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

PART III EXECUTIVE SUMMARY

Acid Deposition Monitoring Network in East Asia
(EANET)

Preface

This is the second *Periodic Report on the State of Acid Deposition in East Asia* describing the outcomes of five years of EANET monitoring activities (2005 – 2009). The report is published in two parts – Part I: Regional Assessment, and Part II: National Assessments. Part I outlines the activities of EANET and provides an assessment of the state of acid deposition. It includes information on related air pollution issues affecting the region based on data acquired from both the network and external literature, and future directions and possible impacts of pollutants on atmospheric environment and ecosystem elements such as soil/vegetation and inland aquatic systems. Also highlighted are quality assurance and quality control (QA/QC) activities designed to ensure reliable data and consistent research activities to be carried out jointly by the EANET community to improve understanding of acid deposition processes in the region. Part I concludes with an outline of future activities aimed at fostering a more integrated approach to acid deposition management in the region. For the formulation of this report, a drafting committee comprised of experts from the participating countries was established to lead the scientific evaluation of monitoring data and the achievements of EANET in its first five years of the Strategy on EANET development (2006 – 2010) with the goals of producing 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, the contents of Part II are not covered in this executive summary.

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

EANET data were exclusively used in the report except when reviewing related studies in EANET countries.

Summary for Policy Makers

Summary of Key Points

The regular phase activities of the Acid Deposition Monitoring Network in East Asia (EANET) was started in 2001 with 10 East Asian countries, and later expanded to also include Cambodia, Lao PDR and Myanmar. Today, it remains the region's most important group for sub-regional cooperation in acid monitoring. The 2010 "Instrument for Strengthening the Acid Deposition Monitoring Network in East Asia" represented a milestone in the history of the organization, and has so far been signed by 9 of the 13 participating countries.

EANET successfully completed its Work Program and Budget in 2011. The most significant achievement of this was the finalization of the Second Periodic Report on the State of Acid Deposition in East Asia (PRSAD2), which provides an assessment of acid deposition conditions and an outline of related air pollution issues affecting the region.

EANET's monitoring work has demonstrated that acid rain remains prevalent across East Asia. The annual average pH of rainwater is lower than 5.0 (the threshold for acid rain) at 60% of monitoring sites, and values of less than 4.6 have been recorded in several locations. Sulphric acid (H_2SO_4) remains the primary contributor to acid rain across the region, therefore, the regional sulphur emission control is still needed to be improved. The control of nitrogen emission is also important in the region in which contribution of nitric acid (HNO_3) to acid rain is almost equal to that of sulphuric acid.

Despite continued acidification in the region, the impact of acid deposition on eco-system functions still appears limited. No decline in tree growth or in the number of species in understory vegetation has been observed during monitoring, and overall forest functions and structures apparently remain sound. Tree decline symptoms observed at some sites were generally considered to stem from pest infestation as a direct cause.

Ozone (O_3) concentration is monitored in Japan, the Republic of the Republic of Korea, Thailand and Russia. Common seasonal variations are observed at these sites – highest values in spring, lowest values in summer, and second-highest values in autumn. Spring maximum concentrations at most sites in Northeast Asia exceed a monthly average of 50 ppb, and are even higher than 60 ppb at some sites. Monthly average O_3 concentrations from 2005 to 2009 were higher than those for the previous five-year period (2000 to 2004) at many sites. Accordingly, it may not be possible at this stage to exclude the direct effects that ozone and other air pollutants exert on trees as contributing factors. The increasing risk that ozone exposure poses to human health and agricultural crops/forest trees remains a source of concern for the region.

A declining trend in lake/stream water pH with a corresponding increase in sulphate concentration was also observed at several sites during the same period. The results of soil and inland water monitoring in some regions showed symptoms of nitrogen saturation/eutrophication due to excess deposition of nitrogen species (nitrates and ammonium), which is a further source of concern. Deposition loads of S and N were very high in these areas.

To clarify the relationship between atmospheric deposition and changes in deposition in soil and inland water, EANET introduced a technique known as catchment-scale analysis, which is based on integrated monitoring of atmospheric deposition, vegetation, soil and inland water, to clarify the ecological impacts of acidification in East Asia.

EANET plays a pivotal role in the global framework for monitoring acid deposition-related chemical species, as its measurements cover areas of exceptionally rapid industrialization and land use change. This makes the organization significant not only regionally but also globally in

elucidating the consequences of atmospheric pollution. In this context, it is important that governments of the participating countries commit to supporting ongoing monitoring regarding the deposition of acid and other related atmospheric air pollutants, and also to extending the assessment of acid deposition monitoring to include other relevant components such as ozone and particulate matter (PM).

There are strong links between acid deposition and climate change. Air pollutants causing acid deposition (such as ozone and aerosols) contribute to climate change, while climate change affects acid deposition through influences such as precipitation variations. Accordingly, extending the assessment of acid deposition conditions to include other relevant air pollutants and climate change should be coordinated in order to improve EANET's treatment of emerging issues.

After more than 10 years of operation, the participating countries now have a basis for considering measures to strengthen the policy relevance of the network's mandate. Accurate monitoring of acid deposition and assessment of the consequences of acid rain on the environment provide opportunities for action. As scientific understanding of the consequences and sources of such deposition becomes more reliable, EANET should consider ways to address the origins of harmful emissions by reaching out to policy developers and decision makers with scientific knowledge and policy advice.

In this sense, the scientific community's role in communicating such information is vital, but it must be presented in a way that can be understood by the general public. Reaching out to society at large by promoting public awareness and common understanding of how atmospheric pollution affects human wellbeing and nature is expected to support policy development toward environmental sustainability in the future.

EANET's scientific and technical capacity has developed significantly during the past 10 years of EANET. In order to further strengthen these areas, the network should consider ways to promote collaborative efforts among participating countries and to improve cooperation with organizations outside the region. A scientific review of the present status of the atmospheric environment in East Asia would also be worthwhile to support discussions on expanding the scope of EANET. The promotion of research activities, including work on modeling and emission inventories, is another possible target for EANET toward further development of the network.

Summary of Technical Evidence

Concentrations in Rainwater and Wet Deposition

The five-year average pH of rainwater for all EANET sites ranges from 4.4 to 6.2. At 26 of the 42 sites (62%), pH values lower than 5.0 (proposed as the threshold value for acid rain) have been recorded. In terms of acid rain causes, sulphuric and nitric acid produced from anthropogenic emissions of sulphur dioxide (SO_2) and nitrogen oxides (NOx), respectively, have been identified as major acidifying species. However, organic acids produced from biogenic emissions are also known to have a significant effect in areas distant from anthropogenic sulphur and nitrogen sources, particularly in tropical and sub-tropical regions. Five-year average rainwater pH values lower than 4.6 have been recorded at specific urban sites in Malaysia, China (Chongqing) and Indonesia, and at several rural/remote sites in the Republic of Korea and Japan. In contrast, average rainwater pH values higher than 6.0 have been seen in China (Xi'an) and Mongolia (Ulaanbaatar). These high values are due to increased contribution of alkaline species such as ammonia (NH_3) from agriculture and calcium carbonate in soil dust, respectively.

In the EANET region, anthropogenic non-sea salt sulfate (nss-SO_4^{2-}) concentration in rainwater is equal to or higher than nitrate (NO_3^-) concentration, meaning that SO_2 emissions are still the major contributor to acid rain. This is particularly true in inland China, where the contribution of NO_3^- is less than 20% of the total for nss-SO_4^{2-} and NO_3^-. In contrast, NO_3^- is relatively high at one Malaysian urban site and a number of sites in Thailand, where NO_3^- accounts for more than 40% of the total for nss-SO_4^{2-} and NO_3^-. Since sulfur emission control is a precedent to nitrogen control in general, the relative significance of nitrogen as an acidifying species in rainwater is predicted to increase in the future in East Asia.

For alkaline species, the contribution of NH_4^+ is higher than that of nss-Ca^{2+} (non-sea salt calcium emitted from terrestrial sources) at many EANET sites (more than 90%) except those influenced by large amounts of soil dust caused in particular by yellow sand. For nitrogenous species, it should be noted that the concentration of NH_4^+ is higher than that of NO_3^- at 78% of sites. Particularly high concentrations of NH_4^+ in rainwater have been observed at some inland sites in China (Xi'an and Chongqing). It should be noted that although NH_4^+ is alkaline in the atmosphere, it turns to NO_3^- in soil and releases H^+ after deposition, thereby contributing to acidification.

Five-year average wet deposition values were calculated by multiplying rainwater ion concentrations by precipitation amounts. Annual precipitation in East Asia ranges from over 2,000 mm in most of Southeast Asia and Japan to less than 500 mm in Russia, Mongolia and China (Xi'an). The three highest totals of hydrogen ion (H^+) deposition are seen in Malaysia (Petaling Jaya), Japan (Ijira), and Indonesia (Jakarta). Deposition of nss-SO_4^{2-} is the highest in China (Chongqing), and that of NO_3^- is the highest at an urban site in Malaysia (Petaling Jaya), reflecting high concentrations in rainwater and precipitation amounts.

In terms of ecosystem impact, effective hydrogen ion deposition (H^+_{eff}) based on the sum of H^+ and NH_4^+ is more significant in soil acidification, since NH_4^+ should be treated as an acidifying species in soil. Higher five-year average deposition values for H^+_{eff} are observed at most Southeast Asian urban sites and in Chongqing, China.

In addition to acidification, eutrophication (nitrogen saturation of soil and water) is also a concern from the viewpoint of ecosystem impacts in East Asia. To determine the risk of nitrogen saturation, the sum of NO_3^- and NH_4^+ depositions (ΣN) were evaluated. Higher depositions of ΣN were observed in Southeast Asian urban areas, inland China and Japan (Ijira).

Even though data for the whole period from 2000 to 2009 are available, statistics covering a 10-year period are generally considered insufficient to support discussion regarding clear trends of

ion concentration in rainwater and wet deposition. Nevertheless, statistically significant cases are seen at some sites for certain species. Significantly increasing trends in both concentration and deposition are seen at the Bangkok site for NH_4^+, which supports increasing trends of ΣN and H^+_{eff}. Some sites in Xiamen and Chongqing in China show increasing trends of NO_3^- concentration. Meanwhile, decreasing trends of nss-SO_4^{2-} in deposition and/or concentration are seen at urban sites in Petaling Jaya in Malaysia and Jakarta in Indonesia.

Concentrations in the Atmosphere and Dry Deposition

Atmospheric concentrations of gaseous pollutants (SO_2, NOx, HNO_3, NH_3 and O_3) and aerosol ion components/mass concentration (PM_{10}/$PM_{2.5}$) are also monitored by EANET. In Northeast Asia, amounts of the gaseous and aerosol species SO_2, HNO_3, NH_3, SO_4^{2-}, NO_3^- and NH_4^+ at several urban and rural sites in China are high compared to those of other areas. Among the more remote sites, amounts of gaseous components are relatively high at Jeju in the Republic of Korea and at Terelj in Mongolia, while amounts of aerosols are similarly high at Jeju and at Oki, Yusuhara and Hedo in Japan. In Southeast Asia, amounts of gaseous species are higher in Petaling Jaya, Metro Manila and Bangkok than in other areas, and amounts of aerosol components are notably high in Hanoi. At more remote sites, Nakhon Ratchasima and Kanchanabri in Thailand show higher concentrations of gaseous and aerosol components. The mass concentration of PM_{10} is observed in China, Japan, the Republic of Korea and Thailand. Values are the highest at urban and rural sites in China, followed by those in Thailand and at Jeju in the Republic of Korea. At most Japanese sites, average monthly PM_{10} concentrations from 2005 to 2009 were lower than those of the previous five-year period (2000 to 2004). $PM_{2.5}$ is monitored only at Oki and Rishiri in Japan among EANET sites.

Ozone (O_3) concentration is monitored in Japan, the Republic of Korea, Thailand and at one Russian site (Mondy). Common seasonal variations are observed at these sites – highest values in spring, lowest values in summer, and second-highest values in autumn. Spring maximum concentrations at most sites in Japan exceed a monthly average of 50 ppb, and are even higher than 60 ppb at some sites. Monthly average O_3 concentrations from 2005 to 2009 were higher than those for the previous five-year period (2000 to 2004) at most sites in Japan.

Five-year mean annual dry deposition amounts for sulphur and nitrogen compounds were calculated using atmospheric concentration data for gases (SO_2, HNO_3, NH_3) and aerosols (SO_4^{2-}, NO_3^- and NH_4^+), and the deposition velocity for each species was estimated from meteorological and land-use data. Estimates were made only for Japanese sites due to issues regarding the availability of hourly meteorological and land-use data. The ratio of dry deposition to wet deposition ranged from approximate parity to less than 30%. Higher total (dry and wet) deposition amounts of nitrogen compounds were observed at Ijira and Banryu, which are classified as rural and urban sites, respectively. Total nitrogen depositions were lower at Rishiri and Ogasawara, which are located far from major nitrogen oxide emission sources. Since the atmospheric lifetime of nitrogen compounds is generally shorter than that of sulphur compounds, the regional characteristics of total nitrogen compound deposition amounts are considered to be at greater influence from local emission sources.

Impacts on Ecosystems

Atmospheric deposition may cause acidification and/or eutrophication of ecosystems. An increasing trend of soil pH has been observed at several sites (e.g., Los Baños in the Philippines and Kanchanaburi in Thailand) over the last decade, while a declining trend is seen at one site (Imsil in the Republic of Korea). However, pH changes do not correspond to those of acid and/or base cations, suggesting the presence of other factors such as the influence of organic matter. Nutrient imbalance may also be a cause of changes in the chemical properties of soil.

No decline in tree growth or in the number of species in understory vegetation was observed during the reporting period from 2000 to 2009, and overall forest functions and structures apparently remain sound. Tree decline symptoms observed at some sites were generally considered to stem from pest damage, although the direct effects of air pollutants such as ozone on trees in the region are still unclear.

A significant decrease in soil C/N content was observed at several sites (Jinyunshan, Xiaoping and Zhuxiandong in China, Bogor in Indonesia, and Banryu in Japan). Simultaneously, the NO_3^- concentration of inland water increased at points in the vicinity of these areas (Jinyunshan Lake, Zhuxiandong Stream, and Patengang Lake near Bogor). Excess NO_3^- in soil, which is indicated by low C/N values, may flow into streams and lakes, leading to increased NO_3 concentrations in inland waters. It may be that high N deposition levels in these areas caused the phenomenon described above. This status in forests is referred to as nitrogen saturation, and terrestrial eutrophication, soil acidification and fresh water eutrophication may occur in stages, causing adverse impacts on ecosystems.

A declining trend of pH in lake/stream water was observed at several sites (Jinyunshan Lake, Xiaoping Dam and Jiwozi River in China, Patengang Lake in Indonesia, and Semenyih Dam in Malaysia) during the period from 2000 to 2009 along with a simultaneous increase in SO_4^{2-} concentrations. This acidification may be related to leaching of SO_4^{2-} from ecosystems. As mentioned above, an increasing trend of NO_3^- concentrations was also observed at one of these sites (Jinyunshan Lake).

Regular monitoring of soil and inland water showed certain symptoms of eutrophication and/or acidification in several areas as mentioned above. In fact, deposition loads of S and N were very high in these places, although no clear link with atmospheric deposition was identified. Accordingly, catchment-scale analysis was also promoted to clarify the relationship between atmospheric deposition and changes in ecosystems and to elucidate the related mechanisms. Both qualitative and quantitative analysis were performed in regard to the results of integrated monitoring, and these included evaluation of atmospheric deposition, vegetation, soil and inland water. More precise evaluation may be carried out in the near future based on the promotion of regular catchment-scale monitoring.

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1. Introduction

EANET has made significant progress and important achievements in data monitoring thanks to the efforts of participating countries. This is the second edition of the *Periodic Report on the State of Acid Deposition in East Asia*, which continues coverage of results relating to the period from 2005 to 2009. The data collected during the first 10 years of EANET's existence enable more precise and definite conclusions to be drawn on temporal and spatial variations in atmospheric deposition. Most of these achievements were made under the *Strategy on EANET Development (2006 – 2010)*, which covered almost all network activities for coordination among the participating countries, the Secretariat and the Network Center.

Inter-laboratory Comparison Projects have been implemented annually to improve quality assurance and quality control (QA/QC) starting with wet deposition. QA/QC plays an essential role in acid deposition monitoring and in other types of environmental evaluation. Related activities are also seen as an important part of EANET's monitoring work in that they ensure the collection of meaningful data. Several documents on QA/QC programs have been developed by EANET to support the provision of reliable data with comparability among participating countries and with information from other monitoring networks outside the East Asian region, such as EMEP (the Co-operative Programme for Monitoring and Evaluation of the Long-range Transmissions of Air Pollutants in Europe) and WMO (the World Meteorological Organization). QA/QC programs also play an important role in EANET activities in participating countries by supporting work at the national level, including the provision of appropriate documentation on QA/QC procedures and the regulation of individual entities. The production of high-quality datasets has been developed through the implementation of such programs and promoted as a major part of the *Strategy on EANET Development (2006 – 2010)*. This approach includes the formulation of SOPs (Standard Operating Procedures) and recommendations for auditing to be realized within the action plan.

Studies on wet deposition, dry deposition and related impacts on ecosystems in East Asia as well as other related research are covered in this report, which deals with most environmental pollutant species. Determining the impacts of acid deposition on the environment, health and ecosystems remains a vital mandate of EANET, whose participating countries conduct ecological impact monitoring work ultimately designed to help assess the impacts of such deposition on soil, forest vegetation, inland lakes and other terrestrial ecosystems. A standard methodology is used for monitoring in all EANET member countries to allow comparison of results at a later stage.

Although not all topics relating to atmospheric environments, emission inventories, modeling and other impact studies may necessarily be directly related to the activities of EANET, they are useful for promoting better understanding of acid deposition and air pollution in East Asia. Accordingly, this report includes reviews of some of these areas. Even if the related data are not fully comprehensive, they are used by EANET for validation and analysis as well as for ecological studies. In addition to data on acidic species, EANET information on surface ozone and PM_{10} mass concentrations (a growing source of concern in regard to health and ecological impacts) is also widely used in research activities. All such information is therefore covered in this report.

Finally, the Eleventh Session of the Scientific Advisory Committee (SAC11) of the Acid Deposition Monitoring Network in East Asia (EANET) adopted the Executive Summary documents during the meeting held in Ho Chi Minh City, Vietnam from 12 to 14 October, 2011.

2. A Decade of EANET Activities

The 2005 – 2009 EANET activity timeframe covered in this report was followed by the period of the Strategy on EANET Development (2006 – 2010), which was approved by the Eighth Session of the Intergovernmental Meeting (IG8) in 2006. The strategy focused on the activities of EANET as a whole, specifying clear targets, activities to be undertaken and completion at the end of 2010. In fact, some related activities had already been started before 2006, and the regular activities of the Secretariat and the Network Center were commenced when EANET was founded over a decade ago. This work has brought EANET closer toward achieving its objectives, and also includes activities relevant to modeling and emission inventories.

Significant achievements were made during the implementation of the Strategy on EANET Development (2006 – 2010). The Secretariat and the Network Center made every effort to carry out all related activities based on the manpower and funding resources available in line with the priorities set by IG8.

