Report of the Long Term National Acid Deposition Monitoring in Japan

(JFY2003-2007)

MARCH 2009

Ministry of the Environment

Government of Japan

Ogasawara acid deposition monitoring site

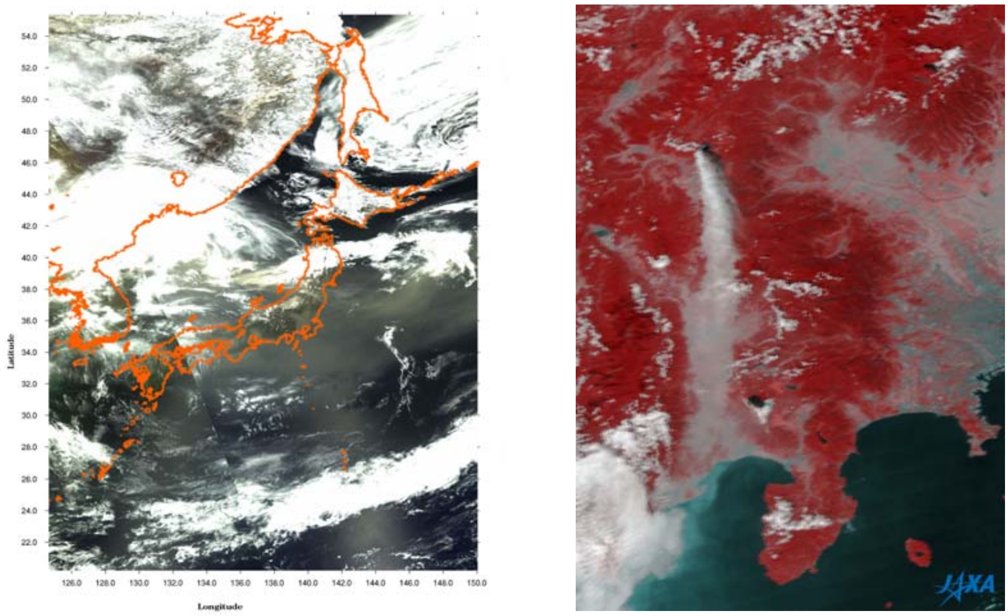
Samplers (Tokyo site)

Soil sampling (Hakusan, Ishikawa)

In-land water sampling
(Nagatomi-ike, Kagawa)

Sampling of river water flow in Ijira Lake
(Koubora gawa)

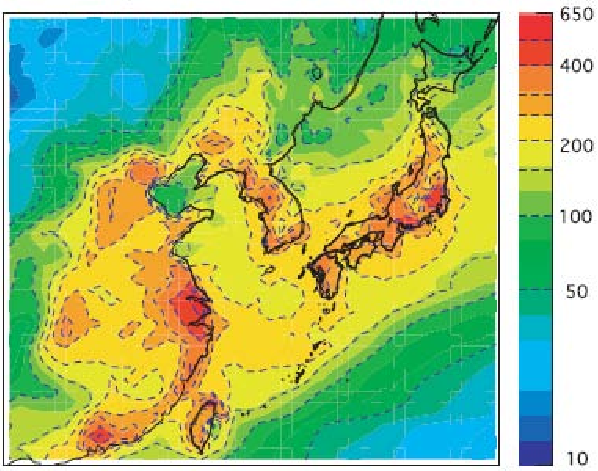
Throughfall sampling at Ijira



Yellow sand from continent
(18 Apr. 2006)

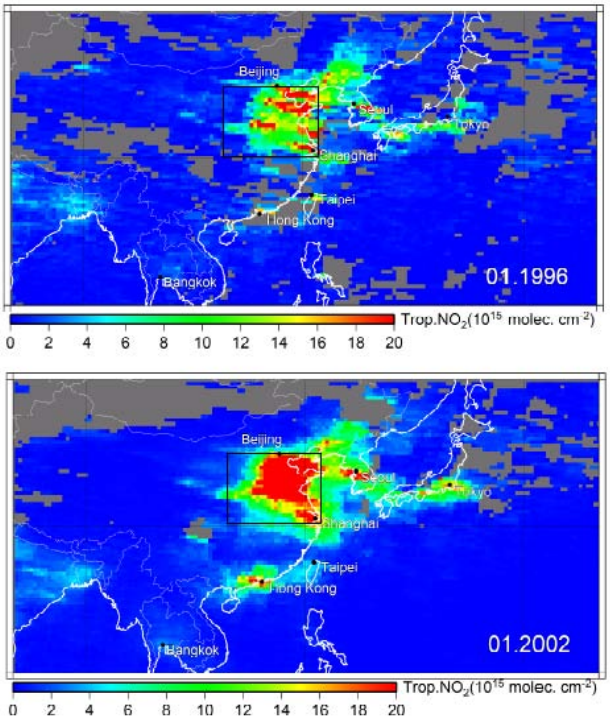
Volcanic fumes from Asama Mt.
(16 Sep. 2004)

Pictures by MODIS mounted in Terra and Aqua earth observation satellite of NASA
(Provided from Japan Aerospace Exploration Agency)

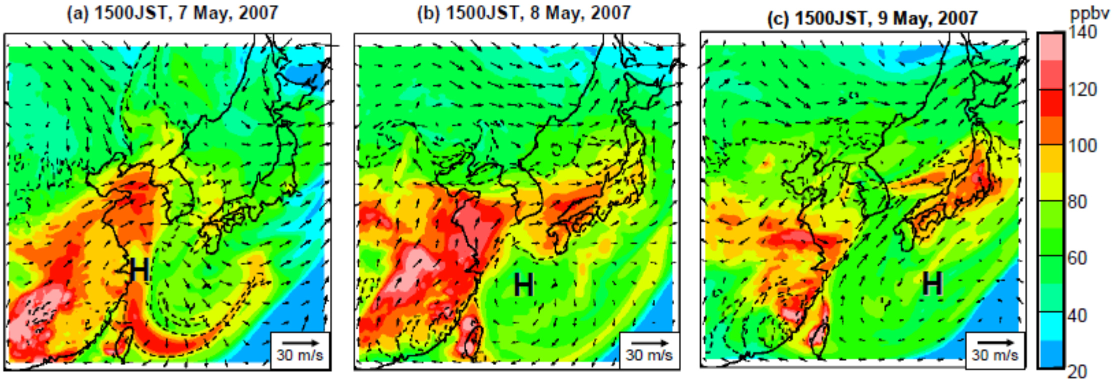


cont = 50, 100, 150, 200, 250, 300, 400, 500 (KgN/km^2/yr)

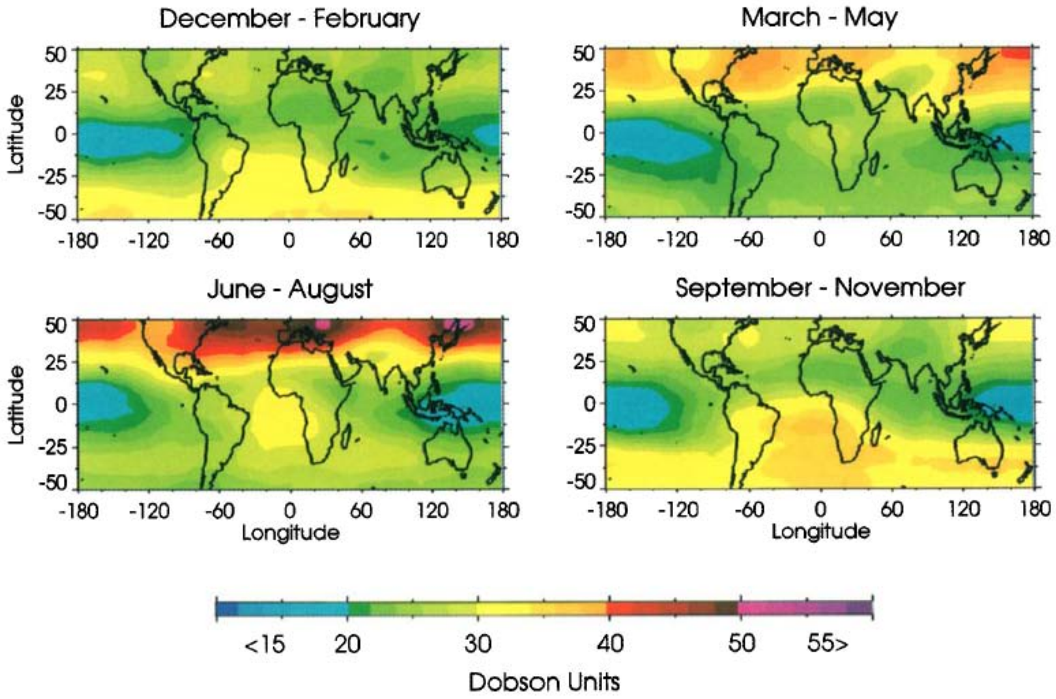
Nitrogen annual deposition quantity distribution (2002)
(Uno, et al., 2007)



Distribution of NO$_2$ concentration in the troposphere in East Asia by GOME
The upper figure shows the average concentration in January 1996 and the lower in January 2002. The grey parts show no data. (Japan Agency for Marine-Earth and Technology, 2005)



The simulation results of the transboundary pollution of ozone in East Asia
(7 - 9 May 2007)
(Ohara, et al., 2008)



The distribution of seasonal global ozone by satellite observation
(Fishman. J, et al., 1997)

INTRODUCTION

About acid deposition, acidification of lakes and forest damage became a diplomatic issue in Europe in the 1960s, and wet acidic air pollution became a problem in Japan in the 1970s. Therefore, the Ministry of the Environment (former Environment Agency) began the acid deposition survey from Japanese Fiscal Year (JFY) 1983 to comprehend the condition and influence of acid deposition in Japan with the aim of preventing harmful influence from acid deposition. Also, in order to maintain the acid deposition monitorihng in wide-area for long term, “The long term monitoring plan of acid deposition” was formulated on March 2002. Based on the plan, the Ministry of the Environment has implemented wet/dry deposition monitoring, land water monitoring for lakes, and soil/vegetation monitoring cooperating with local governments since JFY 2003.

This report shows the results of long term monitoring conducted from JFY 2003 to JFY 2007, in addition, summarizes the results of the intensive survey on Ijira lake catchment area of which the soil is known to be acidified, from JFY 2005 to JFY 2007 focused. In recent years, transboundary air pollution in North-East Asia is pointed out as one of the causes of widening area that photochemical oxidant warnings are announced and increasing its concentration. For these reasons, not only acid deposition but also transboundary air pollution including ozone and aerosol is growing concern. We reviewed the existing study results such as calculation results of atmospheric simulation models as well as the condition of transboundary air pollution.

This report is summarized about the result of study conducted at the acid deposition committee and its atmospheric issues sub-group and ecological impact sub-group established in the Ministry of Environment. The related data was collected and analyzed at Acid deposition and oxidant research center and it organized four working groups (acid deposition analysis, ecological impact analysis, Ijira lake intensive survey, transboundary air pollution) to analyze and review the research results.
We extend a special thank to everyone concerned who cooperated with conducting and summarizing this monitoring survey.

March 2009
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CONTENTS

General overview of results of the survey

1. Overview of the survey

- The Ministry of the Environment has conducted the acid deposition survey in order to figure out the condition and influence of acid deposition in Japan since JFY 1983. Based on the long term monitoring plan of acid deposition which was formulated in March 2002, wet/dry deposition monitoring (31 sites), land water monitoring for lakes (11 sites), and soil/vegetation monitoring (25 sites) have been implemented since JFY 2003.

- This report shows the results of long term monitoring of acid deposition conducted from JFY 2003 to JFY 2007 and summarizes the results of the intensive survey on Ijira lake catchment area of which the soil is known to be acidified, from 2005 to 2007. It also reviews the influence of the transboundary air pollution to Japan.

2. The state of acid deposition and transboundary air pollution

(1) The result of acid deposition monitoring

- The averages of precipitation pH for 5 years at monitoring sits were in the range from pH 4.51 (Ijira lake) to pH 4.96 (Ogasawara). The total average was pH 4.68. Acid deposition is still being monitored. Less than pH3.0, which is thought to give acute damage to plant, was not observed, but it is figured out that strong acidic precipitation less than pH4.0 made up 4.5% in total precipitation at 14 sites which have been conducted daily sampling.

- Precipitation pH showed small changes yearly and relatively low in some monitoring sites in recent years, but it has tendency to remain at the same level in total at the sites which have been observed at for more than 10 years. After 2005, the concentration of nitrate ion in precipitation showed the tendency to increase.

- The yearly amounts of the total acid deposition at 12 monitoring sites in Japan were calculated. Non-sea-salt sulfur was in the range of 16～54mmol m^{-2} y^{-1} and nitrogen was in the range of 22～130mmol m^{-2} y^{-1}.

- The amount of acid deposition by precipitation increased in the side of the Sea of Japan of Honshu from late autumn to spring. Also, the concentration of sulfur dioxide in the atmosphere tended to increase markedly in Sea of Japan side and western area of Japan. In these areas, it would appear that sulfur oxides and nitrogen oxides supplied into the atmosphere increased from late autumn to spring. The pollutants input from tne continent was suggested.

- In Japan, the amount of non-sea-salt sulfate ion and nitrate ion deposited by precipitation was twice or three times larger than those in Europe and the United States. Also, the amount of hydrogen ion deposition was several times higher in Europe and the United States, and the highest among East Asia. It is because Japan has large precipitation and the rate of acid counteracted by basic material in the atmosphere and precipitation is low.

- The quantity of non-sea-salt calcium ion deposition, concentration of PM10 in the atmosphere and calcium ion in aerosol hit the highest in spring all of the country, which suggests the influence of yellow sand.

- The concentration of ozone increases in spring countrywide, which suggests the influence of transboundary air pollution. Also, the change of average value of ozone concentration across the ages tends to increase in the country.

（２） Review of research on transboundary air pollution

- The recent atmospheric simulation models estimated that the rate of the contribution of the transboundary air pollution to the amount of acid deposition in Japan is about 30 – 65% for non- sea-salt sulfur ion and about 35 – 60% for nitrate ion.

- The influence of transboundary air pollution to the concentration of ozone in Japan fluctuates widely depending on land areas, seasons and dates. However, it appears that about 10 -20 % of the monthly average of ozone concentration on Japan's main island in Spring season originates East Asia and estimated that about several ppb influence comes from Europe and the United States in springtime.

- Also, it is predicted that yearly average of ozone concentration in Japan on 2020 will increase 2 – 6ppb compared to 2000 if the emission of air pollutant in China generally continue to increase under the business as usual scenario.

３．Impact to ecosystem

（１）The result of soil/vegetation monitoring

- In 2007, there was the indication that some of the woods declines at 17 out of 25 monitoring sites, but it was not observed that acid deposition or acidified soil mainly caused the declination.

- There is a trend that the pH (H_2O) on the surface and the second layer of the soil in Ijira lake had decreased from 1990 to 2004. The pH in the surface layer was an average of pH 3.9 which indicates that the level of aluminum ion which has impact to growth of plant dissolved out to the soil so that it is necessary to continue to monitor.

- Soil acidification trend was not observed at other observation points in general although some change with the age was observed in a part of monitoring items.

（２）The result of land water monitoring

- The acidity of the surface layer water of the lakes was in the range from pH 5.36 -7.34, and the alkalinity was from 0.017 – 0.512 meq/L. Futago, Meike (Nagano prefecture), Yasya (Fukui prefecture) and Sawano (Kyoto prefecture) lakes were under pH 5.8 and the alkalinity was under 0.030meq/L.. It can be said that they have high sensibility to acid but it was not seen clearly the influence of air deposition at all lakes.

- The acidity at Kamagaya river which flows into Ijira lake had tendency to decrease from 1996 to 2003, but it remains around pH 7 after 2004. The concentration of nitrate ion on the surface layer water at Ijira and its inflow two rivers showed a significant increase.

４．The result of the intensive study at Ijira lake water area

- The acid deposition monitoring survey from JFY 1988 to JFY 2002 found that the physicochemical change such as the decrease of pH value which would present the impact of acid deposition at the rivers input to the Ijira lake and the surrounding soil in Ijira lake catchment area. In order to make clear about the sign of acidify and its process, MOEJ conducted the survey of the inlet and outlet flow in the catchment area and the intensive monitoring at swollen rivers from JFY 2005 to JFY 2007..

- From the following findings i – iii, acidify of the catchment area of Ijira lake was considered to stay continuing to be acidified by the reason that sulfur and nitrogen piled in the soil by acid deposition from the atmosphere flowed out to the rivers.
  - ⅰ The total quantity of nitrogen deposition in Ijira lake catchment area was estimated 18.2 – 28.7 kg-N ha-1 y-1 which exceeded the reference index of nitrate ion outlet to rivers in Europe, 10 kg-N ha-1 y-1. The high concentration of the flow out of the nitrate ion has continued after middle of 1990s. It was considered that this nitrate ion flow out with the large amount of nitrogen deposition contributed to the acidification of the rivers.
  - ⅱ By estimating the material balance at the catchment area, the outlets of sulfur was twice more than the inlets. It was considered that sulfur stored in the soil for years flowed out and contributed the acidification of the rivers.
  - ⅲ By the trend that concentration of sulfate and nitrate ion at the swollen rivers increased, it was indicated that sulfur and nitrate stored on the surface soil around the catchment area flowed out.

- It is difficult to show the progress of future acidification quantitatively because the mechanism of the acidification around Ijira lake catchment area is complicated and it remains a matter to be solved scientifically.

- Although it is considered that the status of acidification may affect to human health and ecosystem immediately, it is needed to conduct survey at any hot spot which would have high risk of acidification since there is concern that other water area same as Ijira lake suffers acidification.

５．Future tasks about prevention of acid deposition

（１）Long term monitoring

- The influence of acid deposition is comprehended by only long term monitoring, and it is possible to appear the influence abruptly by storing over a certain amount of acid material load when lakes and soils have low buffer abilities.

（２）Promotion of international cooperation

- Aiming to extend the action scope of EANET from the conventional acid deposition monitoring to the management of the atmosphere environment in East Asia, it is necessary to establish the international cooperative relationship and promote regional collaboration to prevent air pollution.

（３）Approach to the transboundary air pollution

- The transboundary air pollution from East Asia Continent is pointed out as one of the reason of widening area that photochemical oxidant caution was announced in, rising ozone concentration gradually of and increasing in frequency of the detection of yellow sand. It is needed that the transboundary air pollution monitoring including not only acid deposition but also ozone and aerosol should be conducted.

（４）Promote the investigative study

- In order to comprehend the influence and conditions of long range transboundary air pollutions, it is necessary to promote research studies such as elaborating atmosphere simulation models, making high accuracy in emission inventory, deliberating reference index of sensibility of soil, vegetation and in-land-water and finding out hot spots with high risk in acidification.

１． History of acid deposition survey

Acid deposition is recognized as one of global environmental issues since the substances causing acid deposition are transported over long distance and there is possibility that its influence diffuse to not only inside of the country but also to outside of the country. In Japan, our social concern to acid rain was risen because the fog and drizzle stimulated human eyes or skins and the rain had influence to crops like tobacco culture during rainy season in northern part of Kanto district from 1974 to 1976..

In Europe, it came to the surface that acid deposition acidified lakes in Northern Europe. The treaty for long distance transboundary air pollution was concluded in 1979. Joint monitoring of acid deposition and reducing emission of contaminated materials conducted based on the treaty have been successful.

Meanwhile, the emission of air contaminant has increased with rapid economic growth in East Asia. It is concerned that the transboundary air pollution including acid deposition will become more serious in the future. In order to promote the prevention method for acid deposition based on international cooperation in East Asia, EANET started functioning on a trial basis by Japanese initiative from April 1998 and has been running since January 2001. 13 countries in East Asia are joined now.

○ The domestic research about acid deposition

The Ministry of the environment established the acid deposition prevention committee whose members were consisted of the experts for air pollution, soil/vegetation, and land water, and has continued to conduct the research on acid deposition since 1983 in order to make clear the actual condition and the influence in our country.

As the result, “The general report about acid deposition research” was summed up on June 2004 as the research results conducted for twenty years from 1983 to 2002. It shows the following results.

① The acid depositions as high as those in Europe and the United States are observed nationwide which is pH4.77 on an average. Also it was suggested that the contaminated materials flew into the area on the Sea of Japan side from the continent.

② At this moment, it is not observed that acid depositions damage the ecosystem such as vegetation erosion or acidify the soil.

③ The physicochemical changes of pH seemed to be caused by low level acid deposition at Ijira lake water area, the rivers input to the lake and the surrounding lands where the ecosystem is thought to be fragile against acid depositions. However, these changes do not give any influence immediately to human health and the ecosystem of plants and water plants around the rivers,

As seen above, it is still unclear how acid depositions have an impact to the ecosystem in our country. However, the influence of acid deposition could become obvious in the future if we continue to have acid depositions as it is, because the influence of acid depositions to soil/vegetation and land water is considered to appear after long term period. Therefore, the Ministry of the Environment formulated “The long term monitoring plan of acid deposition” on March 2002 in order to maintain to monitor acid deposition in wide-area for long term, and has conducted the wet/dry deposition monitoring, the land water monitoring for lakes and soil/vegetation monitoring based on this plan.

Table1-1 Number of monitoring site

	Phase 1	Phase 2	Phase 3	Phase 4	—	long term monitoring
Japanese Fiscal year	1983-1987	1988-1992	1993-1997	1998-2000	2001-2002	2003-2007
Wet/dry deposition	14～34	29	48	55	48	31
Soil, vegetation	12	43	88	20	18	25
in land water	133 (screening)	5	33	17	12	11
other		2(snow)				

The relation between acid depositions and observed materials

Air contaminants such as sulfur dioxide or nitrogen oxide are oxidize to sulfur or nitric acids in the air, and come back to the land again (deposition). There are two types in the way of deposition. One is dissolved in rains or snow to be deposited (wet deposition). The other is deposited as the form of gas or particles called aerosol (dry deposition). Acid deposition as one of the environmental problems represents both wet deposition and dry deposition. Scientifically, it is called “acid deposition”.

As the damages by acid depositions, woods are diminishing by acidified soil, land water ecosystems have taken damages by acidified lakes and cultural properties like bronze statues and buildings become damaged. How much they have damage depends on the quantity of acid deposition. So in case of raining, we need to consider not only pH which shows the acid level but also amount of precipitation.

Ozone is known as air contaminant to exert a harmful influence to human health and plant. It is also an acidifying material which acidifies sulfur dioxide and nitrogen oxide very well. Therefore, the generation mechanism of acid depositions has close links to those of ozone.

Also, there are alkaline materials such as ammonia gas in the atmosphere. They generate sulfur ammonia and nitrate ammonia when they react with sulfur and nitric acid. These materials become dry deposition as aerosol or wet deposition dissolved in rains.

As seen above, it is important that “acid depositions” is regarded as the comprehensive air pollution which involves various materials. It is necessary to observe not only precipitation but also gas and related aerosol such as sulfur dioxide, nitrogen oxide, ozone, ammonia and soon.

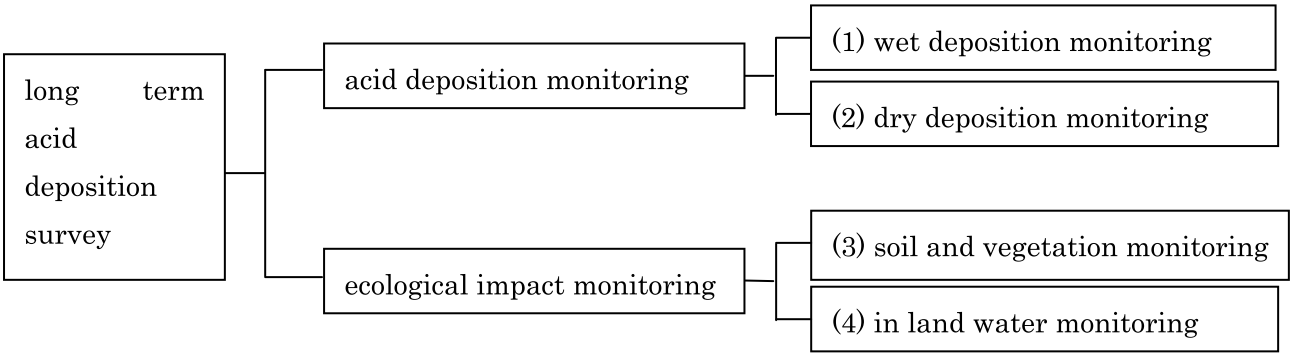
２. The purpose and contents of the research

○ The purpose

The aim of the long term survey of acid depositions is to comprehend the influence of acid deposition early, the long distance transboundary of materials causing acid depositions and the long term trend. Also, it aims to find out the temporal and special changes and its influence in the quantity of acid deposition by monitoring acid deposition and its influence to ecosystem for long time periods with cooperating with EANET in order to predict the influence of acid depositions in the future.

○ The contents of the research

The Ministry of the Environment had conducted wet and dry acid deposition monitoring based on the plan of long term acid deposition survey in order to comprehend the condition of acid deposition, and soil/vegetation monitoring and land water monitoring in order to comprehend the influence of acid depositions to ecosystem from 2003 to 2007.



（１） Wet deposition monitoring

Wet deposition monitoring was conducted at 31 points which were selected to be conducted acid deposition monitoring effectively and efficiently under consideration of the climate, aerial characteristics and the balance. The observation points are classified into 3 areas, which are remote, pastoral, and urban areas in accordance with the EANET wet deposition monitoring manuals (refer to figure 2-1).

The precipitation samples (including snow) were collected with precipitation open collecting devices, which open the covers when it is raining and collect precipitation. The samples were measured and analyzed about 10 items which are hydrogen-ion exponent (pH), electric conductivity (EC), ion concentration (sulfur ion (SO_4^{2-}), nitrate ion (NO_3^-), chloride ion (Cl), ammonium ion (NH_4^+), calcium ion (Ca^{2+}), potassium ion (K^+), magnesium ion (Mg^{2+}) and sodium ion (Na^+).

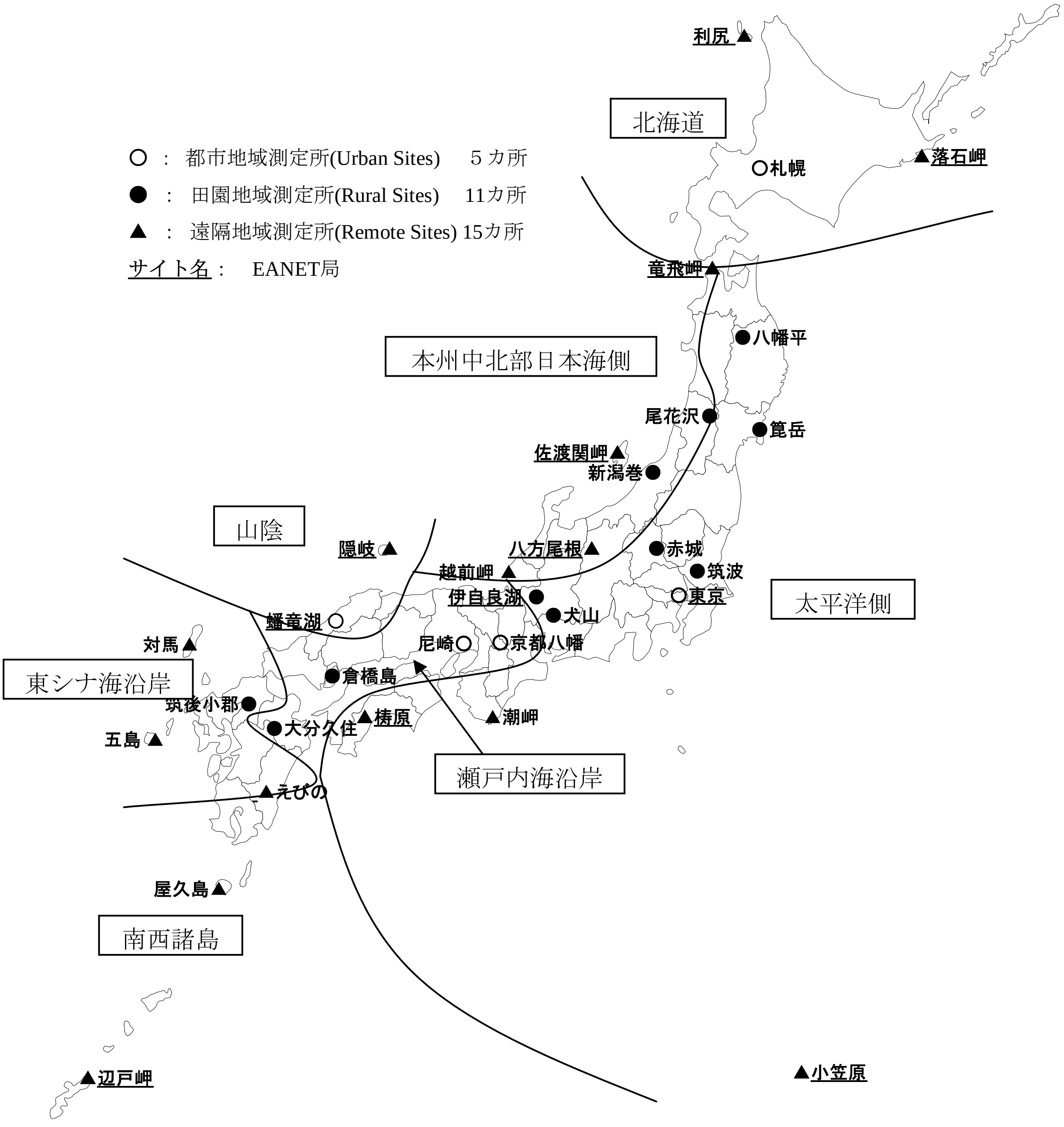


Figure 2-1 Acid deposition monitoring sites

（２） Dry deposition monitoring (atmospheric concentration measuring)

Dry deposition monitoring is basically the method to gain the quantity of dry deposition, but the dry deposition processes are extremely complicated and the measuring method has not standardized yet. Therefore we measured atmospheric concentration from which we are able to estimate the quantity of dry deposition.

At the EANET departments registered in EANET (11 points excluding Tokyo department) out of in the observation points for acid deposition monitoring described (1), we measured continuously each density of sulfur dioxide measured by auto-measuring machines (SO_2), nitric monoxide (NO), nitrogen oxide (NO_X*[1]), ozone (O_3), the particulate materials under 10 μm (PM10). PM2.5[2] was also measured at Rishiri and Oki. Also, the EANET departments except Ochiishi cape analyzed the atmospheric samples vacuumed for two weeks by the filter-pack method, and measured the concentration of particulate components about sulfur dioxide (SO_2), nitric acid (HNO_3), hydrogen chloride (HCl), ammonia (NH_3).
The measuring items per observation points of acid deposition are shown in table 2-1.

（３） Soil/vegetation monitoring

The soil/vegetation monitoring was conducted at total 25 points in 19 areas which were divided into 3 areas. First one was the area mainly focused on the influence to woods, which were mostly natural forests in mountain region which was thought to be affected by external irreversibility. The second one was the area mainly focused on the influence to soil, which were sensible against acid deposition. The third one was the area focused on the influence to land water, which was important to see the influence to land water (refer to table 2-2 for the list of the areas and figure 2-2 for the points distribution). The monitoring was conducted as follows based on “Soil/vegetation monitoring measuring handbook (1998)” and EANET technical manual.

① Forest monitoring

The general survey to forests (per trees (name, breast high diameter, and height)) was conducted once per 5 years, and the declining survey to woods (declination level, records and presumption by pictures) was conducted once a year.

② Soil monitoring

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[1] It is written as “NO_X*” because peroxyacetyl nitrate and a part of nitric acids as well as besides NO_X (NO and NO_2) are to be measured for analysis. Main elements of NO_x at the at the measurement points in urban areas are considered NO and NO_2, therefore, we count the value subtracting NO_x from NO as NO_2 measured value.

[2] To be precise, the particulate materials collected with sizing devices of which trapping efficiency against particles with 10 μm diameter (aerodynamic diameter) is 50%.

The following items were analyzed with the surface (0－10㎝) and next layer (10－20㎝) of soil. It was conducted once per 5 years.

●Required item: moisture content, pH (H_2O) ,pH (KCl), exchangeable base (Ca、Mg、Na、K), exchangeable acid degree, effective cation ion exchangeable content (ECEC), exchangeable AL and H, contained amount of carbonic acid (only in limestone soil).

● Selected items: overall nitrogen content, overall carbon content, phosphoric acid availability, sulfur ion, soil density, soil hardness.

Table 2-1 Measurement items at each observation points

	Name	Prefecture	Category	Wet	period	NOx	SO_2	O_3	PM10	PM2.5	filter pack	WD/WS	Rain	temp/humid	radiation	EANET
1	Rishiri	Hokkaido	Remote	○	Daily	○	○	○	○	○	○	○	○	○	○	○
2	Sapporo		Urban	○	Weekly							○	○	○		
3	Ochiishi		Remote	○	Daily	○	○	○	○	○		○	○	○	○	○
4	Tappi	Aomori	Remote	○	Daily	○	○	○	○		○	○	○	○	○	○
5	Hachimantai	Iwate	Rural	○	Daily							○	○			
6	Nonodake	Miyagi	Rural	○	Daily			○				○	○	○	○	
7	Obanazawa	Yamagata	Rural	○	Weekly							○	○			
8	Tsukuba	Ibaraki	Rural	○	Daily			○				○	○	○	○	
9	Akagi	Gunma	Rural	○	Daily			○				○	○			
10	Ogasawara	Tokyo	Remote	○	Daily	○	○	○	○		○	○	○	○	○	○
11	Tokyo*		Urban	○	Daily						○		○			○
12	Sado-seki	Niigata	Remote	○	Daily	○	○	○	○		○	○	○	○	○	○
13	Maki		Rural	○	Daily			○				○	○	○	○	
14	Echizen	Fukui	Remote	○	Daily							○	○			
15	Happo	Nagano	Remote	○	Daily	○	○	○	○		○	○	○	○	○	○
16	Ijira	Gifu	Rural	○	Weekly	○	○	○	○		○	○	○	○	○	○
17	Inuyama	Aichi	Rural	○	Daily			○				○	○	○	○	
18	Kyoto	Kyoto	Rural	○	Daily			○				○	○	○	○	
19	Amagasaki	Hyogo	Urban	○	Daily							○	○	○	○	
20	Shionomisaki	Wakayama	Remote	○	Daily							○	○			
21	Oki	Shimane	Remote	○	Daily	○	○	○	○	○	○	○	○	○	○	○
22	Banryu		Urban	○	Weekly	○	○	○	○		○	○	○	○	○	○
23	Kurahashi	Hiroshima	Rural	○	Daily			○				○	○	○	○	
24	Yusuhara	Kouchi	Remote	○	Daily	○	○	○	○		○	○	○	○	○	○
25	Chikugo	Fukuoka	Rural	○	Daily			○				○	○	○	○	
26	Tsushima	Nagasaki	Remote	○	Daily			○				○	○			
27	Goto		Remote	○	Daily							○	○			
28	Kujyu	Oita	Rural	○	Weekly							○	○			
29	Ebino	Miyazaki	Remote	○	Daily		○	○				○	○	○		
30	Yaku	Kagoshima	Remote	○	Weekly							○	○			
31	Hedo	Okinawa	Remote	○	Daily	○	○	○	○		○	○	○	○	○	○

＊ : It started in 2007 at Tokyo.

