuel composition such as coal burning. After continuous improvement, SO_2 concentrations in China have been significantly reduced. The SO_2 concentrations at all sites in Japan are very low for a long time. Except for Indonesia, SO_2 concentrations in Southeast Asian countries are generally low.

The Republic of Korea had the highest nationwide O_3 5-year average concentration, and Japan reported the next highest nationwide averaged O_3 concentration. Mongolia had the highest NOx 5-year average concentration, and Philippines reported the next highest.

Mongolia had the highest $PM_{2.5}$ 5-year average concentration at Ulaanbaatar. Jakarta of Indonesia reported the next highest nationwide averaged $PM_{2.5}$ concentration. The $PM_{2.5}$ concentration in Hoa Binh of Vietnam is close to that of Jakarta of Indonesia. Ulaanbaatar of Mongolia also has the highest PM_{10} concentration level with a 5-year mean concentration. The PM_{10} concentration levels in China, Korea, the Philippines, and Thailand were similar.

There are various NH_3 5-year average concentration distributions in East Asia. The spatial distribution of NH_3 in Philippines, Malaysia and Mongolia has a large variation. The 5-year average concentration of NH_3 in the monitoring stations of China and Indonesia is relatively high. The 5-year average SO_4^{2-} concentration was relatively high at Hongwen of China and a site in Vietnam. The 5-year average NO_3^- concentration was relatively high at Hongwen of China and Vietnam. The 5-year average NH_4^+ concentration was relatively high at Hongwen of China and the Republic of Korea.

Substances related to acid deposition, such as precursor gases of acidic substances, ozone as one of the products of atmospheric chemical reactions to form acidic substances in air, and particulate matters as the products of neutralization reactions of acidic substances in the atmosphere, are also the targets of EANET from the early stage. Climate change is obviously not the target of EANET, but air pollutants which can relate to both acid deposition and climate change will be possible targets of EANET in the future. At present, however, acid deposition is still a big environmental problem in EAST Asia. We need to face to the improvement of monitoring activities for acid deposition and related substances.

The shift to electric vehicles and the regulations on the use of low-sulfur fuel in ships (started in January 2020) are expected to continue to reduce SO_2 and SO_4^{2-}, but it is important to continue monitoring and aim for further environmental improvement. It is expected that the use of fossil fuels will decrease rapidly toward a decarbonized society, and the amount of NOx emissions will be further reduced. Changes in NOx concentrations in air can affect the surface ozone concentrations, too. Thus, changes in NOx emissions need to be monitored based on an accurate grasp of NOx and NO_3^- depositions.

Because of the advance of agriculture in Asia, emission of ammonia and ammonium is increasing. Ammonia is an important atmospheric basic-compounds which can neutralize acids. In order to accurately evaluate the concentration of PM, accurate integration of ammonium salt emissions is essential, and more accurate monitoring of NH_3 and NH_4^+ is needed.

Ozone was intensively monitored in the urban area so far from a point of view of the effects on human health. More extensive monitoring of ozone is needed from the viewpoints described above. Ozone is also a secondary pollutant produced in the air by the photochemical reactions of NOx and VOCs; not a pollutant emitted directly by human activity. Thus, it is quite important to utilize simulation-model analyses in addition to field monitoring to know the mechanisms of pollution and to estimate its spatial and temporal distribution. And for the purpose of verification, remote sensing technology including satellite measurements should be taken into view in the near future.

In order to analyze the health effects of PM and to evaluate the route of long-range transport, it is necessary to monitor the chemical components of PM. In addition, dust and sand storm in north-east Asia and haze in south-east Asia should be included in PM monitoring.

Impacts on Ecosystems in East Asia

The EANET has been monitoring various ecosystem components, such as soil, forest vegetation, and inland water, to assess impacts of atmospheric deposition on ecosystems since 2000 when the regular-phase activity started. Additionally, the EANET has been promoting a catchment-scale analysis considering biogeochemical cycles in forest ecosystems since 2010.

Soil monitoring has been conducted at 31 sites from 10 countries including the surveys in 1999 during the preliminary phase. The soil monitoring data since 2000 to 2019 showed the diverse soil characteristics of EANET sites, which were controlled by various factors, such as soil formation process and climatic conditions.

Monitoring on forest vegetation has been conducted at 24 sites in 8 countries. The vegetation monitoring includes implementations of the comprehensive forest survey, tree decline survey, and understory vegetation survey, but the content and frequency of surveys vary widely.