The main results and outputs of these strategy activities are as follows:

- Report on activities to improve data completeness
- Set of standard operating procedures (SOPs)
- New EANET monitoring sites
- Annual data reports
- Updated technical manuals, strategy papers and guidelines for EANET monitoring
- First and second periodic reports on the state of acid deposition in East Asia
- Research projects
- Second report for policy makers (RPM2)
- Bi-annual issuance/publication of the EANET newsletter
- WGFD report on the instrument to provide a sound basis for contribution to EANET
- Capacity building workshops for policy makers, technical training and national workshops on public awareness in participating countries
- Production of CDs, videos, brochures and other public awareness materials relating to acid deposition

EANET's monitoring covers five environmental considerations (wet deposition, dry deposition, soil and vegetation, inland aquatic environments, and catchment analysis), and is conducted according to a set of monitoring guidelines and technical manuals. In 2009, deposition monitoring was conducted for wet deposition at 54 sites (remote: 20; rural: 13; urban: 21), for dry deposition at 44 sites, for soil in 19 areas (27 forests), for forest vegetation in 18 areas (26 forests), and for numerous inland aquatic environments (17 lakes/rivers in 13 participating countries) (Figures 2.1 and 2.2).

The research activities conducted are designed to improve air concentration monitoring methodologies and deposition estimation, build capacity in the development of emission inventories, and promote efforts for the improvement and use of appropriate models to assess and analyze trends in the deposition of acid and other air pollutants on all scales. Joint research projects are also conducted, and a number of such initiatives relating to acid deposition and its effects have been implemented by EANET participating countries.

In addition, the promotion of annual fellowship research programs since 2005 has provided an effective mechanism for encouraging young researchers from across the region to participate in air pollution research activities. The technical capabilities and skills of participating countries in acid deposition monitoring and assessment have been significantly enhanced through a number of EANET activities including individual training at the Network Center, and through the Network Center technical missions to all participating countries with the aim of supporting monitoring performance, laboratory operations, data management and other procedures. Other activities

designed to enhance the skills and knowledge of personnel include national workshops, annual expert meetings and scientific workshops on ecological impacts and other topics related to acid deposition. In cooperation with EANET, the Japan International Cooperation Agency (JICA) conducted the JICA Third Country Training Program in Thailand and the JICA Training Course on EANET in Japan to provide guidance on acid deposition and air quality management. Numerous EANET publications (technical manuals and guidelines, data publications, reports on QA/QC projects, training materials, etc.) have been produced for use by technical staff, specialists and researchers involved in monitoring, data quality control and data management. All of these materials are available on the EANET website (http://www.eanet.cc/product/index.html).

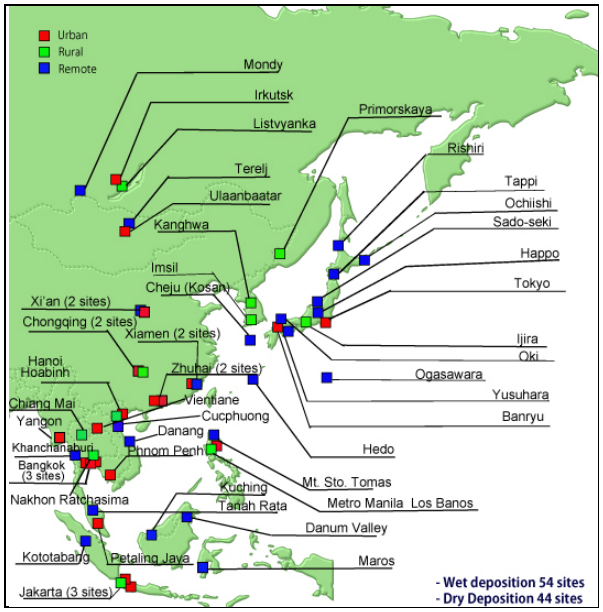


Figure 2.1. EANET deposition monitoring sites (2009)

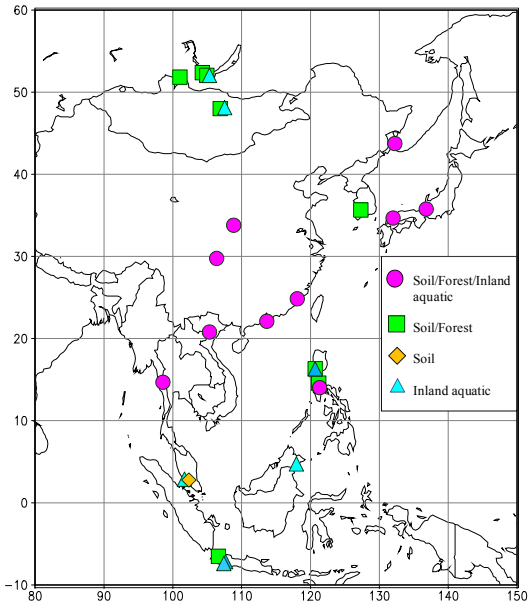


Figure 2.2. EANET ecological impact monitoring sites (2009)

- *Soil: 19 areas (27 forests)*
- *Forest vegetation: 18 areas (26 forests)*
- *Inland aquatic environments: 17 lakes/rivers*

EANET also engages in a range of public awareness activities relating to the deposition of acid and other priority chemical species (including coverage of related effects and control/mitigation measures) targeting a variety of community groups including policy makers, scientists, the general public, young people and school children to promote a common understanding, particularly among school children and teachers, of the atmospheric environmental issues faced by the scientific community and policy makers based on exchanges of information through a network of experts.

3. EANET Data Quality

Chemical analysis and evaluation of Inter-laboratory Comparison Projects are essential in maintaining EANET data quality. Such projects have been implemented annually since 1998 (during the preparatory phase for EANET's establishment) to improve quality assurance and quality control (QA/QC) starting from wet deposition. Today, Inter-laboratory Comparison Projects have been expanded to cover a wider range of fields (wet deposition, dry deposition, soil, and inland aquatic environments, which were the topics of the 12th, 5th, 11th and 10th projects, respectively). The results of wet deposition comparison indicating percentages of flagged data and of those that satisfied the data quality objectives (DQOs) are shown in Figure 3.1.

QA/QC plays an indispensable role in acid deposition monitoring and in other types of environmental evaluation. Related activities are also seen as an important part of EANET's monitoring work to ensure the collection of meaningful data. In such activities, it is especially important that the data obtained meet specified levels of reliability and are accompanied by relevant information on the measurements themselves.

Several documents on QA/QC programs have been developed by EANET to support the provision of reliable data with comparability among participating countries and with information from other monitoring networks outside the East Asian region. Such programs cover activities relating to the components of the measurement/analysis system in general, e.g., field (sampling) sites, laboratories, data management and data reporting processes. QA/QC activities are documented by each relevant entity in line with this EANET guidance.

Appropriate execution of QA/QC programs is regarded as an essential process in obtaining unbiased, accurate and representative data within the EANET framework. Inter-laboratory Comparison Projects are implemented as part of QA/QC programs, and have been conducted annually since the preparatory phase of the network's establishment with all EANET participant laboratories for most environmental measurements in locations where samples are taken. These initiatives provide individual laboratories with opportunities to assess their performance in established analytical procedures and identify problems in order to improve data quality. The ratio of flagged data has decreased since the project system was introduced despite the increased number of laboratories participating, which indicates that most laboratories now satisfy the requirements for the prescribed levels of data quality.

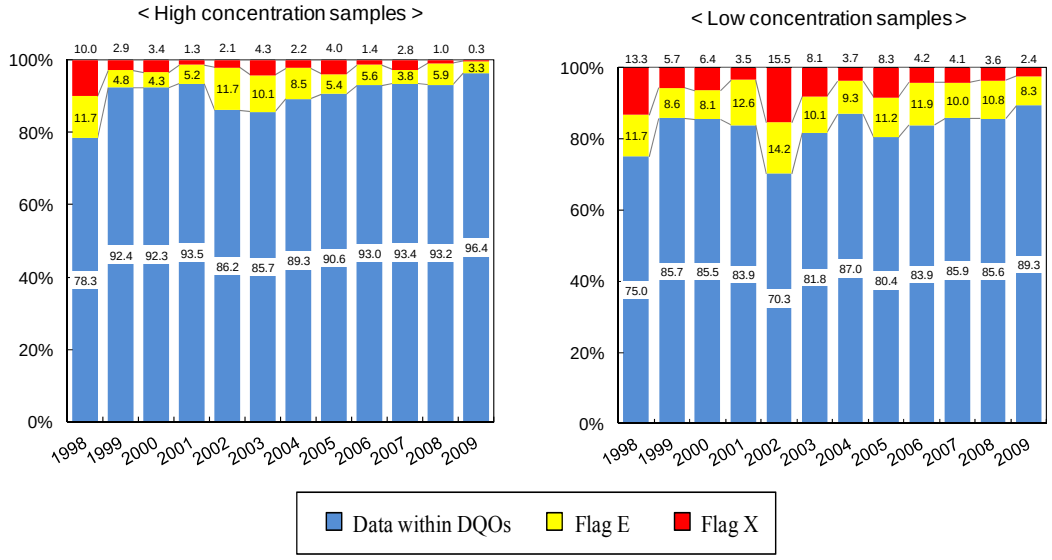
QA/QC programs also play an important role in EANET activities in participating countries by supporting work at the national level, including the provision of appropriate documentation on QA/QC procedures and the regulation of individual entities. The promotion of QA/QC activities was highlighted as a major part of the *Strategy on EANET Development (2006 – 2010)*, including the formulation of SOPs and recommendations for auditing to be realized within the action plan. Further steps to improve such activities are also harmonized with international efforts in the field, and stringent procedures for data management are applied to ensure the accuracy of information.

To promote the collection of high-quality analytical data, Inter-laboratory Comparison Projects are implemented for wet and dry deposition monitoring as well as for soil and inland aquatic environments. For gas concentration monitoring, the filter-pack method is used. Data completeness is also assessed and kept at a high level by conducting equipment maintenance with appropriate frequency and taking steps to reduce human error. The overall quality of monitoring data is improved and maintained based on continuous implementation of these activities.

High-quality datasets have been developed through the implementation of quality assurance and quality control measures at the national level and in the Inter-laboratory Comparison Projects. Since 2000, summaries of annual monitoring results have been published regularly in the Data Report on Acid Deposition in the East Asian Region. All monitoring data provided by EANET

participating countries since 2000 are made available to the public on the EANET website and on compact disc.

Figure 3.1 shows the results of the Inter-laboratory Comparison Projects on wet deposition from 1998 to 2009. The data quality objectives (DQOs) for accuracy have gradually improved since this period.



The flags are indicated according to the degree of deviation to the DQOs.
Flag E: between 15% and 30%,
Flag X: over 30%.

Figure 3.1. Results of the Inter-laboratory Comparison Projects on wet deposition for 1998 – 2009.

4. Wet Deposition

Concept of Acid Deposition

Acids are a group of compounds that release hydrogen atoms in the form of hydrogen ions when dissolved in water. Accordingly, they are intrinsically water-soluble and therefore deeply involved in the processes of atmospheric precipitation. Rather than having specific emission sources, most acids in the atmosphere form chemically during the course of material transport. Major acids include the sulphuric and nitric types derived from sulphur and nitrogen oxides, and acids thus formed acidify atmospheric water droplets such as those of rain. Acidity is commonly expressed on the pH scale, whose variations in a solution can be used to estimate atmospheric water acidification. Another role of atmospheric acids is to transport basic compounds by incorporating bases through acid-base reactions. By way of example, during the atmospheric transport of sulphuric and nitric acids, gaseous ammonia is collected and forms ammonium salts, which enables ammonia to be transported over longer distances. These precipitation acids are eventually deposited onto the earth's surface, causing possible impacts to ecological elements like water, soil, vegetation and materials through acidification, and at the very least disturbing the acid-base balance of such elements.

The present analysis was applied to volume-weighted annual mean concentrations of the analytical ion suite in order to provide a general regional overview of acid deposition. For this purpose, all available annual means since 2000 were subjected to data screening to include only annual mean sets meeting the data completeness criteria for nine ions (NH_4^+, Ca^{2+} (nss-Ca^{2+}), K^+, Mg^{2+}, Na^+, NO_3^-, SO_4^{2-} (nss-SO_4^{2-}) and Cl^-) including H^+ for pH values.

The state of acid deposition was assessed in terms of rainwater pH to provide an easily understandable numerical basis for public discussion of acid rain, and explanations of different pH values were given as a starting point for understanding wet deposition.

On a regional scale, pH values were assessed from a histogram of annual means covering the entire period from 2000 to 2009 (Figure 4.1). All sites that successfully produced at least one full year of measurements among the nine analytical sites monitoring pH during the period are included in the figure, which was eventually based on measurements from 57 sites in all 13 countries. The pH values ranged from 4.18 to 6.94 with an arithmetic mean of 5.07. The histogram is skewed toward higher pHs with a median of 4.94. The values below 4.30 were 4.18 (Jinyunshan, 2001), 4.20 (Haifu, 2008), 4.22 (Nansan, 2000; Haifu, 2009), 4.23 (Petaling Jaya, 2002), 4.25 (Ganghwa, 2005), 4.25 (Petaling Jaya, 2001) and 4.28 (Petaling Jaya, 2003).

In contrast, the values above 6.5 were 6.94 (Jiwozi, 2008), 6.85 (Jiwozi, 2007), 6.72 (Phnom Penh, 2006), 6.69 (Phnom Penh, 2007) and 6.53 (Shuzhan, 2009).

Five-year averages for the period from 2005 to 2009 were obtained for 42 of these 57 sites, and are shown in Figure 4.2 in order of decreasing pH. The values for

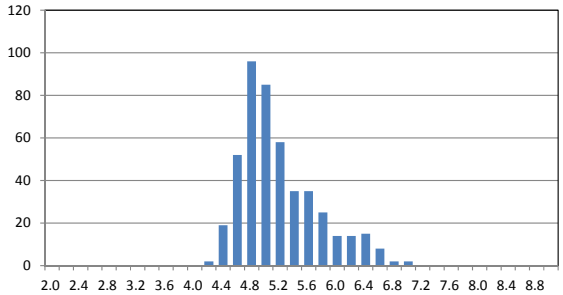


Figure 4.1. Histogram of annual mean pH values for 2005 – 2009.

Cambodia, Lao PDR and Myanmar are excluded because these sites had less than five years of operation due to their time of entry into EANET. The two highest values (6.20 for Jiwozi and 6.19 for Ulaanbaatar) were exceptional considering the distribution, while the remaining pHs show a smooth decrease to the minimum value of 4.42 for Petaling Jaya.

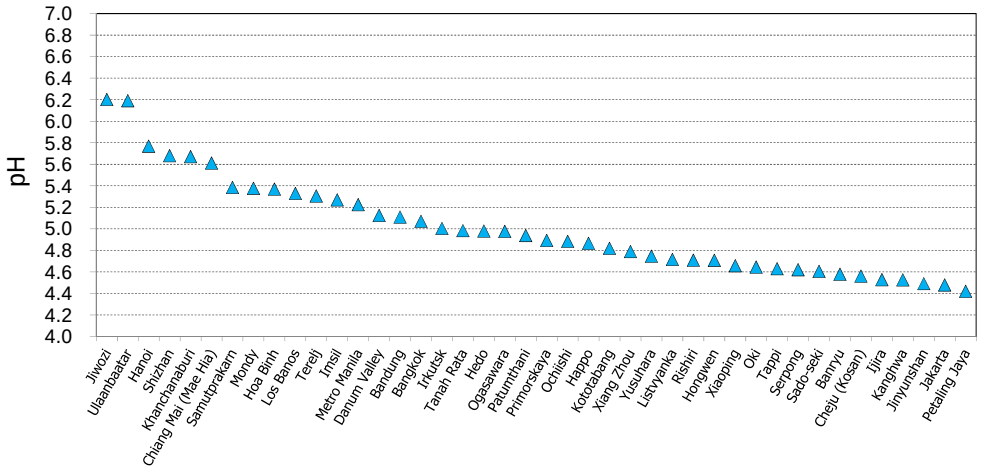


Figure 4.2. Five-year average annual mean pH values for 42 sites.

It is difficult to set a scientifically reasonable pH value to define acid rain, although 5.6 seems to be generally accepted as the standard based on the gas-water equilibrium of carbon dioxide (an acidic gas) in the atmosphere. However, even natural air free from anthropogenic pollution contains acidic gases such as sulphur dioxide due to influences including volcanic emissions and oxidation of maritime dimethylsulfide (DMS, or $(CH_3)_2S$). Some bases are also derived from natural sources such as soil and animals.

Despite the difficulty of defining a specific value, it is useful to set a pH point for preliminary evaluation of observations. Accordingly, a pH of 5.0 is proposed as a round number. In total, 16 of the 42 sites (62%) recorded pHs lower than this guide value. No sites recorded values of 4.0 or less, although the lowest three were less than 4.5.

It is clear that further chemical analysis is necessary to support more detailed discussion of the matter. However, it can safely be said here that precipitation in East Asia is significantly acidic.

Concepts of pAi and pH

pH values should be appropriately interpreted on the basis that they are determined by the nature and balance of acids and bases in aqueous solutions. Specifically, neutralization between acids and bases needs to be quantitatively considered. For this purpose, the metric of pAi has been proposed as detailed below as an indicator similar to pH. In this consideration, two major acids (sulphuric and nitric) are assumed to determine the input acidity (Ai) in order to give an overall picture of acid-base chemistry.

$$pAi = -\log\{[Ai]\} = -\log\{[nss\text{-}SO_4^{2-}] + [NO_3^-]\}$$
$$pH = -\log\{[H^+]\}$$

The two values are complementary to each other: pH is an observable metric that shows acidity after neutralization, whereas pAi is a conceptual parameter corresponding to the level of acidity before neutralization. If no neutralization takes place, the pH and pAi values are identical. pH increases with the addition of bases during the course of neutralization, whereas pAi remains constant throughout.

The pAi-pH concept was applied to five-year annual mean datasets for the period from 2005 to

2009. Figure 4.3 shows that pH ranged from 4.4 to 6.5, while pAi ranged from 3.6 to 5.4. The highest pHs were 6.46 (Yangon), 6.20 (Jiwozi), 6.19 (Ulaanbaatar) and 6.14 (Phnom Penh). In terms of pAi, however, the corresponding values were significantly different at 4.85, 3.78, 4.34 and 4.67. In Jiwozi, for instance, the input acidity would have been considerably higher (possibly as low as pH 3.78 before neutralization), but due to the high degree of neutralization, the eventual pH value was a relaxed 6.20. Shizhan and Jinyunshan had pAi values well below 4.0 at 3.63 and 3.82, respectively. However, different basic contributions seem to have affected the two systems, with the resultant pHs being 5.68 for Shizhan and 4.49 for Jinyunshan.

All plots should be located above the diagonal line of the pAi-pH diagram according to the definition. The points for Kototabang (5.15, 4.82), Danum Valley (5.39, 5.12) and Ogasawara (4.98, 5.04) are well below the line, which could be explained by the possible contribution of organic acids due to the site locations and the significantly high pH values seen.