＊ : Atmosphere concentration and weather items by auto-measuring machines at Ochiishi cape was measured by Center for Global Environmental Research.

Table 2-2 Soil/vegetation monitoring points

	Place	Cat.	Monitor year＊2	Item	Number of plot		
					Woods	Soil	Vegetation
1	Shiretoko National Park (Hokkaido)	Tree	2005	fir	1	2	1
2	Shikotsu Toya National Park (Hokkaido)	Tree	2003	Betula ermanii	1	2	1
3	Towada Hachimantai National Park (Iwate)	Tree	2004	Abies mariesii	1	2	1
4	Bandai Asahi National Park (Niigata)	Tree	2007	beech	1	2	1
5	Nikko National Park (Tochigi)	Tree	2003	beech	1	2	1
6	Chubusangaku National Park (Toyama)	Tree	2005	beech	1	2	1
7	Hakusan National Park (Ishikawa)	Tree	2006	beech	1	2	1
8	Yoshino Kumano National Park (Nara)	Tree	2004	beech	1	2	1
9	Daisen Oki National Park (Tottori)	Tree	2003	beech	1	2	1
10	Ishizuchi Quasi-National Park (Kochi)	Tree	2004	beech	1	2	1
11	Aso Kuju National Park (Oita)	Tree	2005	fagaceae	1	2	1
12	Kirishima Yaku National Park, Yaku Island (Kagoshima)	Tree	2004	cedar	1	2	1
13		Tree	2004	laurilignosa	1	2	1
14	Sekidosan, Horyusan (Ishikawa)	Soil	2005	Red soil, brown forest soil	2	2 × 2	2 × 1
15	Houdouji, Amanosan (Osaka)	Soil	2007	Yellow soil, reddish brown forest soil	2	2 × 2	2 × 1
16	Shimofuridake, Tokusagamine (Yamaguchi)	Soil	2003	Yellow soil, kuroboku soil	2	2 × 2	2 × 1
17	Kashiinomiya, Koshosan (Fukuoka)	Soil	2007	Reddish brown forest soil, brown forest soil	2	2 × 2	2 × 1
18	around Ijirako (Gifu) (Ijira, Yamato) ＊1	water	2006	Ijira catchment brown forest soil, kuroboku soil	2	2 × 2	2 × 1
19	around Banryuko (Shimane) (Banryuko) ＊1	water	2006	Banryuko catchment yellowish brown forest soil, red soil	2	2 × 2	2 × 1

Note) ＊１：Iwami Rinkuu Factory Park: Registered point with EANET

＊２：Year conducted general forest inventories and soil monitorings

Legend symbol

●: Soil/vegetation monitoring points

(Underline shows the areas focused on soil, bold shows EANET registered points.)

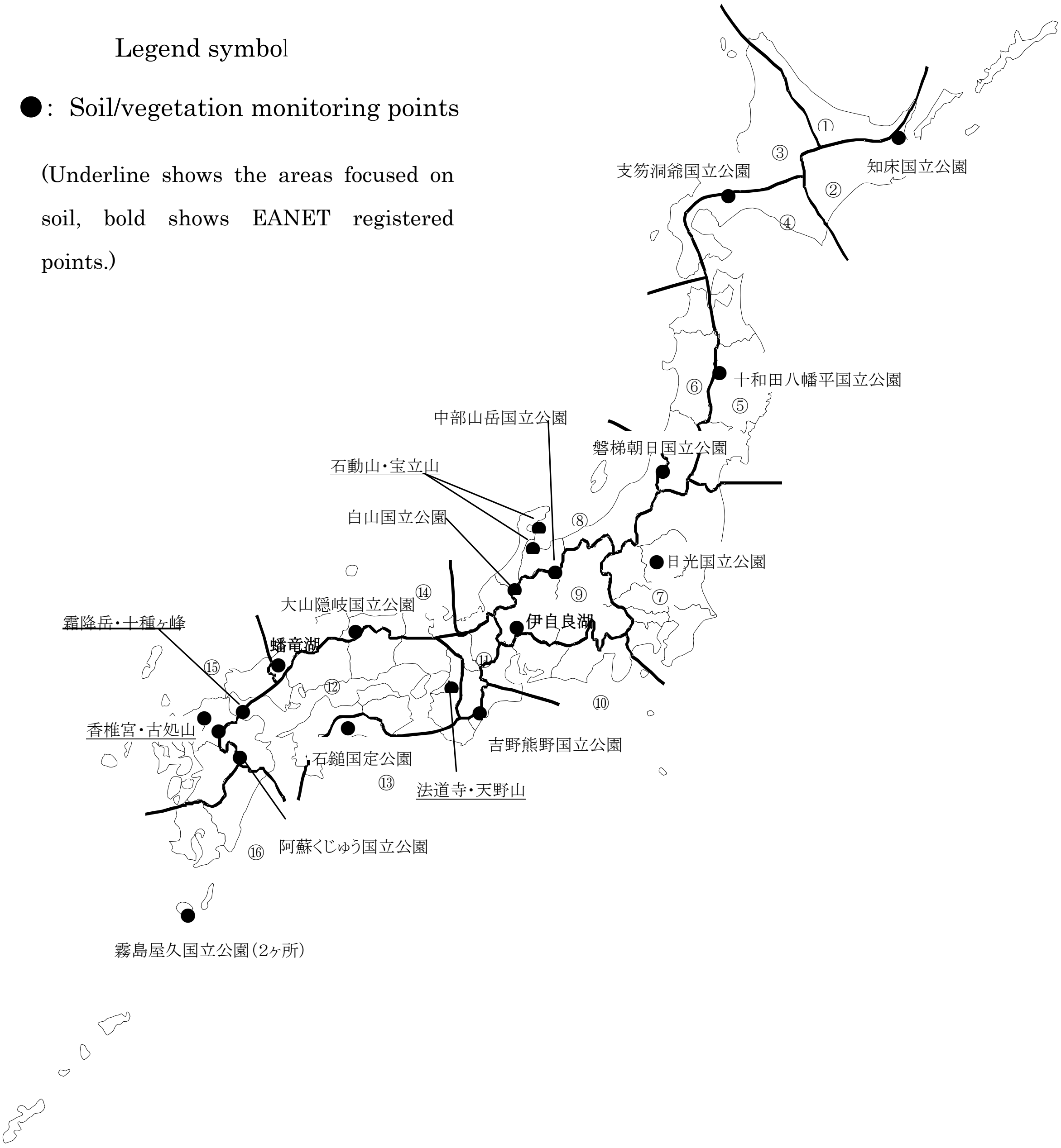


Figure 2-2 Soil/vegetation monitoring points

(The numbers shows the climate division referred by acid deposition monitoring)

（4）Land water monitoring

Land water monitoring was conducted at 11 points which were selected in consideration of the high sensibility against acid deposition, small artificial pollution, and area balance on the basis of "Land water monitoring hand book (February in 2005)" (refer to table 2-3 for the list of the observation points and figure 2-3 for the distribution). The measurement items are following.

① Measurement items for water quality survey 4 times per year: water temperature, pH, electric conductivity, alkali level, NH_4^+, Ca^{2+} , Mg^{2+}、 Na^+, K^+, SO_4^{2-}, NO_3^-, Cl^-, Chl-a, DO (dissolved oxygen)

More than once a year: NO_2^-, PO_4^{3-}, DOC, transparency, water color

② Measurement items for bottom sediment survey (once per 5 years) : SO_4^{2-}、NO_3^-、

Table 2-3 List of lakes for land water monitoring

	Name	Prefecture	year of monitoring of sediment (once five years)	EANET site
1	Imagamioike	Yamagata	2005	
2	Karikomiko	Tochigi	2006	
3	Sankyoike	Niigata	2003	
4	Ohatakeike	Ishikawa	2007	
5	Yashagaike	Fukui	2006	
6	Futagoike	Nagano	2004	
7	Ijirako	Gifu	2006	○
8	Sawanoike	Kyoto	2003	
9	Banryuko	Shimane	2007	○
10	Yamanokuchi-dam	Yamaguchi	2004	
11	Nagatomiike	Kagawa	2007	

In the survey, the samples were collected and analyzed at the local governments concerned, and summarized at Acid Deposition and Oxidant Research Center (including the data checking by the experts at the center and outside specialists). After that, the data was fixed through the inspection at acid deposition investigation meeting (atmosphere and ecosystem committee).

Also, as one of the actions for the Quality Assurance and Quality Control (QA/QC), we sent the precipitation, filter papers of filter pack, simulated samples of land water and soil samples to each laboratory. We conducted the comparative investigations about the analyses (laboratory comparative investigations) and conducted the field surveys about the condition of the ambient surroundings and storage situation of the devices. The next chapter shows the monitoring results (including QA/QC).

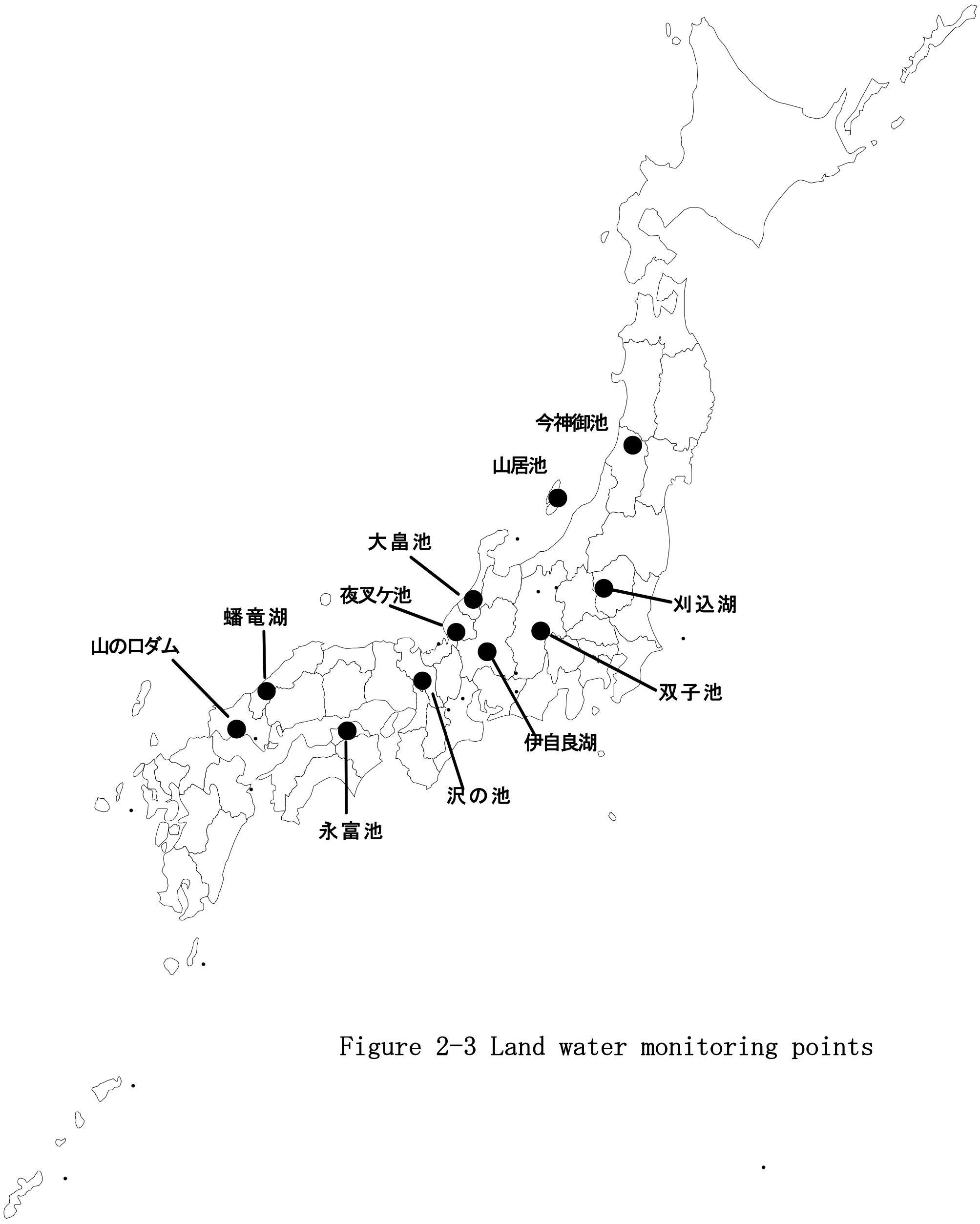


Figure 2-3 Land water monitoring points

3. Monitoring

3.1 Acid Deposition Monitoring

3.1.1 Wet Deposition Monitoring

(1) Trends of the annual mean in wet deposition

The wet deposition monitoring survey for 5 years from 2003 through 2007 has been carried out and clarified the chemical compositions in wet deposition. The obtained data by conducting continually collection and chemical analysis of precipitation samples were collected and comparison between sites was performed. The percentage of the sites which satisfied data completeness criteria for each year were 90% (27/30), 67% (20/30), 70% (21/30), 80% (24/30) and 90% (28/31) in 2003, 2004, 2005, 2006 and 2007 respectively. The numbers of monitoring sites from 2003 through 2006 were 30. In 2007, they were 32 sites, because Tokyo site was newly established in 2007

① Precipitation Amount

The annual precipitation amounts at each monitoring site ranged from 626 mm y^{-1} (Ochiishi in 2004) to 5123mm y^{-1} (Yakushima in 2004). Among the sites, the means of precipitation amount for 5 years at Yakushima (3831mm y^{-1}), Ebino (3117mm y^{-1}) and Yusuhara (2949mm y^{-1}) were much and those at Ochiishi (830mm $y^{-}1$), Rishiri (961mm y^{-1}) and Sapporo (1018mm y^{-1}) were little than other sites.　Then, the tendencies of precipitation amounts were much at the Pacific Ocean side in Shikoku and Kyushu and those in Hokkaido were little.

Moreover, about the variations for five years, an increase or a decrease tendency was not remarkable at every site.

② pH value

The annual mean pH value and the mean pH value for 5 years were shown in Fig .3.1.1. The annual mean pH value at each monitoring site ranged from pH 4.40 (Ijira in 2003) to pH 5.04 (Ogasawara in 2003). Recently, pH value at Rishiri, Sapporo, Goto were becoming low level. The mean pH value for 5 years ranged from pH4.51 (Ijira) to pH 4.95 (Ogasawara), while the overall mean was pH4.68. So, the precipitation in Japan has been still acidifying. Among the sites, the means of pH value at Ogasawara (pH 4.95), Hedo (pH 4.92) and Happo (pH 4.85) were little higher, and those at Ijira (pH4.51), Echizen-misaki (pH4.52) and Niigata-maki (pH4.56) were little lower.

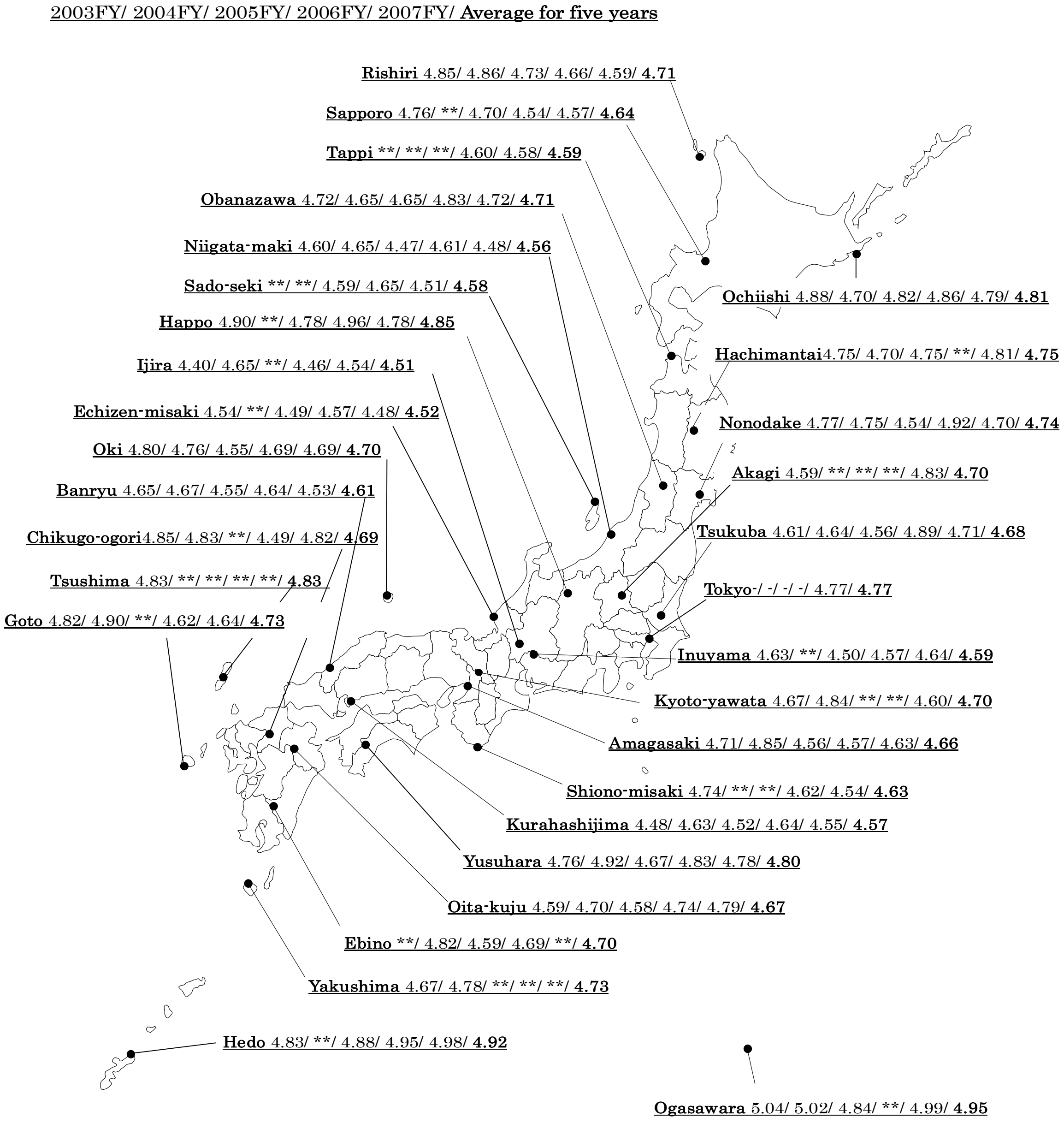
③ Concentrations and deposition amounts of major ion components

pH value is determined by the balance between acids and bases. In the case of wet deposition, sulfuric acid and nitric acid are considered as acids, and gaseous ammonia and basic calcium compounds are considered as base constituents. Therefore, in precipitation, nss-SO_4^{2-} and NO_3^- in precipitation can be considered as acidification indexes, NH_4^+ and nss-Ca^{2+} can be regarded as indexes which control the acidification. Moreover, each wet deposition amount of ion components can serve as effective information, in order to evaluate long-term effects on the ecosystem. The outline of concentrations and deposition amounts of these four main ions components and hydrogen-ion were shown in table.

Data completeness criteria for each site were set up and the data that satisfy the criteria were used for the calculation of monthly and annual means. The data completeness criteria used this report were as follows;

①Percent total precipitation should be 80% or more and percent precipitation coverage length should be 80% or more.

②Contribution of sea salts is 75% or less.



ー : not measured
** : Data do not satisfy criteria for completeness
Notes: Weighted averages for precipitation amounts are calculated

Figure 3-1-1 Annual mean pH value

Table 3-1-1　Concentrations and deposition of main ion components in precipitation

Ion components	Concentration	Deposition
nss-SO_4^{2-}	Range：4.1（2004, Ogasawara）～26.6μmol L^{-1}（2006, Chikugo-ogori、2007, Goto） Mean：13.8μmol L^{-1} Chikugo-ogori and Echizen-misaki were high, and Ogasawara, Yusuhara, and Hedo were low level	Range：5.0（2004, Ogasawara）～67.5mmol m^{-2} y^{-1}（2006, Chikugo-ogori） Yakushima, Ijira, and Ebino were large, and Ochiishi and Ogasawara were small amoumt
NO_3^-	Range：3.2（2005, Ogasawara）～28.8μmol L^{-1}（2007, Banryu） Mean：14.2μmol L^{-1} Tappi, Sado-seki, and Echizen-misaki were high, and Ogasawara, Yusuhara, and Ebino were low level	Range：4.8（2004, Ogasawara）～68.0 mmol m^{-2} y^{-1}（2006, Ijira） Ijira, Yakushima and Echizen-misaki were large, and Ogasawara and Ochiishi were small amount
NH_4^+	Range：3.6（2005, Ogasawara）～37.2μmol L^{-1}（2006, Chikugo-ogori） Mean：15.1μmol L^{-1} Tokyo, Chikugo-ogori, and Sapporo were high, and Ogasawara, Yusuhara, and Hedo were low level	Range：5.7（2004, Ogasawara）～94.2 mmol m^{-2} y^{-1}（2006, Chikugo-ogori） Chikugo-ogori and Ijira were large, andOgasawara and Ochiishi were small amount
nss-Ca^{2+}	Range：0.8（2005, Ogasawara）～11.0μmol L^{-1}（2007, Chikugo-ogori） Mean：3.3μmol L^{-1} Tappi, Chikugo-ogori, and Sado-seki were high, and Ebino, Chikugo-ogori, and Yusuhara were low level	Range：1.7（2005, Ogasawara）～17.9 mmol m^{-2} y^{-1}（2007, Chikugo-ogori） Chikugo-ogori and Happo were large, and Ochiishi, Ogasawara, and Rishiri were small amount
H^+	Range：9.1（2004, Ogasawara）～39.7μmol L^{-1}（2004, Ijira） Mean：20.8μmol L^{-1} Ijira and Echizen-misaki were high, and Ogasawara and Hedo were low level	Range：10.8（2004, Ochiichi）～115 mmol m^{-2} y^{-1}（2004, Ijira） Ijira and Yakushima were large, and Ochiishi, Ogasawara, and Rishiri were small amount

(2) Seasonal variation of wet deposition

In order to evaluation of seasonal fluctuation of wet deposition in each area, at first, 31 monitoring sites were classified into seven areas, Hokkaido, the Sea of Japan side in central and northern Honshu island, the Pacific Ocean side, the Inland Sea of Japan side, San-in, the East China Sea side, and Southwest islands. And then, the analysis on seasonal fluctuation of deposition amounts of each ion components were carried out for every area. Seasonal fluctuations about deposition amount, concentration, and precipitation amount of nss-SO_4^{2-} and NO_3^- which were indexes which contribute to acidification of precipitation, were shown in Fig.3-1-2 and Fig.3-1-3, respectively.

There were most precipitation in July in the Sea of Japan side in central and northern Honshu island, the Pacific Ocean side, the Inland Sea of Japan side, San-in, and the East China Sea side and was most precipitation in June on Southwest islands, because the rainy season has affected these area considerably. The other hand, there was most precipitation amount in September in Hokkaido because of the influence of typhoons.

The nss-SO_4^{2-} concentrations have tended to become high from winter to spring in area except Hokkaido. Moreover, NO_3^- concentrations have trended to become high from winter to spring in area except Southwest islands.

On a national scale, concentrations of nss-SO42- and NO3- increased generally in the periods from winter to spring months whereas nss- SO42- in Hokkaido and NO3- in Southwest islands did not show clear seasonality as the other sites. The deposition showed a steep increase from late autumn to spring in the Sea of Japan side in central and northern Honshu island and San-in. This phenomenon is interpreted as indicating that increasing amounts of acids and its precursors are injected into the atmosphere during this period and suggesting continental pollutants are transported over these regions. In contrast, the deposition in the Pacific Ocean side, the Inland Sea of Japan side, and the East China Sea side had a maximum in July when the precipitation amount peaked in a year. Minimum deposition took place in winter months in the Pacific Ocean side and the Inland Sea of Japan side. In Hokkaido, deposition of nss-SO_4^{2-} and NO_3^- was likely to be lower than that in the other regions throughout the year. Concentration and deposition of NH_4^+ had similar seasonality to those of NO_3^-, which implies these two species followed a common atmospheric process. Non-sea-salt calcium ion showed the maximum in the concentration and deposition in spring time in all the regions except Southwest islands which had a second maximum in this season. This nation-wide seasonality suggested Kosa, or Asian dust, had a significant influence on precipitation chemistry in Japan.

Hydrogen ion concentration showed the maximum in winter in the regions of the Sea of Japan side in central and northern Honshu island, San-in, the East China Sea side, and Southwest islands although no apparent seasonality was noted in Hokkaido, the Pacific Ocean side, and the

Inland Sea of Japan side. The deposition was largest in winter time in the Sea of Japan side in central and northern Honshu island and San-in while the maximum deposition occurred in July in the Pacific Ocean side, the Inland Sea of Japan side, and the East China Sea side.

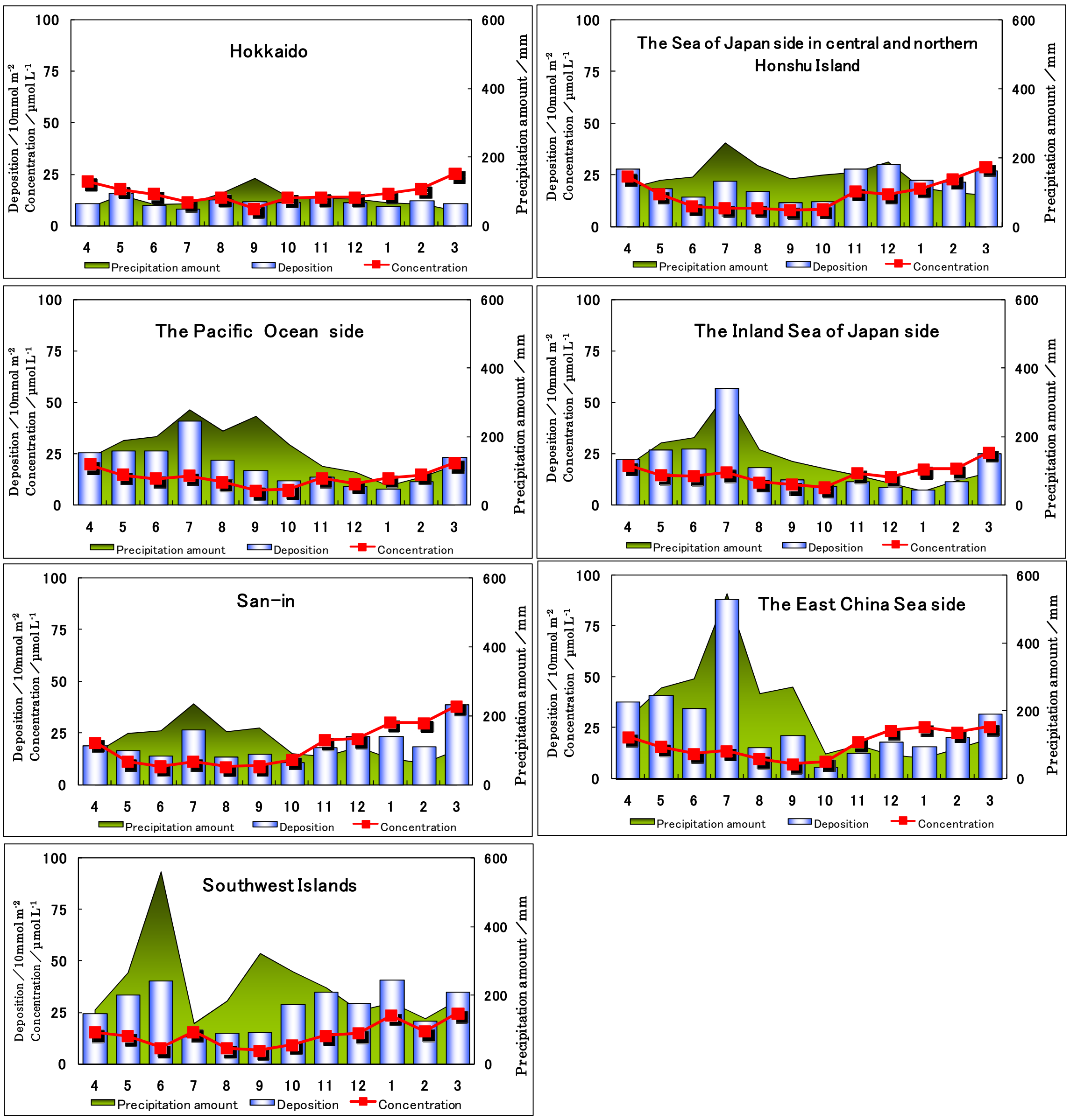


Figure 3-1-2 Regional differences of seasonal variation of nss-SO_4^{2-} concentration and deposition (2003-2007)

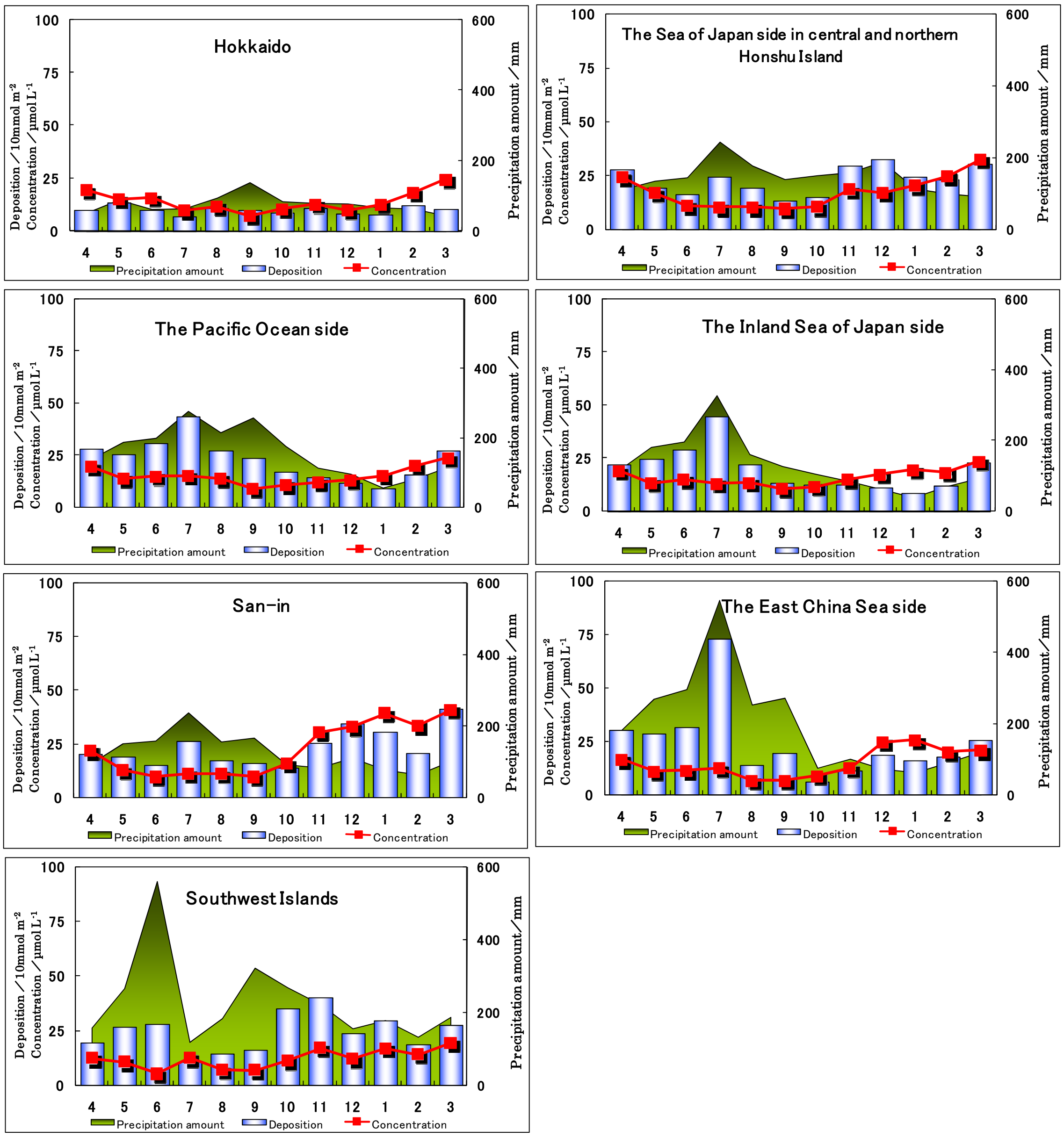


Figure 3-1-3 Regional differences of seasonal variation of NO_3^- concentration and deposition (2003-2007)

(3) Long-term trends of wet deposition

In order to assess long-term trends of wet deposition, annual variations of the national medians for the annual means of rainfall amount, concentrations and deposition of major ions were explored for the sites with more than ten year valid measurements in the period from FY1991 to FY2007.

The medians for the rainfall amount and the concentrations are summarized in Fig. 3.1.4. The precipitation amount fluctuated in FY1990s and approached to a plateau in FY2000s. Deposition of nss-SO$_4^{2-}$ was invariant throughout the period in spite of some variations. Nitrate deposition increased in the middle of 1990s and changed by leveling off thereafter. Ammonium and nss-Ca2+ deposition leveled off during the period with some slight fluctuation. Deposition of H$^+$ was likely to increase from the middle of FY1990s to FY2000 and then showed some fluctuations.

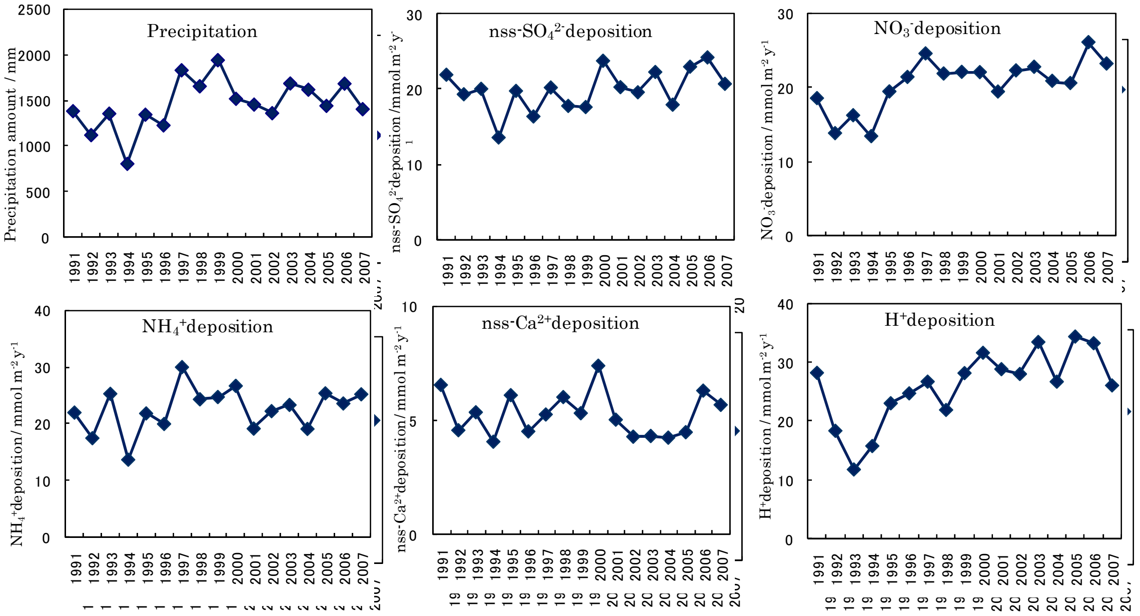


Figure 3-1-4 Trends in annual median of precipitation amounts and deposition of ion components

The annual trends of the medians for pH and concentrations of major ions are illustrated in Fig. 3.1.5. Generally, pH has been rather invariant with some fluctuations. Concentration of nss-SO42- had a decreasing trend up to FY1999, increased in FY2000, and eventually approached a plateau whereas that of NO$_3^-$ has been invariant until FY2004 with a fluctuating trend and increased after FY2005. Ammonium concentration has been generally unvarying with occasional fluctuation and nss-Ca2+ concentration fluctuated in such a manner as it was elevated in FY2000 whereas lowered in FY2003 and FY2004.