Monitoring on inland water chemistry has been conducted in 20 sites from 11 countries. Chemical properties, such as balance of ions, concentration levels, and seasonal changes, varied among the monitoring sites.

Long-term data at two forest catchments, Lake Ijira catchment (IJR) and Kajikawa catchment (KJK), suggested that the catchment biogeochemical cycles have sensitively responded to the recent declining of atmospheric deposition (in particular, SO_4^{2-}) resulting in phenomena indicating recovery from acidification. New catchment analysis sites, Komarovka River (KMR) and La Mesa Watershed (LMW), may also contribute to further understanding of ecological response to atmospheric deposition and climate change.

In order to detect the effects of atmospheric deposition on forest ecosystems, the following points improving the monitoring program and assessment can be recommended:

- To obtain data with low noise factors, such as site disturbance (including local anthropogenic disturbance)
- To improve the system for better data acquisition regarding the control or understanding of anthropogenic disturbances (deforestation, non-execution of monitoring, site changes, etc.)
- To implement monitoring and model analysis considering the differences in time constants, since the time response of ecosystems to atmospheric deposition is different.
- To continue monitoring ecosystems and model analyses using the monitoring results even after the atmospheric deposition has decreased sufficiently.

Especially in the catchment area, it is expected that these combined effects of atmospheric deposition and climate change will be observed as output. Therefore, in order to analyze the effects on forest ecosystems, beside the promotion of catchment area analysis in each region, the following point can be recommended additionally:

- To establish a system that may reset of monitoring sites on soil and forest vegetation in consideration of catchment areas.

These days the effects of atmospheric pollutants, such as ozone and PM, are assumed more serious to forest trees than acid precipitation. Therefore, under the expanded scope of the EANET, ozone and PM should be monitored more extensively to evaluate their risks to the forest ecosystems.

As for inland water environment, although changing patterns of ion concentrations in lakes/rivers in some monitoring sites were well synchronized with those of atmospheric deposition at the nearest monitoring sites, there remains several technical issues that need to be improved.

According to the recommendations described in PRSAD3, the following subject remains as the regional network:

- The number of the EANET sites for ecological monitoring is too small to cover the entire region. A huge variety of forest ecosystems are distributed throughout the region with diversity of climate, geology, vegetation, soil, and human activity. Therefore, a scale-up approach is crucial for a regional risk assessment. Efforts should be made to assess the regional risks of acidification on the East Asian scale to continue the EANET monitoring in the high-risk areas more effectively.

Thus, they are still valid as future recommendations of the EANET. These points should be addressed as quickly as possible.

Relevant Studies on Atmospheric Environment Assessment in the EANET Region

In addition to the effort of EANET, there have been many research activities and organizational initiatives focusing regional air pollution in East Asia as well as in global and hemispherical perspectives. In this part, such research outputs and initiatives in the last few years are reviewed in order to provide useful information on present status of acid deposition and regional air pollution. The covered topics are atmospheric observation including satellitte observation, emission inventory, chemical transport modeling (CTM), ecosystem and human health impact studies, which are not necessarily directly related to EANET, but would be useful for understanding air pollution and acid deposition in East Asia.

Figure 7.1 shows the overall structure of Chapter 6 in the main text. In the chapter, the observation studies, emission inventory studies and CTM are systematically reviewed. In the following part, the ecosystem impact assessment studies and the human health impact studies are reviewed, respectively. In the final part, other international activities in EANET region are introduced and the combined analysis of long-term trends of acid deposition and gas/particles based on ground and satellite measurements and emissions is carried out.

This section does not intend to provide comprehensive review, but the atmospheric observational studies focusing on field campaign and satellite measurement as well as studies on acidification and nitrogen leaching in forest catchments, the Model Inter-Comparison Studies for Asia (MICS-Asia) utilizing EANET data for validation and analysis, ecosystem impact assessment studies on the national and/or regional scales and impacts of $PM_{2.5}$ and ozone on human health are introduced with higher priority. Additionally, some original analysis are conducted in the satellite observation, emission trend and long-term trends of acid deposition and air quality.