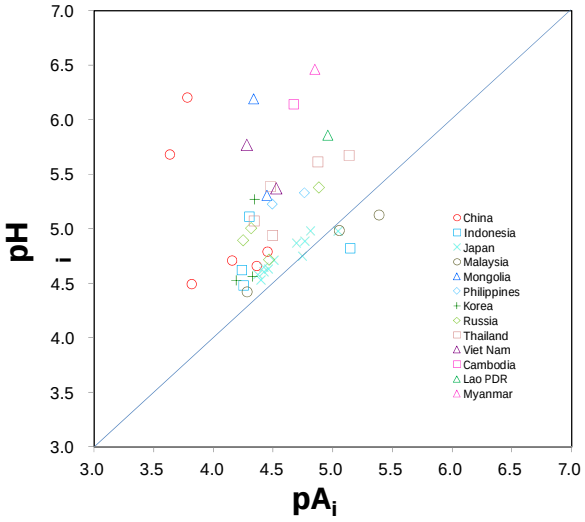


Figure 4.3. pAi-pH diagram for the 13 EANET countries (averaged over 2005 – 2009.

In acid deposition chemistry, the acid pairs NO_3^-/nss-SO_4^{2-}, NH_4^+/nss-Ca^{2+} and NH_4^+/NO_3^- play a highly important role in terms of acidification of the atmosphere and ecosystems. However, other acids (including hydrochloric, hydrofluoric and a variety of organic types) also play significant roles. Here, the relative contribution of the two acids is rationally discussed in terms of corresponding anions.

The relative contribution was quantified in the form of the NO_3^- fraction for the sum of the ion pair NO_3^-/nss- SO_4^{2-} as $[NO_3^-]$ / ($[NO_3^-]$ + [nss- SO_4^{2-}]), which is denoted here as fraction #1 (F1) for simplicity.

$$F1 = [NO_3^-] / ([\text{nss-}\ SO_4^{2-}] + [NO_3^-])$$

The value of F1 was calculated from sulphuric and nitric acids for the five-year annual means of the 42 sites, as these acids played a highly important role in acidification of the atmosphere and ecosystems. The maximum, 0.50, occurred at Petaling Jaya, which means two acids, H_2SO_4 and HNO_3, were equally significant in rain acidification. This value was followed by 0.48, 0.46, 0.45, 0.43, and 0.42 for Pathumthani, Chiang Mai, Bangkok, Vientiane, and Kanchanaburi, respectively. The ratio was lowest, 0.13, for Shizhan, and Jinyunshan (0.16), Jiwozi (0.17), Kototabang (0.18), where H_2SO_4 was four times or more dominant acid than HNO_3 on an equivalent basis.

For the precipitation samples used, a pair of basic compounds (NH_3 and $CaCO_3$) can be represented by NH_4^+ and nss-Ca^{2+}, which suggests the need to define fraction #2 (F2) to assesshe importance of NH_3 relative to $CaCO_3$ as F2 = $[NH_4^+]$ / ([nss-Ca^{2+}] + $[NH_4^+]$).

The F2 values are mostly over 0.50 where the two compounds work equally as bases in the interaction. The only three sites displaying values of less than 0.33 were Jiwozi (0.11), Phnom Penh (0.25) and Shizhan (0.33). The figure for Jiwozi in particular is likely to have been strongly influenced by $CaCO_3$. The maximum was 0.80 for Ijira, and F2 values exceeding 0.70 were also recorded at the seven sites of Serpong (0.75), Xiang Zhou (0.75), Danum Valley (0.72). Thus, ammonia is a more dominant base than $CaCO_3$.

Nitrogen saturation is another significant environmental issue, and a third fraction (F3) was introduced to highlight the relative importance of nitrogen species. F3 is $[NH_4^+]$ / ([NH_4^+] + $[NO_3^-]$).

Among the 45 sites, 10 displayed F3 values lower than 0.50. The lowest was from Petaling Jaya (0.35), and other low values included those at Jakarta (0.40), Oki (0.43), Tanah Rata (0.43), Banryu (0.44) and Tappi (0.55). The maximum was 0.82 for Kototabang, and nine more stations showed values of more than 0.70.

Deposition of Major Ions and Chemical Indices

As with ionic concentrations, the deposition of major ions was also discussed. For deposition, however, some chemical indices (ΣN and H^+_{eff} as defined below) were included in the analysis. Since nitrogen saturation is another environmental issue involved in the relationship with acid deposition and atmospheric deposition, the summation of ionic deposition for ammonium and nitrate was calculated as the wet deposition of inorganic nitrogen: $\Sigma N = NH_4^+ + NO_3^-$, where the chemical symbols denote the deposition amounts of the respective species.

The other index, effective hydrogen ion deposition (H^+_{eff}), is defined as ($H^+ + 2NH_4^+$), where the symbols express the deposition amounts of the respective species on the basis of biological nitrification of ammonium in the soil environment: $NH_4^+ + 2O_2 \rightarrow 2H^+ + NO_3^- + H_2O$.

Since deposition is calculated as the product of concentration and the precipitation amount, the discussion begins with a histogram showing annual rainfall amounts. Six rainfall totals exceeding 4,000 mm yr^{-1} have been recorded since 2000: 5,078 and 4,727 at Mt. Sto. Tomas (2008, 2009), 4,671 and 4,286 at Kuching (2009, 2008), 4,036 at Metro Manila (2000), and 4,009 at Petaling Jaya (2006). The six lowest totals were 69 at Weishuyuan (2001), 83 at Ulaanbaatar (2004), 89 at Terelj (2001), 100 at Jiwozi (2007), 111 at Terelj (2009), and 119 at Mondy (2009). The five-year average annual precipitation amounts are summarized in Figure 4.4.

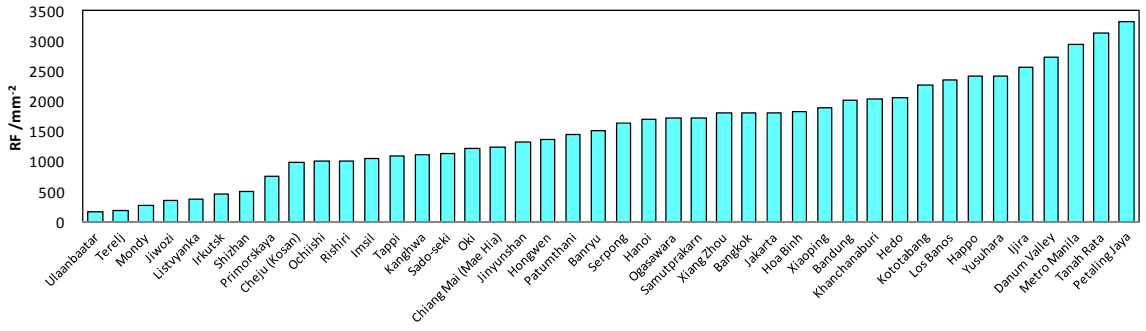


Figure 4.4. Five-year average annual precipitation amounts (2005 – 2009).

The histogram for nss-SO_4^{2-} deposition shows a mono-modal pattern different from that for its concentration. Five deposition amounts exceeding 400 meq $m^{-2}yr^{-1}$ were observed: 583 and 506 at Shizhan (2002, 2003), 478 and 461 at Weishuyuan (2000, 2002), and 457 at Guanyinqiao (2004). The following amounts less than 3 meq $m^{-2}yr^{-1}$ were also reported: 1.0 at Mt. Sto. Tomas (2006), 1.3 at Vientiane (2009), 1.6 at Terelj (2001), and 1.8 and 2.9 at Mondy (2009 and 2008). The five-year average annual deposition amounts of nss-SO_4^{2-} are shown in Figure 4.5.

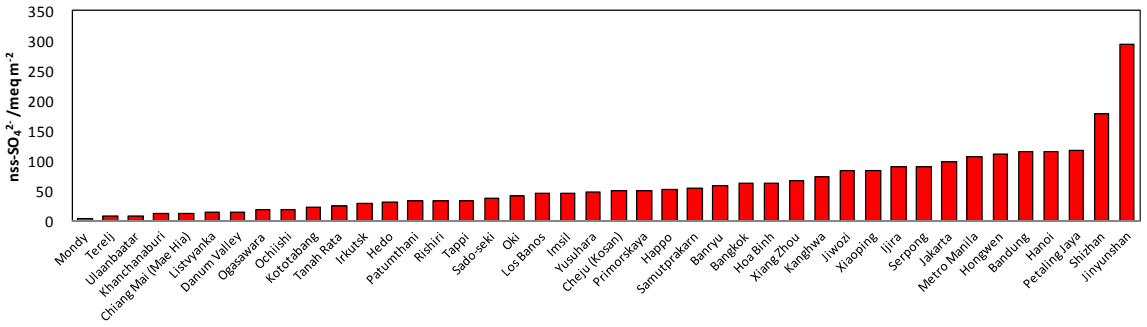


Figure 4.5. Five-year average annual deposition of nss-SO_4^{2-} (2005 – 2009).

Five-year averages of annual deposition for NO_3^-, NH_4^+, nss-Ca^{2+} and H^+ are shown in Figures 4.6, 4.7, 4.8 and 4.9.

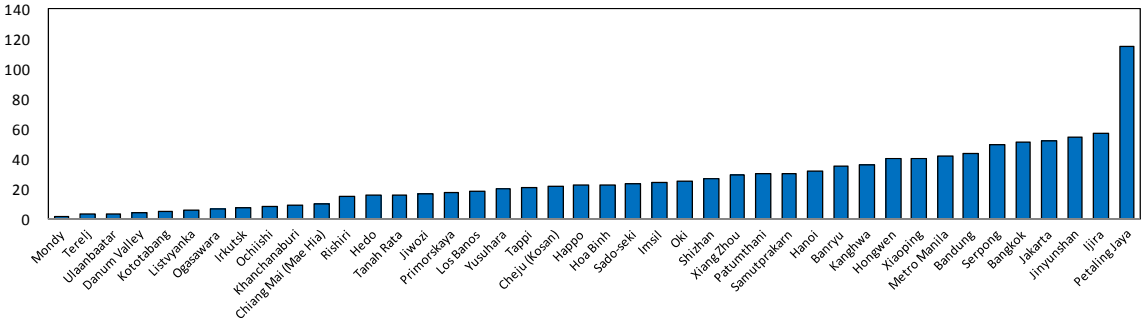


Figure 4.6. Five-year average annual deposition of NO_3^- (2005 – 2009).

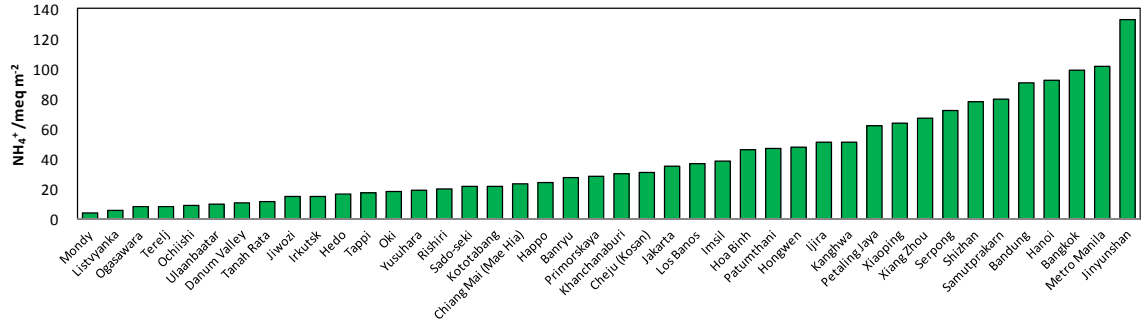


Figure 4.7. Five-year average annual deposition of NH_4^+ (2005 – 2009).

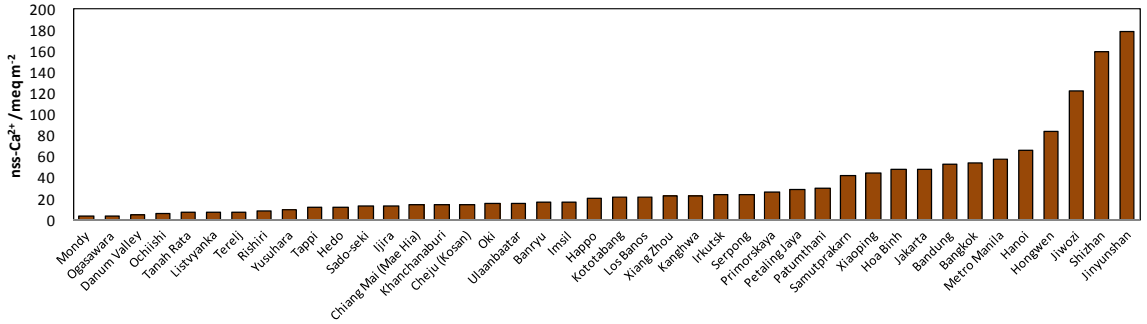


Figure 4.8. Five-year average annual deposition of nss-Ca2+ (2005 – 2009).

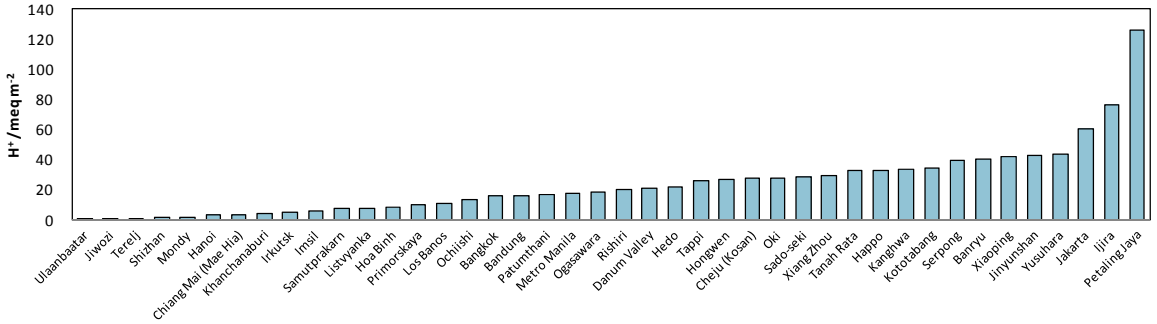


Figure 4.9. Five-year average annual deposition of H^+ (2005 – 2009).

Deposition analysis was extended to two chemical indices (ΣN and H^+_{eff}) to determine possible ecological impacts. Five-year mean figures are useful for the inspection of specific sites to clarify the significance of deposition in EANET countries (Figure 4.10 and Figure 4.11).

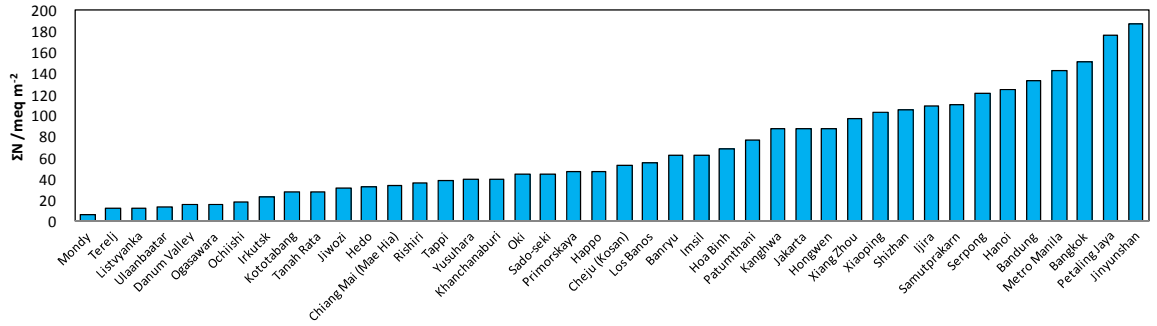


Figure 4.10. Five-year average annual deposition of ΣN (2005 – 2009).

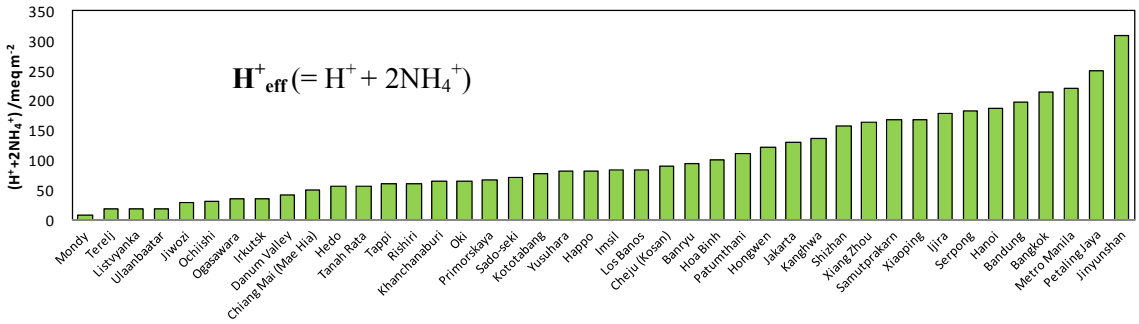


Figure 4.11. Five-year average annual deposition of H^+_{eff} (2005-2009)

Temporal Variations in Concentration and Deposition

Temporal variations in the ionic deposition and concentration of major ions and other indices were explored by applying linear regression analysis to the 2000 – 2009 annual mean figures for concentration and deposition. Statistical testing of the slopes of the regression lines indicated 95% confidence, as shown in Table 4.1. Some sites displayed an increasing trend, while others exhibited a decrease. It is interesting to note that significantly increasing temporal variations for a number of variables were seen in Bangkok. Concentrations and depositions of ΣN and H^+_{eff} show an increasing trend (Figure 4.11).

Although EANET has accumulated up to 10 years of records, the period does not suffice for temporal trend analysis because wet deposition involves a number of factors that vary significantly over time. Accordingly, in contrast to the case with gas and aerosol species, temporal trends in wet deposition may not be clearly identifiable over such short periods. This highlights the indispensable

nature of long-term, high-quality monitoring for symptom detection to allow elimination and mitigation of potential impacts.

Table 4.1. Temporal variations in the ionic deposition and concentration of major ions and other indices

Country	Site	Species	C/D	n	Slope
Thailand	Bangkok	NO_3^-	D	10	3.117
	Bangkok	NH_4^+	C	10	2.739
	Bangkok	NH_4^+	D	10	8.848
	Bangkok	ΣN	C	10	3.986
	Bangkok	ΣN	D	10	13.035
	Bangkok	$(H^+)_{eff}$	C	10	4.738
	Bangkok	$(H^+)_{eff}$	D	10	17.449
Malaysia	Petaling Jaya	nss-SO_4^{2-}	C	10	–1.035
	Petaling Jaya	nss-Ca^{2+}	C	10	–0.374
	Petaling Jaya	nss-Ca^{2+}	D	10	–1.049
Malaysia	Tanah Rata	nss-Ca^{2+}	C	10	–0.268
	Tanah Rata	nss-Ca^{2+}	D	10	–0.716
	Tanah Rata	H^+	C	10	–0.639
Indonesia	Jakarta	nss-SO_4^{2-}	C	9	–5.107
	Jakarta	nss-SO_4^{2-}	D	9	–6.058
Japan	Ijira	nss-Ca^{2+}	C	10	–0.350
	Ijira	nss-Ca^{2+}	D	10	–0.908
Japan	Banryu	NO_3^-	C	10	1.070
	Banryu	NO_3^-	D	10	1.305
China	Xiaoping	NO_3^-	C	10	1.269
	Xiaoping	nss-SO_4^{2-}	C	10	1.352
Thailand	Chiang Mai	NO_3^-	C	9	0.488
	Chiang Mai	ΣN	C	9	0.941
Thailand	Pathumthani	nss-SO_4^{2-}	C	10	–0.930
China	Jinyunshan	NO_3^-	D	9	2.834
Indonesia	Bandung	NH_4^+	C	10	1.928
Japan	Yusuhara	NH_4^+	C	10	0.322

D = deposition, C = concentration

5. Dry and Overall Deposition

Spatial Variations in Air Concentration

Overall deposition amounts vary considerably in EANET regions. In line with these significant spatial differences, analysis of dry deposition in the relevant areas is reported for the two broadly classified sub-regions of Northeast and Southeast Asia.