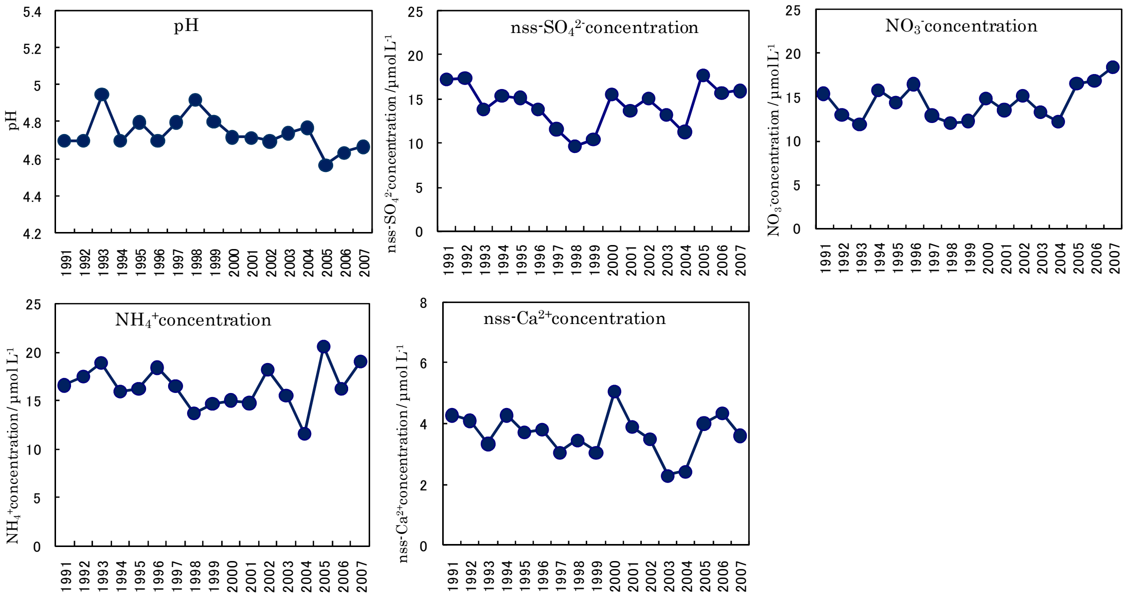


Figure 3-1-5 Trends in annual median of pH value and concentration of ion components

(4) Quality Assurance / Quality Control

To obtain comparable, high quality monitoring data, each participating country is required to carry out acid deposition monitoring using common methodologies as specified in the *Guidelines for Acid Deposition Monitoring in East Asia*, *Technical Documents on Wet Deposition Monitoring in East Asia*. And the inter-laboratory comparison project (round robin analysis survey) was conducted among the analytical laboratories in participating countries of the Acid Deposition Monitoring Network in East Asia (EANET). Some of Japan domestic center or analytical agency participate WMO or EMEP inter-laboratory comparison, and keep analytical quality. Acid Deposition and Oxidant Research Center also direct site audit to monitoring site and analytical laboratory.

1) Inter-laboratory comparison project of wet deposition measurements

The objectives of the project are, through the evaluation of analytical results, analytical equipment and its operating condition and other practices,

i) to recognize the analytical precision and accuracy of the measurement in each participating laboratory,

ii) to give an opportunity to improve the quality of the analysis on wet deposition

iii) to improve reliability of analytical data through the assessment of suitable analytical methods and techniques.

ADORC provide two practice rain sample (high concentration and low concentration) to monitoring center or analytical agency, these analytical data will submitted to ADORC, and assessed by Data Quality Objectives (DQOs : with in ±15%)*.

In this investigate term, analytical data quality keep 99% of DQOs at high concentration samples, and 90% of DQOs at low concentration samples.

*(Flag E will be marked 15% to 30% of DQOs. Flag X will be marked over 30% of DQOs)

2) Site audit of monitoring stations

As part of the QA/QC, ADORC direct site audit to monitoring site and analytical laboratory. EANET site is once two years, other domestic site is once three years. Site audit checked wet sampler condition, surround blockade or source of pollution, collecting sample, and analytical methods.

3.1.2 Dry Deposition Monitoring

(1) Trend of annual mean and seasonal variation in dry depostion

The outline of dry deposition monitoring for major monitoring parameter in 2003 to 2007, was shown in Table 3-1-2. Moreover, temporal variations of monthly mean for SO2, O3 and PM10 and PM2.5 based on the five year estimates were shown in Figure 3-1-6, 3-1-7, and 3-1-8, respectively. The other parameters were referring to the reference data.The annual value and monthly value which were used for analysis were based on the datasets with data completeness of over 70% (An automatic monitor method: one hour value, a filter pack method: two weeks value).

Major measurement parameters

① Automatic Monitor Method

SO_2（12sites）、NOx*（11sites）、O_3（21sites）、PM_{10}（11sites）、$PM_{2.5}$（3sites）

② Filter-pack Method

Concentration of particulate component (SO_4^{2-}、NO_3^-、NH_4^+、Ca^{2+})（11sites）

Concentration of gaseous component （HNO_3、NH_3）（11sites）

About SO_4^{2-} in the particulate component, non-sea salt SO_4^{2-} computed from Na^+ concentration as an index of a sea salt like wet deposition, was used for consideration.

Table 3-1-2　Results of main monitoring items

Monitoring Items	Tendency of annual means and variations of monthly concentrations
SO_2 Automatic Monitor	・Range : <0.1ppb（2003, 2004, and 2007: Ogasawara）～1.2ppb（2005: Yusuhara、2007: Banryu and Ebino） Yusuhara,Ebino, and Banryu were high, and Ogasawara was low level. Mean (2003-2007): 0.6ppb The variations of monthly mean; There were many sites where SO_2 concentrations became high from the late autumn to spring, and those concentrations rose greatly on the western Japan. In Ogasawara, low concentration continued through every year. The high concentrations from the late autumn to spring were considered to be the effect of transboundary air pollution like the seasonal variation of the nss-SO_4^{2-} deposition described by 3.1.1 (2).
NOx* Automatic Monitor	・Range : 0.4ppb（2003, 2004, and 2007: Ogasawara）～4.2ppb（2003: Banryu） Banryu, Ijira and Banryu were high, and Ogasawara was low level. Mean (2003-2007) : 1.7ppb The variations of monthly mean; The high concentrations in the western Japan or along the coast of japan sea from the late autumn to spring were considered to be the effect of transboundary air pollution like the seasonal variation of the NO_3^- deposition described by 3.1.1 (2). The concentration at Ijira was low in winter, and fluctuation of concentration at Rishiri, Ochiishi, Happo, and Hedo were small through every year.
O_3 Automatic Monitor	・Range : 19ppb（2005: Ijira）～60ppb（2004: Akagi、and Happo） Happo and Akagi were high, and Ijira and Kyoto-yawata were low level Mean (2003-2007): 39ppb The variations of monthly mean; O_3 concentration at all sites were high in spring and low in summer, moreover in western Japan, high concentrations of O_3 were appeared in autumn. O_3 concentrations were high at Happo and Akagi where these height were higher than the other sites through every year. The high concentration in spring was considered to be the effect of transboundary air pollution.
PM_{10} Automatic	・Range : 11µg m^{-3}（2003: Ogasawara）～37µg m^{-3}（2005: Hedo） Hedo was high, and Ogasawara and Happo were low level

Monitor	・Mean (2003-2007)：22μg m^{-3} The variations of monthly mean; PM_{10} concentration was high in spring at all sites and there were some sites where the concentration in autumn becomes a little higher. The high concentrations of Ca^{2+} were considered to be effected by yellow sand, because the Ca^{2+} concentration in aerosol were higher in spring than other seasons.
$PM_{2.5}$ Automatic Monitor	・Range：7μg m^{-3} （2004: Ochiishi）～16μg m^{-3}（2005: Oki） It is high in order of Oki, Ochiishi, and Rishiri. ・Mean (2003-2007)：11μg m^{-3} The variations of monthly mean; $PM_{2.5}$ Concentration at Oki is slightly high in spring, and Rishiri and Ochiishi had small fluctuation through every year.
nss-SO_4^{2-} Filter-Pack Method	・Range：0.52μg m^{-3} （2004: Ogasawara）～6.60μg m^{-3}（2005: Banryu） Banryu and Yusuhara were high, and Ogasawara was low level ・Mean (2003-2007)：3.56μg m^{-3} The variations of monthly mean; nss-SO_4^{2-} Concentrations at almost sites were high in summer.
NO_3^- Filter-Pack Method	・Range：0.11μg m^{-3} （2004: Ogasawara）～4.08μg m^{-3}（2007: Tokyo） Hedo and Banryu were high, and Ogasawara and Happo were low level. Tokyo was maximum value, but this data was only for one year. ・Mean (2003-2007)：0.87μg m^{-3} The variations of monthly mean; There were high concentrations in spring at almost sites.
HNO_3 Filter-Pack Method	・Range ：0.03ppb （2003 and 2004 : Ogasawara）～0.93ppb（2007: Tokyo） Yusuhara and Ijira were high, and Ogasawara and Rishiri were low level. Tokyo was maximum value, but this data was only for one year. ・Mean (2003-2007)：0.24ppb The variations of monthly mean; The little high concentrations were seen in a summer. HNO_3 concentration at Ogasawara and Hedo were low and had little fluctuation.
NH_4^+ Filter-Pack Method	・Range：0.08μg m^{-3} （2004: Ogasawara）～1.90μg m^{-3}（2007: Tokyo） Ijira, Banryu, and Yusuhara were high, and Ogasawara and Rishiri were low level. Tokyo was maximum value, but this data was only for one year. ・Mean (2003-2007)：0.83μg m^{-3} The variations of monthly mean; There were high concentrations in summer at almost sites.

NH_3 Filter-Pack Method	・Range：0.18ppb　(2003: Ogasawara)～5.64ppb (2007: Tokyo) Ijira and Banryu were high, and Rishiri, Tappi, and Happo were low level. Tokyo was maximum value, but this data was only for one year. ・Mean (2003-2007)：0.78ppb The variations of monthly mean; The high concentrations from spring to summer were appeared at many sites. Although Tokyo had only one year data, high concentration was continued.
Ca^{2+} Filter-Pack Method	・Range　: 0.02μg m^{-3}　(2004: Ogasawara)～0.57μg m^{-3} (2007: Tokyo) Hedo was high, and Ogasawara was low level. Tokyo was maximum value, but this data was only for one year. ・Mean (2003-2007)：0.24μg m^{-3} The variations of monthly mean; The high concentrations were appeared at almost sites in spring, and the effect of yellow sand was suggested.

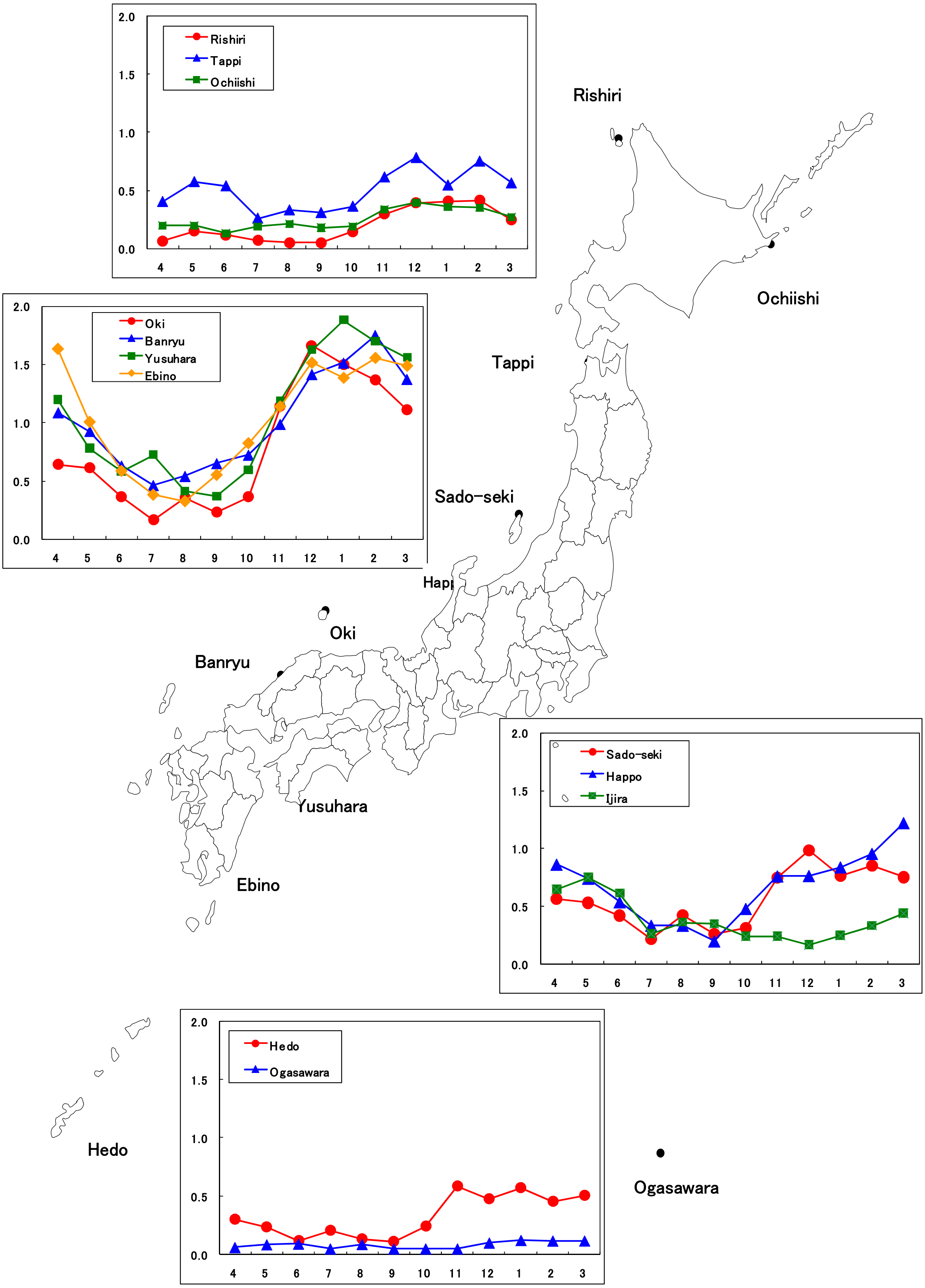


Figure 3-1-6　Temporal variation of monthly mean concentration of SO_2 in2003-2007

（Unit：ppb）

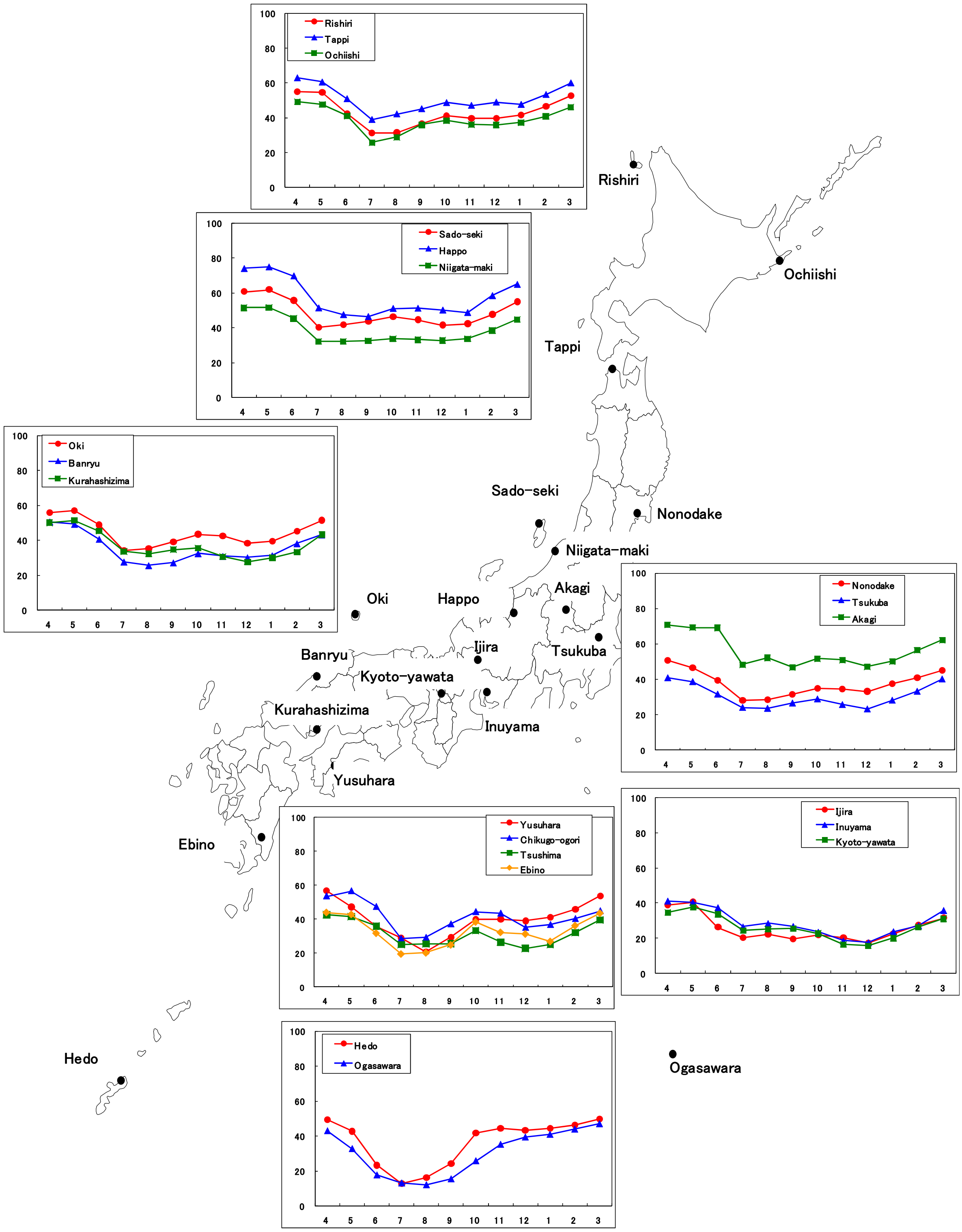


Figure 3-1-7 Temporal variation of monthly mean concentration of O_3 in2003-2007 (Unit:ppb)

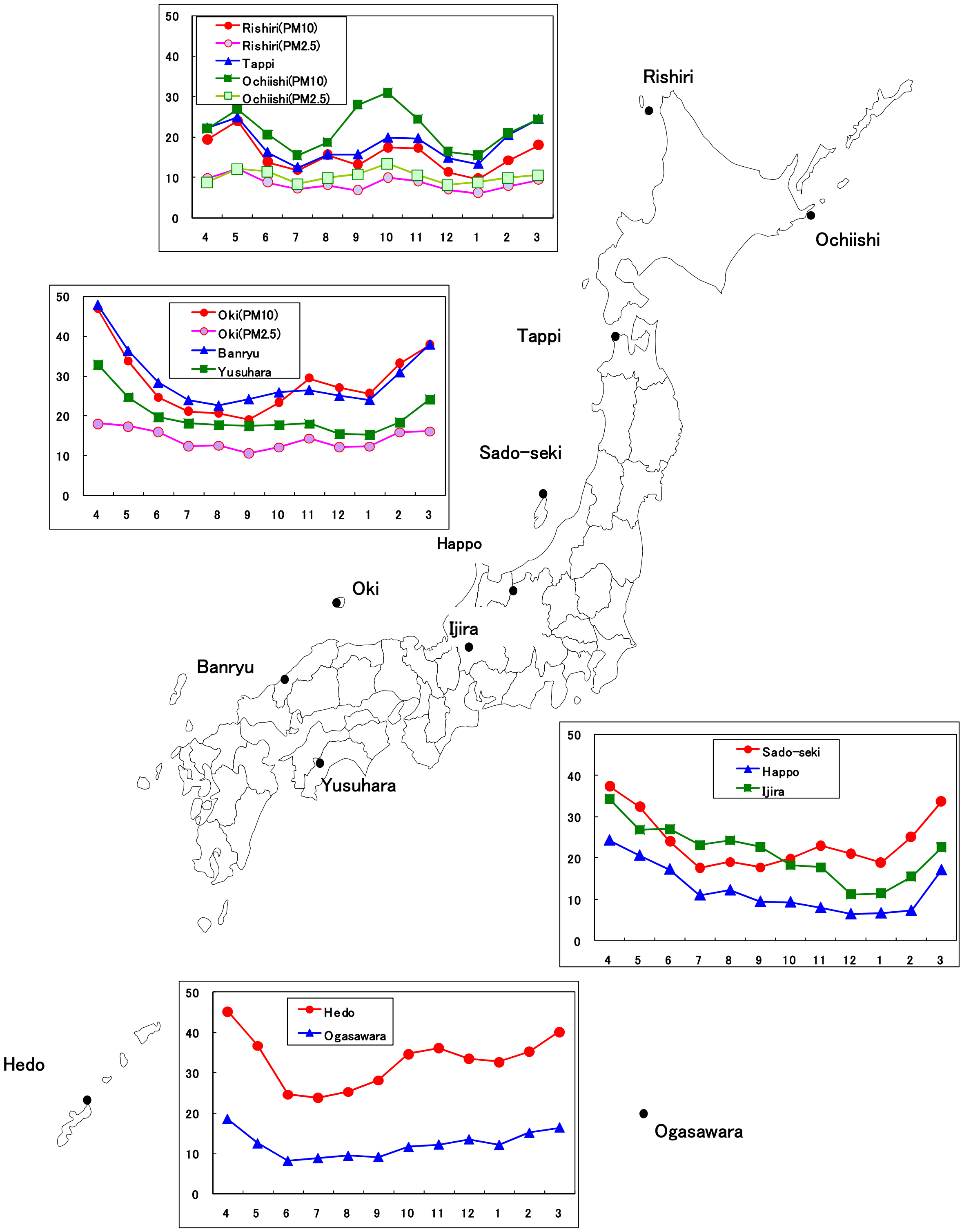


Figure 3-1-8 Temporal variation of monthly mean concentration of PM_{10} and $PM_{2.5}$ in2003-2007

(Unit: μg m^{-3})

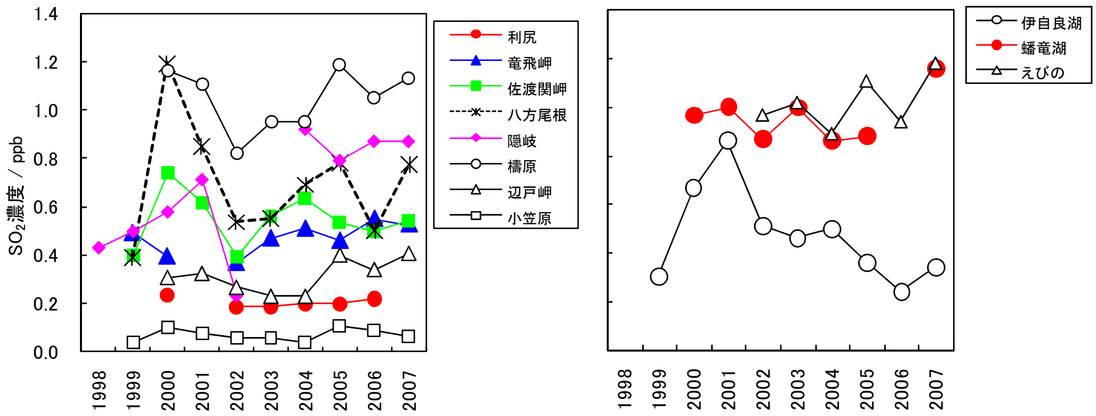
(2) Long-term trends of dry deposition

・SO_2

During the period from 1998 to 2007, temporal variations of SO_2 concentration were illustrated for sites with valid data for more than six years in Figure 3-1-9.

SO_2 concentrations in remote sites were 1.0ppb or less in concentration in general. In it, the concentration in Yusuhara was high and these in Ogasawara and Rishiri located far from the continent were low in this period. In remote sites along the coast of Japan Sea, SO_2 concentrations were likely to decreased from the west side in order of Oki, Sado-Seki, Tappi, and Rishiri, and it was suggested that continental source contribution was larger as the site position was nearer to the continent. On the other hand, in non-remote sites including Ebino near Sakurajima an active volcano, the concentration in Ebino and Banryu were same level that were higher than in Ijira in this period.

In Sado-seki, Happo, Ijira, and Yusuhara, SO_2 concentrations were higher in 2000 and 2001, which would be attributable to the volcanic SO_2 originating the eruption of Mount Oyama on Miyake Island in August, 2000. In consideration of this effect, in recent years, some increasing trends on the concentration with fluctuations were noted in Tappi, Oki, Yusuhara, and Hedo, and the decreasing trend was suggested in Ijira.



［a］：Remote sites ［b］：Non-remote sites

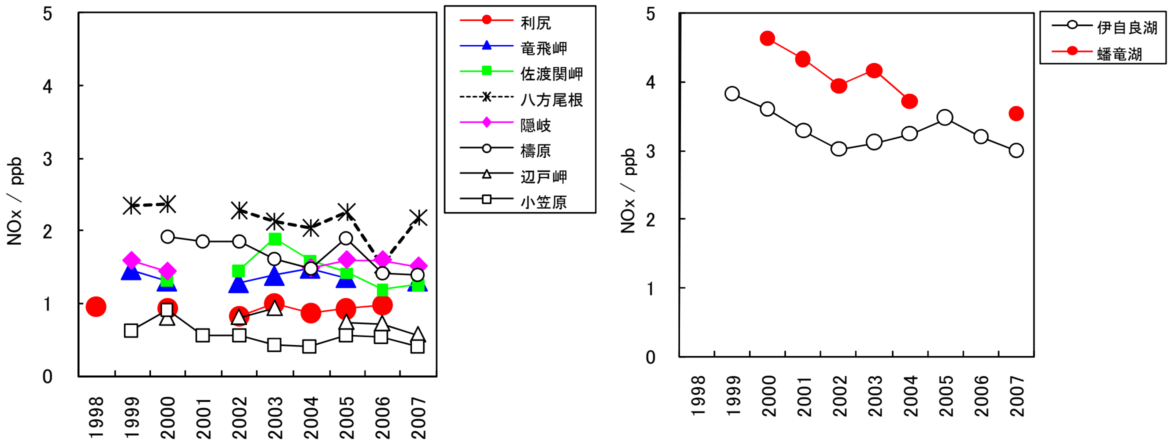
Figure 3-1-9 Trends in annual SO_2 concentration

・NOx*

During the period from 1998 to 2007, temporal variations of NO_X* concentration were illustrated for sites with valid data for more than six years in Figure 3-1-10.

NO_X* concentration in Ijira and Banryu in non-remote site were higher than other remote sites. In remote site, the concentration in Ogasawara and Hedo were low and that in Happo was high. About long-term trend, decreasing trends on the concentration were noted in Yusuhara, Ijira,and

Banryu, and the stable trends were suggested in other sites.



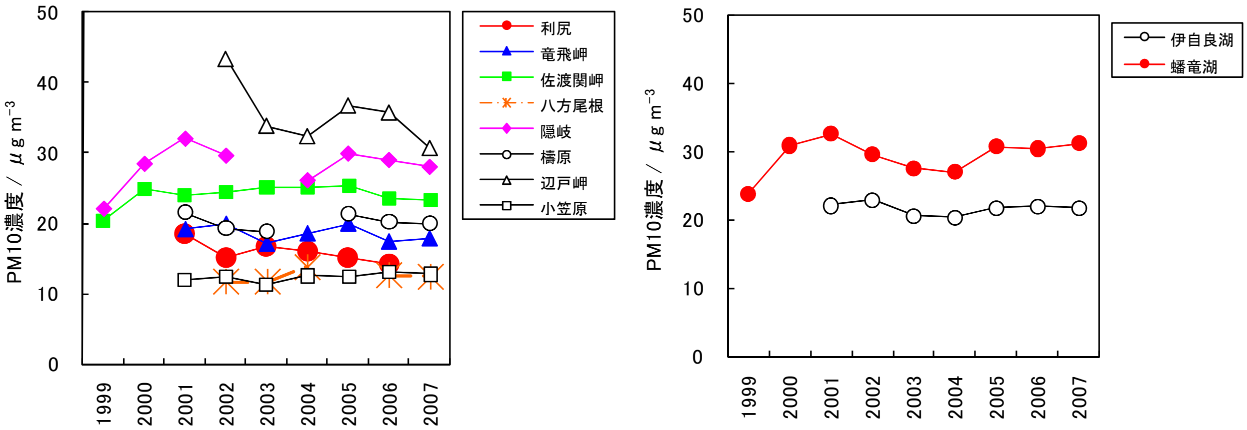
［a］：Remote sites ［b］：Non-remote sites

Figure 3-1-10 Trends in annual NO_X * concentration

・PM_{10}

During the period from 1999 to 2007, temporal variations of PM_{10} concentration were illustrated for sites with valid data for more than five years in Figure 3-1-11.

In remote site, the concentration in Ogasawara and Happo were low and that in Hedo near from the continent was high. Moreover, in remote sites along the coast of the Japan Sea, In remote sites in the Sea of Japan side, PM_{10} concentrations were likely to decreased from the west side in order of Oki, Sado-Seki, Tappi, and Rishiri, and it was suggested that continental source contribution was larger as the site position was nearer to the continent like SO_2 concentration. In non-remote sites, PM_{10} concentration in Banryu was higher than in Ijira in this period. The long-term trends were stable in almost sites except Hedo.



［a］：Remote sites ［b］：Non-remote sites

Figure 3-1-11 Trends in annual PM_{10} concentration

・O_3

Monitoring sites where the valid data was obtained from 1998 in 2007 for seven years or more were classified into the remote (Rishiri, Tappi, Ogasawara, Sado-seki, Oki, Yusuhara, Tsushima, and Hedo), the non-remote (Ijira, Kyoto-yawata, Banryu, Kurahashijima, Chikugo-ogori), the alpine remote (happo), and alpine non-remote (Akagi). Trends of annual mean concentration of O_3 in 2000 to 2007 when the valid data was obtained at 12 or more sites were shown in Figure 3-1-12 and 3-1-13.

Since annual mean concentratios for every classified area were high in order of the alpine, the remote, and the effect from the stratosphere in the alpine were larger than those from the ground level and the disappearance reaction of O_3 by NO in non remote had arisen rather than the remote.

Moreover, O_3 annual mean concentration ia remote, non-remote, alpine remote, and all sites were tended to increase.At the remote sites, O_3 concentrations were likely to decrease in order of the mountainous sites, the Sea of Japan side, and the Pacific Ocean side.

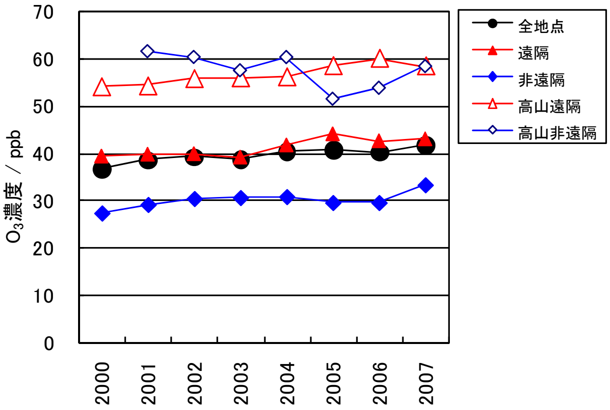


Figure 3-1-12　Trends in annual PM_{10} concentration every regions

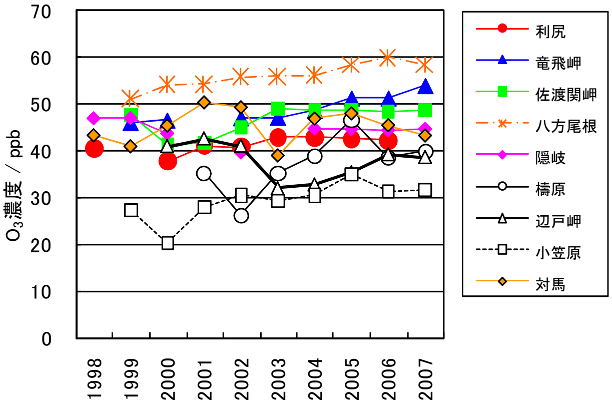


Figure 3-1-13　Trends in annual O_3 concentration

(3) Estimation of Dry Deposition

In 10 sites (EANET sites except Tokyo and Ochiishi) which monitored the atmosphere using the filter-pack method in the 2003 to 2007, the dry deposition (gaseous substance: SO_2, HNO_3, NH_3, ion components in particulate matters: nss-SO_4^{2-}, NO_3^-, NH_4^+) at each site was estimated, and the results were shown in Table 3-1-3. The estimation method was made into the presuming method (the Inferential method) which searched from the product of the ingredient for the concentration in the atmosphere and the deposition rate of the each component. The deposition rate of each component was setting deposition surface to the forest and the grass field using the data of meteorological elements based on the Matsuda's method.

Dry deposition amounts to the forest were much than those to grass field for either the gaseous and particle matter. About the deposition of gaseous substances to the forest, SO_2 depositions at Banryu, Hedo, Sado-seki, and Tappi were larger, and those at Ijira and Rishiri were smaller. HNO_3 depositions at Yusuhara, happo, Sado-seki, Banryu, and Tappi were larger, and that at Ogasawara was small. Moreover, NH_3 deposition showed large amount at Banryu. About the deposition of the particulate matter to the forest, every depositoin at Tappi and Hedo were large, and those at Ogasawara and Ijira were small. The concentration of gaseous and particulate matter at Tappi and Sado-seki were not higher than other sites. But, there was a tendency for a deposition rate to become large depending on an ingredient, so that wind velocity was large. As the results, the large deposition amounts were estimated at both sites where had large wind velocity compared with the other sites. There is a tendency for a deposition rate to become large depending on an ingredient, so that wind velocity is large.

(4) Quality Assurance / Quality Control

Based on the 3rd Scientific Advisory Committee, inter-laboratory comparison for dry deposition started since 2005. Impregnated filters which contain three ions, SO_4^{2-}, Cl^-, and NH_4^+, were prepared and distributed to the participating laboratories by the Network Center (NC). Most of the laboratories participating in EANET activities joined this activity and submitted their analytical results to NC. Obtained results for the amount of SO_4^{2-}, Cl^-, and NH_4^+ on the distributed filters were compared with the prepared values and statistically treated.

Submitted analytical data quality for low concentration sample were keep these quality, 83.3% of DQOs at 1st project, 85.7% of DQOs at 2nd project, and 66.7% of DQOs at 3rd project.

In high concentration samples were keep these quality, 91.7% of DQOs at 1st project, 100% of DQOs at 2nd project, and 95.2% of DQOs at 3rd project.