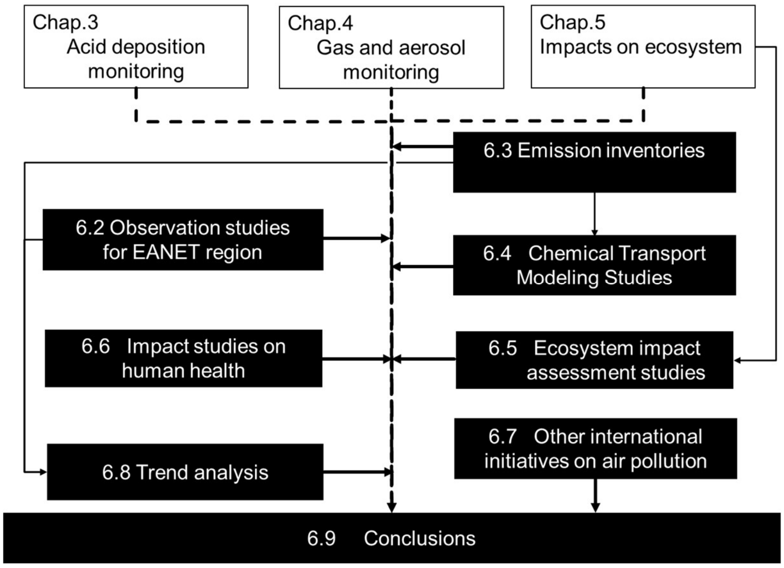


Figure 7.1. Overall strucrure of Chapter 6 (in the main text).

Emission Inventories

East Asia is now the regions of the world with the large air pollutant emissions. Therefore, estimation of emissions in East Asia is important not only for understanding local air pollution, but also for large scale air pollution and global climate change.

Members of NC of EANET have been collaborating with researchers from various research institutes to develop and improve mosaic emission inventories composed of reliable national and regional inventories under the framework of the Model Inter-Comparison Study for Asia (MICS-Asia) project. EANET has provided some assistance with this. As continuous efforts for elaboration of emission inventory are necessary, it is recommended that research activities (e.g., survey of detailed activity data and emission factors) and capacity building for their activities are required in EANET participating countries. In addition, the national emission inventory officially supported and maintained by each participating country is preferable for the air quality management of this area. Therefore, the enhancement of capabilities of each participating country on the development of the national emission inventory should be encouraged through the research and capacity building activities.

Verification, improvement, and updates of emission inventories based on ground and satellite observations, chemical transport modeling, and inverse modeling are essential to reduce uncertainties. This work requires strong collaborative and integrated research among emission inventory developing, monitoring, chemical transport modeling, and inverse modeling groups. These activities will help future QA/QC of national emission inventory in EANET participating countries.

Modeling activities

Chemical transport model (CTM) is an important and powerful tool to grasp the surface concentrations, spatial distribution of deposition, and temporal variation of air pollutants. Therefore, modeling studies based on CTMs validated through EANET monitoring data are strongly recommended to achieve detailed and easy-to-read analysis of spatial and temporal variability in PM, ozone, acid deposition and other monitoring species. However, attention should be paid to the existence of uncertainty existing in CTMs. In order to avoid misinterpretation based on a single CTM, MICS-Asia has been established and it is still in operation. In MICS-Asia, further efforts are planned such as analyzing more specific air pollution events typically occur in East Asia and utilizing model simulated results for impact assessment of ecosystem and human health.

A significant advance has been made in acid deposition modeling in the past 20 years in East Asia. Reliable model analysis results were reported from Southeast Asia, too, in recent years. However, there is still differences among the models. In the recent MICS-Asia studies the participated models performed well for SO_4^{2-} and total ammonium (NH_3 and NH_4^+), but estimation of total nitrate (HNO_3 and NO_3^-) still has uncertainties. Regional scale evaluations of acidification and nitrogen leaching should be still kept going. Collaboration of atmospheric modeling and impact studies should be maintained to make clear the relationship between atmospheric deposition and its impacts to ecosystems. It is recommended for EANET to encourage collaborative works between researchers of different scientific fields such as atmospheric chemistry and ecosystems.

Health Impact studies

Impacts of acid deposition has been mainly investigated from a point of view of impacts on plants. The main substances affecting public health are nitrogen oxides (NOx), sulfur dioxide (SO_2), atmospheric ozone (O_3), and PMs. Epidemiological studies have indicated that symptoms of bronchitis for children was associated with long-term exposure to nitrogen dioxide (NO_2). SO_2 can affect the respiratory system and functions of the lungs, and causes eye irritation. Excessive O_3 exposure can cause breathing problems, aggravate asthma and reduce lung functions. In recent years, fine particles ($PM_{2.5}$) are of significant concern, as these tiny particles penetrate into the lungs, affecting both the respiratory and vascular systems.