Information on dry deposition flux is considered necessary for evaluation at the regional level in addition to information on wet deposition in order to provide more comprehensive data regarding atmospheric impacts on the environment. The EANET network included 44 air concentration monitoring sites in 2009, each with its own set of air concentration compounds depending on the location and the equipment used. The filter-pack method is promoted by EANET as the optimal approach for monitoring gas and aerosol concentrations. To determine levels of nitrogen oxides (NO_x) and ozone (O_3), passive samplers are first used for screening to ascertain average levels of concentration, and automatic air monitors are then promoted for installation in countries with the necessary capacitynd in areas where high concentrations are observed. The Task Force for Dry Deposition Monitoring (TFMDD) indentified related procedures and estimation of deposition velocity values based on a numerical model as well as calculation of dry deposition flux using continuous sets of air concentration and deposition velocity data for monitoring sites. From this implementation, an *ad hoc* database is maintained for parallel direct flux observation, which allows comparison with estimates of dry deposition fluxes and improvement of inferential methods – particularly the numerical model for dry deposition velocity – based on continuous examination.

EANET's Northeast Asian region comprises the eastern part of Russia, Mongolia, China, the Republic of Korea and Japan. Due to the influence of the Pacific Ocean, the Russian Far East has a monsoon climate in which the wind direction in summer and winter is reversed, creating sharp differences in temperature. Mongolia's terrain is characterized by mountains and rolling plateaus, and has a high degree of relief. The country has an average elevation of 1,580 m and an extremely continental climate with long cold winters and short summers during which most of the year's precipitation falls. The country averages 257 cloudless days a year, with average precipitation highest in the North (2,000 – 3,500 mm/yr) and lowest in the South (1,000 – 2,000 mm). Average temperatures over most of the country are below freezing from November to March, and remain around freezing point in April and October. January and February averages of -20°C are common. The climate of China is extremely diverse, ranging from subtropical in the South to subarctic in the North. The country's tremendous differences in latitude, longitude and altitude give rise to sharp variations in precipitation and temperature, creating complex climatic patterns. The Korean Peninsula is under the influence of the monsoon region, and has a temperate climate with four distinct seasons. The movement of air masses from the Asian continent exerts a significant influence on South Korea's weather. Meanwhile, Japan's terrain is around 74% mountainous, and is characterized by mountain ranges running along each of the main islands, scattered plains and intermontane basins. The country is in the temperate zone, and also has four distinct seasons. It is generally a rainy country with high humidity.

Spatial air pollution variations in the Northeast Asian region are presented based on average annual observed values. SO_2, HNO_3, NH_3 and aerosol components are monitored in the same way as SO_4^{2-}, NO_3^- and NH_4^+ with urban, rural and remote area classifications. For most of urban East Asia, the observed NH_3 values are higher than those for SO_2 and much higher than those for HNO_3. This phenomenon is unexpected for city areas because NH_3 is generally found in greater amounts in agricultural regions. NH_3 concentrations were observed in ranges of 7.9 – 11.2 ppb at Hongwen (China), 7.4 – 10.7 ppb at Ulaanbaatar (Mongolia) and 1.8 – 3.8 ppb at Irkutsk (Russia), while SO_2 concentration ranges were 5.2 – 10.9 ppb at Hongwen, 1.8 – 4.8 ppb at Ulaanbaatar and 2.2 – 5.1 ppb at Irkutsk. The urban concentrations at Banryu (Japan) were much lower for SO_2 (0.6 – 1.1 ppb) and NH_3 (1.0 – 1.2 ppb) than for all other urban areas within EANET's East Asian region.

Urban sites: Aerosol concentrations of sulphate (SO_4^{2-}) were higher than those for NO_3^- and NH_4^+ in all urban areas, which indicates further chemical reaction of the primary chemical (SO_2) with NH_4^+ or other available cations to form sulphate compounds. The observation results for 2005 – 2009 showed concentrations that were low for SO_2 but high for SO_4^{2-}. Concentrations of SO_4^{2-} were observed in ranges of 10.58 – 13.38 μg.m^{-3} at Hongwen, 5.11 – 6.55 μg.m^{-3} at Banryu, 4.62 – 5.36 μg.m^{-3} at Tokyo, 1.33 – 5.68 μg.m^{-3} at Ulaanbaatar and 2.13 – 2.91 μg.m^{-3} at Irkutsk.

Nitrate (NO_3^-) concentrations were observed in ranges of 5.81 – 8.10 μg.m^{-3} at Hongwen, 1.35 – 1.57 μg.m^{-3} at Banryu, 3.85 – 4.03 μg.m^{-3} at Tokyo, 0.9 – 1.74 μg.m^{-3} at Ulaanbaatar and 0.61 – 0.89 μg.m^{-3} at Irkutsk. Observed values for NH_4^+ were markedly low at most East Asian urban sites. Although ammonium ions react readily with SO_2 to form sulphate aerosols, the reaction $NH_3(g) + HNO_3(g) \leftrightarrows NH_4NO_3(s)$ can also occur to form ammonium nitrate (NH_4NO_3). This reaction is generally seen only in low-sulphur atmospheric conditions.

Rural sites: Distinctively high SO_2 concentrations (12.2 – 17.8 ppb) were observed only at Jinyunshan (China). Other chemical components (HNO_3 and NH_3) were not monitored at this site or at Imsil and Ganghwa (the Republic of Korea) before 2006. At the latter two sites, low ranges of SO_2 were observed (1.4 – 2.5 ppb and 2.8 – 3.4 ppb, respectively), but the values were higher for NH_3 (3.2 – 9.7 ppb and 5.0 – 6.0 ppb, respectively). All chemical components monitored at Ijira were comparatively low. SO_2 concentrations at the rural sites of Listvyanka and Primosrskaya (Russia) were in the ranges of 1.1 – 6.0 ppb and 0.7 – 1.6 ppb, respectively, while NH_3 values were in the ranges of 0.8 – 4.4 ppb and 1.2 – 3.6 ppb, respectively. HNO_3 concentrations were markedly low for all East Asian rural sites.

Remote sites: The three gas components SO_2, HNO_3 and NH_3 at the remote site in Jeju (the Republic of Korea) were notably higher than those of other remote sites within the East Asian region. The range of observed SO_2 concentrations was 2.2 – 3.4 ppb; for HNO_3, it was 0.5 – 0.7 ppb; and for NH_3, it was 3.7 – 4.9 ppb. These values are similar to the corresponding figures for the Ganghwa site, but higher than those for the Imsil site (the two rural sites in the Republic of Korea). Gas concentrations observed at the six remote sites in Japan varied, with a pattern of increasing values from Rishiri in the North down to Oki in the South except at the two southernmost remote island sites of Hedo and Ogasawara, which showed very low concentrations. The Rishiri site reported the lowest SO_2 concentrations (0.1 – 0.3 ppb), followed by Ochiishi (0.2 ppb), Tappi (0.2 – 0.5 ppb), Sado-Seki (0.4 – 0.6 ppb), Happo (0.4 – 0.8 ppb), Oki (0.6 – 0.9 ppb) and Yusuhara (0.9 – 1.2 ppb). In regard to the two southern islands, Hedo showed very low SO_2 concentrations of 0.2 – 0.4 ppb, and the corresponding levels were negligible at Ogasawara. Ammonia concentrations at all remote sites in Japan varied within the range of 0.2 – 1.2 ppb. Concentrations of HNO_3 at all East Asian remote sites were low (within a range of 0.1 – 0.7), with negligible values observed at Mondy (Russia).

The observed values for the aerosol component SO_4^{2-} were significantly higher than those for NO_3^- and NH_4^+ at all remote East Asian sites. These results indicate that the sources of sulphur may be human activities and natural sulphate. In terms of the four correlated chemical species, i.e., SO_4^{2-}, Na^+, Cl^- and Ca^{2+}, the Japan sites that showed high values for SO_4^{2-} also tended to show high results for Na^+, Cl^- and Ca^{2+}. For example, at the Hedo island site, SO_4^{2-} concentrations were observed in a range of 5.32 – 6.51 μg.m^{-3}, while Na^+, Cl^- and Ca^{2+} were found in ranges of 3.77 – 5.22 μg.m^{-3}, 4.40 – 7.29 μg.m^{-3} and 0.34 – 0.48 μg.m^{-3}, respectively. During the same period, at the Happo inland site, SO_4^{2-} concentrations were observed in a range of 5.32 – 6.51 μg.m^{-3}, while Na^+, Cl^- and Ca^{2+} were found in ranges of 3.77 – 5.22 μg.m^{-3}, 4.40 – 7.29 μg.m^{-3} and 0.34 – 0.48 μg.m^{-3}, respectively. Relatively, the Happo site (located on the mountainous site of central Japan) showed low concentrations for all the components of SO_4^{2-} (0.59 – 2.79 μg.m^{-3}), Na^+ (0.25 – 0.29 μg.m^{-3}),

Cl^- (0.01 – 0.03 µg.m^{-3}) and Ca^{2+} (0.07 – 0.17 µg.m^{-3}). At the Jeju site in the Republic of Korea, SO_4^{2-} concentrations showed no relationship with those of Na^+, Cl^- and Ca^{2+}. The SO_4^{2-} concentrations were markedly high for a remote area (4.68 – 8.05 mg.m^{-3}), while those of Na^+, Cl^- and Ca^{2+} were observed in low ranges (0.35 – 0.51 µg.m^{-3}, 0.13 – 0.32 µg.m^{-3} and 0.05 – 0.11 µg.m^{-3}, respectively). The measured concentrations of all these chemical components were markedly low at Terelj (Mongolia) and Mondy (Russia). SO_4^{2-}, Na^+, Cl^- and Ca^{2+} were observed at Terelj in ranges of 0.85 – 1.42 µg.m^{-3}, 0.08 – 0.14 µg.m^{-3}, 0.19 – 0.36 µg.m^{-3} and 0.27 – 0.42 µg.m^{-3}, respectively, and at Mondy in ranges of 0.22 – 0.49 µg.m^{-3}, 0.04 µg.m^{-3} (negligible), 0.03 µg.m^{-3} (negligible) and 0.03 – 0.07 µg.m^{-3}, respectively. It should be noted that for other particulate components, e.g., Mg^{2+} and K^+, the concentration values were within the Ca^{2+} ranges seen at all remote East Asian sites.

The concentration distributions of other important aerosol components, e.g., nitrate (NO_3^-) and ammonium ions (HN_4^+), were markedly low at all remote sites.

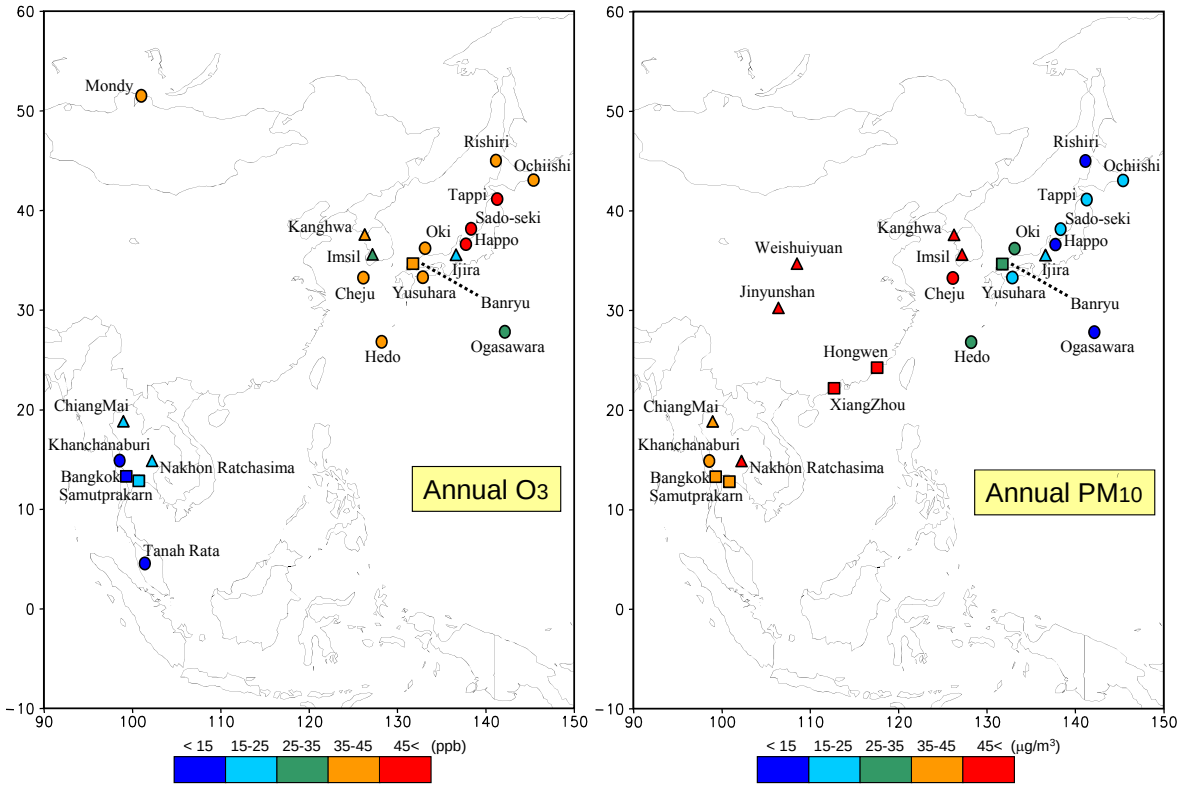


Figure 5.1. Spatial distribution of O_3 and PM_{10} concentrations at EANET sites. The values are annual averages for 2005 – 2009. *Square, triangle and circle symbols denote urban, rural and remote sites, respectively.*

Figure 5.1 illustrates the spatial distribution of five-year average O_3 and PM_{10} concentrations at EANET sites. Values for NO_2, NOx and PM_{10} are high at sites in China and lower at those in Japan, most of which are classified as remote. Annual O_3 concentrations were higher at mountainous Japanese and Russian sites.

Southeast Asia straddles two geographic regions. Its mainland part includes Cambodia, Laos, Myanmar, Thailand, Vietnam and peninsular Malaysia, while its maritime part comprises Indonesia and the Philippines. The climate in Southeast Asia is mainly tropical, i.e., hot and humid all year round with abundant rainfall.

The results of spatial variation analysis for air concentrations at most urban sites in South Asian regions for 2005 to 2009 showed NH_3 values that were generally three times higher than those for SO_2. Conversely, values for HNO_3 were noticeably low in urban areas, which are thought to be a result of reduced conversion of NOx into HNO_3. For rural areas, the high concentration of NH_3 is thought to stem from the use of fertilizers in hot and humid conditions, as agriculture is a major activity in most Southeast Asian countries.

Although Jakarta (Indonesia) and Samutprakan (Thailand) monitored only SO_2, the observed values were noticeably high in ranges of 7.4 – 11.0 ppb and 4.8 – 6.6 ppb, respectively. However, the SO_2 concentration in Bangkok fell from 7.3 ppb in 2005 to 1.9 ppb in 2009, which may have been a result of a national policy to improve vehicle fuel quality. At other urban sites at Petaling Jaya (Malaysia), Metro Manila (Philippines), Bangkok (Thailand) and Hanoi (Vietnam), SO_2 concentrations were observed in ranges of 1.8 – 6.3 ppb, 1.2 – 3.1 ppb, 0.8 – 7.3 and 1.3 – 3.9 ppb, respectively. The measured values of NH_3 were in ranges of 6.1 – 16.9 ppb, 3.8 – 12.5 ppb, 7.8 – 10.2 ppb and 2.8 – 5.0 ppb at Petaling Jaya, Metro Manila, Bangkok and Hanoi, respectively. Reported HNO_3 amounts were markedly low at all urban sites. This component has a short life in the atmosphere as it forms readily in humid conditions and is then washed out by heavy monsoon rains in the tropical region.

Variations in aerosol compositions at EANET urban sites in South Asia are also observed. Sulphate component amounts are proportionately much higher than those of the other two acid components, whether the site is located on high land (Hanoi), in a hot and humid central region (Bangkok), on the Malay Peninsula (Petaling Jaya) or on Pacific islands (Metro Manila). SO_4^{2-} concentration ranges were 2.26 – 3.32 $\mu g.m^{-3}$ at Petaling Jaya, 2.37 – 3.31 $\mu g.m^{-3}$ at Metro Manila, 2.13 – 4.71 $\mu g.m^{-3}$ at Bangkok, and 8.77 – 20.35 $\mu g.m^{-3}$ at Hanoi. It is noted that amounts of ambient aerosols in Hanoi fell markedly between 2005 and 2009. The measured values for the other components of NO_3^- and NH_4^+ were small.

Spatial variations in chemical compositions are seen among Southeast Asian rural areas. The high ammonia concentrations observed at all EANET rural sites indicate significant effects of nitrogen constituent emissions from agriculture in the vicinity of the monitoring areas and a weaker influence from mass transport of sulphur components from urban areas. The concentration ratio of NH_3/SO_2 at Serpong (Indonesia) was 7, while the corresponding values for other sites were 11 for Los Baños (Philippines), 6 for Chiang Mai (Thailand), and 1.5 for Hoa Binh (Vietnam). The high amounts of NH_3 and the low amounts of HNO_3 and NH_4^+ are considered to have slowed reverse atmospheric oxidation of NH_3 to HNO_3 and NH_4^+. The acid chemical distribution at all Southeast Asian remote sites followed the same pattern as that seen at urban and rural sites.

NO_2 and NOx concentrations were high at urban sites in Indonesia and Thailand, while annual O_3 concentrations were low in Thailand, and moderate annual PM_{10} concentrations were observed at urban sites in the country.

Temporal Variations in Air Concentration

In Northeast Asia, the presence of sulphur components at all monitoring sites in China (Xiamen-Hongwen and Xiang Zhou), Mongolia (Ulaanbaatar and Terelj) and Russia (Irkutsk, Listvyanka, Primorskaya and Mondy) showed distinctively seasonal cyclical patterns during the period 2005 – 2009. The observed concentrations of SO_2 and SO_4^{2-} were low in mid-summer and high in winter, which indicates high energy utilization in low-temperature conditions. SO_2 concentrations in China showed some temporal decrease, but values in Mongolia and Russia fluctuated during the period. The maximum concentration of SO_2 at the Russian urban site of Irkutsk was four to five times higher than that at the remote site of Mondy. In fact, the SO_2 concentration at Mondy was quite small in 2009 (0.1 ppb), showing a significant decrease from the 2005 value of 1.6 ppb.