Table 3-1-3 Dry Deposition

Site	Year	Gaseous substance (mmol m^{-2} $year^{-1}$)						Particulate matter (mmol m^{-2} $year^{-1}$)					
		Forest			Grass field			Forest			Grass field		
		SO_2	HNO_3	NH_3	SO_2	HNO_3	NH_3	nss-SO_4^{2-}	NO_3^-	NH_4^+	nss-SO_4^{2-}	NO_3^-	NH_4^+
Rishiri	2003	4.0	7.2	4.7	2.2	2.0	2.7	7.7	4.5	8.6	0.8	0.4	0.9
	2004	4.1	5.1	5.2	2.1	1.4	3.0	7.1	4.6	6.5	0.6	0.4	0.6
	2005	3.9	7.0	4.6	2.1	1.9	2.7	8.9	4.5	9.1	0.8	0.3	0.9
	2006	4.3	4.9	4.5	2.3	1.3	2.3	7.2	3.9	7.3	0.7	0.3	0.7
	2007	**	**	**	**	**	**	**	**	**	**	**	**
Tappi	2003	15.9	19.4	5.2	7.3	5.4	3.9	13.5	11.6	19.2	1.0	0.7	1.4
	2004	16.9	21.2	5.2	7.2	5.9	3.5	17.9	14.0	19.3	1.1	0.7	1.3
	2005	14.1	22.0	2.3	6.2	6.1	1.7	21.8	16.7	25.3	1.6	0.9	1.8
	2006	16.6	16.7	2.8	8.0	4.7	2.1	19.1	16.2	22.7	1.4	0.9	1.7
	2007	15.2	20.7	4.1	7.8	5.8	3.2	22.9	18.3	23.5	1.5	1.0	1.7
Ogasawara	2003	7.6	2.0	2.3	2.7	0.5	1.2	1.5	1.1	1.0	0.4	0.2	0.3
	2004	7.6	2.3	3.4	3.3	0.6	2.0	0.5	0.3	0.4	0.1	0.1	0.1
	2005	9.9	2.5	7.1	3.7	0.7	4.0	1.6	0.7	1.0	0.4	0.1	0.2
	2006	**	**	**	**	**	**	**	**	**	**	**	**
	2007	1.9	2.6	5.1	0.7	0.7	2.8	4.3	1.9	3.0	0.7	0.3	0.5
Sade-seki	2003	12.5	18.3	3.3	5.4	5.0	2.4	10.1	6.5	8.3	0.7	0.4	0.7
	2004	**	**	**	**	**	**	**	**	**	**	**	**
	2005	16.1	28.8	4.2	7.0	7.9	3.2	15.4	7.7	14.3	1.3	0.5	1.3
	2006	20.7	23.2	4.4	8.1	6.4	3.2	12.7	7.8	12.0	1.0	0.5	1.1
	2007	22.5	20.3	4.5	9.6	5.6	3.5	18.9	13.3	17.4	1.3	0.8	1.3
Happo	2003	11.8	21.8	6.8	5.5	6.1	3.1	8.7	3.2	10.9	0.8	0.2	1.2
	2004	15.9	23.4	5.3	6.8	6.6	2.9	11.0	4.0	13.0	1.0	0.3	1.4
	2005	16.5	14.9	6.9	7.4	4.2	3.7	12.3	3.5	14.2	1.2	0.2	1.6
	2006	10.9	23.6	5.1	5.5	6.5	2.8	9.5	1.6	10.8	1.0	0.1	1.2
	2007	10.6	24.5	4.2	5.1	6.8	2.3	9.7	1.8	12.0	1.0	0.2	1.3
Ijira	2003	3.9	4.5	8.9	1.4	1.2	3.3	1.7	0.4	3.1	0.5	0.1	1.0
	2004	4.2	7.0	8.5	1.6	1.9	3.5	2.5	0.6	3.7	0.9	0.2	1.3
	2005	3.4	7.5	7.9	1.4	2.0	3.3	2.6	0.5	3.9	0.9	0.2	1.4
	2006	3.0	6.6	7.6	1.3	1.8	3.2	2.3	0.5	3.4	0.9	0.1	1.4
	2007	2.3	5.1	5.9	1.0	1.4	2.7	2.0	0.5	3.0	0.8	0.2	1.3
Oki	2003	12.0	7.2	5.0	5.4	2.0	3.2	11.5	5.7	8.6	1.1	0.4	0.9
	2004	12.7	8.4	5.8	5.6	2.3	4.0	10.7	6.9	12.8	1.1	0.6	1.4
	2005	15.8	16.4	5.5	6.7	4.5	3.9	17.5	8.9	19.8	2.0	0.7	2.4
	2006	15.5	15.9	4.1	7.2	4.3	3.2	16.8	9.0	20.1	1.9	0.7	2.3
	2007	**	**	**	**	**	**	**	**	**	**	**	**
Banryu	2003	18.5	12.7	10.3	8.2	3.5	6.1	8.3	4.3	8.1	1.0	0.4	0.9
	2004	21.6	19.8	13.1	9.0	5.4	7.8	11.7	6.8	15.3	1.5	0.6	2.0
	2005	16.9	23.0	8.3	8.1	6.3	5.3	14.4	8.0	19.4	2.1	0.7	2.7
	2006	21.4	20.9	9.9	9.5	5.7	6.1	15.6	8.5	20.8	2.3	0.8	3.0
	2007	21.2	21.2	8.8	9.5	5.8	5.5	15.6	8.2	20.1	2.0	0.8	2.7
Yusuhara	2003	11.7	16.7	5.4	5.8	4.6	3.0	8.3	2.8	10.3	1.4	0.4	1.7
	2004	11.7	19.0	5.3	5.2	5.2	2.7	11.6	2.3	13.4	1.8	0.3	2.2
	2005	12.3	25.5	3.6	6.4	7.2	2.2	11.9	2.7	15.0	2.5	0.4	3.1
	2006	17.8	23.7	4.8	8.2	6.7	2.4	11.1	2.4	14.8	2.2	0.3	2.8
	2007	17.5	25.3	7.3	8.5	7.1	3.5	12.5	2.5	15.5	2.3	0.4	3.0
Hedo	2003	21.1	8.9	5.0	9.0	2.4	3.9	16.3	17.4	16.7	1.4	1.2	1.4
	2004	28.9	14.2	6.7	12.6	3.9	5.2	9.6	6.6	7.9	0.9	0.5	0.7
	2005	17.1	11.4	5.9	7.5	3.2	4.8	26.0	14.9	22.0	2.4	1.0	2.0
	2006	13.8	8.6	5.4	6.4	2.4	4.3	24.4	14.9	19.4	2.3	1.1	1.9
	2007	12.4	12.0	7.9	5.6	3.3	6.2	26.2	14.3	24.2	2.2	1.0	2.3

** Data was rejected because of less than 70% of valid data in the period

3.1.3 Synthetic consideration to acid deposition

(1) Total Deposition

Total deposition of sulfur and nitrogen were calculated as the sum total value of these wet depositions and dry depositions.

Wet deposition were used the actual measurement value shown at 3.1.1.

Dry deposition were used the value which computed from the weight average of the dry deposition of a forest and a grassy field estimated at 3.1.2 (3) with a land use rate around about 1km of monitoring site.

The distribution of the total deposition of sulfur and nitrogen were shown in Figure 3-1-14 and 3-1-15. The deposition of sulfur and nitrogen were corresponding to sulfur dioxide and nitrogen oxide, respectively, which were the main causative agents of acid deposition.

In addition, the calculated periods were covered from 2003 to 2007, and the data which did not meet the standard of annual completeness were not used.

The total sulfur deposition was calculated from the sum of wet deposition of nss-SO_4^{2-} in precipitation and dry deposition of SO_2 in gaseous substances and nss-SO_4^{2-} in particulate matters.

In addition, total SO_4^{2-} depositions including contribution of a sea salt SO_4^{2-} were published to reference data. Five years mean of total sulfur deposition except ss-SO42- deposition were ranged from 16 to 54 mmol $m^{-2}y^{-1}$ at the sites estimated these total deposition, those at Banryu and Ijira were large, and those at Rishiri and Ogasawara were small. The small total sulfur deposition at Rishiri and Ogasawara were suggested that these sites were located far from the source area.

About contribution of the wet and dry deposition to total deposition, wet deposition was larger at Ijira, Rishiri, Happo, and Ogasawara, dry deposition was larger at Tappi, Hedo, and Oki, and both deposition were almost same at other sites. Although the contribution rate of dry deposition changed with sites, the variation by the difference in a year is small (standard deviation per average value was 0.05-0.23). So, the ratio of wet deposition to dry deposition was stable at the same site. Moreover, the rate of the particulate matters occupied to dry deposition were about 40 % on the average.

When the air pollutants were moved long-range trans-boundary, these were changed to a particulate matters in many cases, and as mentioned above, the contribution of the particulate matters in dry deposition could not ignored , and the influence of the long-range trans-boundary in dry deposition were suggested.

The total of nitrogen was calculated from sum of wet deposition of NO_3^- and NH_4^+ in precipitation and dry deposition of gaseous HNO_3 and NH_4^+, particulate NO_3^-, NH_4^+.

The five years mean of total nitrogen deposition were ranged from 22 to 130mmol m^{-2} y^{-1}, and these at Ijira and Banryu were large, these at Ogasawara and Rishiri were small.

So, Ogasawara and Rishiri sites could not be easily influenced by the long-range trans-boundary from a continent and had not large source around there. Between the sites in the Sea of Japan side and the Pacific Ocean side, the differences in the total deposition were not found clearly.

About contribution of the wet and dry deposition to the total deposition, the wet deposition at Ijira, Rishiri, Ogasawara, Happo, Sado-Seki, Banryu, and Hedo were larger, the dry deposition was larger at Tappi, and both deposition were almost same at the other sites. Since many sites had dry deposition which were occupied about 40% or more of the total deposition, it was suggested that observation of only wet deposition was insufficient for grasp of the nitrogen deposition from atmosphere. Except Ijira located in central Honshu island, Ogasawara and Rishiri which had no source around there and could not be easily influenced by the long-range trans-boundary from a continent, showed the tendency for contribution of the dry deposition occupied in the total deposition to be small.

At the site except Rishiri, Ijira, Hedo, and Ogasawara, gaseous HNO3 was a substance with the biggest contribution to dry deposition of nitrogen.

On the other hand, in addition to gaseous HNO_3, contribution of particulate NH_4^+ was also large at Tappi, Oki, Banryu, and Yusuhara, and also contribution of particulate NO_3^- was also large at Tappi. Many sites which had larger dry deposition of NH_4^+ were located the Sea of Japan side, and since the deposition of particulate nss-SO_4^{2-} were large at these sites, the relevance between these substances was also suggested. Except Ijira, wet and dry deposition of sulfur showed significant positive correlation (1% of percentage of risk), and also indicated significant positive correlation with wet and dry deposition of nitrogen, respectively. Although the causative agent of wet and dry deposition were not originates same source and not conveyed in the same course necessarily, such correlation has suggested that both sulfur and nitrogen were influenced by the source of the industrial area and metropolis where these components were discharged in large amounts.

In CASTNET (Clean Air Status and Trends Network) of U.S. Environmental Protection Agency, the total deposition of sulfur and nitrogen were estimated at five areas (20 sites) in eastern USA.

About dry deposition, since the estimating method for a deposition velocity was differed, strict comparison was not completed, but the average value in 2003 to 2006 was compared by reference.

The sulfur deposition in the eastern USA was ranged from 14 to 38 mmol m^{-2} y^{-1} (mean value : 26 mmol m^{-2} y^{-1}, and those in Japanese sites except Ogasawara and Rishiri, had exceeded this range. Since the sulfur deposition on CASTNET was contained a sea salt components, the total deposition of non-sea salt components were expected still lower. About total deposition of nitrogen, since CASTNET did not calculate a gaseous NH_3, It compared with Japanese data except for gaseous NH3. The nitrogen deposition in the eastern USA were ranged from 26 to 56 mmol $m^{-2}y^{-1}$ (mean value: 45 mmol $m^{-2}y^{-1}$), and those in Japanese sites except Ogasawara, Rishiri, and Hedo, had exceeded this range, and those mean value was 71mmol m^{-2} y^{-1}.

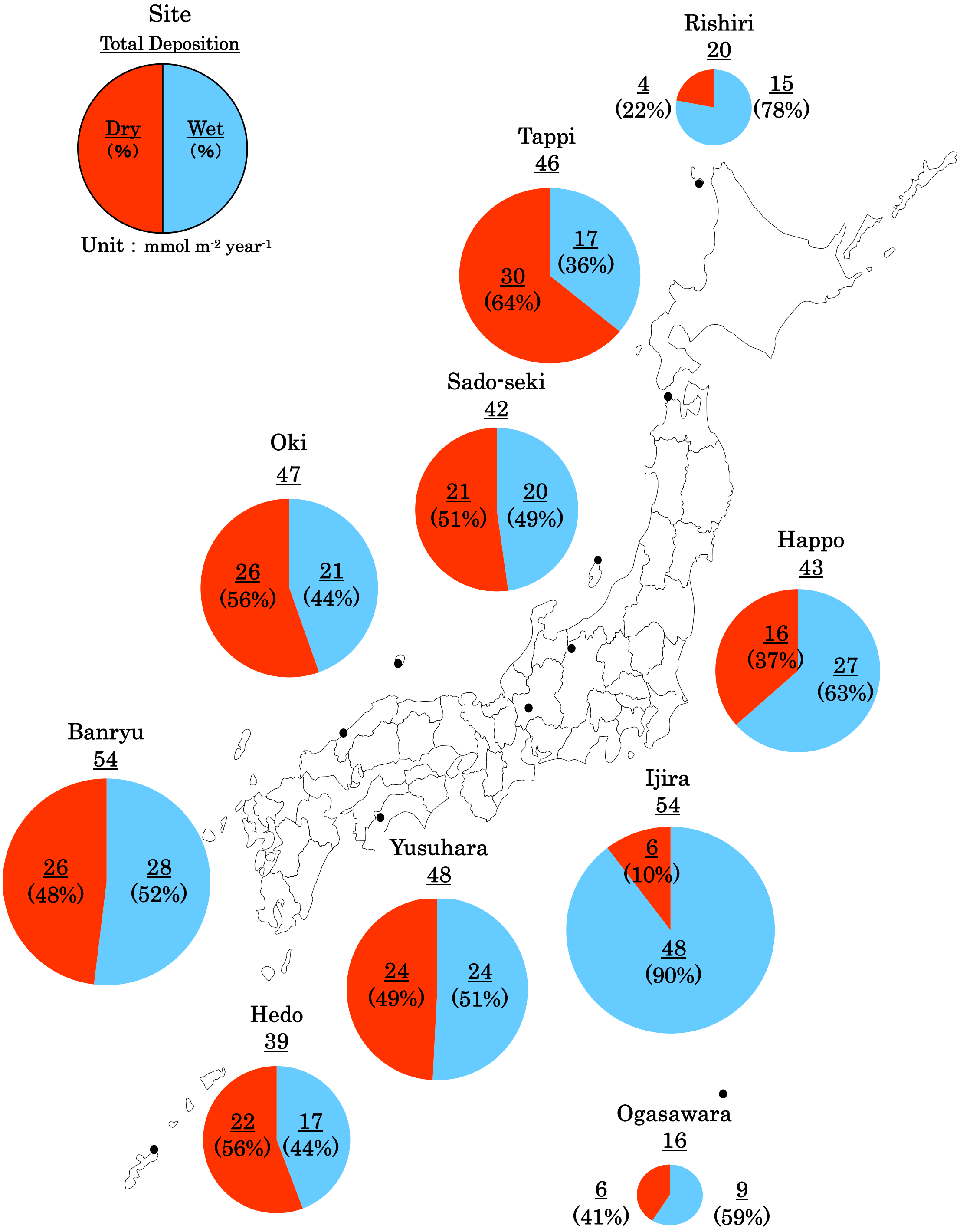


Figure 3-1-14 Average of Annual Total Sulfur Deposition derived from non sea salt (2003-2007)

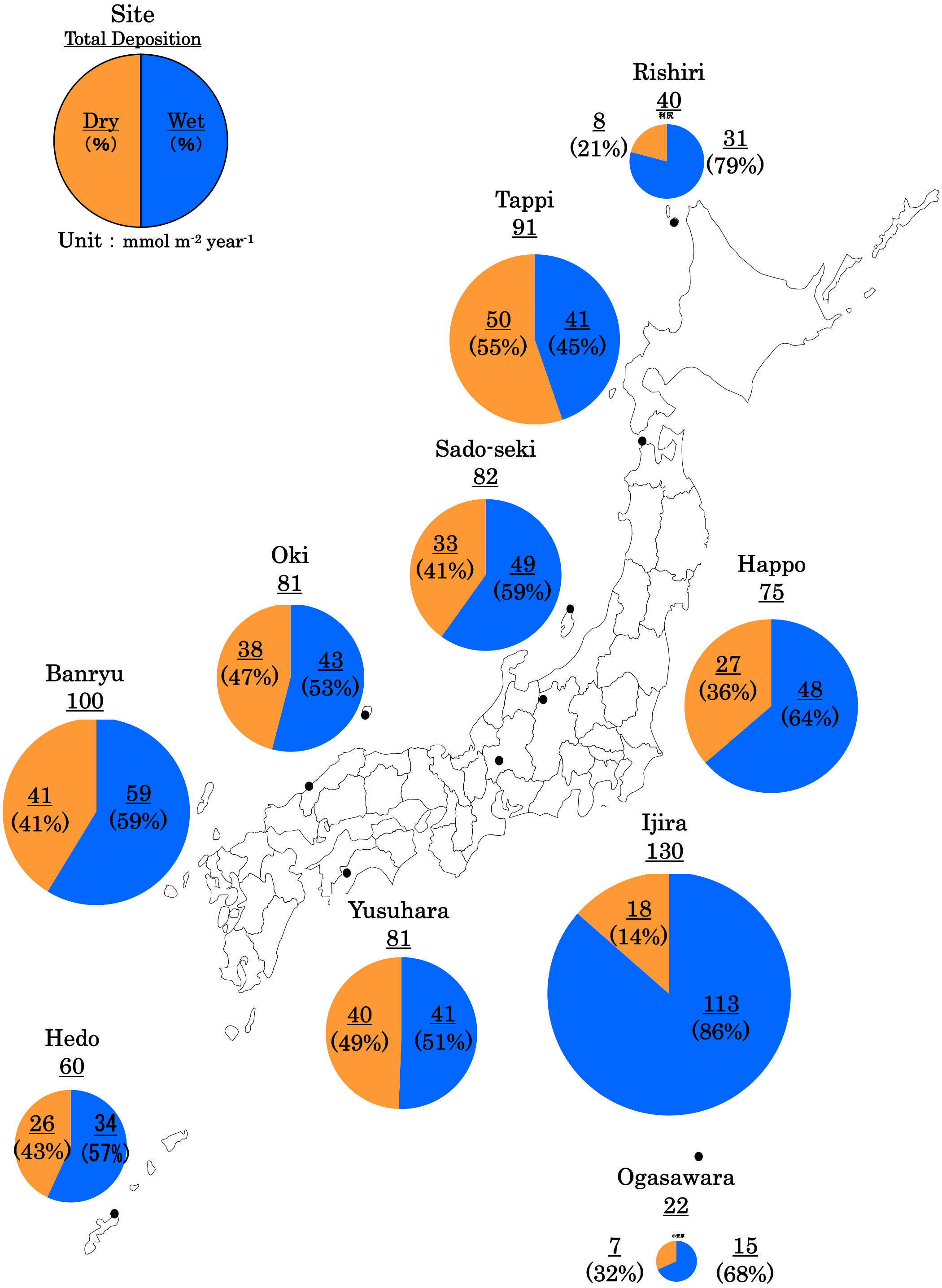


Figure 3-1-15　Average of Annual Total Nitrogen deposition (2003-2007)）

(2) Observation and its cause of low pH precipitation

① The appearance range of pH value in 2003-2007

Since a pH value in the precipitation is equalized so that the sampling period becomes long, in order to grasp the low pH precipitation, the pH value of a sample with a short sampling period could be applied. The histogram is shown in Figure 3-1-16 about the distribution of precipitation pH value monitored daily with 9,022 samples at 14 sites (Rishiri, Ochiishi, Tappi, Ogasawara, Sado-seki, Happo, Echizen-misaki, Shiono-misaki, Oki, Yusuhara, Tsushima, Goto, Ebino, Hedo) from 2003 to 2007

pH of most frequency was 4.8 and these arithmetic averages were pH 4.76. The appearance ranges of pH were 3.35-8.18, and range width was 4.83. If this is converted into the concentration of the hydrogen ion leading to acid, it differs also 68,000 times.

The number of precipitation sample which had less than pH four was 409, and these rate were 4.5% of the whole.

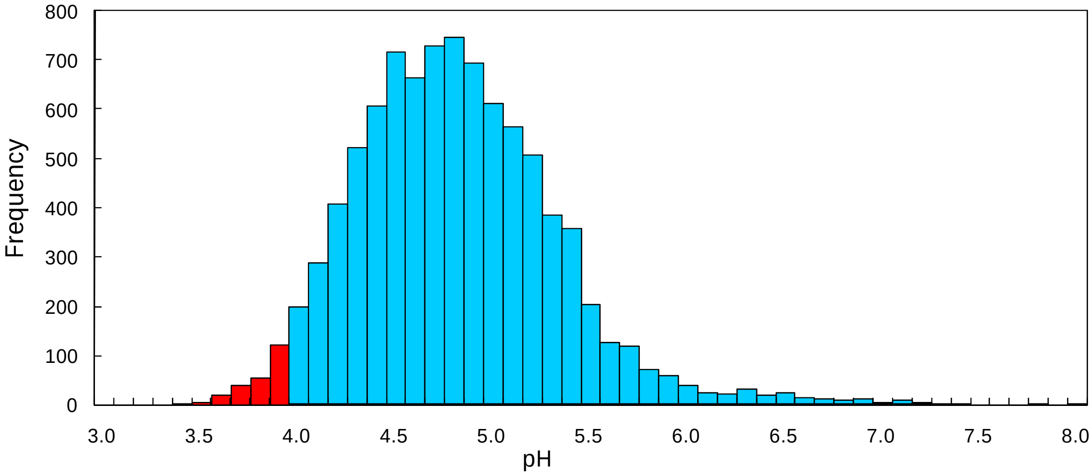


Figure 3-1-16 pH distribution of daily precipitation (2003-2007)

② The site and the season which less than pH four rain was observed

We considered about the site and the season which had 409 precipitations with less than pH four. 240 (59%) samples among 409 samples were monitored at the following 4 sites among 14 sites; Echizen-misaki, Shiono-misaki, Yusuhara, and Ebino. Moreover, the precipitation had less than pH four was observed 3 times even in Ogasawara which was a remote site. About the rates of observation less than pH four precipitation, there are most rates of winter (December to February) at 40%, and following, spring: 24%, summer: 19%, autumn: 17%. In Japan, precipitation had less than pH four had been always observed.

Table 3-1-4 The frequency of less than pH four precipitation
in annual and every season

Site	All season			Spring			Summer			Autumn			Winter		
	n	N	n/N(%)	n	N	n/N(%)	n	N	n/N(%)	n	N	n/N(%)	n	N	n/N(%)
Rishiri	8	670	1.2	2	149	1.3	5	123	4.1	0	178	0.0	1	220	0.5
Ochiishi	10	472	2.1	4	133	3.0	3	108	2.8	2	148	1.4	1	83	1.2
Tappi	22	494	4.5	5	109	4.6	9	132	6.8	3	143	2.1	5	110	4.5
Ogasawara	3	624	0.5	1	169	0.6	0	125	0.0	0	183	0.0	2	147	1.4
Sado-seki	16	562	2.8	7	116	6.0	3	162	1.9	4	143	2.8	2	141	1.4
Happo	15	879	1.7	6	209	2.9	2	237	0.8	3	226	1.3	4	207	1.9
Echizen-misaki	71	872	**8.1**	15	200	7.5	9	177	5.1	9	182	4.9	38	313	12.1
Shiono-misaki	53	679	**7.8**	10	188	5.3	14	196	7.1	15	172	8.7	14	123	11.4
Oki	29	669	4.3	8	156	5.1	2	152	1.3	3	148	2.0	16	213	7.5
Yusuhara	67	741	**9.0**	15	191	7.9	10	218	4.6	12	160	7.5	30	172	17.4
Tsushima	25	450	5.6	3	115	2.6	8	166	4.8	1	103	1.0	13	66	19.7
Goto	28	511	5.5	8	134	6.0	4	152	2.6	3	102	2.9	13	123	10.6
Ebino	49	725	**6.8**	12	203	5.9	8	211	3.8	11	145	7.6	18	166	10.8
Hedo	13	674	1.9	2	183	1.1	0	150	0.0	4	171	2.3	7	170	4.1
Total	409	9022	4.5	98	2255	4.3	77	2309	3.3	70	2204	3.2	164	2254	7.3

(3) Analysis of trans-boundary pollution using the monitoring data of O_3, SO_2, and so on

① Relationship between the longitude and the particulate SO_4^{2-} concentration

A range of the annual mean concentration of particulate SO_4^{2-} from J.F.Y. 2003 to 2007 is summarized as follows; J.F.Y. 2003: 1.60 µg/m^3 - 4.78 µg/m^3, average 3.32 µg/m^3, (10 sites), J.F.Y. 2004: 0.71 µg/m^3 – 5.17 µg/m^3, average 3.38 µg/m^3, (10 sites), J.F.Y. 2005: 1.36 µg/m^3 – 6.98 µg/m^3, average 4.73 µg/m^3, (10 sites), J.F.Y. 2006: 2.38 µg/m^3 – 6.60 µg/m^3, average 4.46 µg/m^3, (10 sites), J.F.Y. 2007: 2.37 µg/m^3 – 6.32 µg/m^3, average 4.46 µg/m^3, (11 sites). The sites are mainly located in remote area. This concentration level is almost the same as that observed in urban/rural areas in Japan (2.26 µg/m^3 - 6.54 µg/m^3, average 4.45 µg/m^3) (32 sites)（3-year mean from J.F.Y. 2003 – 2005） (Aikawa et al., 2008), indicating that the particulate SO_4^{2-} concentration over Japan is affected by not only the domestic contribution in Japan but also the contribution from a wider area.

Figure 3-1-19 shows the relationship between the longitude of the site and the particulate SO_4^{2-} concentration. A 5-year mean, the highest and the lowest annual mean concentrations are plotted in the vertical axis. Figure 3-1-19 indicates that the particulate SO_4^{2-} concentration is higher at the site located near the Asian Continent. Aikawa et al. (2008) demonstrates that the similar longitudinal gradient in the particulate SO_4^{2-} concentration measured at the above 32 sites located in urban/rural areas (Fig. 3-1-20). Further, as shown in Figure 3-1-19, the particulate SO_4^{2-} concentration level in Japan is lower than that in the Republic of Korea.

Conclusively, the atmospheric physics/chemistry concerning the particulate SO_4^{2-} formation, transport, diffusion, and deposition in a wider area as well as the domestic contribution in Japan are profoundly involved in the determination of the particulate SO_4^{2-} concentration over Japan.

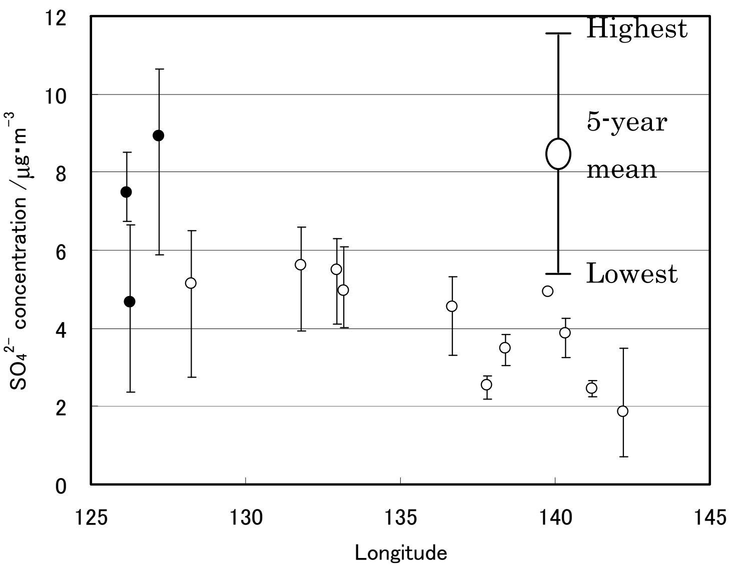


Figure 3-1-19 Relationship between the longitude of the site and the particulate SO_4^{2-} concentration. A 5-year mean value, the highest and the lowest annual mean concentrations are plotted in the vertical axis.

Solid circle shows the site in the Republic of Korea.

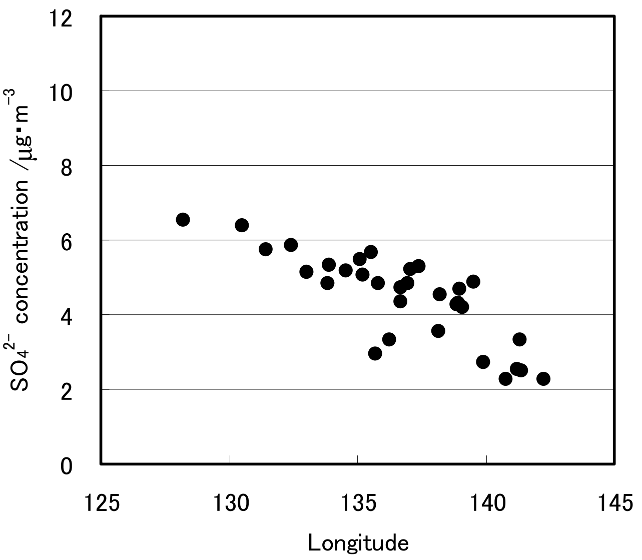


Figure 3-1-20 Relationship between the longitude of the site and the particulate SO_4^{2-} concentration measured at the 32 sites located in urban/rural areas in Japan. 3-year (J.F.Y. 2003 –

2005) mean concentration is plotted in the vertical axis.

② Behavior of nss-SO_4^{2-} in wet deposition and particulate matters in Hedo, Okinawa.

Hedo was located as a background site on the East China Sea, and was suitable site to grasp the effect of trans-boundary air pollution. Trans-boundary air pollution was analyzed from the observation result at the National Hedo acid deposition monitoring station and Hedo atmosphere and the aerosol observation station of National Institute of Environmental Studies.

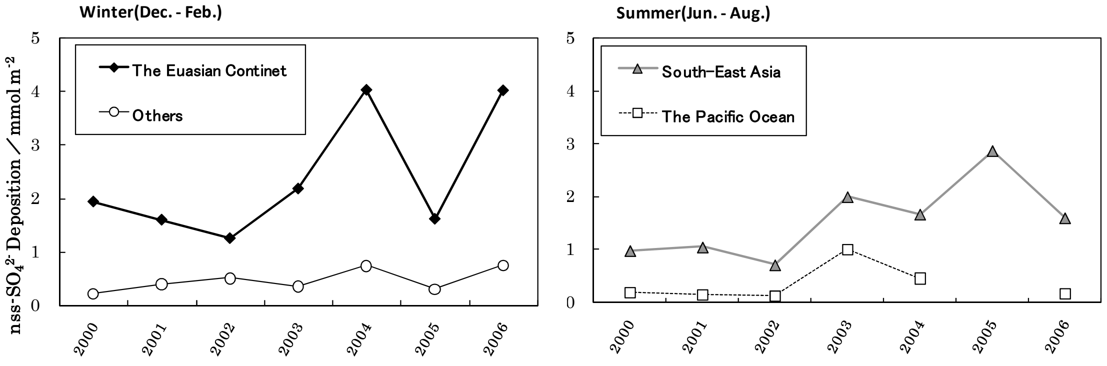
About the daily sample collected using wet-only sampler (more than 5mm day^{-1}) in winter (December to February) and in summer (June to August),

From the analysis result of the back trajectory method using an analysis program (NIES-CGER METEX), the nss-SO_4^{2-} deposition vie each transportation routes which were classified into continent side and others in winter, and Southeast Asia side and the Pacific Ocean side, were calculated (Figure 3-1-21). In summer, using data totaled except for the sample influenced by the typhoon.

In winter, the nss-SO_4^{2-}deposition from the continent side were increasing around from 2003 like the northern Japan mentioned below.

On the other hand, in summer, the nss-SO_4^{2-}deposition from the Southeast Asia side was increasing, and although nss-SO_4^{2-}concentration was low, since there was much precipitation, there was comparatively much deposition and it also had the year which is equal to the dposition in winter.

From these things, Okinawa area located in low latitude as compared with the Honshu island and near the Southeast Asia area, was suggested to be received the effect of the trans-boundary air pollution not only from China but also from Southeast Asia.



Moreover, at Hedo atmosphere and an aerosol observation station, concentration is measured for every hour in China etc. in recent years paying attention to SO2 which the amount of discharge is increasing, and nss-SO_4^{2-} in aerosol.

Hourly trends of SO_2, O_3, and nss-SO_4^{2-} from 12 to 23 March, 2005 were shown in Figure 3-1-22.

The concentration of SO_2 and O_3 monitored by the automatic monitor at Hedo site were increasing from the night on 17 to 18 March.

In other periods, although SO_2 concentrations were very low with 1 or less ppbv, these went up about 5 ppb from 17 to 18 March.

Moreover, O3 concentration went up to near the 80ppbv on March 18, although these were about 40 ppbv the previous day .

Furthermore, this was interlocked with, and nss-SO_4^{2-}(p) concentration rose rapidly and exceeded 20μg m^{-3}.

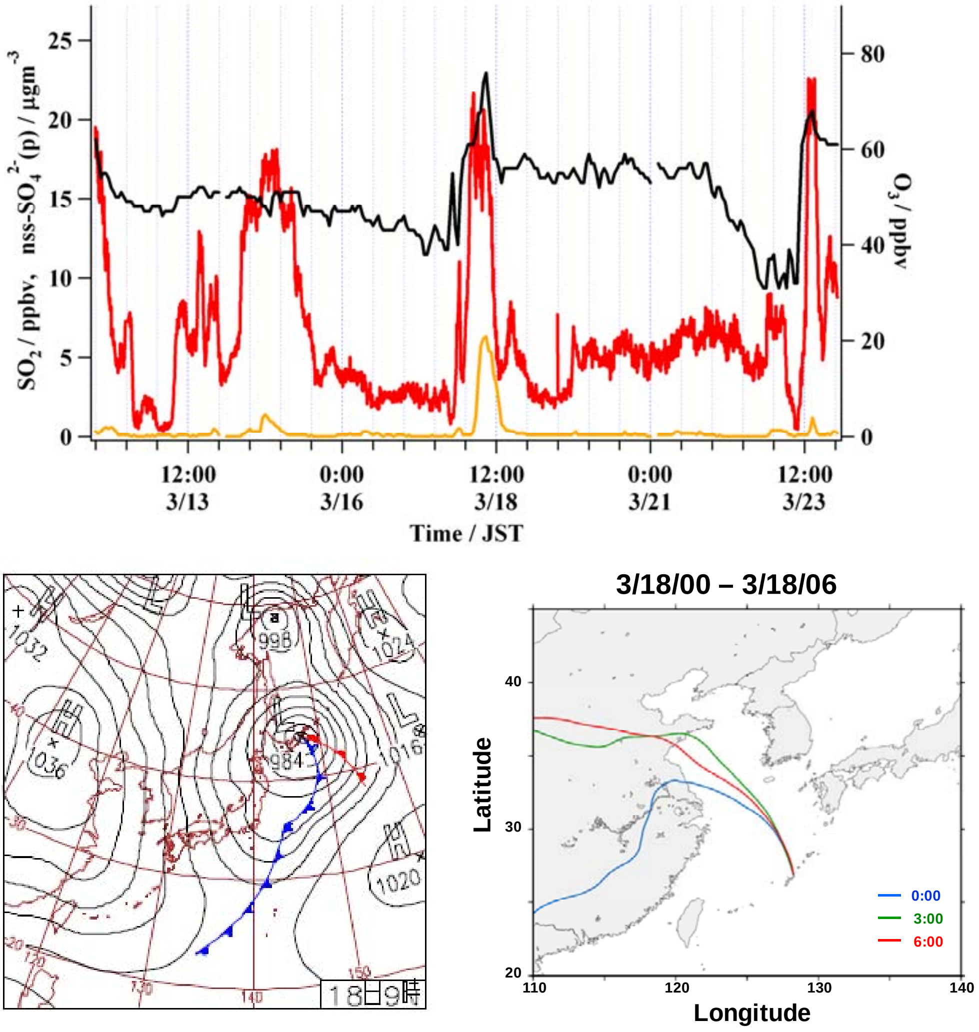
In a weather chart, in the stage at 9:00 a.m. on March 18, a front passes through near Okinawa, it is moving to the Pacific Ocean, and the high pressure has moved to the south of Beijing after it.