Even if there is still uncertainty at the moment, air pollution measures are required considering the adverse health effects at present and expected in the near future in East Asia. During the past 20 years, participating countries of the EANET have made great efforts to reduce emissions of acid deposition and related pollutants through methods such as effective policies, advanced technologies and best practices. Therefore, assessment on effectiveness of various measures contributing to emission reduction of acid deposition and related air pollution considering the adverse effects caused by multiple air pollutants is necessary, which could figure out applicable measures to further improve air quality.

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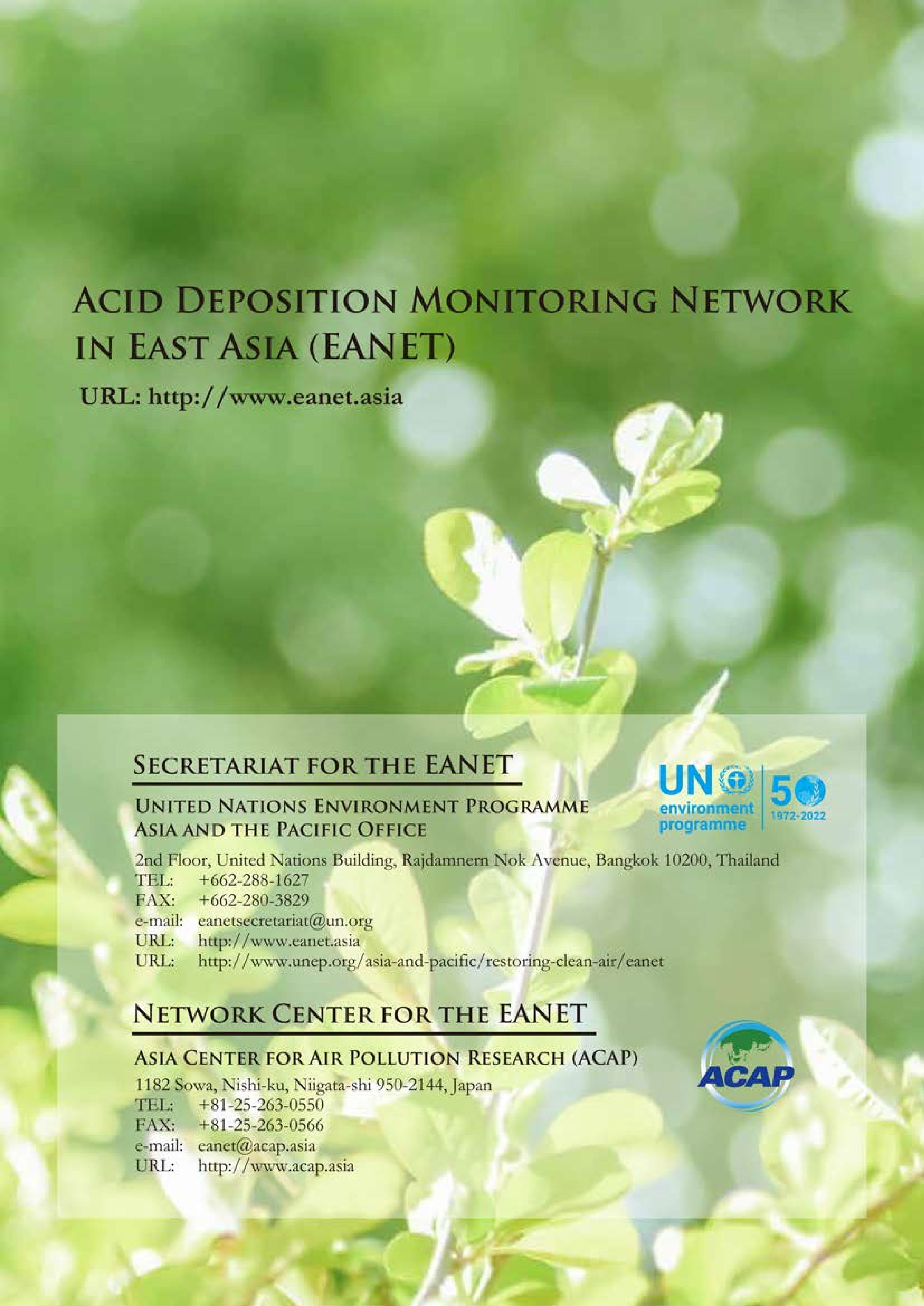

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