The Imsil, Ganghwa and Jeju monitoring sites in the Republic of Korea also showed fluctuating cyclical patterns of sulphuric gas and aerosol presence. Sulphur component concentrations reached their maximum values in winter (around January) and their minimum values in summer except in 2005 and 2007. The SO_2 concentration at Ganghwa for this period was noticeably low in mid-2007, while sulphate concentrations conversely peaked at this time. At the remote site of Jeju too, the observation value for SO_2 in 2008 was very low (showing a monthly average of just 0.1 ppb) compared to those of other years, while the concentration of sulphate components was exceptionally high in the same year.

Although sulphuric gas and aerosol concentrations in Japan showed similarities with those of other Northeast Asian sites, where values exhibit seasonal variability, there was a delay of two to three months in the sulphate peak after the SO_2 concentration reached its maximum value. For the remote island site at Ogasawara, observed SO_2 concentrations were almost all negligible during the period 2005 – 2009 except in 2008.

HNO_3 and NO_3^- values also showed seasonal fluctuations. The rural site at Primorskaya (Russia) exhibited a clear cyclical pattern as components reached their maximum values in winter and their minimum values in mid-summer. Concentrations of nitrate components were noticeably low at Terelj (Mongolia) and at Irkutsk, Listvyanka and Primorskaya (Russia) in certain years. Accordingly, there appears to be no relationship between these components and the large existing amounts of HNO_3. Generally, NO_3^- concentrations were three to four times higher than those of HNO_3.

A clear pattern of seasonal variation for NH_3 and NH_4^+ is seen at most northern inland Asian sites except those in Russia (Listvyanka, Primorskaya and Mondy). NH_3 concentrations were high in winter and became very low in summer, while the corresponding values for NH_4^+ showed an opposite trend. The high release of NH_3 during the warm season is considered to be a result of agricultural activities, while the high NH_4^+ values seen in winter are attributed to slow biological reaction. It is also observed that amounts of NH_3 and NH_4^+ release do not differ significantly among urban, rural and remote sites. In terms of the seasonal variation trend for NH_3 and NH_4^+ concentrations at the Pacific island monitoring sites (the Republic of Korea and Japan), the values for both were also high in winter and low in summer. However, in terms of quantity, no significant difference in either component was seen amongrban, rural and remotes sites.

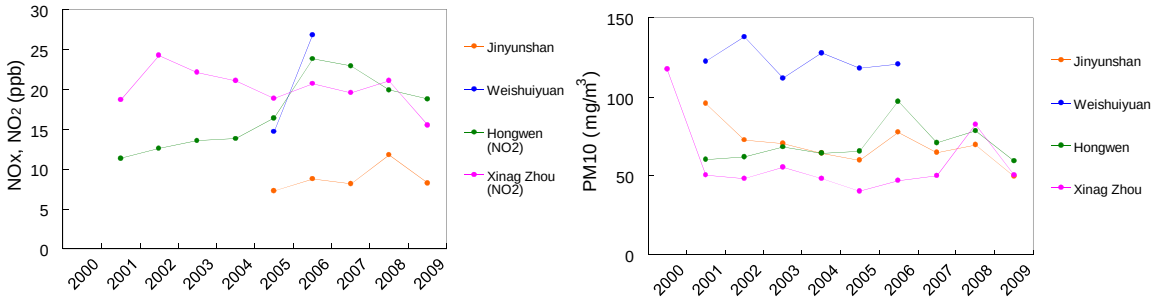


Figure 5.2. Annual variations of NOx, NO_2 and PM_{10} concentrations in China for the period 2001 – 2009.

Figure 5.2 shows annual variations of NOx, NO_2 and PM_{10} concentrations in China for the period 2001 – 2009. The values for NO_2 and PM_{10} show a decreasing trend, especially since 2006, which may imply recent air quality improvement. On the other hand, as shown in Figure 5.3, average monthly O_3 concentrations from 2005 to 2009 at many Japanese sites were higher than those seen from 2000 to 2004. This may be partially due to long-range transportation of oxidants in Northeast Asia. At most Japanese sites, the average monthly PM_{10} concentrations from 2005 to 2009 were lower than those seen from 2000 to 2004. The reduced high-percentile concentrations observed in spring are mainly attributable to the influence of monthly and annual average concentrations

(Figure 5.4).

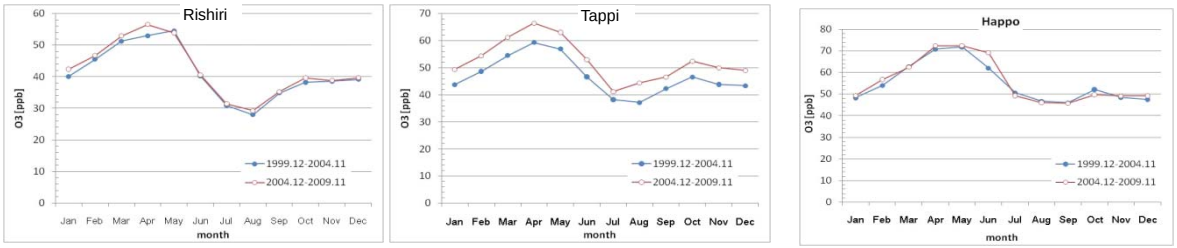


Figure 5.3. Comparison of five-year average monthly O_3 concentrations between the periods 1999 – 2004 and 2005 – 2009 at Rishiri, Tappi and Happo in Japan.

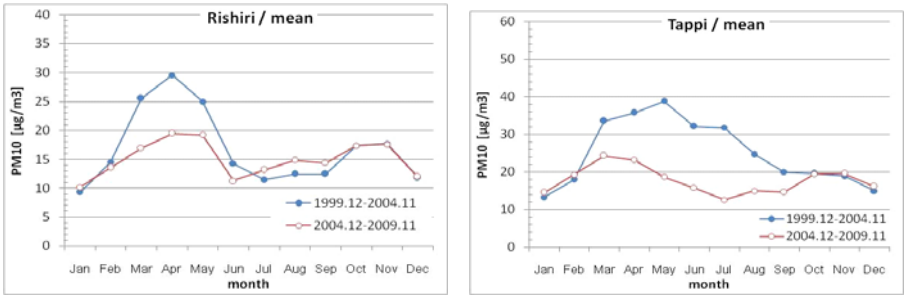


Figure 5.4. Comparison of five-year average monthly PM_{10} concentrations between the periods 1999 – 2004 and 2005 – 2009 at Rishiri and Tappi in Japan.

All urban EANET sites in the Southeast Asian region showed significant SO_2 and SO_4^{2-} emission reductions during the period 2005 – 2009, which may be related to the pollution reduction policies implemented in most cities in the region. Comparison of average measured values from 2005 with those of 2009 shows significant SO_2 reductions from 7.3 to 1.9 at Bangkok, from 2.4 to 1.3 at Hanoi, from 2.7 to 0.9 at Hoa Binh, from 2.7 to 0.9 at Metro Manila (2008), from 3.8 to 1.8 at Petaling Jaya, and from 11.0 to 8.3 at Jakarta. A reduction was also seen in the related chemical species of SO_4^{2-} during the period 2005 – 2009 in approximately the same magnitude as that of SO_2 for all urban sites except Bangkok, where no clear trend of reduction was observed.

Concentrations of SO_2 and SO_4^{2-} at rural and remote sites are likely to exhibit seasonal independency because the climatology of Southeast Asia is invariant, especially in countries near the equator such as Malaysia and Indonesia. It is therefore presumed that fluctuations in ambient concentrations stem from mobile and industrial emissions.

The concentrations of HNO_3 and NO_3^- at the inland monitoring sites in Thailand and Vietnam are more affected by seasonal variations than those located close to the Pacific Ocean due to the inland climate, which is characterized by dry and wet seasons. The observed concentrations are low during the hot and wet season (around the middle of the year) and high during the cool and dry season (around the end of the year). The low concentrations are believed to stem from the washing-out effects of heavy monsoon rains. Conversely, countries close to the Pacific Ocean (such as Malaysia, Indonesia and the Philippines) are under the influence of rainforest climatology in which the ambient temperature does not vary significantly and high monthly rainfall totals are seen. No clear seasonal fluctuations in concentration are observed. It should be noted that levels of HNO_3 and NO_3^- for the remote site at Mt. Sto. Tomas (Philippines) were very high in 2008. For the remote site at Kanchanaburi (Thailand), concentrations were measured only three times a year (in March, July and November) in consideration of the practicality of transporting a mobile monitoring unit to the site, which is located in a high mountainous area.

Observed concentrations of NH_3 and NH_4^+ at urban, rural and remote sites in Thailand followed

seasonal variation patterns influenced by the tropical climate rather than by agricultural cultivation cycles. High concentrations are seen in the dry season and very low concentrations are seen in the wet season, even though cultivation is carried out all year round, because the climate is warm throughout the year. At the rural site of Nakhon Ratchasima, NH_3 concentration was very low compared to that of NH_4^+. This indicated a weaker impact from urban pollutants created by photochemical reaction.

The monitoring sites at urban Hanoi and rural Hoa Binh in Vietnam also showed cycles of seasonal variation in 2005 and 2006. However, concentrations of NH_3 from 2007 to 2009 remained constantly high throughout the year, while those of NH_4^+ were noticeably low. This was considered to stem from the expansion of city communities in recent years and the consequently increased effects of photochemical reaction.

For sites located near the Pacific Ocean, seasonal variation cycles of chemical concentrations were seen (although not as clearly as those of Thailand and Vietnam) at urban Petaling Jaya and rural Tanah Rata (Malaysia) and at rural Serpong (Indonesia). Concentrations of NH_3 and NH_4^+ were high in the middle of the year and low toward the end of the year. The cycle pattern differed from those seen in inland Southeast Asia, where heavy rainfall occurs in June and July. Due to the characteristic climatology of the region around the equator, heavy rainfall in Malaysia and Indonesia occurs in November and December. It should be noted that the chemical concentration at the remote Danum Valley site (Malaysia) increased substantially toward the end of 2009.

The concentrations of NH_3 and NH_4^+ observed at urban Metro Manila, rural Los Baños and remote Mt. Sto. Tomas in the Philippines generally did not show seasonal variations during the period 2005 – 2009. In 2008, the values for both components were exceptionally high at certain times of the year.

In the Southeast Asian region, O_3 concentrations at some sites in Thailand showed an increasing trend, but no significant trends were seen in other data. Variations in PM concentration values during specific periods are notable because severe adverse atmospheric effects have been reported during haze events in the dry season.

Characteristics of Overall Deposition

Figure 5.5 shows the mean dry, wet and total deposition amounts for sulphur and nitrogen compounds for Japanese EANET sites, CASTNET sites in the US and EMEP sites in Europe. It can be seen that the five-year average total deposition amounts of sulphur and nitrogen compounds in Japan were higher than those of the other networks. This was due to the higher precipitation amounts and high nss-SO_4^{2-} concentrations of wet deposition observed in Japan. The higher deposition amounts of sulphur compounds were also possibly caused by long-range transboundary air pollution and natural emission sources, such as volcanic eruptions in the country. Total deposition amounts at some sites in Japan have increased remarkably in recent years. Increasing SO_2 and NOx emissions in East Asia result in larger amounts of atmospheric deposition, making overall deposition in Japan higher than that in other networks.

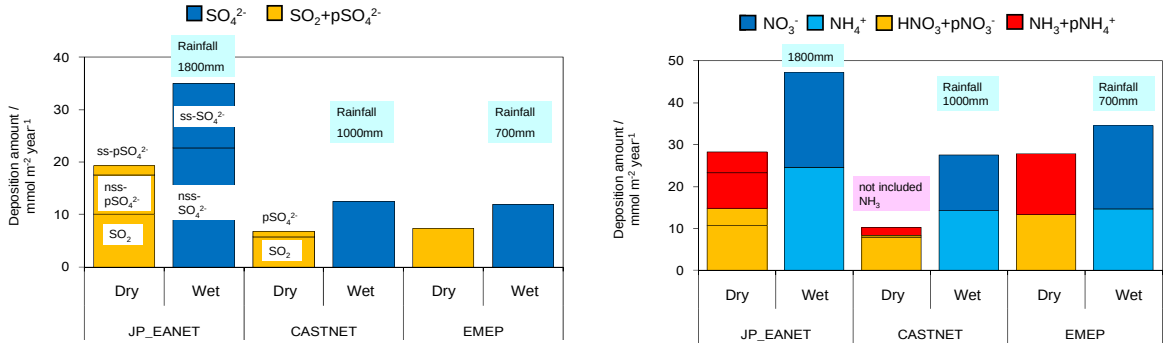


Figure 5.5. Comparison of dry, wet and total deposition amounts of sulphur and nitrogen compounds for Japanese EANET sites, CASTNET sites in the US and EMEP sites in Europe (averages for 2003 – 2007).

6. Impacts on Ecosystems

Trends in Soil, Vegetation and Inland Aquatic Environments

Clarifying acid deposition's impacts on ecosystems is a vital mandate of EANET. Against this background, network countries conduct ecological monitoring of soil, forest vegetation and inland aquatic environments at strategic locations in line with EANET prescriptions and guidelines. Based on data accumulated over the previous decade, trends in soil chemistry, forest vegetation and inland water chemistry were investigated.

Soil trends: Soil is monitored several times over a period of 10 years (2000 – 2009) at 20 sites encompassing around 40 plots and 200 subplots in the seven countries of China, Indonesia, Japan, the Republic of Korea, the Philippines, Russia and Thailand. Mean values of chemical presence in soil showed large inter-site variations. Values at a depth of 0 – 10 cm ranged from pH 4.0 to 6.8 for H_2O, pH 3.4 – 5.8 for KCl, and 0.05 – 26.0 $cmol_c\ kg^{-1}$ for ex-Ca^{2+}. The coefficient of variance (CV) in the regional variation was 18% in terms of pH for H_2O at 0 – 10 cm.

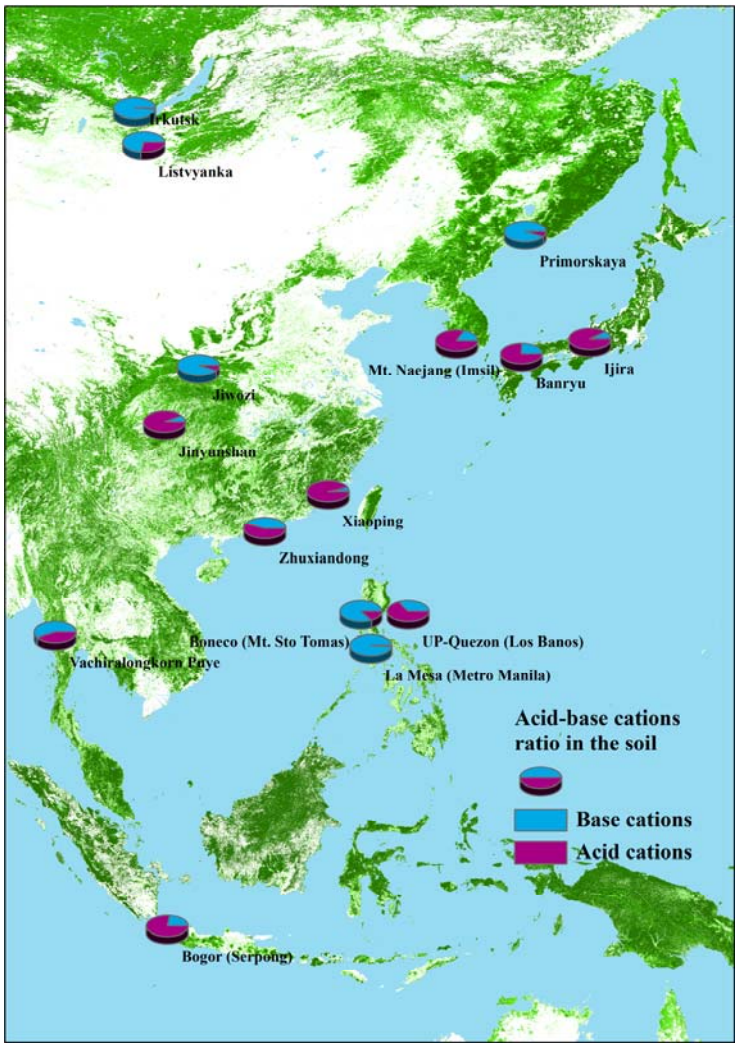


Figure 6.1. Ratio of base cations (Ca^{2+} + Mg^{2+} + Na^{+} + K^{+}) to acid cations (H^{+}+ Al^{3+}) in exchangeable ions in surface (0 – 10 cm) soil. Representative sites for each area are displayed. Dense green shading indicates tree cover. Values are averages for the period 2000 – 2009.

Figure 6.1 shows regional variations in the ratio between exchangeable base cations (Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}) and acid cations (H^{+}, Al^{3+}) adsorbed in the negative charge of surface clay minerals or organic components of soil. This ratio indicates the buffering capacity of soil against atmospheric acid or base depositions. Base cations were dominant in the soil at sites including those in Russia, Jiwozi (China), Vachiralongkorn (Thailand), Boneco (Mt. Sto. Tomas, Philippines) and La Mesa (Metro Manila, Philippines), whereas acid cations showed significant dominance at many other sites. In general, these regional variations mainly stem from soil type differences depending on geological and climatic conditions as well as topographical and biological factors over a long time scale.

In the variation, the pH of H_2O at a depth of 0 – 10 cm showed a correlation with the value for Ex-Ca^{2+} (Figure 6.2 a). This relationship indicates that base cation concentrations are crucial for soil pH in the area except for Russia. In general, high Ex-Ca^{2+} concentrations and pH values were observed on basic rock such as limestone (e.g., at Vachiralongkorn). The buffering capacity against acid deposition should be much higher in the area. Meanwhile, as shown in Figure 6.2 b, the pH of H_2O under the influence of base cations strongly regulates the solubility of aluminum ions (Al^{3+}), which are harmful to plant roots (Ulich, 1990). Over 70% of samples showed pH values of less than 5.4 with aluminum present in the form of Al^{3+} (Figure 6.2 b). A pH decline may lead to increased Al^{3+} presence at many monitoring sites, although such increases may also be related to site characteristics.

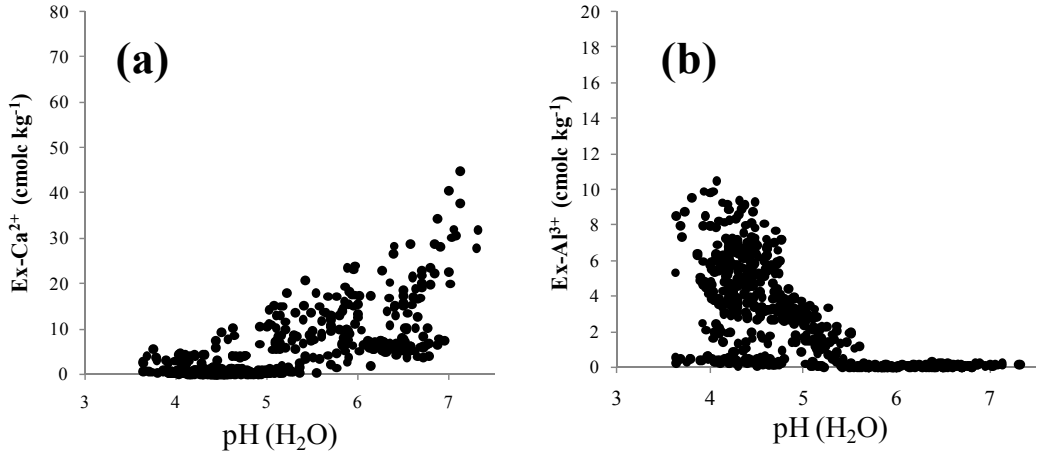


Figure 6.2. Scatter plots for pH and Ex-Ca^{2+} (a) and for pH and Ex-Al^{3+} (b) at depths of 0 – 10 and 10 – 20 cm from 2000 to 2009 for all monitoring site except those in Russia.