At this time, the isobar is running to north and south, and it has become a situation in which an air mass tends to carry out an advection current from the direction of a continent.

Furthermore, when the remains line of a back style is calculated using NOAAHYSPLIT4, it turns out that the advection current of the air mass has been carried out from the East Coast coastal area in China from Shanghai to the Santo peninsula in the Okinawa district after front passage.

It is 5 conjectured that SO_2, O_3, and the concentration rise of nss-SO_4^{2-}(p) which were observed from these things on March 18 were brought about with the air mass of the China origin.

in addition to this, the same phenomenon was seen several times, and when an air mass reached from the area along the shore from Shanghai to the Santo peninsula in China, nss-SO_4^{2-}(p) concentration was high.

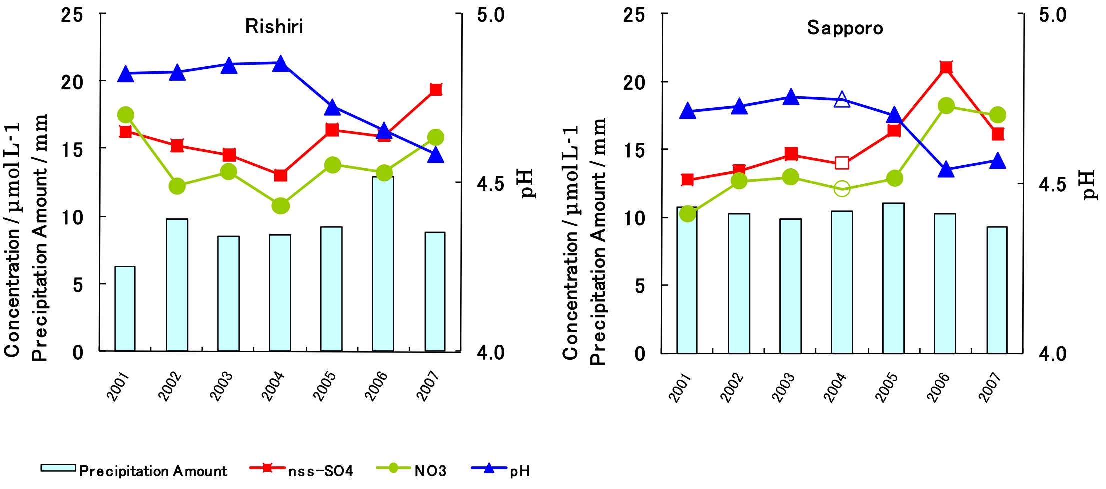


Figire 3-1-22 Upper：Trends of SO2, O3, and nss-SO42- at Hedo acid deposition monitoring site and atmosphere and aerosol monitoring station
Lower-berth left: the weather chart at 9:00 a.m. on 18 March
Lower-berth right: result of back trajectory analysis (calculation result on the basis of 0:00, 3:00, and 6:00 a.m. on 18 March.)

③ Behavior of wet deposition of components in northern Japan

In northern part of Japan, for example Hokkaido, since concentration of precipitation components is lower than the other regions and the precipitation amount is low, wet deposition amount is not so large. It was reported that contribution of emission sources in China toward the deposition amount in these regions is larger than the other regions and that the influence is more remarkable especially in winter season. To examine the effect of long-range trans-boundary air pollution, we analyzed behavior of wet deposition of components in northern Japan.

Major objects of the analysis are the dates of Rishiri and Sapporo station located on the side of Japan Sea which are subject to the effect of long-range trans-boundary air pollution. We analyzed them from year 13 downward, avoiding the influence of eruption of Miyake volcano in year 12. Figure 3-1-23 presents the annual change of precipitation amount, nss-SO_4^{2-}, NO_3^- and pH. In both stations, pH value showed a slight rising trend from year 13 to 15, but while pH value declined significantly after year 17 downward, concentrations of nss-SO_4^{2-} and NO_3^- showed a increasing trend and deposition amounts of nss-SO_4^{2-}, NO_3^- and H^+ also increased: this is similar to the annual change of deposition amount of nss-SO_4^{2-} in Hedomisaki in winter season. As regards to these trends of wet deposition in recent years, precipitation samples with low pH value (4.2 or less) which were able to be identified where they come form by the backward trajectory suggested a possibility of the effect of long-range trans-boundary air pollution.



④ State of long-range trans-boundary air pollution of yellow sand and O_3.

Since yellow sand was observed at more than 50 meteorological observatories from Tohoku to Okinawa in 26 and 27 May year 19 and a photochemical oxidant advisory was issued in 7 prefectures of the western Japan in 27 May year 19, we analyzed the state of the air pollution in this period. The objects for the analysis were Tappi, Sado-seki, Ijira, Oki, Yusuhara and Ogasawara.

Figure 3-1-24 shows changes of concentrations of PM_{10}, O_3 and SO_2 from 25 to 30 May.

The concentrations of PM_{10}, O_3 and SO_2 at Oki, Yusuhara, Ijira and Sadosekimisaki changed almost synchronously from 26 to 28 May, and the concentrations of PM_{10} and SO_2 at Yusuhara and Ijirako showed synchronic change with double peaks in the period. The maximum concentrations of PM_{10} in this period were about 300µg m^{-3} in the 4 regions and the maximum concentrations of O_3 showed more than 100ppb in the same period at Oki and Yusuhara as remote site. The maximum concentration peak appeared earlier in western region in this order Oki, Yusuhara, Ijira and Sado-seki, considering the influence of prevailing west wind in 26 to 27 May. The concentrations of PM_{10} and O_3 rose synchronously and the maximum concentration of PM_{10} was about 70µg m^{-3} at Tappi in 26 May. The concentrations of PM_{10} and SO_2 changed almost synchronously and the maximum concentration of PM_{10} was over 200µg m^{-3} at Ogasawara from 28 to 30 May. The double peaks of PM_{10} and SO_2 observed at Yusuhara and Ijira was appeared after a delay of about 2 days at Ogasawara. The concentration of O_3 rose to 50 ppb and kept same level while SO_2 began to be detected and PM_{10} showed high concentration. Yellow sand was observed by Light Detection And Ranging (LIDAR) at Matsue, Niigata and Sendai during the high concentration of PM_{10}, suggesting the increase of PM_{10} resulted from yellow sand. We could understand to catch the huge yellow sands covering from Tohoku to western Japan and transported to Ogasawara keeping high concentration by this monitoring. Increase of the concentration of O_3 and SO_2 linked with the observation of yellow sand and prevailing west wind suggested the influence of long-range trans-boundary air pollution.

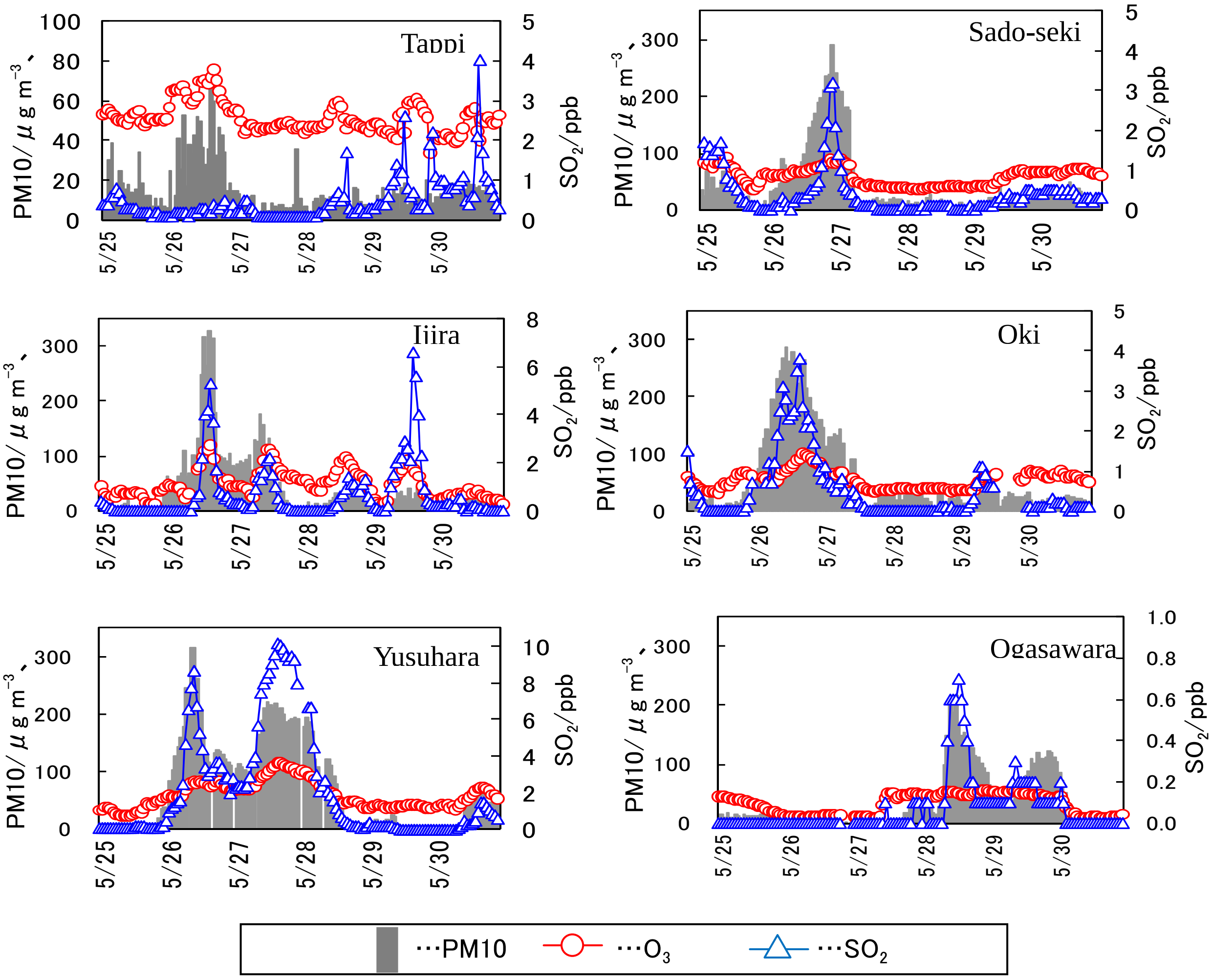


Figure 3-1-24　Trends of PM_{10}、 O_3, and SO_2 concentrations through 25 to 29 May, 2007

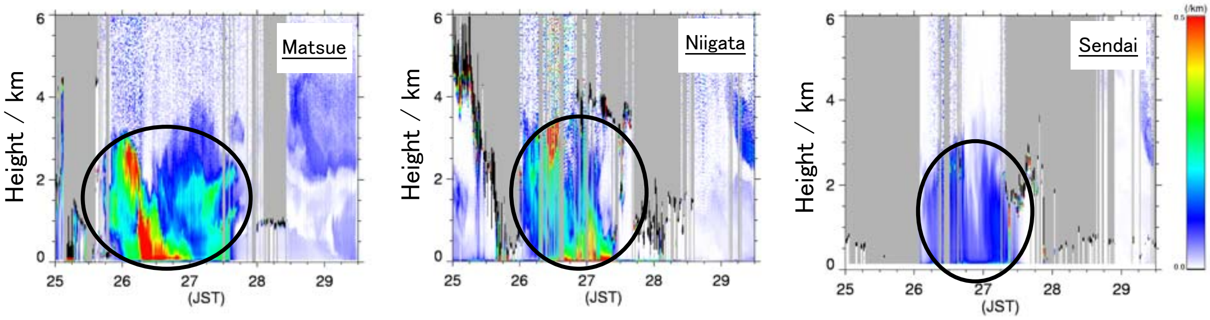


Figure 3-1-25　Observation results using LIDAR system at Matsue, Niigata, and Sendai through 25 to 29 May, 2007

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3.2 Results of Ecological Impact Monitoring

3.2.1 Results of Soil and Vegetation Monitoring

(1) Results of soil and vegetation monitoring from 2003 to 2007

The objectives of this survey were to obtain baseline data on soil and vegetation and to make early detection of acid deposition impacts on forest ecosystems. Twenty-five sites (50 soil plots) of 19 areas were established in conservation areas such as national parks (NPs) or natural heritages focusing on impacts on trees or in the areas with red-yellow soils focusing on impacts on soil. Soil surveys and forest vegetation surveys were carried out there.

1) Acid buffering capacity of soil and effective factors to the variability

Forest soils in Japan are generally acidic. Chemical properties of soil depend on its formation process and effective factors to the formation. Acidic soil can be produced even under natural conditions without anthropogenic effects. Therefore, chronological changes of chemical properties and their degrees will be evaluation criteria for assessment of impacts on soil. In this section, acid buffering capacity of soil was evaluated taking soil types in the respective sites into consideration. The pH (H_2O) and exchangeable (Ca+Mg+K)/Al (equivalent ratio) of the surface soil of each site were shown in Fig. 3-2-11.

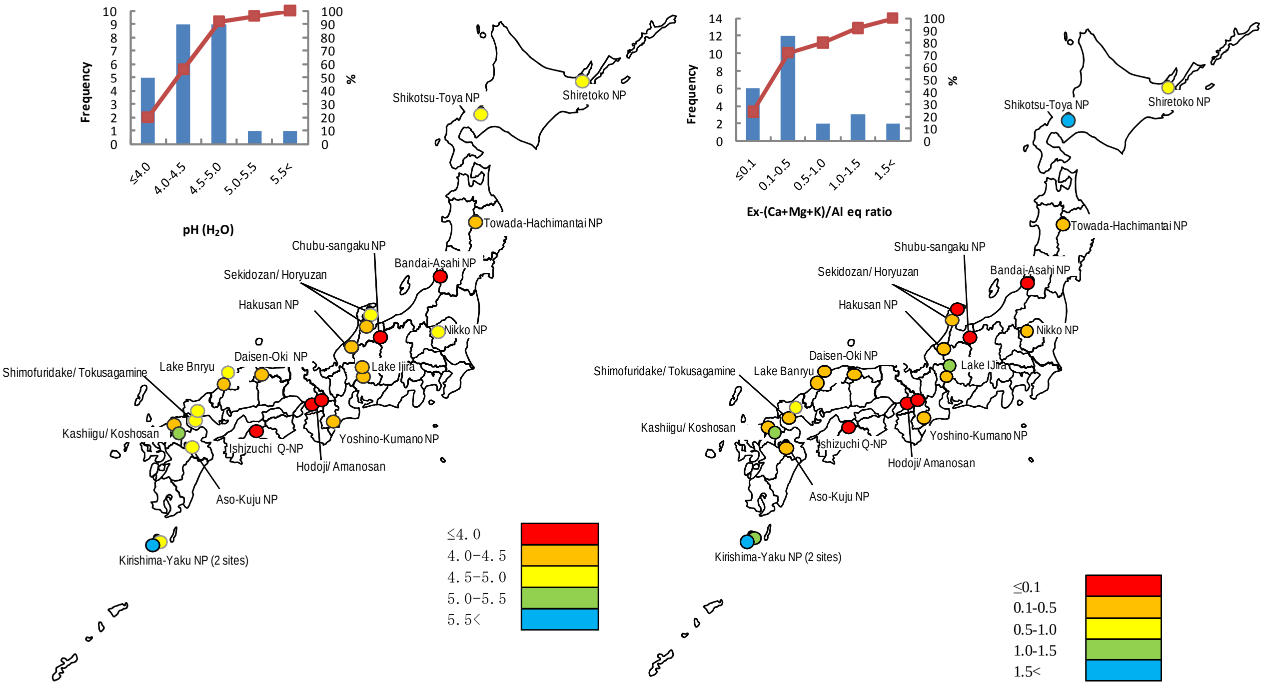


Fig. 3-2-1-1 The pH (H_2O) (left) and exchangeable (Ca+Mg+K)/Al (equivalent ratio) (right) of surface soil in each site

The soil pH (H_2O) was lower than or equal to 5.0 in all the sites except for a laurel forest of Yakushima Island or Mt. Koshosan. The pH (H_2O) was lower than or equal to 4.0 especially in Bandai-Asahi National Park (brown forest soil), Chubu-Sangaku NP (podzolic soil), Ishizuchi Quasi-NP (brown forest soil), Houdouji Temple (red-yellow soil), and Mt. Amanosan (brown forest soil). The pH (KCl) was lower than 4.0 in all the sites except for the laurel forest of Yakushima Island (4.8), Mt. Koshosan (4.5), Nikko NP (4.1), and Yoshino-Kumano NP (4.1). Especially in Ishizuchi Quasi-NP, Houdouji Temple, and Bandai-Asahi NP, significantly lower pH was recorded: pH (H_2O) was 3.8, 3.8, and 3.9, and pH (KCl) was 3.1, 3.2, and 3.4, respectively.

The equivalent ratio of exchangeable (Ca+Mg+K) to exchangeable Al (Ex-BC/Al ratio), which can be considered as an indicator of acid buffering capacity, showed a similar tendency to pHs: the values in the sites with significantly lower pH (< 4.0), such as Mt. Amanosan, Ishizuchi Quasi-NP, Bandai-Asahi NP, and Houdouji Temple, were 0.06, 0.07, 0.09, and 0.09, respectively. Moreover, the Ex-BC/Al ratio showed a relatively high positive correlation with pH (H_2O) and pH (KCl) (r: 0.611 and 0.574, respectively).

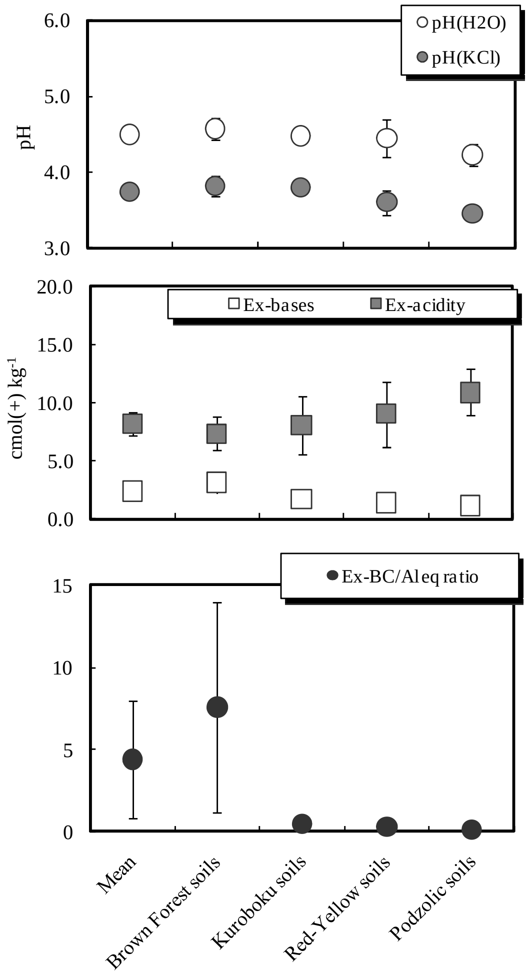


Fig. 3-2-1-2 Chemical properties of each soil Great Groups

Based on an acidification experiment of various soil types, the Synthesis Report on Japan Acid Deposition Surveys (2004) pointed out that the Ex-BC/Al ratio can be changed by a pH 4.0 acid precipitation in case of the soil whose Ex-BC/Al ratio was lower than 0.1. Therefore, in the sites with lower pH and Ex-BC/Al ratio, changes in soil chemical properties should be monitored continuously.

According to the Unified Soil Classification System of Japan-2nd Approximation- (Japanese Society of Pedology, 2002), soils in the survey sites can be classified into four Great Groups, namely, Brown Forest soils (14 sites), Kuroboku soils (4 sites), Red-Yellow soils (4 sites), and Podozolic soils (3 sites). The mean values and standard errors of representative parameters among each soil Great Group were shown in Fig. 3-2-1-2. In pH (H_2O), Podozolic soils <

Red-Yellow soils ≈ Kuroboku soils ≈ Brown Forest soils, and in pH (KCl), Podozolic soils < Red-Yellow soils < Kuroboku soils ≈ Brown Forest soils. Exchangeable acidity was from 3 to 10 times larger than sum of exchangeable base cations (Ca, Mg, K, and Na) in all the soil Great Groups, reflecting low pH of the soils. Especially in Podozolic soils and Red-Yellow soils, the difference between the sum of base cations and the acidity was significantly larger, suggesting less base cations and more acidity.

In Ex-BC/Al ratio, Podozolic soils (0.1) < Red-Yellow soils (0.3) ≈ Kuroboku soils (0.5) << Brown Forest soils (7.6). The larger mean value of Brown Forest soils can be attributed to a base-rich soil in the laurel forest of Yakushima Island. When this significantly larger value is removed, the mean value will be 1.2. Variation among Brown Forest soils was quite large. As mentioned above, the survey sites showing relatively high acidity, represented by Ishizuchi Quasi-NP, Bandai-Asahi NP, Mt. Amanosan, could also be found.

Soil monitoring was carried out taking effective factors to variability of the obtained data into account. As the effective factors in the field sampling, several sampling levels, such as “area”, “soil type (site)”, “plot”, and “subplot”, were considered. Cumulative contributions of these sampling levels to the data variability were shown in Fig. 3-2-1-3.

Area	18
Soil type	1 or 2
Plot	2
Subplot	5
Repeat analysis	2 times

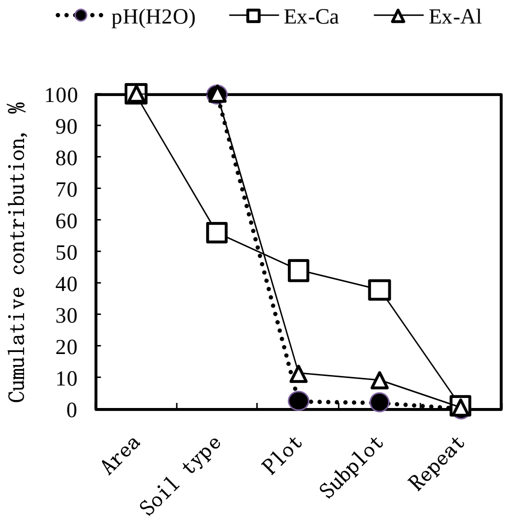


Fig. 3-2-1-3 Soil sampling system (left) and cumulative contributions to variance in chemical properties of surface soils (right). The data in Mt. Koshosan, which showed significantly larger variations among plots and subplots, were not included in the analysis.

In soil pH and Ex-Al, indicators of acidity, “soil types” had much effect on the total variability, reflecting difference of soil acidity in the respective soil types. In Ex-Ca, “subplots” and “area” had much effect on the total variability; the former one might reflect spatial heterogeneity of soil, such as thickness of organic layer, and the latter one might reflect geology or climate than soil type.

2) Evaluation of soil acidification trends

Most survey sites focusing on soil acidification were established in 2001 and Red-Yellow soils or similar types of soil were selected with their reference types of soil in each area. According to the Long-Term Monitoring Program, the second surveys were carried out for all the sites until 2007, while the sampling intervals were varied from 2 to 6 years. The mean values among the respective plots (mean of five subplots) in these sites were shown in Table 3-2-1-1.

Table 3-2-1-1 Changes of chemical properties of surface soils (0-10 cm) in Red-Yellow soils

Area	Site	Soil type	Plot	Survey year	pH		ECEC	BS	Ex-(Ca+Mg+K)/Al eq ratio
					H_2O	KCl	cmol(+) kg^{-1}	%	
Ishikawa Pref.	Sekidozan	Brown forest soils	1	2001	4.4	3.7	15.29	8.02	0.08
				2005	4.3	3.5**	14.82	10.83	0.12
			2	2001	4.5	3.8	13.16	11.19	0.13
				2005	4.4	3.5**	13.50	12.38	0.15
	Horyuzan	Red soils	1	2001	4.6	3.8	17.17	9.87	0.10
				2005	4.6	3.6	17.20	8.50	0.09
			2	2001	4.6	3.8	19.67	12.38	0.13
				2005	4.6	3.6**	19.18	10.51	0.11
Osaka Pref.	Amanosan	Reddish brown forest soils	1	2001	4.2	3.4	9.16	9.07	0.11
				2007	4.1	3.4	9.03	5.22*	0.06*
			2	2001	3.8	3.1	11.10	6.55	0.08
				2007	4.0	3.3	10.47	4.72	0.05
	Hodoji	Yellow soils	1	2001	3.9	3.2	11.29	5.24	0.06
				2007	3.8	3.1	10.51	9.12*	0.10*
			2	2001	4.0	3.3	8.31	7.40	0.09
				2007	3.9	3.2	9.13	8.14	0.09
Shimane Pref.	Lake Banryu	Brown forest soils	1	2001	4.8	3.9	4.30	17.07	0.21
				2006	4.9	3.8	5.13	30.39*	0.53
			2	2001	4.7	3.7	4.86	27.10	0.44
				2006	4.8	3.9*	4.83	21.97	0.33
	Iwami-rinku FP	Red soils	1	2001	4.3	3.5	8.56	16.51	0.22
				2006	4.5	3.7	7.58	15.65	0.21
			2	2001	4.2	3.3	10.61	10.72	0.12
				2006	4.4	3.6	10.53	12.60	0.16
Yamaguchi Pref.	Tokusagamine	Kuroboku soils	1	2001	4.6	3.8	11.87	10.08	0.12
				2003	4.5	3.9*	11.43	13.03	0.20
			2	2001	4.6	3.8	12.22	13.09	0.20
				2003	4.6	3.9	11.39	10.01	0.13
	Simofuridake	Yellow soils	1	2001	4.9	3.9	5.18	31.14	0.77
				2003	4.9	3.9	4.87	36.01	0.84
			2	2001	4.8	3.9	4.45	25.95	0.47
				2003	5.0	4.0	4.05	36.11	0.97
Fukuoka Pref.	Kashiigu	Reddish brown forest soils	1	2001	4.2	3.4	19.00	13.04	0.17
				2007	4.1	3.3	18.71	8.57	0.09
			2	2001	4.5	3.5	27.90	25.03	0.42
				2007	4.4	3.4	23.62	15.13	0.18
	Koshosan	Brown forest soils	1	2001	4.8	3.9	14.23	51.29	>1.0
				2007	4.5	3.6	8.35	40.22	0.84
			2	2001	6.5	5.6	30.54	90.49	>1.5
				2007	6.1	5.4	25.50	78.40	>1.5

Note. Superscripts, * and **, indicate significant difference at 5% level and 1% level, respectively, by the t-test for the mean values of subplots.

Most plots showed no clear acidification trend but two sites in Ishikawa Pref. showed significant decrease of pH (KCl). Soil acidity in these sites may increase, since pH (KCl) indicates potential acidity of soil. In Plot 1 of Mt. Amanosan (Osaka Pref.), pH (H_2O) and pH (KCl) showed mostly same levels but base saturation and Ex-BC/Al ratio showed significant decrease. In these sites, Ex-BC/Al ratio was low, approx. 0.1, and therefore, the soils may be sensitive to acid deposition, as mentioned above. The continuous surveys are necessary in these sites. On the other hand, the opposite tendency, increase of parameters, was also found; base saturation and pH (KCl) at Lake Banryu (Shimane Pref.), pH (KCl) at Mt. Tokusagamine (Yamaguchi Pref.), and base saturation and Ex-BC/Al ratio at Houdouji Temple (Osaka Pref.). Especially around Lake Banryu, changes in conditions of the catchment area may be related to the data, since forest managements for pine wilting decease have been carried out for the last decade.

The changes in pH (H_2O) and Ex-Al of the surface soil horizon (3 - 5 cm depths) and the subsequent soil horizon (10 – 15 cm depths) were shown for the long-term monitoring plots in the Lake Ijira Catchment site. The pH (H_2O) decreased continuously in the surface horizon and subsequent horizon at all five plots for 14 year from 1990 to 2004, while some fluctuations could be found. The mean pH (H_2O) of the surface layer (collected by the fixed depth, 0 - 10 cm) among two plots for the EANET monitoring were 4.3 (3.9 - 4.4) in 2006, and it can be clarified that these plots were also acidified similarly to the long-term plots. The Ex-Al, an indicator of acidity, showed increasing trends in most plots especially for the surface horizon, and in a few plots for the subsequent horizon.

In the Lake Ijira catchment, as described in the section 3.3, a wet deposition amount of H^+ is the highest level in Japan, and the river water chemistry showed several acidification phenomena, such as, temporary SO_4^{2-} leaching, decrease of the water pH, and continuous increase of the NO_3^- concentration. Therefore, changes in the soil chemical properties can be considered as a part of the acidification process in the whole catchment area.

Since soil pH may decrease due to a rapid uptake of nutrient bases during plant growth periods, it may be necessary to evaluate the respective contributions of H^+ production in the ecosystem and H^+ input from the atmosphere to the soil acidification in detail. However, as shown in the section 3.3, the tree growth rates in the Lake Ijira catchment showed decreasing trends during the sampling period of soil. The rapid decrease of soil pH observed here cannot be explained only by plant uptake. In the catchment, since forest managements such as thinning had not been carried out, the management conditions should also be considered. However, taking the highest level of wet deposition amounts in Japan, the high acid loading seems to be a main cause of soil acidification. The soil pH (H_2O) decreased to the significantly lower level, 3.9 in average in the surface soil. In the buffering process of soil, buffering reactions by Al-(hydro)-oxide may occur mainly in the pH range, and Al^{3+} toxic to plant growth can be leached into the soil water. Therefore, it may be necessary to monitor effects of the soil acidification on plant physiology in future.

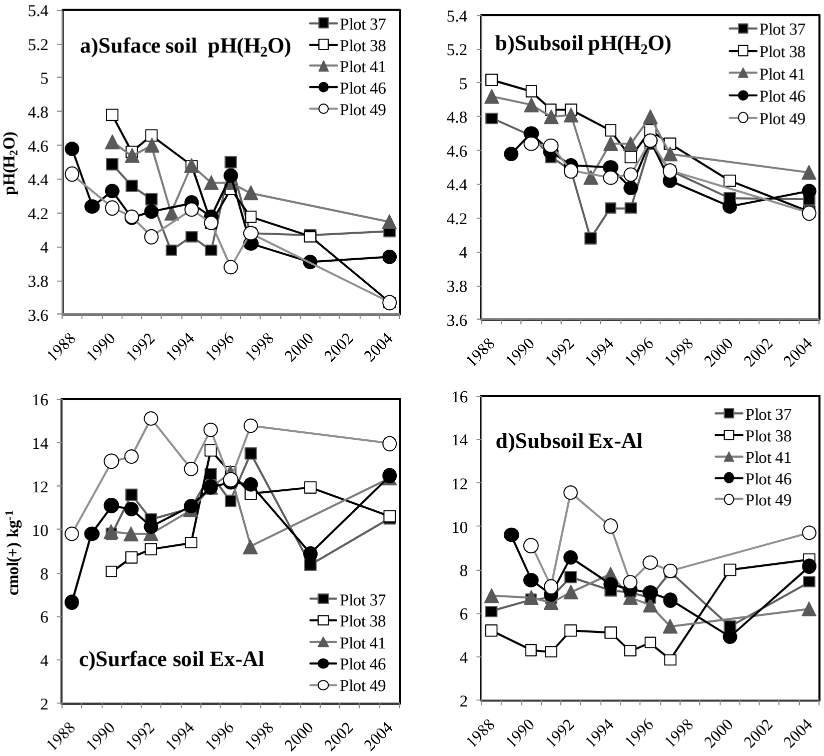


Fig. 3-2-1-4 Chronological changes in pH (H_2O) and exchangeable Al of soils at long-term monitoring plots in the Lake Ijira Catchment site

Surface layer, until 3 – 5 cm depths; subsequent layer, until 10 – 15 cm depths. The measurement of pH(H_2O) was done by the following soil:solution ratio: 1:5 until 1992, and 1:2.5 from 1993.

3) Assessment of tree decline conditions

Figure 3-2-1-5 showed the mean values of “Dieback of stem” and “Density of foliage”, which were observed as indicess of tree decline symptoms. The indices were recorded according to 5- or 4-class decline scales, from 0 (healthy) to 5 (dead) in case of “Dieback of stem” or 4 (dead) in case of “Density of foliage”. The larger values showed clearer decline symptoms.

Some decline symptoms were observed in 17 sites among 25 sites, while small changes of tree forms in Mt. Sekidozan and Mt. Amanosan were not included in the figure. The cumulative values of two parameters were the largest in Chubu-Sangaku NP and then in Yoshino-Kumano NP, where more than half of trees showed some symptoms. In Yoshino-Kumano NP, typhoons may be a main cause of the symptoms, which were observed in *Fagus crenata* and *Abies homolepis*. In Chubu-sangaku

NP, the cause has not been identified but small precipitation amounts during the growing period in 2007 were pointed out. In other sites also, the declining trees, which were mainly caused by acid deposition or soil acidification, were not identified.