As for temporal trends in pH (H_2O), base saturation and exchangeable acidity at depths of 0 – 10 cm, pH continuously increased in China at Jinyunshan, in the Philippines at Mt. Makiling-1 (in the area of Los Baños) and at UP-Quezon (Los Baños) and in Thailand at Vachiralongkorn Dam-1, and decreased only in the Republic of Korea at Mt. Naejang (Imsil). Base saturation decreased significantly in China at Jinyunshan from 2003 to 2006, at Jiwozi from 2006 to 2009 and at Zhuxiandong from 2003 to 2006 and in the Republic of Korea at Mt. Naejang (Imsil) from 2001 to 2004, whereas values increased in Japan at Banryu-1. In the Philippines at UP-Quezon (Los Baños), base saturation significantly decreased from 2000 to 2005 and increased from 2005 to 2009. Exchangeable acidity significantly increased in China at Jiwozi from 2006 to 2009 and at Xiaoping from 2002 to 2005, in Indonesia at Bogor (Serpong) from 2001 to 2006, in the Republic of Korea at Mt. Naejang (Imsil) from 2000 to 2004, and in the Philippines at UP-Quezon (Los Baños) from 2000 to 2009.

The changes in pH noted above did not correspond to those of acid and/or base cations, suggesting the effects of other factors such as organic matter. Nutrient imbalance may also be a cause of changes in the chemical properties of soil. A significant reduction in soil C/N (total-C/total-N) was

observed in China (Jinyunshan, Xiaoping, and Zhuxiandong), Indonesia (Bogor) and Japan (Banryu), although total-C and N concentrations were not mandatory items in soil monitoring. The changes in the C/N ratio may be related to nitrogen saturation as discussed below.

Vegetation trends: The eight countries of China, Indonesia, Japan, the Republic of Korea, Mongolia, the Philippines, Russia and Thailand monitored for impacts on forest vegetation. The monitoring plots varied in terms of species composition, and included monocultures (pure stands), mixed stands dominated by a single species, and highly mixed stands where no species dominated. The pure stands mostly consisted of conifers such as *Pinus elliottii, Pinus massoniana* and *Chamaecyparis obtusa*. There were also pure stands of *Michelia macclurei* and *Acacia confusa*. Pure stands accounted for 33% of the plots established, while the rest were mixed stands (67%) with highly variable species compositions, however, dipterocarps species are the dominant species at the UP Quezon site in the Philippines and in Thailand. While, the dominant species in China and Japan is the genus *Castanopsis*.

An overall vitality decline due to dense planting was observed for *Pinus armandi* at Jiwozi (Dabagou, China) after 2003, while a decline due to pest infestation was observed for *Pinus massoniana* at Zhuxiandong-1 from the same year. A decline in *Acacia confusa* at Zhuxiandong-2 was also reported**.** In Primorskaya, Russia, all monitored trees showed symptoms of decline due to pest infestation in 2006. Likewise, occasional pest and disease infestation reduced the vitality of *Chamaecyparis obtusa* at Lake Ijira in Japan. In the Philippines, Thailand and Indonesia, tree decline was not seen during the period 2000 – 2009, probably due to dense canopy cover in forest areas. However, in 2008, tree decline caused by termite infestation was reported in the Philippines. There was no evidence of decline attributable to air pollution or acid deposition. Decreases in biometric figures for trees at certain monitoring plots in the region were attributed to natural phenomena such as typhoons, pest infestation, infectious tree diseases and other anthropogenic factors.

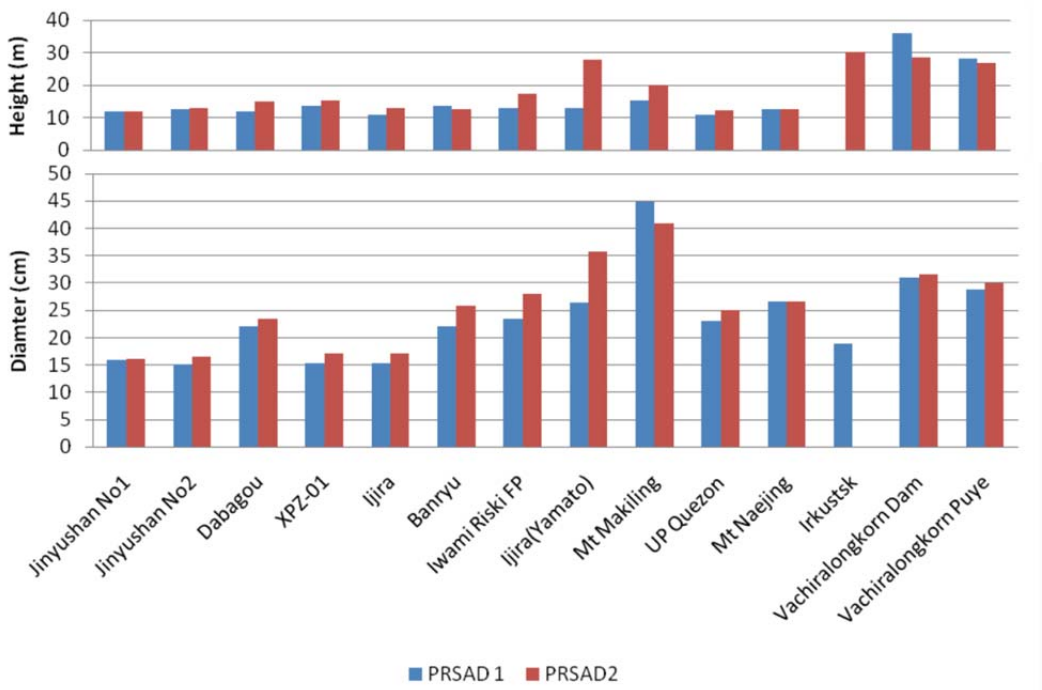


Figure 6.3. Changes in diameter at breast height (DBH: bottom) and average height (top) from the period 2000 – 2004 (PRSAD1) to 2005 – 2009 (PRSAD2). Dabagou and XPZ-01 (Xiaoping) are plots in Jiwozi and Zhuxiandong, respectively.

Based on data accumulated over the previous 10 years, increased DBH and height values were noted in comparison between the 2000 – 2004 (PRSAD1) and 2005 – 2009 (PRSAD2) periods, except for an average diameter reduction at Mt. Makiling in the Philippines (Figure 6.3). This decline in diameter may have been due to the death of large canopy trees in the area. The number

of understory species increased between PRSAD1 and PRSAD2 except in Vachiralongkorn Puye. The increase in recruitment (new species) between these two periods indicated that overall forest functions and structure were maintained. Most recruitment was of the general type.
Overall, there was no decline in growth or new recruitment at EANET ecological monitoring sites between the two reporting periods. The decline symptoms observed were often temporary in nature and caused by biotic causal agents.

Inland aquatic environment trends: As of 2009, there were 18 EANET sites for monitoring of inland aquatic environments – 4 in China, 2 each in Indonesia, Japan, Malaysia, the Philippines and the Russian Federation, and 1 each in Lao PDR, Mongolia, Thailand and Vietnam. Six rivers and twelve lakes have monitoring sites. Among the lakes, six are artificial (reservoirs/dams) and six are natural.

Based on data accumulated over a period of 10 years, significant trends are observed at several sites. The data obtained from quarterly sampling were applied for the non-parametric Seasonal Mann-Kendall method, and the results are shown in Table 6.1 (several new sites were not included in the trend analysis).

Table 6.1 Trends of inland water chemistry.

Country	site	pH	EC	Alkalinity	SO_4^{2-}	NO_3^-
China	Jinyunsyan Lake	-3.34 ***	4.34 ***	-2.69 **	3.44 ***	2.50 *
	Xiaopin Dam	-2.35 *	0.06	-0.91	2.40 *	0.21
	Jiwozi River	-2.11 *	2.92 **	3.46 ***	3.68 ***	0.87
	Zhuxiandong Stream	0.39	-1.88	-0.38	-2.27 *	-0.09
Indonesia	Patengang Lake	-2.36 *	-1.79	-1.40	2.35 *	1.23
Japan	Ijira Lake	1.49	0.65	3.19 **	1.06	-3.01 **
	Kamagatani River flowing river to Ijira Lake	3.05 **	1.79	3.81 ***	1.25	-1.39
	Kobara River flowing river to Ijira Lake	2.47 *	1.83	3.09 **	2.34 *	-1.70
	Banryu Lake	-0.67	2.47 *	-0.72	1.17	2.60 **
	Banryu Lake 3	-0.67	2.20 *	-0.49	1.57	1.86
Malaysia	Semenih Dam	-2.87 **	0.27	0.46	2.09 *	-1.59
Mongolia	Terelj River	0.60	1.53	2.69 **	1.37	-1.04
Phillipines	Pandin Lake	-1.09	-0.89	-1.40	3.27 **	3.91 ***
Russia	Pereemnaya River	-0.94	4.46 ***	0.59	4.70 ***	2.63 **
	Komarovka River	-0.37	0.49	-3.32 ***	2.92 **	0.49
Thailand	Vachiralongkorn Dam 1 Ban Pong Chang	0.87	-2.98 **	0.57	-2.07 *	1.81
	Vachiralongkorn Dam 2 Ban Pang Pueng	1.45	-3.39 ***	1.06	-1.34	2.44 *
Vietnam	Hoa Binh Reservoir	0.36	1.61	-1.37	1.61	1.70

*Note: Values show Z scores from the seasonal Mann-Kendall test. Negative and positive values show declining and increasing trends, respectively. Asterisks show levels of significance: *: P < 0.05; **: P < 0.01; and ***: P < 0.001. Significant declining and increasing trends are highlighted in pink and blue, respectively.*

The pH of inland water declined significantly at the five monitoring sites of Jinyunshan Lake, Xiaoping Dam, Jiwozi River, Patengang Lake and Semenyih Dam. In these acidified lakes/rivers, significant simultaneous increases were also seen in SO_4^{2-} concentrations. The concentration of NO_3^- also increased significantly at Jinyunshan Lake. It is possible that leaching of SO_4^{2-} and/or NO_3^- caused leaching of H^+, resulting in water acidification. However, water chemistry seems to have recovered during this period in the Lake Ijira catchment area, which had been reported as acidified and nitrogen-saturated (Nakahara *et al*., 2010, *Biogeochemistry*).

Relationships Linking Ecological Trends

Although data on atmospheric deposition and ecological considerations are collected on a mutually independent basis, correspondence among the results is seen at several sites where surveys on the items detailed above have been conducted.

As described previously, the pH of soil collected at China's Jinyunshan site increased, while that of inland water in the area decreased. Adsorption of anions by soil may be generally regulated by soil pH, as soil can have anion exchange capacity. In general, when soil pH is low and free H^+ exists in a soil solution, adsorption of SO_4^{2-} is enhanced. Conversely, with increased soil pH, adsorbed SO_4^{2-} is released with H^+ into soil water, and subsequently into lake/river water. Probably, this is why the soil pH trend corresponded closely to that of lake water chemistry in the case of Jinyunshan Lake.

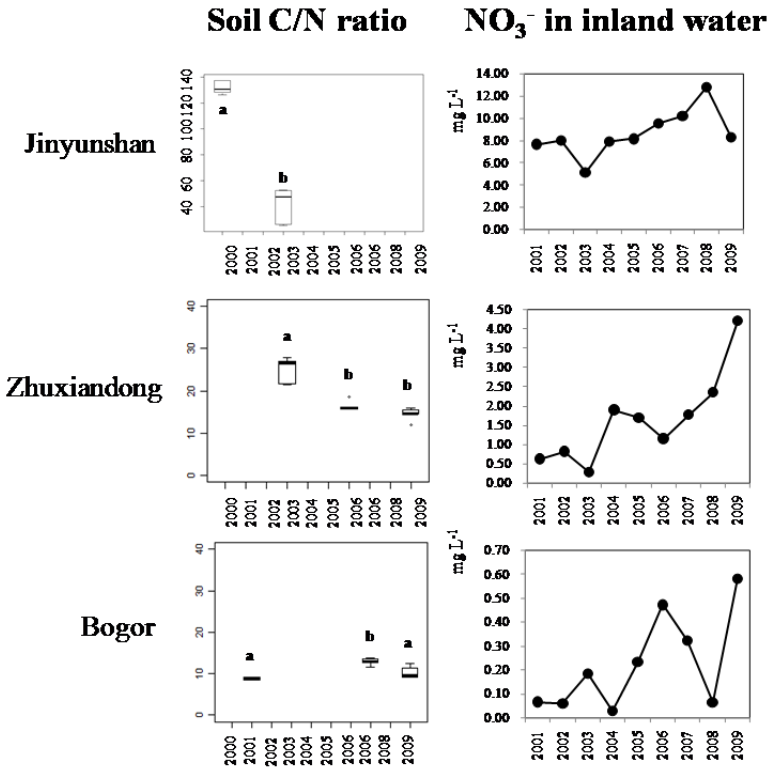


Figure 6.4. Trends in the C/N ratio in surface soil and NO_3^- concentration in inland water. In Zhuxiandong, the sampling point for inland water was changed from a lake location to a stream location in 2004. The data for inland water in Bogor are from Patengang Lake.

At several monitoring sites, temporal variations in soil C/N and NO_3^- concentration in lake or stream water can be compared. As mentioned above, soil C/N decreased significantly at Jinyunshan, Xiaoping, Zhuxiandong, Bogor and Banryu. In the same period, NO_3^- concentrations in lake/stream water were seen to increase at Jinyunshan Lake, Zhuxiandong Stream and Patengang Lake (Figure

6.4). Excess NO_3^- in the soil solution, which is indicated by low C/N values, may flow into streams and lead to increased NO_3^- concentration in lakes. As described above, reduced base saturation was observed at Jinyunshan and Zhuxiandong, probably because NO_3^- leaching was accompanied by base cations in the soil solution (Binkly, 1987). In forests, C/N content in leaf litter or top mineral soil is a good indicator of the ecosystem's N status. NO_3^- leaching increases with higher levels of N deposition in many European forests where low C/N values in leaf litter are observed, creating a state known as nitrogen saturation. Terrestrial eutrophication, soil acidification and fresh-water eutrophication occur in stages as a result of excessive N input, which may lead to this nitrogen saturation in forest areas. A tendency of nitrogen saturation has been reported in subtropical China (Chen and Mulder, 2006, Fang *et al*., 2008, and Fang *et al*., 2009) and in the Lake Ijira catchment area of Japan (Nakahara *et al.*, 2010).

Meanwhile, at UP-Quezon (Los Baños), forest rehabilitation was observed as young trees (DBH < 5 cm) became dominant in 2002, while the number of medium-sized trees (DBH > 7.5) increased in 2008. As described above, soil pH (H_2O) increased continuously during the same period. Forest growth may lead to increased soil pH as a result of greater amounts of organic matter, which may explain the increase in base saturation with exchangeable acidity observed here. This suggests that forest conditions (growth or degradation) are also key factors controlling the chemical properties of soil.

Catchment-scale Analysis in EANET Countries

The anticipated progress of monitoring in the coming years is expected to create a growing need for careful discussion of the relationships linking ecosystem components, including soil, vegetation, inland water and the atmosphere. Catchment-scale monitoring plays a role in supporting this requirement.

Joint research projects have been conducted at tropical rainforest, tropical seasonal forest and temperate coniferous forest sites at Danum Valley in Sabah State (Malaysia) Sakaerat in Nakhon Ratchasima Province (Thailand) and Kajikawa in Niigata Prefecture (Japan). As a result, the biogeochemical characteristics of East Asian forest ecosystems and the possible impacts of atmospheric deposition have been clarified and reported in international journals. Based on the scientific/technical experience gained through these projects, the *Guideline for Catchment-scale Monitoring in East Asia* was developed and endorsed by the Scientific Advisory Committee at its 10th session in 2010, and is expected to be used in regular catchment-scale monitoring in EANET countries. With this development, the monitoring initiatives of EANET take on a new dimension in further elucidating the acid deposition phenomenon and its concomitant effects in the East Asian region.

7. Related Studies in EANET Countries*

Project Activities Related to EANET Studies

Acid deposition monitoring in EANET countries provides important information helping to clarify the effects of acidification and the long-term trends of causative agents and other related air pollutants. The long-term monitoring data accumulated are useful for evaluating impacts on ecosystems and human health. Accordingly, field campaigns investigating acid deposition and regional air quality in East Asia are conducted as related studies. Sites for such work include central eastern China and the Pearl River Delta in southeastern China, Gosan on Jeju Island in the Republic of Korea, Fukue Island in western Japan and Cape Hedo in southern Japan. Initiatives include the Atmospheric Brown Cloud (ABC) project in South Asia (in the tropical Indian Ocean, the Arabian Sea and the Bay of Bengal), which also includes parts of Northwest Asia (Saudi Arabia, Pakistan, Afghanistan) and the Aerosol Characterization Experiment (ACE) project encompassing the Yellow Sea, the East China Sea, the seas around northern Japan and the region downwind of Japan. Investigation of O_3, CO, black carbon (BC), organic carbon (OC) and aerosols has shown that their emissions have significant impacts not only on primary pollutants but also on secondary pollutions such as O_3.

Acidification and nitrogen leaching in Japanese catchment areas have also been noted at sites such as Lake Ijira in central Japan. Deposition of H^+ and nitrogen in the Lake Ijira catchment area began to increase in 1994, and sulphur deposition had decreased until the same year (Nakahara *et al*., 2010). It was suggested that these trends were partly responsible for those seen in stream water pH, NO_3^- concentration and SO_4^{2-} concentration during the corresponding periods. As a result, river water acidification was reported in the three adjoining prefectures of Gifu, Nagano and Niigata in central Japan. Acidification trends were seen in rivers originating in these areas, which were dominated by low acid-neutralizing-capacity (ANC) geological materials such as granite, rhyolite and chert. In fact, river water in these areas had low pH, EC and alkalinity, reflecting the geology of the respective regions (Matsubara *et al*., 2010).

The facts suggest that high acid deposition loads may cause acidification of surface water, particularly in acid-sensitive catchment areas. Accordingly, areas characterized by low-ANC geology and high acid deposition should be carefully monitored as acidification hotspots. Nitrogen leaching hotspots should also be considered based on vegetation characteristics and nitrogen deposition amounts, although it is still uncertain whether the threshold value of 10 $kg.ha^{-1}.yr^{-1}$ is directly applicable to Japanese catchment areas. Nonetheless, in addition to the implementation of continuous monitoring in acidified catchment areas, identification of hotspots should also be promoted taking into account the distribution of geology, soil, vegetation and atmospheric deposition.

Emission Inventories

As a result of rapid development in industry and commerce, anthropogenic emissions in the Asian region are higher than those in Europe and North America, and will continue to increase in the future (Akimoto, 2003). It is therefore essential to develop emission inventories of air pollutants in Asia as efficient tools for managing the air quality and gridded emission datasets used in atmospheric chemistry model simulations. Essentially, emission amounts for each country are estimated based on combinations of activity statistics (such as those on fuel consumption, industrial production and heads of livestock) and source- or region-specific emission factors. Gridded data are developed by allocating emissions to grid cells using the spatial distribution of the population and the road network.

**It should be noted that some external scientific data and the conclusion in this chapter do not represent the official views of EANET.*

Large power stations and industrial plants are often treated as large point sources (LPSs), and emissions from these points are allocated to the grids in which the plants are located. In this way, the emission inventory details the amounts and types of air pollutants released into the atmosphere by source category for a specific geographic area over a particular time period.

Ten years ago, the number of emission inventories for the Asian region was very limited. Those developed more recently include REAS (the Regional Emission Inventory in Asia; Ohara *et al.*, 2007), GAINS (Greenhouse Gas and Air Pollution Interactions and Synergies; Klimont *et al.*, 2009), and INTEX-B (Intercontinental Chemical Transport Experiment-Phase B; Zhang *et al.*, 2009). Figure 7.1 compares Asian emissions of NO_x and SO_2 for these inventories from 1980 to the latter half of the 2000s.

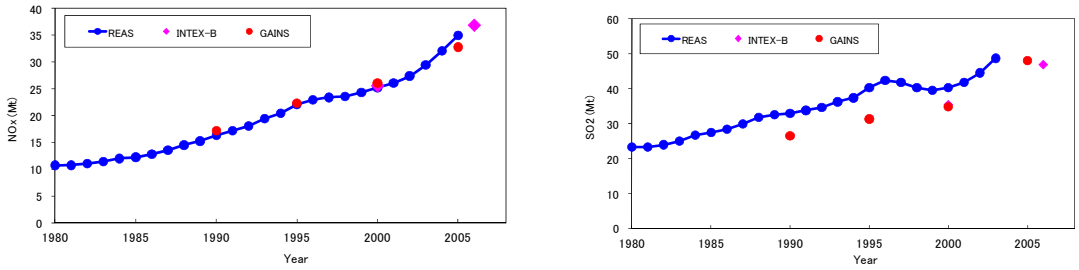


Figure 7.1. Time series of NO_x and SO_2 emissions in Asia.

In Asian total NO_x emissions for 2000 and 2005, the values of REAS, GAINS and INTEX-B are within a 10% variance (25.1 – 26.1 Mt for 2000 and 32.9 – 36.8 Mt for 2005). Total emissions of NO_x in Asia showed a monotonic increase during the period 1980 – 2006 with no dips, in contrast to the pattern of SO_2 emissions. According to the REAS inventory, emissions increased by a factor of 3.5 from 1980 to 2005, with values of 10.7 Mt in 1980 and 35.0 Mt in 2005. NO_x emissions in China increased dramatically by a factor of 5.0 from 1980 to 2005, with higher growth seen after 2000 (involving increase by a factor of 1.7 over only five years (Ohara *et al*., 2007)). These trends of China's NO_x emissions in REAS were consistent with those in other inventories, including GAINS, INTEX-B and Chinese groups' results (Hao *et al*., 2002; Tian *et al*., 2005). Historical estimates of SO_2 (1990 – 2006) in Asia show similar patterns in all studies, although absolute values vary significantly. According to the REAS inventory, total emissions of SO_2 in Asia increased from 1980 to 1996, but then decreased until 1999, reflecting a reduction in fuel consumption due to the Asian economic crisis. After 2000, SO_2 emissions grew at a phenomenal rate. In the period 1980 – 2003, they increased by a factor of 2.2, and notably by factors of 3.2 in India and 2.5 in China (Ohara *et al*., 2007). For SO_2 emissions in 2000 in Asia, the GAINS value (34.9 Mt) was almost the same as the INTEX-B value (35.5 Mt) but lower than REAS (41.2 Mt). In more recent years, the value for GAINS (48.1 Mt) for 2005 was similar to that for INTEX-B (47.0 Mt) for 2006.

In addition to Asian emission inventories, datasets from global inventories are also often used for analysis of air quality in the Asian region, not only as input data for global atmospheric chemistry modeling but also for the comparison of amounts and trends of emissions between Asia and other regions. Global emission inventories that comprehensively cover pollutants including NO_x and SO_2 during the period 1980 – 2010 are summarized in Table 7.1.

Table 7.1 Global emission inventories for anthropogenic emissions from 1980 to 2010.

Inventory	Year	Spatial resolution	Author, website
ACCMIP	1980 – 2010	0.5 deg.	Lamarque *et al*., 2010 http://ether.ipsl.jussieu.fr – ECCAD
ACCity	1980 – 2010	0.5 deg.	http://ether.ipsl.jussieu.fr – ECCAD
RCPs	2000 – 2010	0.5 deg.	http://ether.ipsl.jussieu.fr – ECCAD
EDGAR v3	1990, 1995, 2000	1 deg	Olivier *et al*., 2005; http://edgar.jrc.ec.europa.eu
EDGAR v4.1	1970 – 2005	1, 0.5, 0.1 deg.	http://edgar.jrc.ec.europa.eu
HYDE	1980, 1990	1 deg	Van Aardenne *et al*., 2001 http://www.pbl.nl/en/themasites/hyde/index.html
RETRO	1980 – 2000	0.5 deg.	http://retro.enes.org/data_emissions.shtml
GAINS	1990 – 2010	-	http://gains.iiasa.ac.at/gains

For global NO_x emissions, all inventories show a slight increase during the period 1980 – 2010. Conversely, most inventories of SO_2 indicate a significant decrease in global emissions during the same period. However, as indicated in Figure 7.1, Asian emissions of NO_x and SO_2 increased significantly during the period 1980 – 2010. Accordingly, there is an urgent need for accurate estimation of Asian emissions.

The global and regional emission inventories described above were developed by scientific researchers. In addition to these data, national emission inventories are also reported, mostly by individual governments. One advantage of government-based inventories is that they are sometimes based on more detailed country-specific information such as emission factors and activity data that are not easily accessible by researchers. Another significant advantage is that they are intended for official use. It is highly desirable to unify researcher-based and government-based inventories because it is now recognized that long-range trans-boundary air pollution must be taken into account in many cases even in the evaluation of urban and industrial air pollution. However, no harmonized system for the compilation of inventories that are transparent and comparable enough to be agreed upon among East Asian countries has yet been established. In order to deepen understanding of the situation of regional air pollution in East Asia and develop the atmospheric environmental management capabilities of individual countries, there is a need to establish a system in which inventories can be improved continuously in collaboration with administrative agencies and inventory experts in East Asia.

Chemical Transport Modeling Studies

The Model Inter-comparison Study for Asia (MICS-Asia) was an initiative designed to help develop a better common understanding of the performance and uncertainties of chemical transport models (CTMs) in East Asia. MICS-Asia II was carried out during the period from 2003 to 2007 in association with ACAP and IIASA as part of EANET's additional activities. The study's output was published in the form of several papers in Atmospheric Environment (an international journal on research in the field of air pollution and its societal impacts) in 2008. The scope of MICS-Asia II was expanded to include nitrogen compounds, O_3 and aerosols in addition to the sulphur compounds studied in the precedent MICS-Asia I initiative, and four months in different seasons were examined (March, July and December 2001, and March 2002). Newly obtained EANET observational data from these years were fully utilized in the form of a region-wide database for comparison. Nine regional modeling groups participated in the studies.

Comparison of simulated and observed monthly mean values of SO_2 indicated spatial variability at higher levels over China and moderate levels over Japan, reflecting the emission intensities of the respective regions. Systematic over-prediction at Weishuiyuan in China and Ijira in Japan was

considered to likely stem from the rather coarse resolution of the models and the resultant difficulty of distinguishing the gradient for rural sites that may be located in or near grid cells containing significant sulphur sources. The ensemble mean correlation coefficient over all available sites for the combination of the four periods was 0.51. Similar comparisons were also performed for depositions of sulphur, nitrate and ammonium, and for concentrations of O_3. Figure 7.2 shows the ensemble mean monthly mean total deposition amounts of sulphur in the form of sulphate for March and July 2001. Wet deposition of sulphate and dry deposition of SO_2 are the two dominant modes of sulphur deposition. The majority of deposition occurs in southern China, over the western Pacific and in Japan during spring. In summer, the high-deposition regions move northward, creating one deposition center in northern China and another in the southwestern area of the country. For nitrate, the ensemble results show that in spring, high wet deposition is seen in Japan, southeastern China,

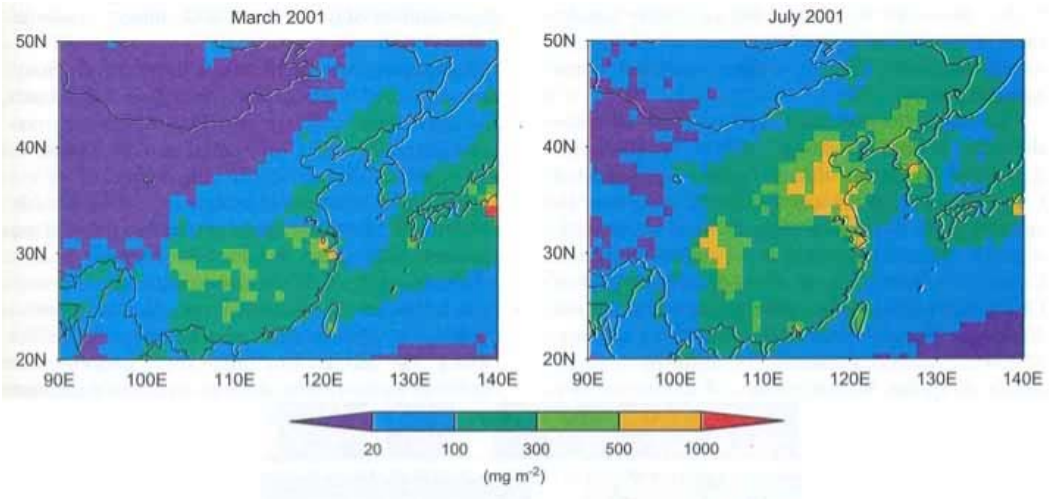


Figure 7.2. Ensemble mean monthly mean total sulphur deposition amounts (mg m^{-3} as sulphate) for March and July 2001. *(Carmichael et al., 2008).*

parts of northeastern China, and the Republic of Korea. In summer, the NO_3^- high wet deposition center moves northward following the motion of the rain belt in China, and also toward parts of Southeast Asia where the wet season is prevalent.

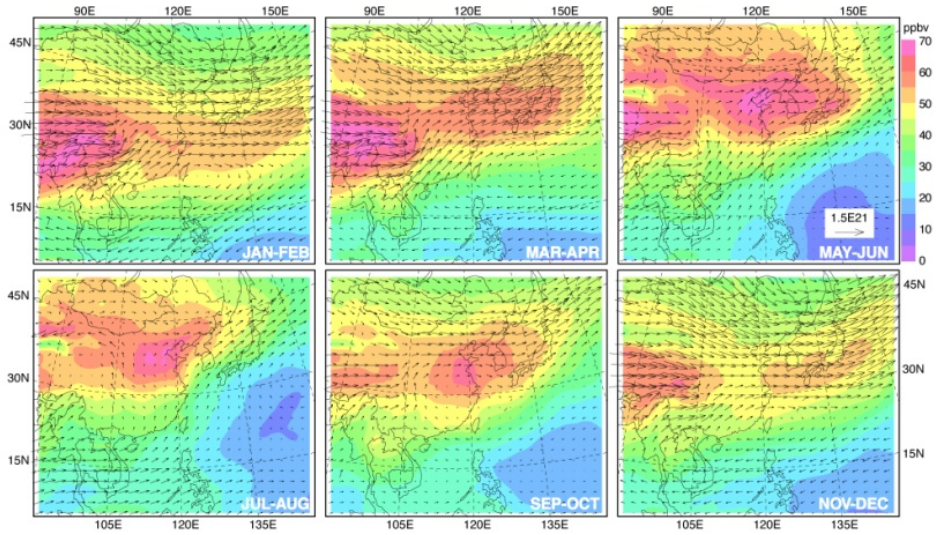


Figure 7.3. Spatial distribution of surface ozone concentration (ppbv) and ozone flux (arrows) in the boundary layer in East Asia. *(Yamaji et al., 2006).*

Regional spatial distribution and seasonal variations in surface ozone in East Asia have been simulated in several regional CTM studies. Yamaji *et al*. (2006), for example, showed simulated

seasonal variations in the spatial distribution of surface O_3 and ozone flux in East Asia (Figure 7.3). In spring (March – June), high O_3 continental outflow covers the Korean Peninsula and Japan, and the concentration of O_3 transfer from eastern China to Japan is 55 – 60 ppbv with a spring maximum at around 35 – 40°N in line with actual observations (Tanimoto *et al*. 2005). In July – August, the southern part of the Korean Peninsula, Japan and coastal parts of southern China are under the effects of a marine air mass, reducing O_3 concentrations to 25 – 40 ppbv. Additionally, continental Southeast Asia is in the wet season under the influence of marine air, and O_3 concentrations are generally very low. In the wintertime, O_3 concentrations around 40°N from Mongolia and northern China to Japan remain relatively low (around 30 – 50 ppbv) because photochemical O_3 production is suppressed by weak solar radiation in the area. O_3 concentrations in the northern part of the Indochina Peninsula are markedly high from January to April due to continental inflow and emissions from biomass combustion during the dry season (Pochanart *et al.*, 2003).

In regard to trends, several published papers have covered modeling analysis of the concentration and deposition of sulphur, nitrogen and O_3 compared to EANET monitoring data. Lu *et al*. (2010) reported that simulation trends for total sulphur concentration in China mirrored inter-annual variations of SO_2 emissions. Total sulphur increased from 2001 to 2006, leveled off in 2007, and fell dramatically in 2008, reflecting the decreasing trend of SO_2 emissions in China due to the penetration of flue gas desulfurization (FGD) in large power plants. For nitrogen, Morino *et al.* (2011) reported that wet deposition rates of NO_3^- across Japan increased during the period 1989 – 2008 at rates of 2 – 5%/yr depending on the region. Sensitivity simulations indicated that the increase of NO_3^- wet deposition rates was mostly (61 – 94%) explained by increased emissions of NO_x in China. Ge *et al*. (2011) studied the impact of the East Asian summer monsoon on long-term variations in acid deposition in Central China and suggested that changes in meteorological factors play an important role in addition to changes in emissions. For O_3, Tanimoto *et al*. (2009) reported that six sea-level EANET sites in Japan showed increasing trends of between 0.41 and 1.00 ppbv yr^{-1} (average 0.63 ppbv yr^{-1}, 1.2% yr^{-1}) in observation, and 0.17 to 1.14 ppbv yr^{-1} (average 0.70 ppbv yr^{-1}, 1.4% yr^{-1}) in simulation. It is suggested that the increasing trend of O_3 over Japan may be due to higher anthropogenic emissions in East Asia, but large inter-annual variability obscures the long-term trend. Kurokawa *et al*. (2009) suggested that large year-to-year variations in O_3 over Japan are strongly influenced by meteorological variability, which controls the direction of Asian continental outflow.

One of the most important areas of study based on chemical transport modeling involves source-receptor (S/R) relationships. Figure 7.4 shows the S/R relationships of sulphur and nitrogen depositions (Lin *et al.*, 2008b). Long-range transport from industrial areas of China accounts for a significant percentage (>20%) of anthropogenic sulphur and reactive nitrogen depositions throughout East Asia. At the same time, northwestern China receives approximately 35% of its sulphur load and 45% of its nitrogen load from emissions elsewhere – mainly from the Indian subcontinent. For receptor areas in Southeast Asia except Thailand and parts of India, the dominant sources (>50%) are emissions from Thailand, Southern China and southeastern China. In addition, the contributions of NOx emissions from international shipping are considerable for nitrogen deposition over Southeast Asia, especially in the Philippines. Within China, local emissions are the dominant source of sulphur deposition in southwestern and southern areas, and for reactive nitrogen loading over these two regions, sources in southeastern China account for up to 20 – 30% of the total. Excluding volcanic sources, sulphur contributions in Japan are estimated to be 37% from domestic sources, 31% from China, 7% from the Korean Peninsula, and 18% from other overseas sources. For Vietnam, the figures are 11% from Vietnam, 51% from China, 10% from Thailand, 5% from other overseas sources, and 3% from international shipping.

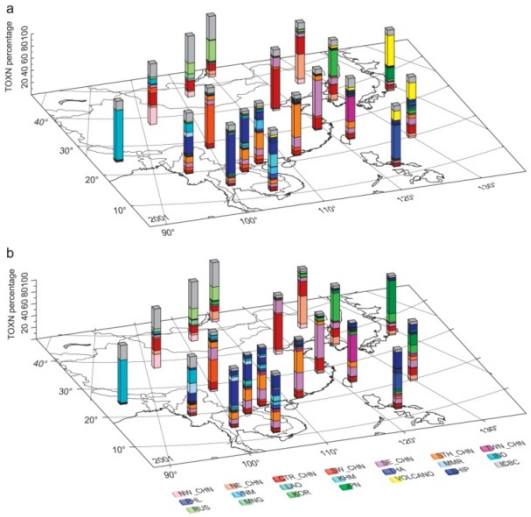


Figure 7.4. Region-to-region source-receptor relationships for (a) total sulphur and (b) total reactive nitrogen deposition. Vertical bars show the origin of substances deposited in each receptor region. Values show percentages of total deposition. *(Lin et al., 2008b.)*

The source-receptor relationship for surface O_3 in East Asia for 2000 – 2005 has been analyzed using the tagged tracer method with a global chemical transport model called CHASER (CHemical Atmospheric general circulation model for Studies of the atmospheric Environment and Radiative forcing; Nagashima *et al*., 2010). The source region was separated into 45 regions including 22 regions each in the planetary boundary layer/free troposphere and the stratosphere. In spring, domestic contribution of O_3 accounted for about 15% of the total in northeastern China (CH-NE), 40% over China's North central plain (CH-NCP), 60% in the Yangtze River Basin, 50% in southeastern China (CH-SE), and about 20% on the Korean Peninsula (KOR) and in central Japan (JPN-E+W). In CH-NE and CH-SE, a large contribution of nearly 20% from eastern Siberia and the Indochina Peninsula was identified. The contributions of China to the Republic of Korea and Japan were estimated as 20 and 12%, respectively. The contribution of intercontinental transport increased with latitude, with figures of 21% for CH-NE and 13 – 14% for CH-NCP, KOR and JPN-E+W. In summer, although the contribution of intra-continental trans-boundary transport was still significant, domestic contribution increased significantly while inter-continental transport and stratosphere decreased significantly. The study also showed that for hourly mean concentrations of surface O_3, domestic sources accounted for more than 50% in the very high concentration range of >90 ppbv even in spring, and that the relative impact of overseas sources was substantial in the mid-high concentration range of 60 – 90 ppb.