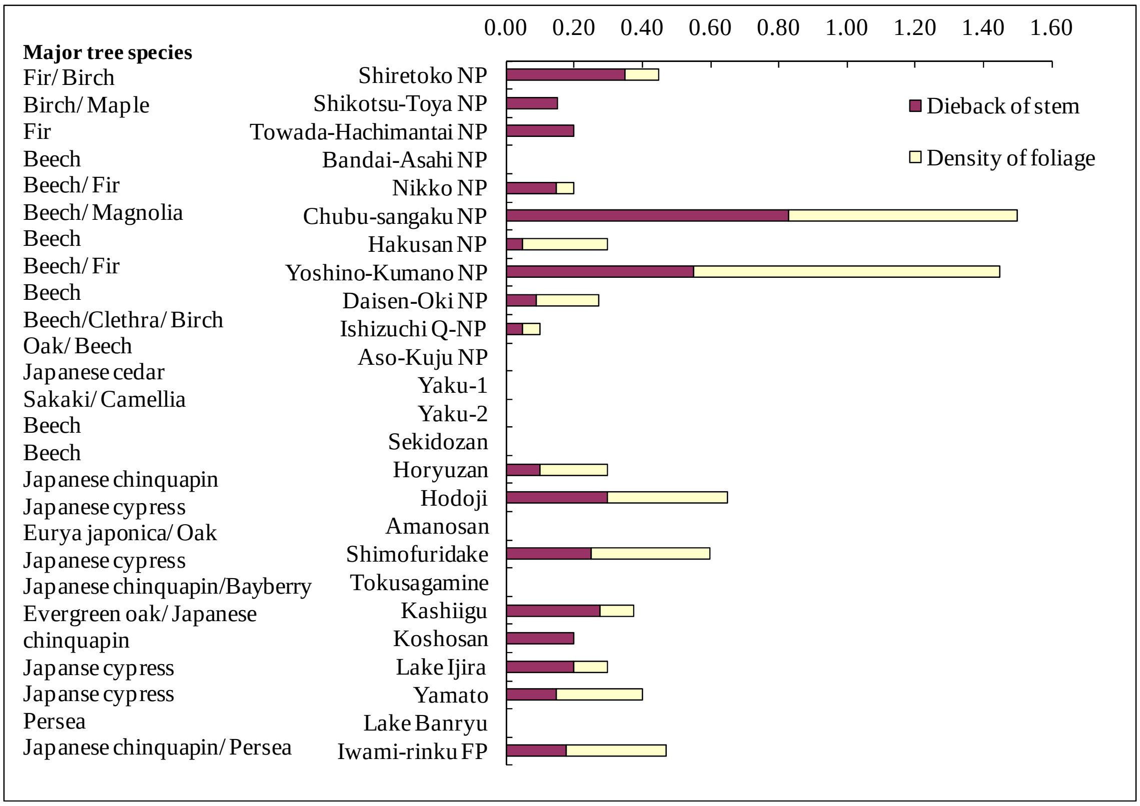


Figure 3-2-1-5 Major tree species and conditions on "Dieback of stem" and "Density of foliage" in 2007.

The column graph shows cumulative values of the means of two parameters in the respective plots (the larger values shows clearer decline symptoms).

Chronological changes in "Vitality of trees" recorded as the total index of tree decline were shown in Fig. 3-2-1-6. The tree decline symptoms that were processed from 2004 to 2005 seemed to be recovered in the later years in Shikotsu-Toya NP, Nikko NP, Daisen-Oki NP, and Ishizuchi Quasi-NP. It was suggested that damages by typhoons in 2003 were recovered after a few years. It is difficult by the observation to clearly distinguish changes by acid deposition or air pollution from changes by meteorological events/conditions, such as typhoons, cold summer, and draught stress, especially in the previous surveys until 2002, because the observation was done only once 3 – 5 years. However, the observation has been done at least once a year since 2003 according to the Long-Term Monitoring Plan, and therefore, it may be possible to distinguish effects of meteorological events/conditions. In the Lake Ijira catchment, a thinning of the dense forest as a forest management was done just after the observation of 2006, and the recovery has already been observed in 2007.

On the other hand, it seemed that the tree decline symptoms were gradually processed in Shiretoko NP, Chubu-Sangaku NP, Yoshino-Kumano NP, Hakusan NP, and Houdouji Temple. In these sites, suppression by other surrounding trees or damages by strong winds were suggested as possible main causes. However, the soil pH (H_2O) was significantly low in Houdouji Temple as described above. It can be suggested that continuous observations should be done carefully in these sites.

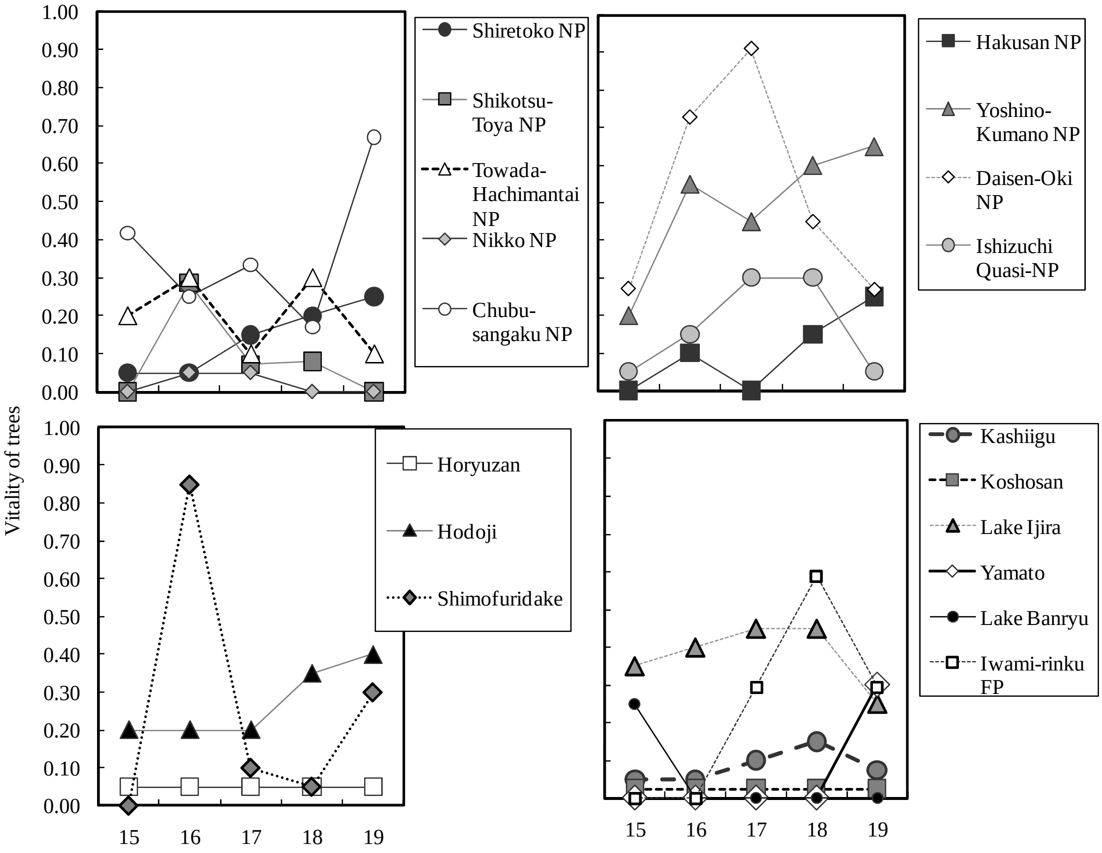


Fig. 3-2-1-6 Chronological changes in "Vitality of trees" (the larger value shows severer decline.)

As a part of forest vegetation monitoring, description of trees (measurement of tree size or growth) and surveys on species composition in understory vegetation have also been done and the data has been accumulated. At the present, the first surveys have just finished in the last five years, and it is be expected that growth rates of trees in the respective sites will be evaluated in future.

(2) Quality assurance/quality control (QA/QC) of the data

The data quality is assured through the audit of the national center of Japan, ADORC, and in this occasion, design of the monitoring plots and sampling procedures in the sites and conditions of the laboratories are confirmed. As for analytical precisions of soil, variations in the repeatability condition, within-laboratory reproducibility condition, and reproducibility condition were significantly small and their precisions might be enough high according to the results of the inter-laboratory comparison projects on soil. The reliable data may be obtained in the surveys.

3.2.2. Results of Monitoring on Inland Aquatic Environment

(1) Results of monitoring on inland aquatic environment from 2003 to 2007

In this survey, 11 lakes and their inflow/outflow rivers with low EC and low alkalinity, which may be susceptible to acidification, were selected as the monitoring sites. The mean values of the representative parameters from 2003 to 2007 in the respective monitoring sites were shown in Table 3-2-2-1. The alkalinity in most sites was lower than 0.200 mmol$_c$ L^{-1}. Lake Futagoike (Oike), Lake Yashagaike, and Lake Sawanoike may be the most sensitive to acid deposition, showing the lowest level of alkalinity and pH; lower than 0.030 mmol$_c$ L^{-1} and lower than pH 5.8, respectively. In all the lakes including these sensitive ones, obvious effects of acid deposition were not identified. On the other hand, alkalinity and pH in Lake Nagatomiike were significantly higher than other lakes. Lake Sankyoike and Lake Banryu had large effects of sea salts and the water was dominated by Cl$^-$ and Na$^+$. The NO3- concentration was the highest in the surface water of Lake Ijira and inflow rivers among the monitoring sites. The features of the catchment areas were shown in Table 3-2-2-1 (b).

Table 3-2-2-1(a) Mean values of representative parameters in the monitoring lakes from 2003 to 2007

Location	Lake or River name	Point	pH	EC	Alkalinity	SO_4^{2-}	NO_3^-	Cl^-	NH_4^+	Na^+	K^+	Ca^{2+}	Mg^{2+}
Prefecture				mS m^{-1}	mmol$_c$ L^{-1}	μmol$_c$ L^{-1}							
Yamagata.	Imagamioike	Surface	6.37	3.90	0.071	80.7	*1	175.3	*1	226.1	11.1	26.0	43.3
Tochigi	Karikomiko	Surface	6.91	3.25	0.156	115.2	5.3	20.0	1.4	131.0	14.0	117.8	14.3
Nagano	Futafoike(Oike)	Surface	7.02	1.86	0.112	33.4	15.9	11.3	0.7	46.1	6.4	107.4	15.8
	Futagoike(Meike)	Surface	5.73	0.73	0.020	29.3	4.1	10.0	0.7	13.7	4.1	22.1	7.5
Niigata	Sankyoike	Surface	6.87	8.49	0.109	92.8	2.1	542.6	1.1	510.9	30.9	81.8	113.8
Ishikawa	Ohataike	Surface	6.53	4.29	0.134	33.5	8.6	186.5	2.5	197.1	25.8	65.0	70.1
Fukui	Yashagaike	Surface	5.36	1.64	0.017	42.7	7.5	61.3	4.9	59.7	8.5	20.3	18.2
Gifu	Ijira	Surface	7.03	4.24	0.163	108.8	31.0	64.7	3.5	91.9	7.9	153.3	110.7
	Kamgatani river	Inflow	6.96	4.43	0.140	136.2	39.6	63.2	*1	93.8	7.3	150.4	122.8
	Kobora river	Inflow	6.72	3.86	0.125	101.6	33.3	68.9	*1	102.5	6.5	100.5	116.5
Kyoto	Sawanoike	Surface	5.70	1.73	0.021	32.2	*1	83.4	*1	65.7	6.4	30.1	30.6
Shimane	Banryu	Surface	7.06	10.65	0.166	88.1	1.7	634.3	1.8	605.5	47.6	66.1	141.0
	Banryu No.3	Surface	7.05	10.60	0.173	89.9	2.1	624.5	1.4	604.4	47.9	72.2	142.5
Yamaguchi	Yamanokuchi dam	Surface	6.64	6.02	0.107	97.8	9.7	285.8	*1	332.4	25.1	65.6	72.2
Kagawa	Nagatomike	Surface	7.34	9.53	0.512	169.5	25.4	155.2	5.2	339.3	25.4	416.4	104.8

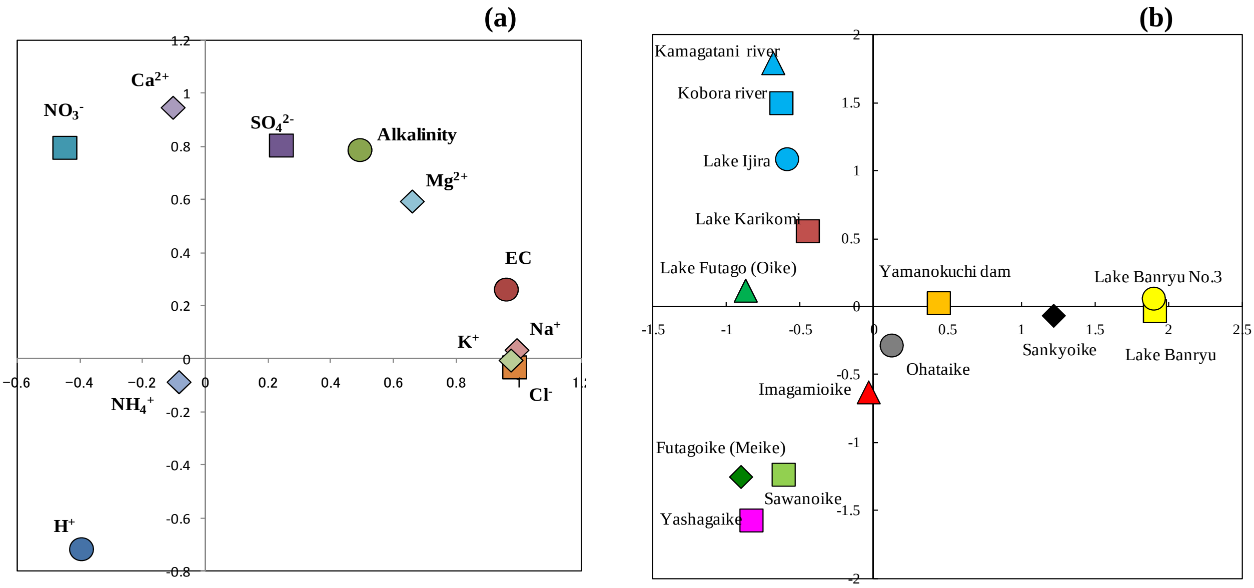
*1:Less than detection limit

Table 3-2-2-1(b) Features of the respective catchment areas

Location	Lake name	Surface stratum	Soil type	Vegetation
Yamagata	Imagamioike	Partially consolidated	Drained podsol	*Fagus crenata, Sasa kurilensis*
Tochigi	Karikomiko	Rhyolite, Andesite	Aquic and Drained podsol	*Abies mariesii, Abies veitchii*
Nagano	Futafoike	Lava	Aquic humic podsol	*Smilacina viridiflora, Betula ermanii*
Niigata	Sankyoike	Neogene period sediment	Brown forest soil	*Quercus crispula, Quercus serrata*
Ishikawa	Ohataike	Rhyolite, Andesite	Drained brown forest soil	*Castanea, Quercus crispula*
Fukui	Yashagaike	Sandstone, Shale, Chert	Brown forest soil	Fagus crenata, Lindera umbellata var. membranacea
Gifu	Ijirako	Chert	Brown forest soil	*Pinus densiflora*、*Chamaecyparis obtusa*
Kyoto	Sawanoike	Chert	Drained brown forest soil	*Pinus densiflora, Quercus serrata*
Shimane	Banryuko	Quaternary period sediment	Residual regosol	*Pinus densiflora, Pinus thunbergii*
Yamaguchi	Yamanokuchi dam	Rhyolite	Brown forest soil	*Rhododendron reticulatum*、 *Pinus densiflora*
Kagawa	Nagatomike	Diorite	Drained brown forest soil	*Quercus acutissima, Quercus serrata,*

The principal component analysis (PCA) was applied to the mean values of the monitoring sites, and loadings of two major factors (a) and principal component scores (b) were shown in Fig. 3-2-2-1. The first factor loading (X axis) was 45.0% and the second factor loading (Y axis) was 33.9%, and more than 75% was explained by two axes. The data of Lake Nagatomiike with significant high alkalinity and Ca2+ was omitted in this analysis. The PCA was applied only to the acid sensitive lakes with low alkalinity (< 0.200mmolc L-1), and therefore, characteristics of the respective lakes/catchments were confirmed in detail.

Fig. 3-2-2-1 Factor loadings (a) in the principal component analysis and principal component scores (b) of the respective monitoring lakes



As for the loadings of two major factors (see Fig. 3-2-2-1 a), the first factor loadings (X axis) of EC and many ions such as sea-salt-origin ions, Mg^{2+} and K^+ showed positive high values, indicating amounts of dissolved ions in the water. The second factor loadings (Y axis) of Ca^{2+} and alkalinity showed high positive values, while the loading of H^+ showed negative value, indicating acidity of the water. Moreover, the loadings of SO_4^{2-} and NO_3^- showed positive high values, suggesting effects of these ions.

As for the scores of each monitoring site (see Fig. 3-2-2-1 b), Lake Sankyoike and Lake Banryu showed high values on X axis because of their high EC, Na+, and Cl- suggesting effects of sea salts. The third quadrant may show the category of acid-sensitive lakes, whose acidity is high and ion concentration is low. The waters at Lake Yashagaike, Lake Futagoike (Meike), and Lake Sawanoike, which were located in the third quadrant, were high acid sensitive and therefore, acidification of these lakes should be carefully monitored. Lake Imagamioike was also located in the third quadrant and might be relatively acid sensitive. On the other hand, Lake Ijira, which has been acidified, was located in the second quadrant, showing less acidity compared with the lakes such as Lake Yashagaike. However, concentrations of SO_4^{2-} and NO_3^- at Lake Ijira were very high, suggesting effects of NO_3^- leaching by N saturation and high SO_4^{2-} leaching, as discussed later in the section 3.3. Lake Ijira should also be carefully monitored, since the acidification status may be different from those at the third quadrant. Lake Karikomi may have effects of high concentrations of SO_4^{2-} and Ca^{2+}.

(2) Long-term acidification trends of inland waters

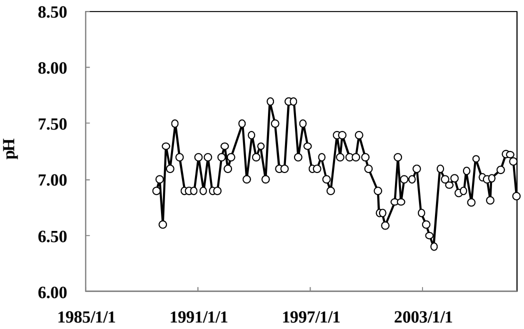
A long-term trend analysis was done for major parameters by using the seasonal Mann-Kendall tests (Hirsch et al, 1982) as shown in Table 3-2-2-2. The analysis was applied only for several long-term monitoring sites.

Table 3-2-2-2 Long-term trend analysis of major parameters in the surface water or inflow rivers of the monitoring lakes

Lake or River name	Duration	N	Z-score										
			pH	EC	Alkalinity	SO_4^{2-}	NO_3^-	Cl^-	NH_4^+	Na^+	K^+	Ca^{2+}	Mg^{2+}
Hutagoike(Oike)	1998-2007	30	1.56	1.83	1.35	4.94***	0.52	-0.31	-0.29	2.35**	-1.74	2.18*	1.26
Hutagoike(Meike)	1998-2007	30	-0.98	-1.77	0.73	3.53***	-1.66	-0.73	-1.83	2.94**	-1.84	0.05	-0.64
Sankyoike	2001-2007	27	-1.52	0.95	0.96	0.00	0.24	0.87	-0.78	0.16	-0.48	0.39	0.00
Yasyagaike	2000-2007	41	-1.81	-1.10	-2.27*	-1.23	1.55	-2.58**	-0.66	-2.00*	0.97	-1.17	-0.91
Ijirako	1988-2007	76	-0.74	5.68***	5.45***	0.87	5.43***	2.3*	1.89	0.89	-0.56	3.60***	2.55*
Kamagatani river	1988-2007	77	-2.07*	3.28**	1.19	1.03	6.00***	2.49*	-0.11	0.96	-0.05	1.49	1.68
Kobara river	1988-2007	57	-1.00	5.43***	1.66	2.20*	5.36***	2.28*	-0.41	2.87**	1.82	2.20*	3.16**
Banryuko	1989-2007	76	2.49*	5.14***	5.63***	-0.47	0.84	4.96***	-1.98*	6.00***	1.16	-0.97	3.61***
Banryuko No3.	1989-2007	76	2.40*	5.71***	4.60***	0.00	-0.11	4.28***	-1.61	5.61***	0.83	0.13	3.11**

Notice: Asterisks, *, **, and ***, shows significant levels, $P < 0.05$, 0.01, and 0.001, respectively, by long-term trend analysis of seasonal Mann-Kendall tests using 3 or 4 times yearly seasonal data.

Kamagatani River (an inflow river to Lake Ijira) showed a significant declining trend of pH ($p < 0.05$). As suggested in the Synthesis Report on Japan Acid Deposition Surveys (2004), a remarkable pH decline was seen since 1996 at Kamagatani river, but the trend became mostly flat since 2004 (Fig. 3-2-2-2 (a)). On the other hand, a significant increasing trend of pH was found in Lake Banryu (Fig. 3-2-2-2 (d)). A significant declining trend of alkalinity was seen in Lake Yashagaike, while the increasing trend was seen in Lake Ijira and Lake Banryu (Fig. 3-2-2-2 (e)).



Fif. 3-2-2-2(a)

pH changes at Kamagatani river

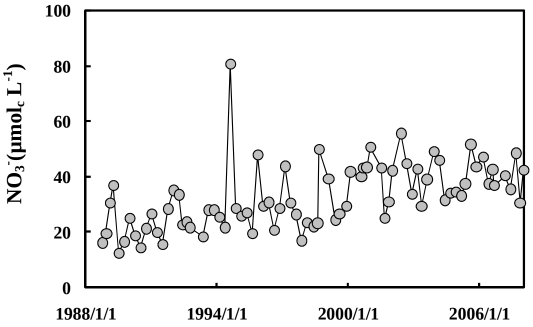


Fig. 3-2-2-2(b)

NO_3^- changes at Kamagatani river

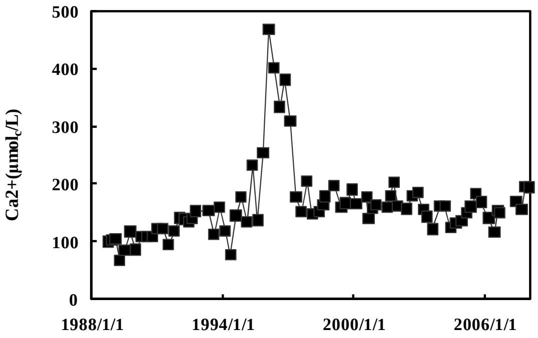


Fig. 3-2-2-2(c)

Ca^{2+}changes at Lake Ijira

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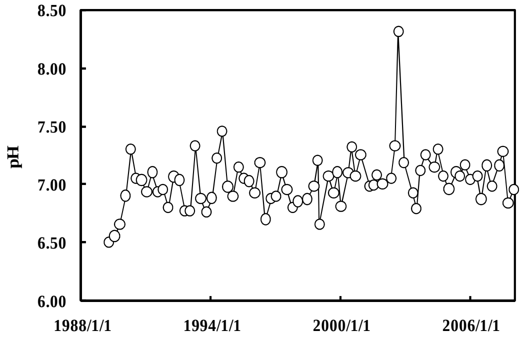

Fig. 3-2-2-2(d)

pH changes at Lake Banryu

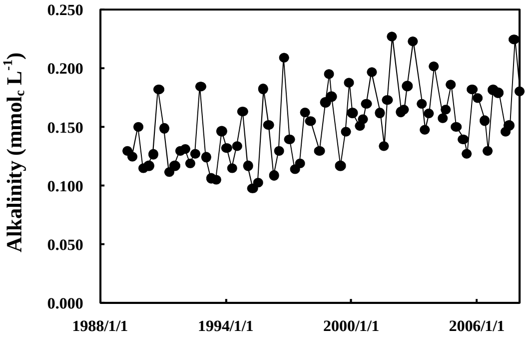

Fig. 3-2-2-2(e)

Alkalinity changes at Lake Banryu

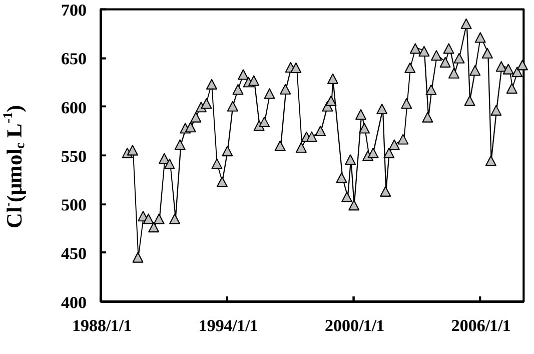

Fig. 3-2-2-2(f)

Cl- changes at Lake Banryu

At the Lake Ijira catchment, a significant increasing trend of NO_3^- concentration was seen in the surface water of the lake and two inflow rivers (Fig. 3-2-2-2 (b)). As mentioned above, the pH of Kamagatani River showed the declining trend, while the alkalinity of the lake surface water showed the increasing trend. Moreover, concentrations of base cations such as Ca^{2+} and Mg^{2+} also showed increasing trends in the lake surface water and Kobora River, another inflow river (Fig. 3-2-2-2 (c)).

Significant increasing trends of SO_4^{2-} concentration was seen in Lakes Futagoike (twin lakes, Oike and Meike). At the Lake Oike, concentrations of Na^+ and Ca^{2+} showed a significant increasing trend. At the Lake Banryu, where the alkalinity shows the increasing trend, Na^+, Cl^-, Mg^{2+} showed the increasing trend (Fig. 3-2-2-2 (f)). Sea-salt-origin ions or ions derived from anthropogenic activities seemed to be increasing there, while Ca^{2+} concentration showed no clear trend.Concentrations of Na^+ and Cl^- showed a significant declining trend at the Lake Yashagaike, which was classified into the most acid-sensitive lakes (see Fig. 3-2-2-1).

(2) Quality assurance/quality control (QA/QC) of the data

The data quality is assured through the audit of the national center of Japan, ADORC, and in this occasion, sampling procedures at the sites and conditions of the laboratories are confirmed. Results of the inter-laboratory comparison projects for the last five years were shown in Fig. 3-2-2-3. Through all the projects, the data more than 90% satisfied the data quality objectives. The laboratories seemed to keep relatively high data quality for the period. It was clarified that cations (low concentration or divalent ions) showed flagged data more frequently than pH, EC, and anions and larger variations among the laboratories. Efforts should be made for the data quality of cations.

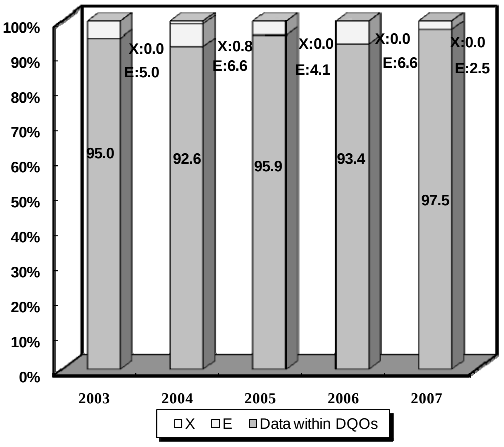

Fig. 3-2-2-3 Changes of number of flagged data in the inter-laboratory comparison projects for the last five years.

3.2.3 Discussion on ecological effects of acid deposition

As described above, during the last five years, obvious acidification trends and related tree decline phenomena were not reported in most sites, while soil acidification was found in the Lake Ijira catchment. Since a few plots' pHs were lower than 4.0 in the Lake Ijira catchment, the soil acidification and its effects on trees should be monitored and carefully evaluated. As described later in the section 3.3, acidification phenomena have been reported recently in the Lake Ijira catchment, suggesting effects of acid deposition. In some areas whose soil or geology may have low acid buffering capacity, a long-term acidification trend of rivers was reported. In this sub-section, referring the previous research facts, hot spots, the area with high risk of acidification, were identified based on soil or geological features and acid deposition amounts.

(1) Identification of acid-sensitive areas

As for spatial distribution of sensitivities to acid deposition, Environment Agency of Japan and Japanese Society of Soil Science and Plant Nutrition have compiled "the Map for assessing susceptibility of Japanese soils to acid precipitation (1:2,000,000)" in 1983 (Fig. 3-2-3-1). In the map, Japanese major soil types were classified into 6 classes and a distribution of each class was shown on a soil map of Japan (the classification was updated and merged into 3 classes in 1994). According to the map, soils with low acid buffering capacity, such as regosols (immature soils), red-yellow soils, and brown forest soils (dry type), are distributed mainly in Southeast Japan. The map was created based on acid buffering capacity of soils. However, chemical components of rocks or minerals as a parent material of soil, their weathering rate, and tolerance of ecosystems (vegetation) may be related to the sensitivity of each area.

Taking the questions above into account, Yoshinaga et al. (1994) created a map of relative sensitivity to acid deposition for Japan Islands by using Digital National Land Information according to the method of Stockholm Environment Institute (SEI method). In the SEI method, four parameters, including geology, soil, land-use, and precipitation are taken into account and 5-class evaluation (from 1 to 5) are applied to each parameter. As a result, the areas with the following conditions become the most sensitive; whose precipitation amount is more than 1200 mm, dominated by coniferous species, and covered by soils derived from silicate-rich rocks with low weathering rate. The most sensitive points with the highest score, 5, can be found mostly in the area, where granite rocks are distributed, mostly in Southeast Japan. The sensitive points showing the score, 4, were distributed also in granite-rock areas or coniferous forests in Hokkaido. These high-sensitive areas included the areas covered by granite rocks in Kii Peninsula, Chubu (Central) Mountains including Gifu Prefecture, and Asahi Mountains in the north part of Niigata Prefecture, in addition to Southeast Japan. In the SEI method, the precipitation amount, 1200 mm, was used as a threshold but most part of Japan may have larger precipitation. Therefore, difference cannot be clarified by the precipitation, and this may be one of problems on applicability of this method to Japan.

Based on the previous approaches above, as acid-sensitive ecosystems, the areas, where silicate-rich rocks such as granite are distributed, and also which were mainly covered by acid-sensitive soils,

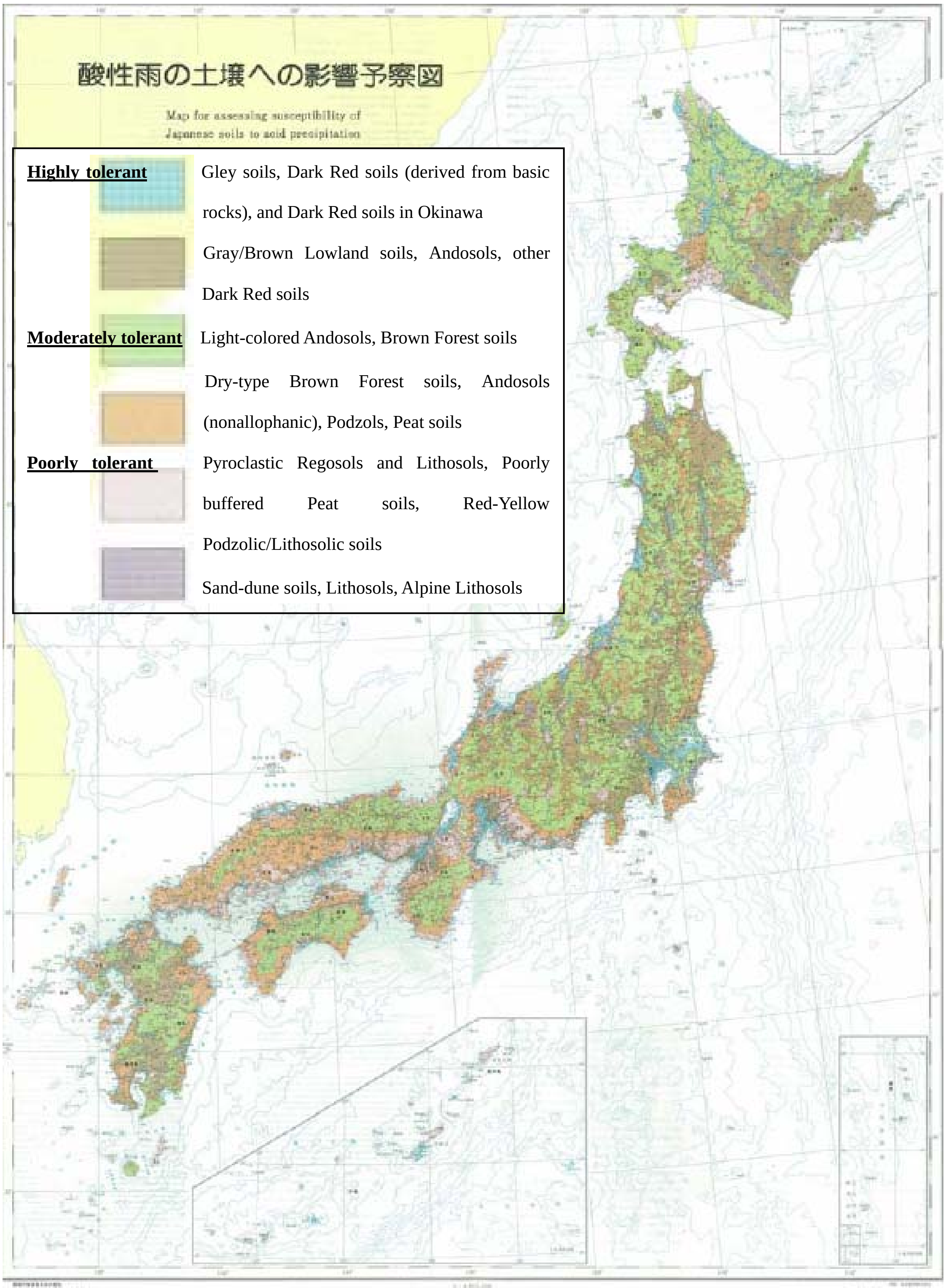

Fig. 3-2-3-1 Map for assessing susceptibility of Japanese soils to acid precipitation (Environment Agency of Japan and Japanese Society of Soil Science and Plant Nutrition, 1983)

such as immature soils, red-yellow soils, and brown forest soils (dry type), can be identified.

Although the map by Yoshinaga et al. does not include Yakushima Island in South Kyushu, the island is famous for granite. Vegetation types should also be considered but most part of Japanese forest is covered by planted coniferous species, such as *Cryptomeria japonica* or *Chamaecyparis obtusa*. Further investigation is necessary on evaluation of vegetation distribution.

(2) Distribution of wet deposition

Effects of acid deposition on ecosystems may become obvious in the areas, which are highly acid-sensitive and also whose deposition is significantly large. Since it takes a long time for changes in soil chemical properties, deposition amounts in the past such as 1960s' or cumulative amounts should also be considered. At present, deposition amounts in the past were not available enough but surrounding areas of mega cities or industrial areas should be taken into consideration.

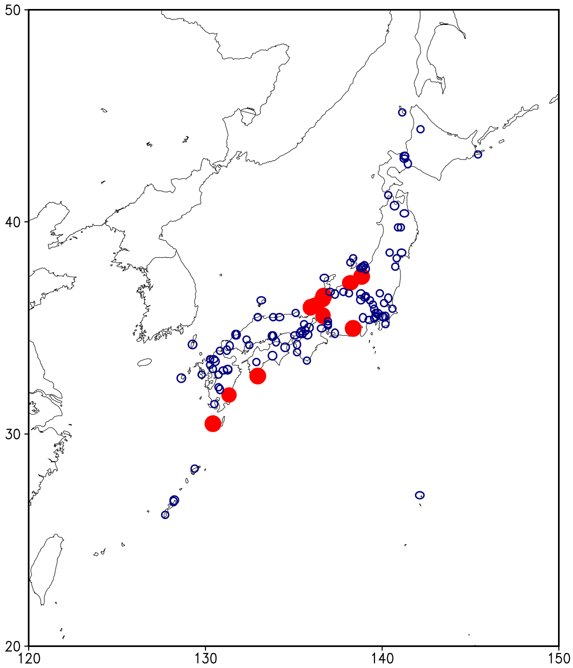

Fig. 3-2-3-2 Distribution of the monitoring sites with high wet H^+ deposition in Japan The wet deposition of H^+ at the red colored sites is within 10% highest among 120 sites.

Distribution of the monitoring sites with high wet H^+ deposition was shown in Fig. 3-2-3-2. The map was created based on the surveys by Japan Environment Laboratories Association (JELA) (1999 – 2004) and MOEJ (1998 – 2004) for evaluation of regional watershed acidification, which was discussed in the section 3.3. Although contribution of dry deposition should also be considered, a spatial distribution of wet deposition was discussed tentatively in this evaluation because of its high data availability. The sites with high wet H^+ deposition were distributed in Chubu region including Lake Ijira, Shizuoka, and the coastal area along the Sea of Japan, Shikoku Island, South Kyushu, and Yakushima Island. As described in the section 3.3, the deposition characteristics are different among these sites. In the south part of Japan, such as Shikoku Island, South Kyushu, and Yakushima Island, the concentration was relatively low but the precipitation amount was significantly high, and therefore, the deposition amount was the highest levels in Japan. On the other hand, in Chubu region, including Lake Ijira and the coastal area along the Sea of Japan, both concentration and precipitation were relatively high, and therefore, the deposition amount was the highest levels in Japan. These deposition characteristics should also be considered for the evaluation in future.

(3) Hot spots to be monitored

As highly acid-sensitive ecosystems, the forest areas, where silicate-rich acid rocks such as granite are distributed and which is covered by immature soils, red-yellow soils, or brown forest soils (dry type), can be considered and these areas may be distributed in the north part of Niigata Prefecture, Chubu region, Kii Peninsula, and Southeast Japan. Taking the wet deposition amount into account, south part of Chubu region including Lake Ijira, the coastal area of Chubu region along the Sea of Japan including Niigata Prefecture, South Kyushu, and Yakushima Island can be identified as hot spots. As discussed later in the section 3.3, the long-term declining trend of the river water pH was reported in Gifu Prefecture and the north part of Niigata Prefecture. Especially in Niigata Prefecture, the relationship with geology was obvious. In these areas, acidification on the watershed scale, including soil and river, should be considered. Possibility of long-term soil acidification should be checked in the areas and an investigation of the soil plots surveyed in the past can be one of the options. In case of Yakushima Island, the soil layer is relatively thin, and also the direct runoff may have large contribution to water discharge taking steep geographical conditions of the island into account. It may be possible that much water not enough neutralized in the soil layer is flowed into streams/rivers in such a condition. Effects on streams/rivers and aquatic organisms should mainly be considered there.

In addition to the hot spots identified above, North Kyushu and Seto Island Sea region should also carefully evaluated, although these regions were not identified as high-deposition areas. Because major industrial areas are located in these regions and it seemed emission from these areas were significantly much in the past.

Discussion above mainly focused on the watershed/catchment scale acidification including soil and river. However, direct effects of acidic/oxidizing pollutants on trees should also be considered for identification of hot spots. Taking recent increasing trends of ozone concentrations and widespread Photochemical Oxidants Warning into account, direct exposure effects of ozone on plants should be evaluated. Calculation of AOT40 using the available ozone data and measurement of ozone concentrations in forest area by using passive samplers should be considered. Through the activities, the areas, where the ozone concentration is higher than certain threshold, should be identified, and then, the monitoring system focusing on ozone effects, such as observation of specific visible injury, should be developed in future.

References

1) Yoshinaga et al. (1994) Mapping of relative sensitivity to acid deposition for Japan Islands by utilizing Digital National Land Information. *Japanese Journal of Soil Science and Plant Nutrition*, 65, 565-568.

2) Hettelingh et al., 1991. Mapping critical loads for Europe, CCE Technical Report, No.1, p.86, Coodinate Center for Effects, National Institute of Public Health and Environment Protection, Bilthoven, Netherland.

3.3 Results of Ijira Intensive Surveys

3.3.1 Acidification phenomena in Lake Ijira catchment

The Synthesis Report of Japan Acid Deposition Surveys (2004) reported that simultaneous changes in chemical properties of inflow rivers and soil were recorded in Lake Ijira catchment in Yamagata City, Gifu Prefecture, where wet deposition amounts of acid substances were the highest level in Japan. In this sub-section, acidification phenomena and discussions on the mechanism, which were reported in the Synthesis Report, will be summarized, and the discussion point of the intensive survey will be clarified.

(1) The highest level of wet deposition in Japan

It was pointed out that wet depositions at the Lake Ijira catchment were the highest level among the monitoring sites in Japan. Especially for nss-SO_4^{2-} and H^+, wet deposition amounts at Lake Ijira catchment were the highest among the sites. Annual total deposition of N was estimated to be approximately 19 kg-N ha^{-1} y^{-1}, which exceeded 10 kg-N ha^{-1} y^{-1}, a threshold value of the dissolved inorganic N (DIN) input at which NO_3^- output increase in river water in Europe and the U.S.

(2) Low acid neutralizing capacities in terms of geology/soil conditions and river water chemistry

Bedrock geology of the Lake Ijira catchment is dominated by chert, which is a silicate-rich rock. It can be considered that the acid neutralizing capacity (ANC) of the catchment is relatively low geologically. In fact, as described in the section 3.2, soils in the Lake Ijira catchment have been acidified continuously since the survey started. Although an internal acid production by plant uptake or the forest management condition should also be considered, the large wet deposition may affect soil chemistry.

Geology and soil conditions may closely related to acidification on the catchment-scale. It is pointed out that acid rocks, such as granite and ryholite, and chert, which are rich in silicate and poor in base cations, and soils derived from these rocks may have low ANC.

River water chemistry can be considered as a result of chemical reactions under the geology and soil conditions in the catchment and material cycles in the soil-plant ecosystems. The inland water, whose alkalinity as an indicator of ANC is lower than 200 μmol_c L^{-1}, may be susceptible to acid deposition. It was reported that alkalinity was relatively high in most rivers of Japan, 550 μmol_c L^{-1} in average of 225 rivers, and the acid-sensitive rivers with the alkalinity < 200 μmol_c L^{-1} was limited (Kobayashi, 1960). However, the alkalinity of Kamagatani River, one of the inflow rivers of Lake

Ijira, was very low, approximately 120 $\mu mol_c\ L^{-1}$, which can be considered as acid-sensitive also in terms of the river water chemistry.

(3) Specific ion leaching triggered by meteorological events, such as cold summer, and drought/torrid heat of summer

At Kamagatani River flowing into Lake Ijira, the high-concentration leaching of NO_3^-, SO_4^{2-} and Al and the decline of pH was observed from 1994 to 1996, as shown in Fig. 3-3-1-1. The NO_3^- concentrations have been increased, and an effect of high N deposition was suggested.

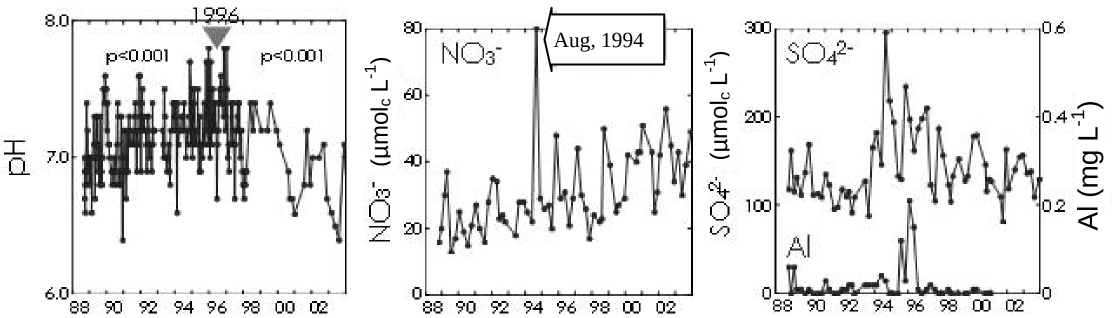

Fig. 3-3-1-1 Chronological changes of concentrations of pH (left), NO_3^- (middle), and SO_4^{2-} and Al (right)

Moreover, it was estimated by the preliminary budget analysis that the output of SO_4^{2-} from the river exceeded the input to the catchment, and it was suggested that S accumulated in the catchment has been leached into the river. In fact, extractable S in the soil was single- or double-digit larger than those in other previous study catchments by MOEJ, suggesting high S content in the soil.

As a possible mechanism of high-concentration ion leaching, reduction of tree growth caused by the cold summer in 1993 and the drought and torrid heat of summer in 1994 might induce changes in N cycle processes and affect the river water pH. Chronological changes in mean basal area increments (BAI) for two tree species, *Cryptomeria japonica* and *Chamaecyparis obtusa*, which were thinned by the forest management in the catchment, were shown in Fig. 3-3-1-2. Both species showed significant reduction of BAI twice, since 1982 and 1994.

Although effects of the growth reduction since 1982 was not clarified in this study (started in 1988), N uptake must be reduced by the growth reduction since 1994. It can be considered that the reduction of N uptake caused NO_3^- leaching into the river and leaching of H^+ produced with NO_3^- decreased the river water pH. Moreover, rapid mineralization of soil organic matters induced by precipitation after drought in 1994 may also contribute to NO_3^- production. After this period, seasonality of NO_3^- concentration in the river was lost, and high NO_3^- concentrations could be seen even in summer, where N uptake by plants must be active. Thus, the N cycle in the catchment may be changed into the condition that NO_3^- can be easily leached into the river.

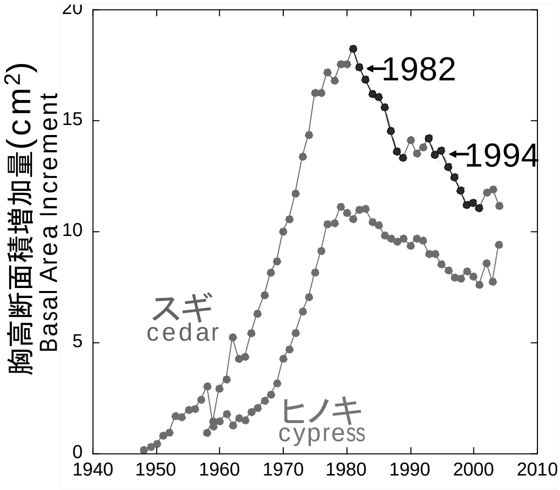

Fig. 3-3-1-2 Chronological changes on basal area increment of Japanese cedar (*Cryptomeria japonica*) and Japanese cypress (*Chamaecyparis obtusa*) in the Lake Ijira catchment. The plots for the respective species were the mean of 50 trees.

Moreover, it may be possible that mineralization of organic S was also accelerated by the drought event as well, although the detailed mechanism needs further discussion. Since H^+ can be produced also in the process of the organic S mineralization, this might contribute to decrease of the river water pH. After high-concentration leaching of the ions, a positive correlation between NO_3^- and SO_4^{2-} was seen, suggesting simultaneous leaching of these ions, although the positive correlation could not be seen in usual.

(4) Summary of hypothesis on acidification process in the Lake Ijira catchment

A hypothesis on the catchment-scale acidification and processes of ion leaching could be summarized as shown in Fig. 3-3-1-3. In the catchment ecosystems, which received high acid loads, meteorological events functioned as a trigger of changes in material cycles. In particular, the N cycle changed from the N-retaining condition into the N-releasing condition (Nakahara et al, submitted). Then, acidification proceeded in the catchment. The process can be understood as follows:

i) Wet deposition in the Lake Ijira catchment is the largest level in Japan
ii) Soil pH has decreased, and its effects on trees are concerned.
iii) The Kamagatani catchment, one of the sub-catchments, has an acid-sensitive catchment with low alkalinity, reflecting geology and soil conditions.
iv) It was estimated that the output of SO_4^{2-} from the river exceeded the input from the

atmosphere to the catchment.

v) N deposition exceeds the threshold value of DIN input at which NO_3^- output increases in the river. In fact, the NO_3^- concentration increases year by year.

vi) Tree growth, the main sink of N in the ecosystems, decreased due to meteorological events, such as cold summer and drought in mid-1990s. Reduction of N uptake by trees and enhanced mineralization of organic N after the drought caused high-concentration NO_3^- leaching, accompanied by much H^+ production and leaching in the catchment resulting declining of the river water pH.

vii) The NO_3^- leaches into the river even during the tree-growing season in summer.

viii) It is possible that mineralization of accumulated organic S is also accelerated by drought causing high-concentration leaching of SO42- in mid-1990s.

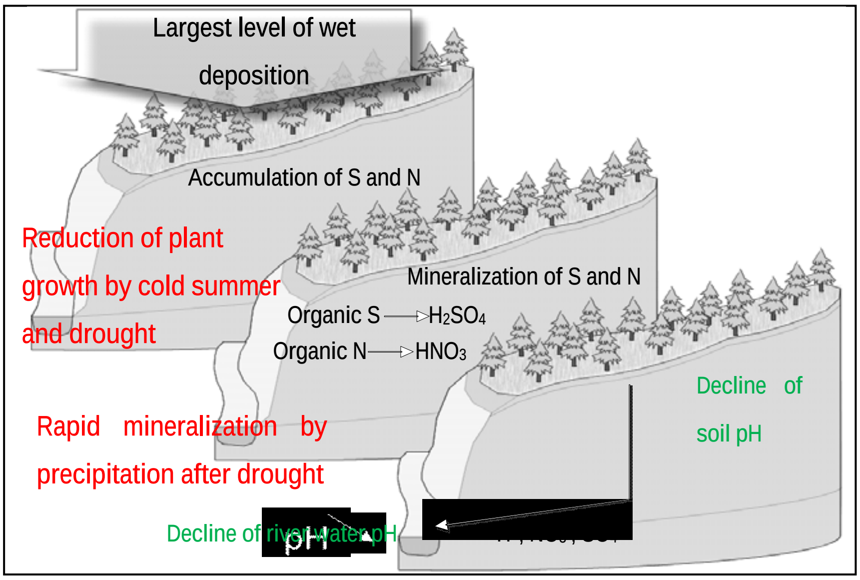

Fig. 3-3-1-3 Hypothesis of acidification process in the Lake Ijira catchment

Acidification process in the Lake Ijira catchment has been clarified by the Synthesis Report and subsequent discussions. However, to discuss the process above precisely, total input including wet and dry depositions, plant growth/uptake rate in the catchment-scale, and input-output budget should be precisely estimated. So far, enough data sets have not been obtained for precise quantitative discussion only by the regular monitoring; dry deposition has not been considered enough, river chemistry and discharge were measured only four times a year, and growth rate was measured. Taking the questions above into account, the Ijira intensive surveys, “Evaluation of mechanism on catchment-scale acidification”, was implemented from 2005 to 2007 to clarify the acidification process more quantitatively based on biogeochemical cycle analysis.

3.3.2 Summary methods of the intensive surveys

Intensive surveys on input and output were carried out in the Lake Ijira catchment to estimate total input (deposition) from atmosphere and output from the river. Intensive sampling of the river water was also conducted during heavy rain events to clarify runoff characteristics, and the acidification process was discussed in detail. Moreover, in other catchments/watersheds than the Lake Ijira, such as mountainous lakes/ponds and streams located in granite area, declining trends of pH and/or alkalinity were reported, and it was suggested that cumulative inputs (depositions) of acid substances might exceed the neutralizing capacity of these catchments. Therefore, to discuss the possibility of spread acidification of watersheds by atmospheric deposition, analysis of public data was also conducted for distribution of precipitation and wet deposition in the whole country and long-term trends of river water pH in the Chubu region. The measurement items are as follows:

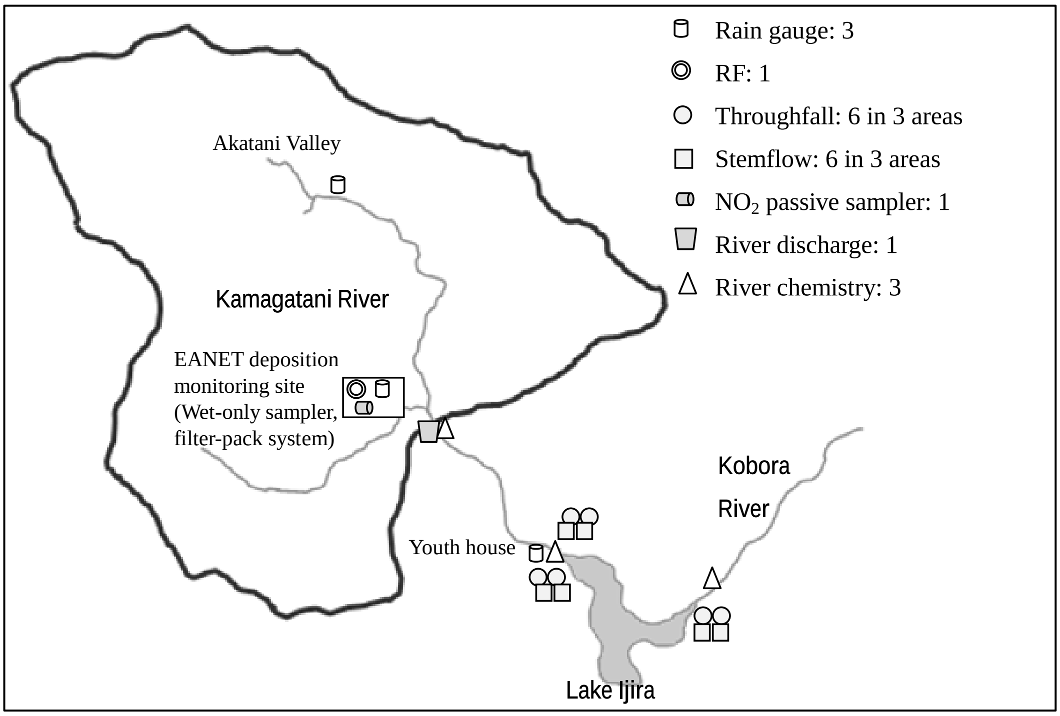

Fig. 3-3-2-1 Layout of the collectors in the Lake Ijira catchment

The area surrounded by bold line shows the catchment (approx. 298 ha) for budget analysis.

(1) Input-output surveys (see Fig. 3-3-2-1)

The following items were measured to estimate total input to the catchment and output from the river.

- For input:

  Rainfall outside the forest canopy, throughfall, stemflow, precipitation amount, NO2 measurement by a passive sampler

- For output:

  Discharge from the river, river water chemistry (2 points at Kamagatani River and 1 point at Kobora River)

Water samples were collected every two weeks and analyzed for cations and anions. Alkalinity, total dissolved Al, SiO2, and total organic carbon (TOC) were also analyzed for the river waters. As for estimate of total input, data on wet deposition by the wet-only precipitation sampler and dry deposition by the filter-pack method at the Lake Ijira EANET deposition monitoring site and also data of the passive sampler (for NO_2) were used.

(2) Intensive sampling of the river water during heavy rain events

Intensive sampling of the river water was implemented by using an automatic water sampler twice during heavy rain events. The water samples were collected at one-hour interval for two days (48 hours), respectively, from 18 to 20 July 2006 and from 13 to 16 July 2007. Cations, anions, alkalinity, total dissolved Al, SiO2, and TOC were analyzed.

(3) Spatial evaluation of wet deposition in the whole country

Wet deposition amount at the Lake Ijira catchment is the highest level in Japan, because the ion concentrations are relatively high and also the precipitation amounts are significantly large, approx. 3000 mm. As for the possibility of widespread acidification of watersheds, the precipitation data at Automated Meteorological Data Acquisition System (AMeDAS) stations, and wet deposition data at stations by MOEJ (from 1998 to 2004) and stations by JELA (from 1999 to 2004) were synthetically analyzed to identify spots to be monitored in terms of wet deposition, and the position of the Lake Ijira catchment in the whole country and other stations with high-wet-deposition were identified.

(4) Trend analysis and field measurements of rivers and streams in Niigata and Gifu prefectures

Long-term acidification has been reported for the mountainous rivers, which were located in areas dominated by acidic rocks such as granite, in Nagano Prefecture (Kurita and Ueda, 2006). To discuss possibility of more widespread acidification of watersheds in Chubu region, long-term trend analysis on river water pH was carried out by using the Public Water Body data in cooperation with Niigata and Gifu prefectures.

3.3.3 Summary of the survey results

The major results of the intensive surveys can be summarized as follows:

(1) Acid loads in the Lake Ijira catchment and its effects on river water chemistry

1) Input-output surveys

Changes in monthly precipitation amounts in the Lake Ijira catchments and the water level at Kamagatani River were shown in Fig. 3-3-3-1 for the period from August 2005 to November 2007. The precipitation amounts here included partly snowfalls in winter. The rain gauges were installed in three different altitudes, namely, Akatani Valley (upper slope), EANET station (middle slope) and Youth house (lower slope). Seasonal variation of the monthly precipitation amounts showed similar patterns among the positions, although the precipitation amounts are usually larger at the Akatani Valley on the upper slope. Changes in Kamagatani River reflected the precipitation pattern. The water year used for the following analyses was defined as starting at the beginning of November based on the seasonal variation of precipitation and water level.

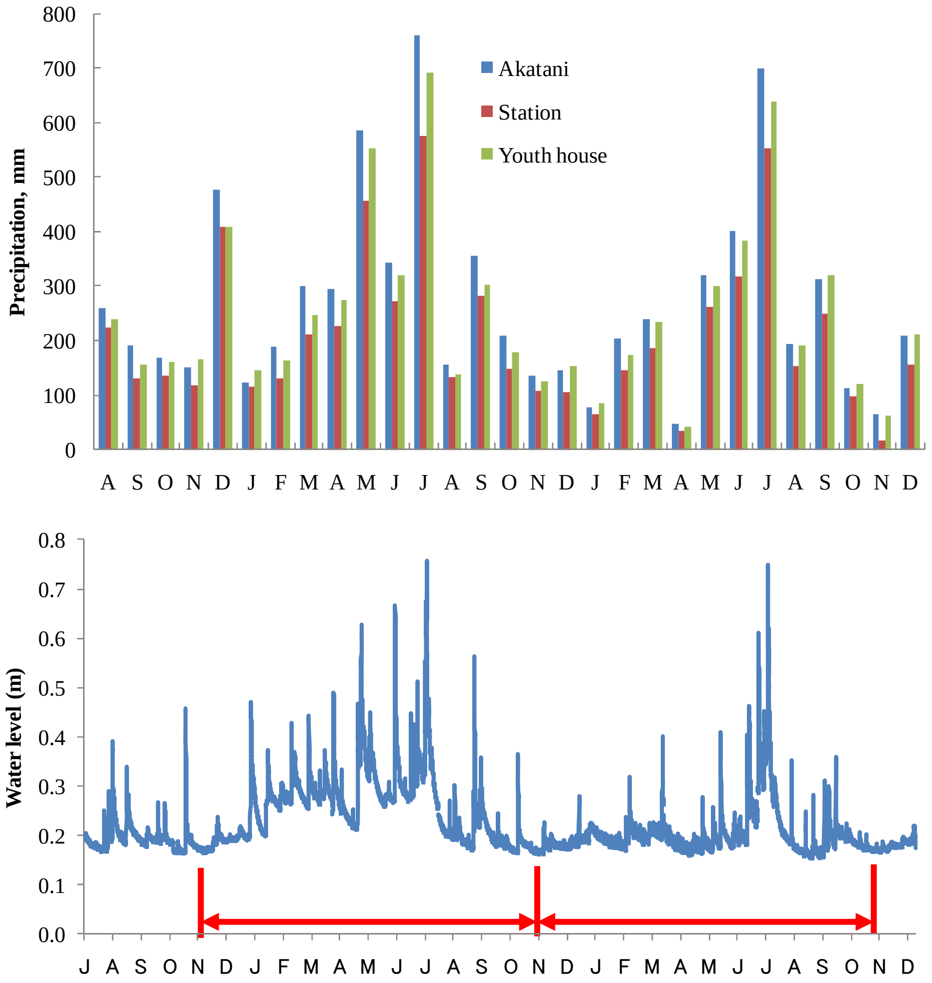

Fig. 3-3-3-1 Changes in precipitation and water level, from August 2005 to November 2007 Arrows show the water years.

Water balance in the Kamagatani sub-catchment was shown for two water years in Fig. 3-3-3-2, which was calculated based on the precipitation and water level in Fig. 3-3-3-1. Three slope positions showed different precipitation amounts and the precipitation was the highest at Akatani Valley on the upper slope, the second at Youth house on the lower slope, and then the lowest at EANET station on the middle slope. Since EANET station is located in a small valley of the branch river of the Kamagatani River, the precipitation amount may be underestimated there. The precipitation amount was approximately 800 mm lower in the water-year 2006-2007 than in the water-year 2005-2006. Runoff rate was approximately 77% and 49% in the first and second water years, respectively. The runoff rate in the first water year may be a reasonable value taking evapotranspiration rate into account, although the runoff rate in the second year seems to be low. Water balance may fluctuate depending on balance between precipitation and evapotranspiration in each water year. However, it can be considered the water balance applicable to the budget analysis was obtained at least for the first year, although long-term monitoring may be necessary for precise water balance.

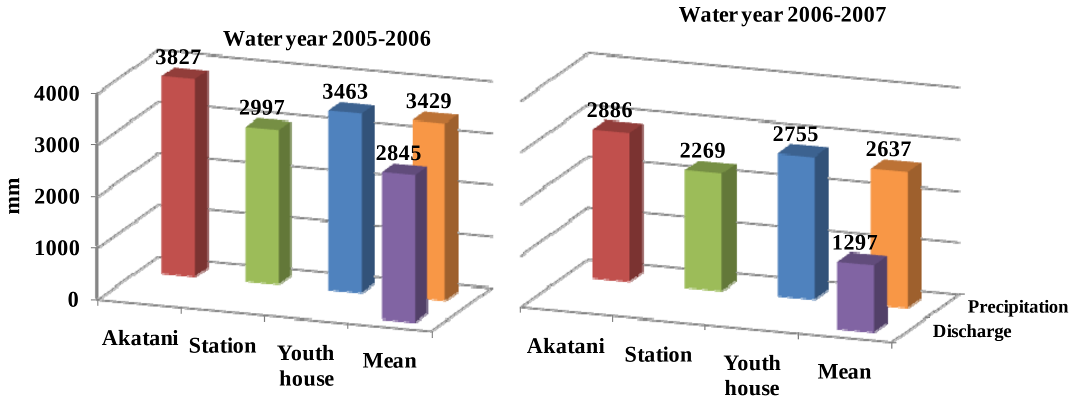

Fig. 3-3-3-2 Water balance in the Kamagatani sub-catchment

The water year 2005-2006: 364 days; the water year 2006-2007: 365 days

Total deposition (wet and dry) and the contribution rate of dry deposition were shown for the respective water years in Fig. 3-3-3-3. Wet depositions in the Lake Ijira catchment are the largest level in Japan because of the high precipitation amounts. Moreover, as described above, the precipitation amounts recorded at EANET station may be underestimated. Taking distribution of the precipitation amounts above into account, wet depositions were corrected by using the mean precipitation amounts of three slope positions. Therefore, the wet depositions in Fig. 3-3-3-3 were larger than those at EANET station. It was clarified that the contribution rates of dry deposition to the total deposition were relatively small, 5 – 25%. For calculation of dry deposition, NO_2 measured by the passive sampler was also taken into account for estimation of the NO_3^- deposition.

It has been pointed out that N deposition was significantly large in the Lake Ijira catchment. The estimated dissolved inorganic N input was 28.7 and 18.2 kg-N ha^{-1} y^{-1} in the first and second water years, respectively. The values significantly exceeded 10 kg-N ha^{-1} y^{-1}, the threshold discussed in Europe and U.S., at which NO_3^- output increases in stream water. It was suggested that N saturation accompanying acidification of the catchment would be possible.

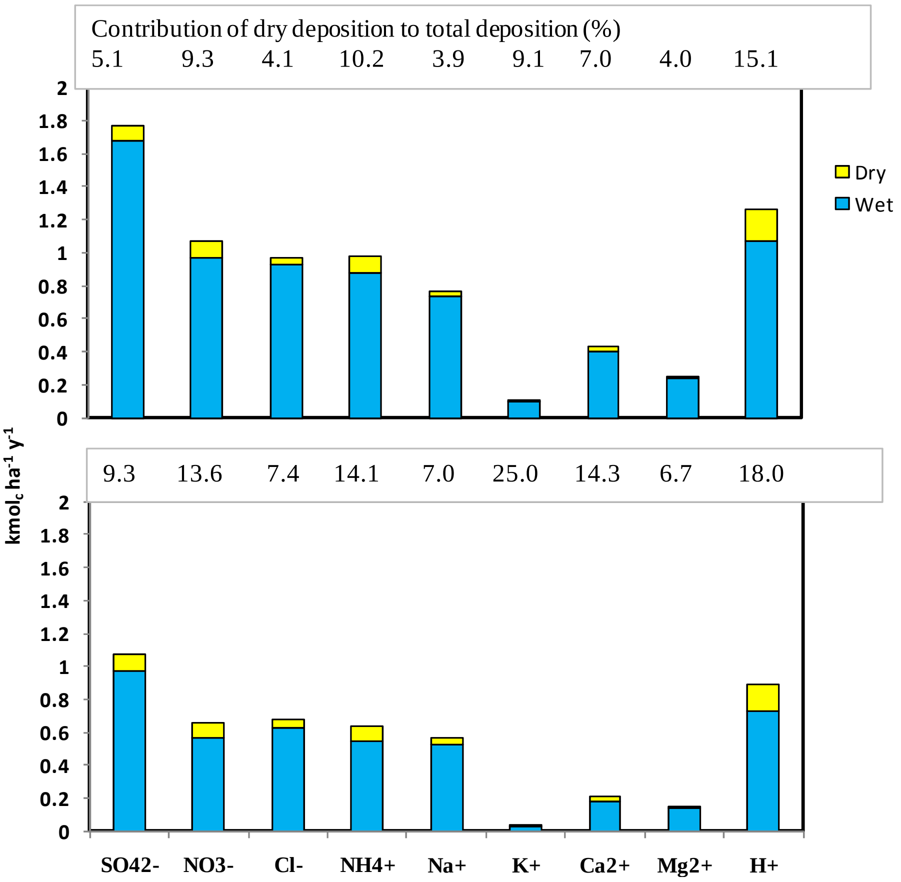

Fig. 3-3-3-3 Total deposition and the contribution rate of dry deposition in the Kamagatani catchment Wet, wet deposition; Dry, dry deposition. Wet depositions were calculated by using air concentration measured at EANET station and the mean precipitation amounts of three different slope positions. NO_2 measured by the passive sampler was included in the NO_3^- deposition.

The throughfall-stemflow method cannot precisely estimate the ions, such as NH_4^+ and K^+, which may have large effects of canopy interactions, namely, uptake, consumption or leaching of ions on forest canopy. In the surveys, wet deposition and dry deposition was estimated independently in addition to the throughfall-stemflow method, and therefore, possibility of uptake or leaching of the ions on forest canopy can be discussed more practically. Since measurement of throughfall and stemflow was implemented in the forest area surrounding the Lake Ijira, variations derived from positional or topographical differences with EANET station might be included in the throughfall-stemflow data. However, it was suggested that most of the total deposition was loaded onto the forest floor in SO_4^{2-} and NO_3^- as reported in previous literatures. Uptake, consumption or leaching of the ions on the forest canopy was not mostly found especially in the second water year, while some uncertain differences were found in the first water year. On the other hand, it was suggested that a part of NH_4^+ and H^+ was absorbed, consumed or neutralized on the forest canopy and much amount of K^+ and Ca^{2+} was leached from the forest canopy. It was pointed out that estimate of dry deposition was essential for precise discussion of material cycles in ecosystems including the forest canopy.

2) Intensive sampling during heavy rain events

Specific variation of ion concentration was found in the intensive sampling during heavy rain events in summer. Results of the intensive sampling in 2006 were shown in Fig. 3-3-3-4.

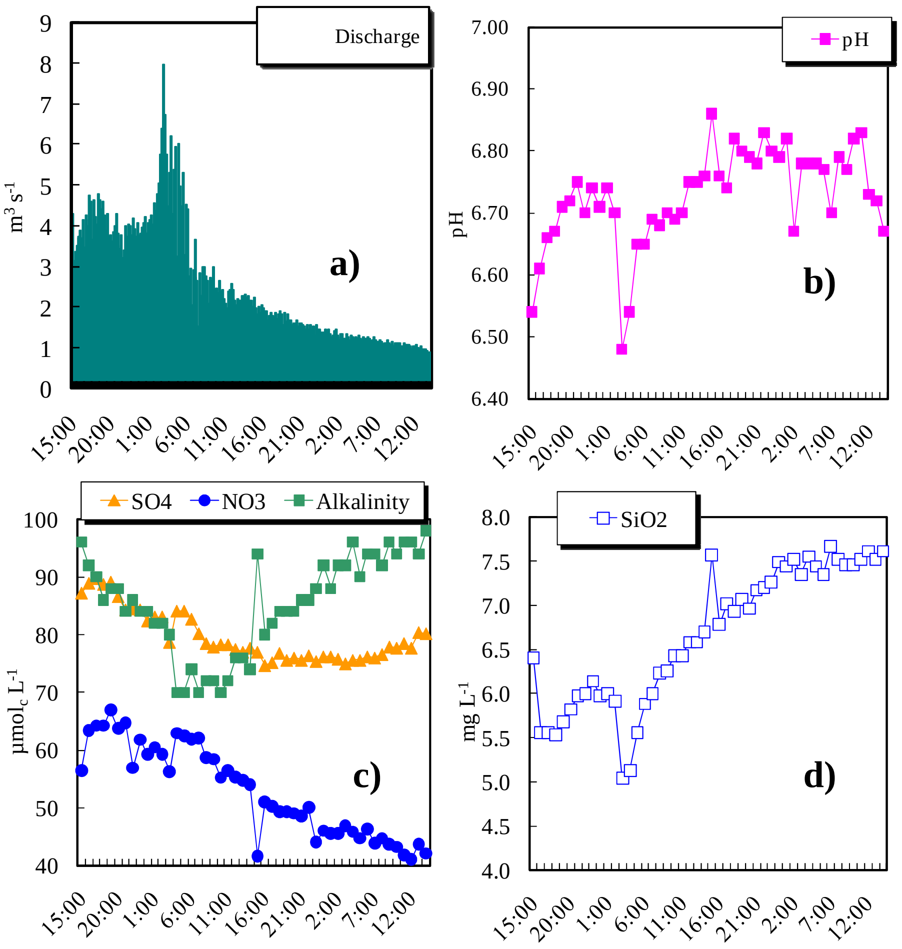

Fig. 3-3-3-4 Variation of ion concentration during a heavy rain event in 2006

(From 15:00, July 18, to 14:00, July 20 (at one-hour interval))

The sampling started at 15:00 in July 18, when the discharge had already increased into a certain level. The river water discharge started increasing again from around 1:00 in July 19 and then reached the peak at around 15:00. The pH decreased quickly with increase of the discharge and then increased gradually with decrease of the discharge. At the same time, alkalinity showed the similar pattern to that of pH. On the other hand, ion concentrations of NO_3^- and SO_4^{2-} decreased tentatively with the peak discharge but increased quickly and then decreased with decrease of the discharge. As a result, positive correlation between NO_3^- and SO_4^{2-} concentrations can be found during the peak discharge (from 15:00 July 18 to 12:00 July 19), while no correlation is found in ordinary condition. Moreover, a concentration of SiO_2, which is an indicator of rock weathering and included much in deep groundwater, decreased with increase of the discharge similar to other ions. The similar phenomena can be seen also in the intensive sampling in 2007.