Ecosystem and Human Health Impact Assessment Studies

Ecosystems are complex and dynamic, with nutrients continually cycling through biotic and abiotic processes. Although the magnitude and characteristics of the nutrient cycle depend on meteorological, edaphic and vegetative conditions, the rate of the internal cycle is generally much higher than that of atmospheric deposition. As the effects of such deposition on ecosystems therefore manifest themselves over the long term, ongoing observation is necessary to detect changes in material flows and in water/soil properties. In addition to the long-term monitoring of various parameters in catchment areas, biogeochemical models are also expected to be useful in clarifying the mechanisms of ecosystem structural and functional change resulting from acid deposition and in predicting their future status under certain scenarios based on atmospheric deposition induced by developments in social and economic situations. Biogeochemical models are classified into three groups: those originating from soil chemical process models, soil biological process models and plant circulation models. These various types were reviewed in terms of their objectives, the processes involved, time scale and spatial resolution, and include ILWAS (the

Integrated Lake Water Acidification Study; Goldstein *et al*., 1985), NUCSAM (the Nutrient Cycling and Soil Acidification Model) and RESAM (the Regional Soil Acidification Model; de Vries *et al*., 1998). Some models were modified for evaluation of the biogeochemical cycle in forest ecosystems with the C and N cycle in above-ground vegetation taken into consideration. By way of example, the CENTURY model originally developed for estimation of organic matter formation and mineralization in soil now includes the processes of the biogeochemical cycle for carbon, nitrogen, phosphorus and sulphur in forest ecosystems as well as in cropland and grassland ecosystems (Kirschbaum and Paul, 2002; Peng *et al.*, 1998). Plant circulation models include FOREST-BGC (Running and Coughlan, 1988; Running and Cower, 1991) and PnET-CN (the lumped parameter model of carbon (C), water and nitrogen (N) interactions in forest ecosystems; Aber *et al*., 1997), which simulate the carbon and nitrogen cycle with consideration of processes such as photosynthesis, respiration, allocation and litterfall. The PnET model was also modified to create an integrated biogeochemical model (PnET-BGC) that included soil chemical processes. PROFILE (Warfvinge and Sverdrup, 1992) is a steady-state model of soil chemical processes that considers the weathering rate of each particular mineral with kinetic modeling assuming reactions with H^+, H_2O, CO_2 and organic acid.

Monitoring has been carried out to clarify the status of acid deposition and the terrestrial ecosystems on which people depend for healthy living. As air pollution characteristics differ widely by area regardless of national borders, they affect all flora and fauna in terrestrial and inland aquatic water ecosystems to some extent. One causative factor in forest decline is aluminum toxicity in acidic soil, which harms fine roots and hinders tree growth (Ulrich *et al.,* 1980). However, several other factors are also at work, including insects and fungal diseases, meteorological damage from influences such as frost and drought, air pollution, acid deposition and ozone exposure. In this way, multiple causes and complex mechanisms are involved in ecosystem damage.

Acidification risk is also recognized as being linked to SO_2, NOx, NO_3 and NH_4 concentrations. As control efforts in regard to sulphur oxides have increased in many countries, other sources of these acidified substances are now considered. A situation of high loads exceeding the N demand of forest growth is known as nitrogen saturation in forest ecosystem terms. High levels of N deposition stimulate transpiration from cedar needles even in dry soil on Japan's Kanto Plain. In this way, our environment still faces a risk of gradual acidification and eutrophication. Accordingly, soil maps are updated with the latest information from monitoring data and modeling in the interests of reducing the risk of environmental deterioration.

Ozone is also harmful to vegetation, materials and human health (WHO, 2003 and 2005). Effects are acute responses to the exposure to ozone (WHO, 2008); adverse effects on pulmonary function and respiratory symptoms, lung inflammatory reactions, increase in medication usage, hospital admissions and death rates. Other likely influences include chronic effects associated with ozone exposure such as reduced lung function, development of atherosclerosis and asthma, and reduced life expectancy, although evidence for these considerations is less conclusive, mostly due to the limited number of epidemiological studies conducted (WHO, 2005). To provide adequate protection of public health, WHO sets the air quality guideline (AQG) value for ozone at 100 $\mu g.m^{-3}$ (50 ppb) as an 8-hour daily average, which is estimated to cause a 1 – 2% increase in daily mortality (WHO, 2005). The Clean Air Act in the US requires the US Environmental Protection Agency (USEPA) to set the national ambient air quality standard (NAAQS) for ground-level ozone at 75 ppb as an 8-hour daily average (3-year average of the 4^{th}-highest daily maximum 8-hour average must not exceed 75 ppb) (USEPA, http://epa.gov/air/criteria.html).

The effects of particulate matter (PM) on the respiratory system are also emphasized. WHO (2006) reported effects related to short-term exposure include lung inflammatory reaction, respiratory symptoms, adverse effects on the cardiovascular system, increased medication usage, higher hospital admissions and premature mortality; effects related to long-term exposure include increased incidence of lower respiratory symptoms and chronic obstructive pulmonary diseases,

reduced lung function and life expectancy. WHO set the AQG value of 10 $\mu g.m^{-3}$ as the annual mean for $PM_{2.5}$ to represent the lower end of the range over which significant effects on survival were observed in the American Cancer Society (ACS) Study (WHO, 2005; Pope *et al.,* 2002). The AQG for $PM_{2.5}$ is also set at 25 $\mu g.m^{-3}$ as the 24-hour mean (99^{th} percentile (3 days/year)) since at low levels there is less concern about remaining episodic excursions. The Clean Air Act in the US required USEPA to set NAAQS for $PM_{2.5}$ at 15 $\mu g.m^{-3}$ as the annual mean (the 3-year average must not exceed this value and 35 $\mu g.m^{-3}$ as the 24-hour mean (the 3-year average of the 98^{th} percentile must not exceed this value) (USEPA, http://epa.gov/air/criteria.html).

Some studies have assessed the risk to human health by air pollutants in areas including the East Asian region. Several indices are used for assessing the effects of ozone on human health, such as SOMO35 (accumulated ozone concentrations above a threshold of 35 ppb), EU60 using a threshold of 60 ppb, and USEPA80 evaluating the exceedance of 80 ppb.

To evaluate global mortality caused by long-term exposure to ozone and $PM_{2.5}$ for 2000, Anenberg *et al*. (2010) used global atmospheric chemical transport model (CTM) simulations and considered health impact functions. Figure 7.5 shows the estimated annual mortalities attributed to ozone. The estimates were reduced by approximately 30% for assumed low-concentration thresholds of 33.3 ppb for O_3 and 5.8 $\mu g.m^{-3}$ for $PM_{2.5}$.

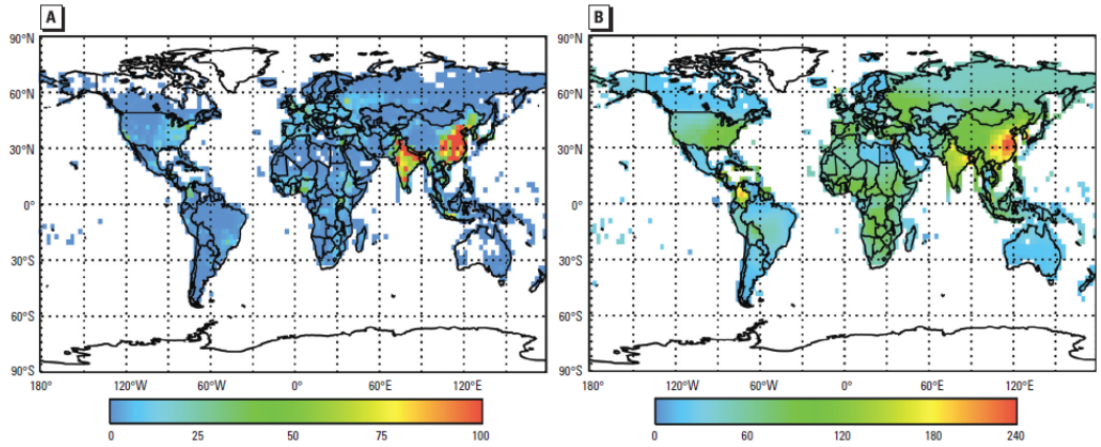


Figure 7.5. Estimated annual premature mortalities attributed to anthropogenic O_3 when no upper or lower concentration threshold is assumed, for respiratory mortalities per 1,000 km^2 (A) and rate of respiratory mortalities per 10^6 people (B). *(Anenberg et al. 2010)*

The above health risk assessment studies are mainly based on epidemiological researches in Europe and the US. Accordingly, there is a urgent need for implementing such researches in East Asia in consideration of regional specific ozone/PM concentrations and related responses so that uncertainty in risk estimation can be reduced.

Ozone causes reduction in crop yields, tree growth and carbon sequestration, and modification in species composition (Ashmore, 2005; USEPA, 2006). In terrestrial ecosystems, the most significant direct effects of ozone are those on leaf physiology and plant growth, and many other indirect effects on ecosystems flow from these direct effects (The Royal Society, 2008).

The development of fundamental data sets such as those on crop distribution and crop calendars is paramount for more reliable risk assessment of crop yield loss as well as the exposure-response functions of crops in East Asia.

The first decade of EANET's operations was successful in terms of wide-ranging provision of data for the international scientific community. As the network produces data with a level of high-quality suitable for scientific research, EANET is expected to play an important role in future successful operations relating to international science.

8. Summary and Recommendations

The 10th anniversary of the Acid Deposition Monitoring Network in East Asia (EANET) was marked by the adoption of instrument to strengthen the organization's activities.

8.1 Summary

Present Status of Acid Rain in East Asia

The five-year average pH of rainwater is less than 5.0 at over 60% of all EANET monitoring sites, and is even less than 4.6 at several sites. It can therefore be said that acid rain still prevails in East Asia. Sulphric acid is more significant than nitric acid as an acidifying species, which means that SO_2 emissions are still the major contributor to acid rain in these areas. In contrast, contribution of nitric acid is relatively high in several sites, where NO_3^- accounts for more than 40% of total for nss-SO_4^{2-} and NO_3^-. Since sulfur emission control is a precedent to nitrogen control in general, the relative significance of nitrogen as an acidifying species in rainwater is predicted to increase in the future in East Asia.

Although ammonium ions (NH_4^+) are a buffering species for acids in rainwater, they change to nitrate (NO_3^-) in soil and release hydrogen ions (H^+) after deposition, thereby contributing to acidification. From the viewpoint of ecosystem impact, effective hydrogen ion (H^+_{eff}) deposition (considering both H^+ and NH_4^+) in rainwater is more significant than pH in soil acidification. Relatively high deposition levels of H^+_{eff} were observed at most urban sites in southern East Asia.

In addition to acidification, eutrophication (nitrogen saturation of soil and water) is of concern from the viewpoint of ecosystem impacts in East Asia. To evaluate the risk of nitrogen saturation, the sum of NO_3^- and NH_4^+ depositions (ΣN) was evaluated. Higher deposition of ΣN was observed in Southeast Asian urban areas and at several sites in Northeast Asia.

The 10-year period of EANET monitoring completed so far is generally considered an insufficient length of time to support discussion of clear trends of ion concentration in rainwater and wet deposition. Nevertheless, statistically significant increasing and decreasing trends of concentration and deposition are seen at several sites for particular species. The continuation of EANET's monitoring activities is crucial in elucidating clear trends of acid rain over the next decade in response to emission control policy.

Trend of Ozone Concentration

Ozone (O_3) concentration is monitored in Japan, the Republic of the Republic of Korea, Thailand and Russia. Common seasonal variations are observed at these sites – highest values in spring, lowest values in summer, and second-highest values in autumn. Spring maximum concentrations at most sites in Japan exceed a monthly average of 50 ppb, and are even higher than 60 ppb at several sites. Monthly average O_3 concentrations from 2005 to 2009 were higher than those for the previous five-year period at most sites in Japan.

Impacts on Ecosystems

Atmospheric deposition may cause acidification and/or eutrophication of ecosystems. Soil acidification is not generally observed at EANET monitoring sites, although a decreasing trend in soil pH was seen at one site in Northeast Asia. In several plots at particular sites in Southeast Asia, an increasing trend of soil pH has been observed for the last decade.

In contrast, a significant decrease in the carbon-to-nitrogen ratio (C/N) in soil, suggesting increased

nitrogen input, was observed at several plots in both Northeast and Southeast Asia. Simultaneously, the NO_3^- concentration of inland water increased around these sites. Excess NO_3^- in soil may flow into streams and lakes leading to increased NO_3^- concentration in inland waters, which may in turn suppress tree growth and cause degradation in the future.

No decline in tree growth or in the number of species in understory vegetation was observed during the reporting period from 2000 to 2009. Tree decline symptoms observed at some sites were generally considered to stem from pest infection. In addition to the future potential risk of nitrogen saturation mentioned above, the direct effects of air pollutants such as ozone on trees remain a source of concern for the region. The relatively high and rising trend of ozone concentration observed at EANET sites in Northeast Asia may pose a potential ecological risk to forest trees.

Catchment-scale analysis, which is intended to clarify the relationship between atmospheric deposition flux and deposition variations in soil and inland water, is now implemented by EANET. An integrated monitoring approach for atmospheric, vegetation, soil and inland water should be promoted in the future to provide a clearer view of ecological impacts in East Asia.

8.2 Recommendations

Technical Recommendations

Common QA/QC procedures are strongly recommended in the interests of obtaining datasets with greater degrees of comparability and reliability. To make EANET data more useful for scientific analysis, sampling frequencies and other monitoring protocols should be made to match those specified in related technical manuals. In order to increase the transparency of monitoring techniques adopted by participating countries, a national monitoring plan covering areas such as site selection, sampling/analytical methodologies and monitoring intervals should be created within a certain timeframe and submitted to the Network Center to be shared. It is also advisable to establish data checking procedures, include appropriate calibration and maintenance of analytical instruments for the monitoring of gases and particulate matter (PM) among future activities, establish an EANET-wide ozone monitoring system with calibration traceable to international standards for automatic instruments, and improve inland aquatic environment monitoring through repeated measurement to promote the evaluation of reproducibility for soil and vegetation observation.

In order to clarify the status of the atmospheric environment over the whole EANET region, establishment of additional monitoring sites is also recommended. The nomination of domestic monitoring sites is one possible way to achieve this. Continuous monitoring over future decades is highly recommended for EANET. The conditions surrounding monitoring sites on local and regional scales should be checked on an ongoing basis, and equipment maintenance should also be conducted to ensure uniform conditions for sample collection and analysis.

QA/QC of air concentration monitoring is a highly important consideration for EANET. Management of certified standard gases, calibration of O_3 based on international traceable standards and inter-laboratory comparison with a reference method of PM monitoring should be promoted in the future. To avoid filter pack artifacts, the adoption of annular denuders and/or diffusion scrubbers should be considered for monitoring innovation, and the introduction of automatic instruments for real-time air concentration monitoring should be encouraged in the meantime. Dry deposition monitoring is a top priority in measuring and reporting meteorological parameters and land surface characteristics to enable the calculation of dry deposition. Real-time measurement of gas concentrations in air using automatic instruments is also highly recommended.

Careful selection of air concentration monitoring sites in consideration of site representativeness and compatibility for air quality and dry deposition flux measurement is advisable.

In the area of ecological impact monitoring, atmospheric deposition monitoring should be carried out in the same catchment area, and both wet and dry deposition fluxes can be estimated as input parameters according to the *Guideline for Catchment-scale Monitoring in East Asia* recently compiled and approved by EANET. As increased emissions of nitrogen oxides are predicted in East Asia, EANET monitoring and the identification of potential environmental risk hotspots are recommended to support consideration of both acidification and eutrophication. Taking such hotspots into consideration, a strategic monitoring framework should be designed in the near future. It is also recommended that ozone concentrations and tree health be monitored simultaneously in forest areas using automatic instruments to identify higher concentrations during the daytime.

Several regional-scale emissions inventories in Asia have been widely utilized as model input for the analysis of air concentrations and depositions of acids, ozone and their precursors. It is hoped that emission inventories for each participating country will be compiled and made available to the scientific community for use in the interpretation of spatial distribution, temporal variation and long-term trends of atmospheric and rainwater compositions and depositions. Researcher- and government-based inventories should be harmonized as far as possible through cooperation between administrative agencies and inventory experts in each country. It is also recommended that EANET consider the issue of co-benefits through activities such as discussions on how acid deposition and air quality relate to climate change.

As the presence of EANET sites is still very sparse from a regional perspective, chemical transport modeling is the only way to give an overview of the spatial distribution of concentrations, depositions and temporal variations of monitored species. Modeling activities based on global and regional chemical transport models validated using EANET monitoring data by scientists in participating countries should be encouraged to promote understanding of the causes of spatial and temporal variability in monitoring species. More effort should be made in Southeast Asia for better tuning of models. Ensemble means of multi-models are considered more robust than single-model predictions for spatial and temporal variations and source-receptor relationships. It is highly recommended that scientists in the countries in question join projects such as MICS-Asia (the Model Inter-Comparison Study for Asia) in order to provide a wider range of modeling results and support more transparent discussions on the present status of the atmospheric environment.

Now that the first 10-year period of EANET's acid deposition monitoring is complete, there is a need for critical evaluation of the potential environmental risk posed by acidification in East Asia. Thorough review of experiences in Europe and North America in contrast to the situation in East Asia should be conducted to highlight potential risks from acidification in the latter. It is also recommended that eutrophication be included among the targets of EANET monitoring and that potential environmental risk hotspots be identified, as mentioned previously. In this regard, the accuracy of ammonia emission inventories should be improved.

The high levels of uncertainty seen in estimates of the health-related and agricultural impacts of O_3 and PM necessitate more extensive epidemiological study in each EANET country. The use of national monitoring network data on O_3 and $PM_{2.5}$ is recommended to support more accurate estimation of these impacts.

Assessment of acidification and eutrophication risk as well as ozone and PM concentrations is considered useful in raising awareness of the importance of atmospheric pollution issues in the EANET community, and should be implemented in all participating countries.

General Recommendations

The global spread of atmospheric pollution calls for consideration of new issues such as the link between urban air pollution and regional/hemispherical air pollution as well as the relationship between air pollution and climate change. Acid deposition is thus closely linked to many other environmental issues. It shares precursor emissions with ozone and aerosols/particulate matter, meaning that measures to mitigate the phenomenon can also be expected to provide co-benefits in terms of suppressing other atmospheric pollutants. Ozone, aerosols and other acid deposition-causing air pollutants contribute to climate change, which in turn influences acid deposition and its effects through phenomena such as precipitation change. Accordingly, extended assessment of the state of acid deposition, including consideration of other relevant atmospheric pollutants and climate change, should be coordinated to improve EANET's response to emerging issues.

There is an urgent need to secure achievements that will sustain EANET's development, as well as demand to strengthen the basis of the network's operations. For this purpose, the following targets should be pursued:

・Improvement of acid deposition monitoring, including consideration of other relevant components such as ozone and particulate matter (PM), with increased transparency
・Extended assessment of the state of acid deposition, including consideration of other relevant atmospheric pollutants
・Promotion of research activities, including the development of modeling and emission inventories
・Establishment of an epistemic community and promotion of public awareness to achieve a common understanding on atmospheric pollution
・Enhancement of the policy relevance of activities relating to the provision of policy advice and information based on sound science and assessment
・Strengthening of technological support and capacity by enhancing cooperative efforts among participating countries
・Review regarding the present status of the atmospheric environment in East Asia and discussion of future scope expansion
・Enhancement of collaboration with organizations outside the region

To achieve these goals, stakeholders on all levels, including policy makers, the scientific community, business operators and the public, must make a firm commitment to an improved ethic of stewardship.

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