Leaching through shallow soil layers or surface runoff may largely contribute to the discharge during heavy rain events due to increase of groundwater level. Therefore, N distributed mainly in shallow soil layers can be leached into the river as NO_3^-. So far, many reports pointed out that SO_4^{2-} concentration decreased with increase of the discharge. However, in the Lake Ijira catchment, SO_4^{2-} and NO_3^- were leached simultaneously with increase of the river water discharge. Therefore, it was suggested that leaching from shallow soil layer largely contributed to SO_4^{2-} leaching during heavy events. The leaching of NO_3^- and SO_4^{2-} may also contribute to the temporary acidification during heavy rain events. As described below, S output from the river is more than input (deposition) in this catchment, suggesting leaching of accumulated S. Further investigations are necessary for chemical form of accumulated S and its source.

3) Evaluation of the material budget

The input-output budget of major ions in the Kamagatani sub-catchment was shown in Fig. 3-3-3-5. In the catchment, the output of SO_4^{2-} was estimated to be approximately 190% and 170% of the input in the water years 2005-2006 and 2006-2007, respectively. It was clarified that SO_4^{2-} was discharged from the catchment more than its input. In the first water year, the budget of Cl^-, which may not mostly be utilized in ecosystems, was not balanced. Even if the input and output of Cl^- were balanced, the corrected output of SO_4^{2-} became 110% of the input.

In Europe, it was reported that SO_4^{2-} was still being discharged from streams and rivers in the area which suffered high acid deposition loads even now, while the deposition has already decreased by reduction of the emission. It was suggested that accumulated S in the past was discharged in such areas. In the Kamagatani sub-catchment, the similar phenomenon may occur. The output pattern of SO_4^{2-} during heavy rain events suggested contribution of accumulated S in the catchment. Moreover, outputs of Ca^{2+} and Mg^{2+} were more than 900% of the inputs, suggesting reduction of ANC of soil, while the river water was well neutralized.

Reflecting much N deposition, certain amount of NO_3^- was leached into the river even in summer when plant N uptake must be active, suggesting acid loads to the river with the NO_3^- leaching. In fact, NO_3^- concentration in the river water increased for the last decade and the pH showed a declining trend. Possible acidification due to NO3- leaching should be monitored continuously in future.

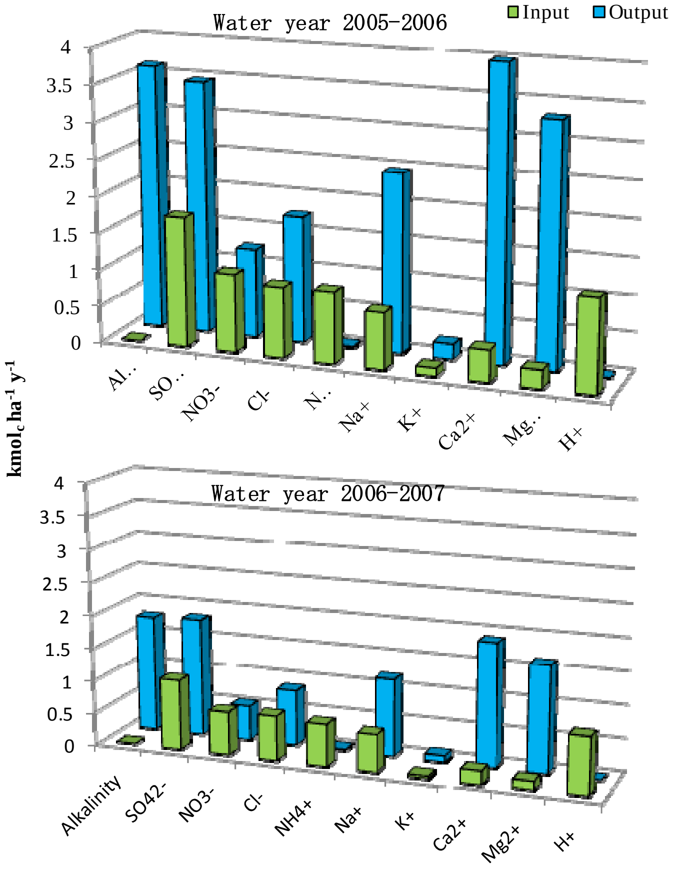

Fig. 3-3-3-5 Material budget in the Kamagatani sub-catchment

For precise discussion on the catchment-scale acidification by acid deposition, sources of H^+ including acid deposition and internal production in forest ecosystems as well as sink of H^+ in the ecosystems should be taken into account and calculation of H^+ budget in the ecosystems may be informative (van Breemen et al., 1983). In most catchments of Japan, production of alkalinity is larger than H^+ load derived from an external source, acid deposition; ANC in forest ecosystems is quite large (Tokuchi and Ohte, 1998). However, as shown in Table 3-3-3-1, H^+ load derived from acid deposition is larger than alkalinity production, ANC of the river water, suggesting possible acidification of the catchment.

Table 3-3-3-1 External H^+ load and alkalinity production in the Kamagatani sub-catchment

Water year	H^+ load		Alkalinity production
	kmol$_c$ ha^{-1} y^{-1}		
2005-2006	4.04	>	3.61
2006-2007	2.12	>	1.78

Note: External H+ load was calculated based of N budget and transformation, and budgets of SO_4^{2-} and H^+. Alkalinity discharge was assumed as the net production here.

4) Summary

The intensive survey above provides not only quantitative data which can support the previous discussion but also important facts for future catchment analysis. The main outcomes are as follows:

i. Total deposition was precisely estimated based on wet deposition taking account of spatial variation of precipitation in mountainous area and dry deposition calculated from air concentration measurement. It was clarified that N deposition in the Kamagatani sub-catchment was significantly larger than the threshold value, 10kg-N ha^{-1} y^{-1}, at which NO_3^- discharge might be accelerated; it was 28.7kg-N ha^{-1} y^{-1} in the first water year.

ii. Specific trends on SO_4^{2-} discharge during heavy rain events suggested contribution of accumulated S in shallow soil layers in the catchment.

iii. The material budget in the Kamagatani sub-catchment showed that the output of S from the river was larger than the input, also suggesting contribution of accumulated S to the stream acidification.

iv. The NO_3^- discharge derived from much N deposition as well as accumulated S may contribute to the stream acidification.

The outcomes above concluded that accumulated S, which might be derived from atmospheric deposition in the past, has been discharged into the stream in the Lake Ijira catchment, while the N deposition, which exceeded the capacity of the ecosystems, has caused continuous acidification of the catchment.

(2) Possible widespread acidification of watersheds

1) Characterization of the Lake Ijira catchment and extract of hot spots based on the spatial distribution of wet deposition in Japan

We deposition amounts of nss-SO_4^{2-}, NO_3^-, NH_4^+ and H^+ at the Lake Ijira station were within 10% highest among 120 stations in Japan according the recent data from 1998 to 2004. In particular, the wet deposition of H^+ was the highest in Japan. Much wet deposition amounts of acidic components at the Lake Ijira station can be attributed to high precipitation amounts, 15% highest in Japan, and their relative high concentrations. The concentrations of nss-SO_4^{2-}, NO_3^- and H^+ were 27%, 19% and 2% highest in Japan, respectively (Fig. 3-3-3-6).

On the other hand, the areas with high acid deposition load comparable to the Lake Ijira station were also identified by the analysis as shown in Fig. 3-3-3-7. Wet deposition of nss-SO_4^{2-} was high in the area along the Sea of Japan in Chubu and Chugoku region and Kyushu region in addition to Chukyo Industrial area and its surrounding area including the Lake Ijira station. The NO_3^- deposition was high in the area along the Sea of Japan in Chubu and Chugoku region, Tokyo, and the north Kanto region. The NH_4^+ deposition was high in the area along the Sea of Japan in Chubu region, Tokyo metropolitan area and the north Kanto region in addition to the Chukyo Industrial area and its surrounding area. The H+ deposition was high in the area along the Sea of Japan, Tokai region, and Kyushu region. Risk assessment of widespread watersheds acidification needs to discuss conditions of the ecosystems, such as geology, soil condition, vegetation, and topography.

The assessment above was based on the recent deposition data but cumulative deposition amounts for the last several decades should also be considered for possible effects on the ecosystems. In particular, S deposition must be significantly larger during the high-growth period of Japan in 1960s than present.

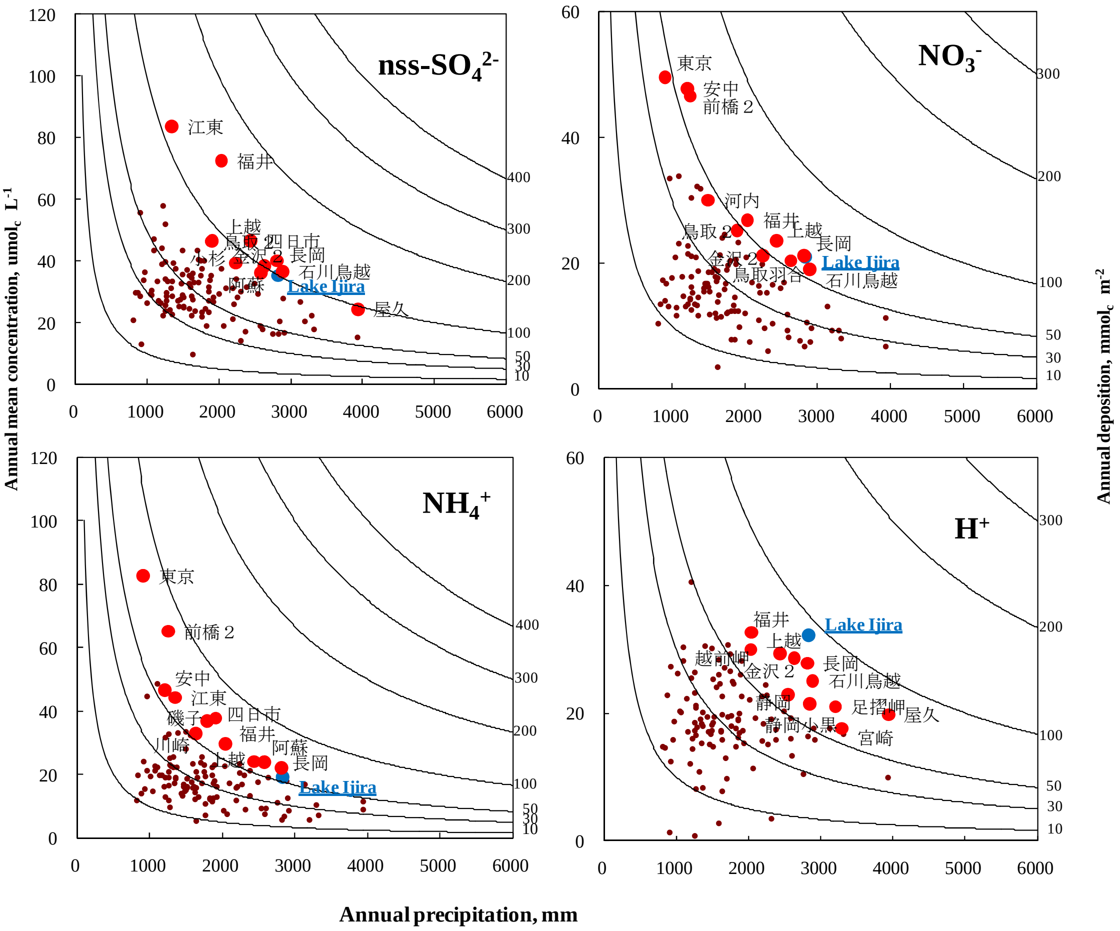

Fig. 3-3-3-6 Relationship between precipitation, concentration, and wet deposition of nss-SO_4^{2-}, NO_3^-, NH_4^+, and H^+ at the monitoring sites by JELA (1999-2004) and MOEJ (1998-2004)

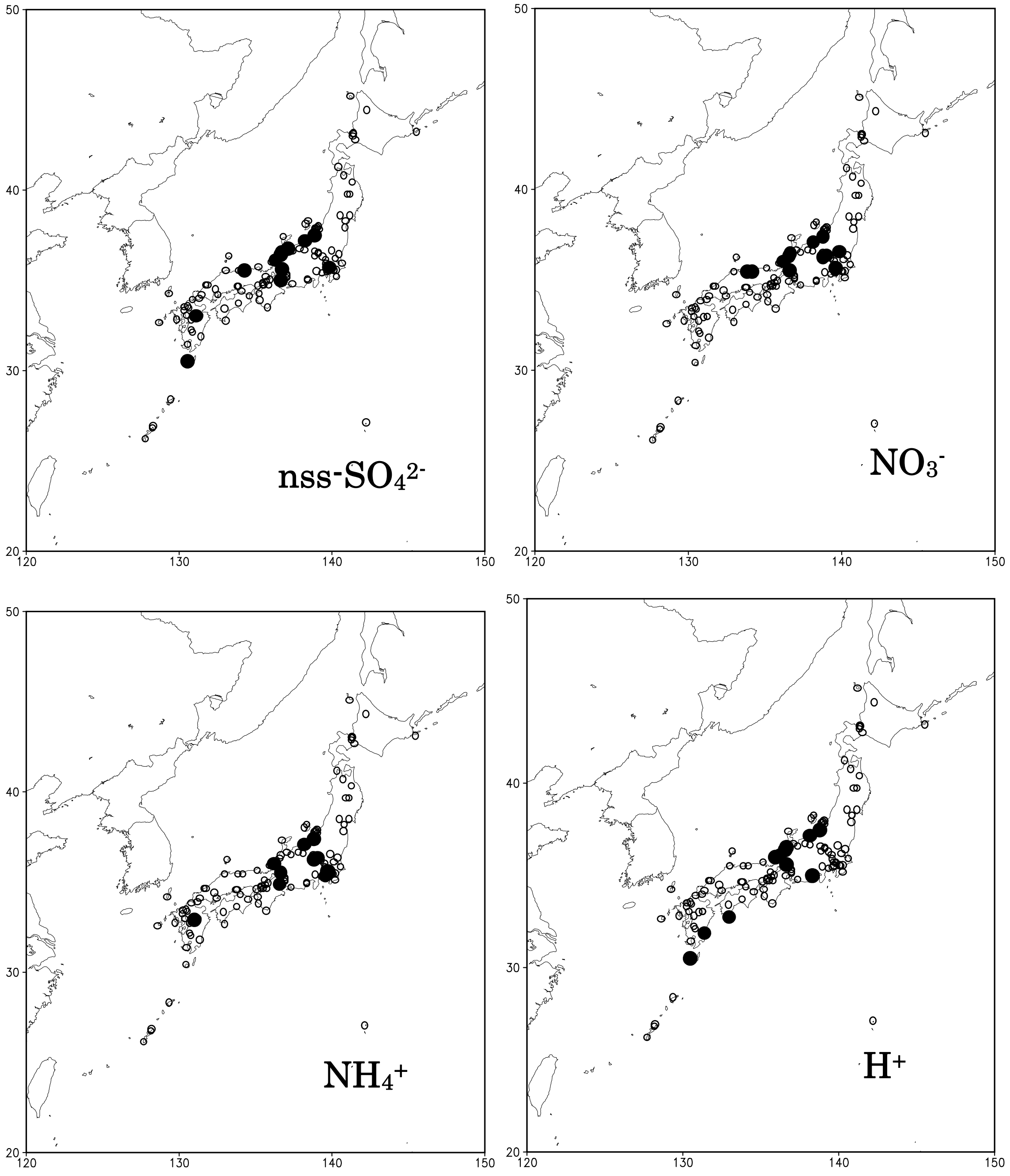

Fig. 3-3-3-7 Spatial distribution of wet deposition of nss-SO_4^{2-}, NO_3^-, NH_4^+, and H^+ (based on Fig. 3-3-3-6) Large black circle shows the sites within 10% highest among the sites.

2) Acidification trends in rivers of Chubu region

Thirty four points in Niigata Prefecture and 40 points in Gifu Prefecture, whose BOD was lower than 1.0 mg L^{-1}, were selected from the total 140 points and 120 points respectively for the trend analysis (Fig. 3-3-3-8).

The temporal trend analysis of the river water pH was carried out for the data from the mid 1980s to 2003 in the points above using the Seasonal Mann-Kendall method. For the points with the declining trend of pH, ion concentrations and alkalinity were analyzed by field sampling and literature study on land-use was carried out. Moreover, effects of hot spring were investigated by sampling along the river flow in the rivers having suspicious for hot spring contamination.

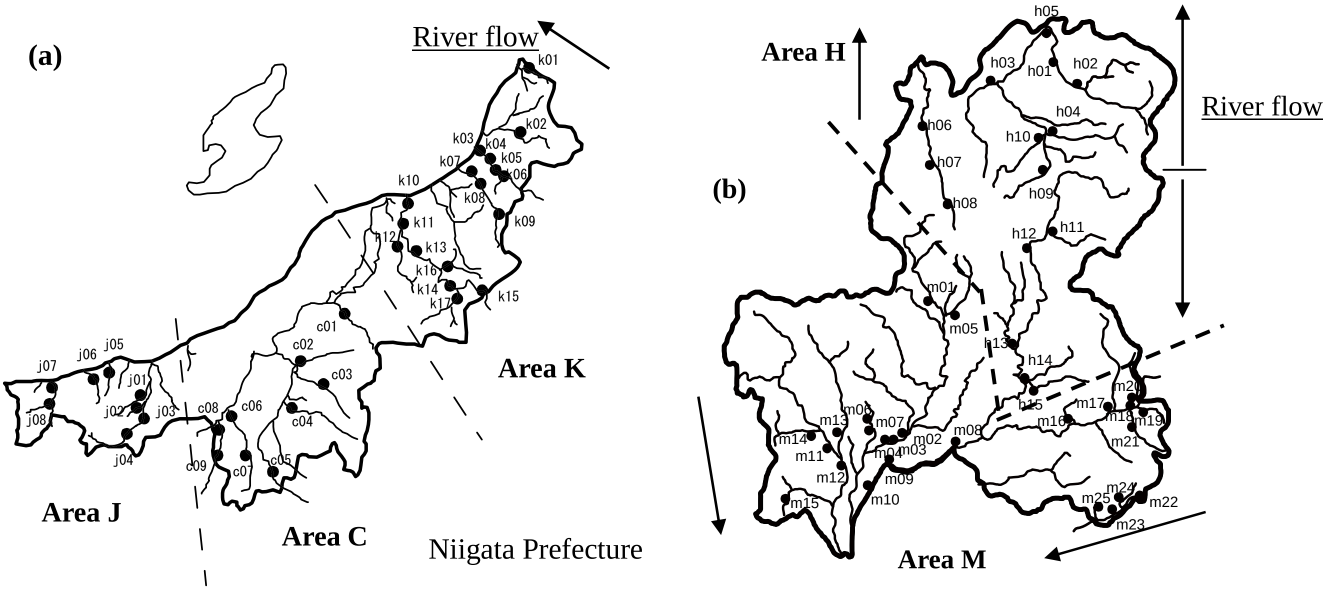

Fig. 3-3-3-8 Points for the trend analysis of river water pH in Niigata Prefecture (a) and Gifu Prefecture (b)

The points, whose BOD was lower than 1.0 mg L^{-1}, were selected as the points with the minimum local contamination. Arrows show direction of the river flow.

The long-term acidification trend of the river water pH was found by the temporal trend analysis at 11 points in Niigata Prefecture and 13 points in Gifu Prefecture as shown in Table 3-3-3-2. In particular in Niigata Prefecture, all the points with the declining pH trend are located especially in the northern part, area K (Ka-etsu). In Gifu Prefecture, locality of the trend was not obvious and the declining trend was found mostly in the upstream rivers.

Table 3-3-3-2 The long-term trend of the river water pH in Niigata Prefecture (a) and Gifu Prefecture (b)

(a)

Area	River system	River name	Points	n	Zscore
K	Ou River	Omata	k01	213	-2.45*
	Miomote River	Miomote	k02	213	-3.06**
	Ara River	Aara	k03	211	-4.92***
			k04	215	-4.71***
			k05	213	-7.34***
			k06	215	-8.22***
	Tinai River	Tainai	k07	205	-1.25
			k08	211	-2.27*
			k09	215	-1.42
	Agano River	Agano	k10	213	-0.28
			k11	213	-0.95
			k12	214	-1.12
			k13	215	0.24
			k14	213	-3.53***
			k15	213	-3.58***
		Araya	k16	214	-3.31***
		Tokonami	k17	212	-3.78***
C	Shinano	Ikarashi	c01	214	-1.29
		Hama	c02	213	0.13
		Sanashi	c03	213	-0.64
		Mikuni	c04	196	1.03
		Uozu	c05	212	0.11
		Kisyosu	c06	215	2.65**
			c07	212	0.66
		Nakatsu	c08	214	7.50***
			c09	209	3.54***
J	Ski River	Yashiro	j01	212	0.58
		Sibue	j02	215	2.90**
		Seki	j03	215	1.49
			j04	214	5.50***
	No River	No	j05	215	1.08
	Haya River	Haya	j06	213	-0.95
	Hime River	Hime	j07	212	0.65
			j08	214	0.38

*＝p<0.05 **＝p<0.01 ***＝p<0.001

(b)

Area		River system	River name	Point	n	Zscore
H		Takahara River	Takahara	h01	191	-3.00**
				h02	191	-2.99**
		Miya River	Odori	h03	96	3.24**
			Kohachiga	h04	191	-3.08**
			Miya	h05	191	-1.70
		Sho River	Sho	h06	96	-2.87**
				h07	64	-1.55
				h08	64	-1.61
		Miya River	Miya	h09	191	-6.65***
			Kawakami	h10	96	1.55
		Kiso River	Osaka	h11	96	-0.78
			Hida	h12	192	-1.68
			Maze	h13	186	0.01
			Shira	h14	96	5.76***
			Kuro	h15	96	5.15***
M	Western part	Nagara River	Nagara	m01	192	3.42***
				m02	192	-5.78***
				m03	192	-3.95***
				m04	192	-3.77***
			Yoshida	m05	192	6.00***
			Ijira	m06	192	8.53***
				m07	192	0.489
		Kiso River	Kiso	m08	192	-6.28***
				m09	192	-0.79
				m10	192	-1.48
		Ibi River	Ibi	m11	192	-5.03***
				m12	192	-2.63**
			Neo	m13	192	-2.62**
			Kasu	m14	188	1.27
			Makita	m15	192	2.37*
	Eastern part	Kiso River	Nakanokata	m16	96	3.11**
			Tukechi	m17	96	1.35
			Kawaue	m18	96	1.39
			Ochiai	m19	96	2.16*
			Kiso	m20	192	0.89
			Nakatsu	m21	96	2.98**
		Yahagi River	Yahagi	m22	191	3.33***
				m23	192	-4.28***
			Kamimura	m24	180	2.86**
			Azuma	m25	96	3.07**

*＝p<0.05 **＝p<0.01 ***＝p<0.001

Note: n, number of the data; Z-score shows the trend and its strength, the positive value shows increasing trend and the negative value shows declining trend; Hatched cells show significant declining trends.

According to the field surveys in Niigata Prefecture, alkalinity and EC were lower than 0.200 mmol$_c$ L^{-1} and 10 mS m^{-1} respectively, suggesting their high acid sensitive, in most points of the area K, whose upstream area was dominated by granite. On the other hand, alkalinity and EC were higher, suggesting low acid sensitive, in area J and area C (Jo-etsu and Chu-etsu), whose upstream area was dominated by andesite or limestone. The long-term trend of the river water pH may reflect the bedrock geology. In Gifu Prefecture also, acid-sensitive geology, such as granite, rhyolite and chert, can be found. Although locality of the pH trend relationship between the geology and the river water chemistry were not obvious, the long-term declining trend of pH was observed in the upstream points, such as h02, h09, and m11.

The river water chemistry may have large effects of land use in the watershed. Moreover, in case of Japan, there are many volcanoes, and hot springs or mineral springs may exist in upstream area of mountains. The investigation along the river flow near hot springs found clear effects of hot spring on the river water chemistry in c06, c07, c08 and c09 of Niigata Prefecture and h02 of Gifu Prefecture, in which EC and SO_4^{2-} concentration increased significantly near hot springs. On the other hand, in k01, k06, k16 and k17 of Niigata Prefecture and h09 and m11 of Gifu Prefecture, it was suggested that effects of hot spring drainage and mines were minimum. Moreover, 90% of the watersheds were occupied by forest and the percentage of agricultural field was significantly low in these points.

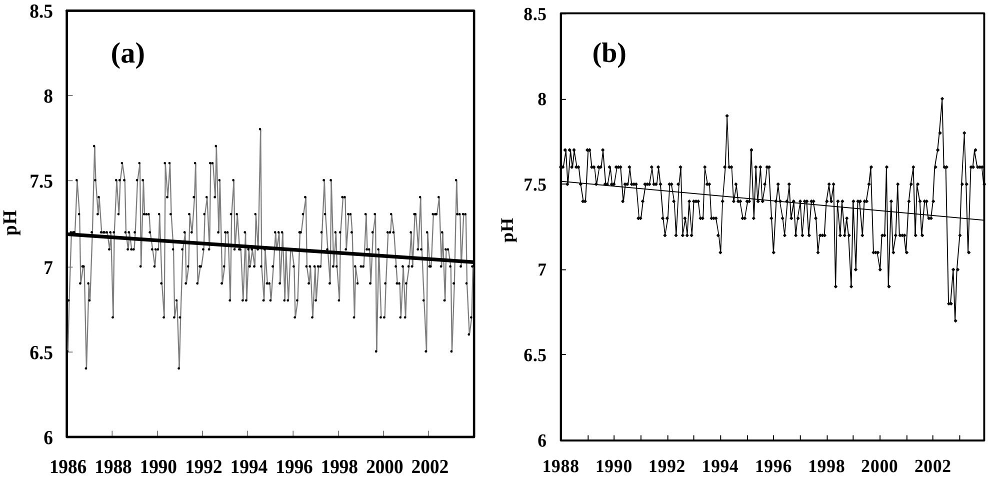

Fig. 3-3-3-9 Typical declining trends of pH in a) k16, Kotobukibashi Bridge in Araya River, Niigata Prefecture and in b) m11, Okajima Bridge in Ibi River, Gifu Prefectrure

The typical declining trends of pH in the points with minimal effects of land use and hot spring were shown in Fig. 3-3-3-9. Most points of Niigata Prefecture shows relatively monotonous declining trend as seen in k16 of Fig. 3-3-3-9 a). In case of Gifu Prefecture, as seen in m11 of Fig. 3-3-3-9 b), the pH declined clearly after 1996, which was similar to the phenomenon observed in the Kamagatani River flowing into the Lake Ijira. However, slight increase can be seen after 2000, which was different from the Kamagatani River. The upstream area of Araya River (k16) was dominated by granite, and the area of Ibi River was dominated by sedimentary rocks including chert and granite. As the seasonal variation, the lowest pH was recorded in Niigata and Gifu prefectures in April or May, snow-melting season of high mountainous area,, which may reflect so-called acid shock phenomena.

As shown in the spatial distribution of wet deposition in Japan, the area along the Sea of Japan represented by Niigata Prefecture and Chubu region represented by Gifu Prefecture can be characterized as the area with high wet deposition load. The trend analysis above was not a direct evidence of acid deposition impacts on the river water chemistry. However, the natural environmental factors such as hot spring and volcano and the anthropogenic factors such as land-use change, agriculture and mining cannot explain adequately the long-term declining trend of pH in several points of Niigata and Gifu prefectures. It may be possible that similar phenomena occur in other acid-sensitive areas. Acidification of streams or rivers should be further focused on. Moreover, similarity between the long-term declining trends in Gifu Prefecture and the trend in the Lake Ijira should be investigated.

3.3.4 Current status of watershed acidification in Japan and future directions of the catchment analysis

Synthesis analysis based on the previous monitoring activities and the intensive surveys suggested that atmospheric deposition of S, N, and H^+, which is the highest level among Japan, affected the river water chemistry through biogeochemical cycles in forest ecosystems in the Lake Ijira (Kamagatani) catchment.

Based on precise estimates of wet deposition and dry deposition, N deposition in the catchment was significantly higher than the threshold value, , at which NO_3^- leaching into streams or rivers would be accelerated in Europe and U.S. In fact, NO_3^- concentration in the river showed the increasing trend and no seasonality with relatively high concentrations even in summer, when vegetation N uptake must be active. The river pH showed declining trend. It was suggested that significant high N deposition contributed to the river acidification. Although the trigger of the phenomena may be meteorological events in the mid-1990s, the catchment is continuously acidified by NO_3^- leaching derived from high N deposition, which may exceed the capacity of the ecosystems.

On the other hand, S output exceeded the input in the catchment. It was suggested that S accumulated in the past was discharged from the catchment, possibly contributing to the acidification above. Since concentrations of NO_3^- と SO_4^{2-} increased simultaneously with increase of the water discharge during heavy rain events, it was suggested that the accumulated S was discharged with N from shallow soil layers.

The Lake Ijira catchment is a typical forested catchment in Japan, where Japanese cedar and cypress were planted, and the similar acidic rocks are distributed in many areas. The acidification of watersheds may not be a specific phenomenon in this area. The acid-sensitive catchment, high deposition load, and meteorological events as a trigger functioned as the necessary conditions for the acidification. The spatial distribution of wet deposition indicated existence of the areas with high deposition load comparable to the Lake Ijira. In particular, in the area along the Sea of Japan in Chubu region, the wet deposition amounts are recently the highest level in Japan. Moreover, the long-term declining trends of the river water pH in Niigata and Gifu prefectures indicated possible acidification of watersheds in the areas with acid-sensitive geology and high acid deposition.

As described in the section 3.2, taking these phenomena into consideration, the area with acid sensitive ecosystems and high acid deposition should be identified as hot spots, and possible effects of acid deposition on the river water chemistry should be investigated in further detail. Moreover, since soil pH decreased to lower than pH 4.0 in the Lake Ijira catchment, leaching of Al^{3+}, which is toxic to plant growth, may increase accordingly. Continuous monitoring including effects on trees may be necessary.

As the reasons why the acidification trends could be identified in the Lake Ijira catchment, in addition that the area had necessary conditions as a hot spot, it can be pointed out that an integrated monitoring including atmospheric deposition, soil, vegetation, and inland water has been implemented here. The catchment-scale analysis of biogeochemical material cycles may allow more quantitative discussion on effects of acid deposition on forest ecosystems. For further development of the catchment-scale analysis as the integrated monitoring in future, the following subjects should be discussed.

1) Strengthening of the stream/river monitoring and promotion of catchment analysis

   The monitoring on streams and rivers should be more focused on, which may reflect changes in material cycles more directly. Taking hot spots into account, widespread information should be accumulated. As for the Lake Ijira catchment, monitoring in the Kamagatani River will be carried out continuously at two-week interval, and the long-term assessment of the river water chemistry and input-output budget will be carried out. In future, more effective monitoring methodologies should be discussed with development of the catchment-scale model.

2) Development of effective methods to estimate forest growth and nutrient uptake

   Tree growth and nutrient uptake may be closely related to acidification of the catchment as discussed above. To evaluate acidification derived from external H^+ sources such as acid deposition quantitatively, it is important to estimate plant growth and nutrient uptake in the whole forest area. The precise estimation of forest growth requires detailed and continuous field observations. In future, more effective and simplified estimation methodologies should be developed for the catchment analysis.

3) Collaboration with existing study sites

   Research outcomes in existing study sites managed by universities and research organizations should be referred for further collaboration between the Long-term Acid Deposition Surveys and the research activities. Cooperative intensive observations or standardization of the monitoring methodologies should be promoted. In future, synthesis assessment of Japanese watersheds including the Lake Ijira catchment should be discussed. Through the assessment, the position of the catchment analysis should be clarified in the Long-term Acid Deposition Surveys.

4) Dissemination of the catchment analysis to the East Asian region

   The catchment analysis will be an effective tool for assessment of ecological impacts in the East Asian region, where emission of acid substances is being increased. Studies of the catchment analysis should be promoted in the region.

5) Development of the catchment-scale model

   Quantitative data obtained through the intensive surveys, the data in existing study sites in Japan, and case studies in the East Asian region should be integrated for development of the catchment-scale model including comparison of existing models. For validation of the acidification mechanism in the Lake Ijira catchment, modeling may be essential. The data obtained by 1) and 2) above will be informative as validation data of the model.

Taking the subjects into account, the catchment-scale analysis should be promoted as the integrated monitoring and assessment methodologies for impacts of acid deposition on ecosystems.

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3.4 Comparison with foreign monitoring data

EANET was established in 1998 and started its regular phase of operation in 2001. The monitoring results were reported every year as the data report. In 2007, “Periodic Report in the State of Acid deposition in East Asia”1) was published, which described the outcomes of five-years' EANET's monitoring activities(2000-2004), and was the first report with comprehensive analysis and assessment of acid deposition in East Asia.

The European continent has long been covered by the Co-operative Programme for Monitoring and Evaluation of Long-range Transmission of Air Pollutants in Europe (EMEP), and the monitoring in North American region has been also continuously conducted by the National Atmospheric Deposition Program (NADP) etc.

In this section, the state of acid deposition in Japan was compared with those in East Asia, Europe and North America using these monitoring data above mentioned.

3.4.1 Atmospheric deposition

(1) State of wet deposition

The state of wet deposition in Japan, East Asia, North America and Europe was analyzed. The data in non-urban sites of this survey in Japan and the data in non-urban sites of Acid Deposition Survey conducted by Japan Environmental Laboratories Association2)~5) were used for comparison with EANET6), NADP7), and EMEP8) data, which represent the data in East Asia, North America, and Europe respectively. The data that satisfy the completeness (%TP and %PCL) above 80% during the period from April 2003 to March 2007 for Japan and from January 2003 to December 2007 for other regions were used for the calculation of annual means.

Figure 3-4-4-1 shows the relationships between ion concentrations and precipitation amounts of 10, 25, 50, 75, and 90 percentiles on deposition flux in Japan, North America, and Europe. The range of precipitation amounts in Japan were larger than those in North America and Europe, and the median value in Japan was 1680mm y^{-1}, which was 1.7 times larger than in North America and 2.5 times than in Europe. nss-SO_4^{2-} concentrations in Japan were distributed in narrow range with high concentrations compared with Europe and North America, and the 10 percentile value was the same concentration of the median value in Europe and North America. The median concentration in Japan was 1.6 times higher than in North America and 1.3 times in Europe. Median value of deposition amounts of nss-SO_4^{2-} in Japan exceeded 20 mmol m^{-2}, which was three times larger than in Europe and North America. The range of NO_3^- concentrations in Japan and North America were within those in Europe, and the median value in Europe was the highest among the three regions and that of Japan was about 70% of Europe. However, because of large precipitation amounts, deposition amounts of NO_3^- in Japan was about two times larger than those in Europe and North America.

Acidity of rain water in Japan were distributed in narrower range than in Europe and North

America. The range of pH values in Japan were the latter half of 4. However pH values in Europe and North America were distributed between middle of 4 and middle of 5, which were wider range than in Japan. The median pH value was 4.7 in Japan, 4.9 in North America and 5.0 in Europe, which mean the concentration of H^+ in Japan were 1.7 times and 2.2 times higher than in North America and Europe, respectively. Therefore the median value of H^+ deposition amounts in Japan was 6 times and 3 times larger than in North America and Europe, which could be explained by considering the difference of the concentrations of acidity and precipitation amounts. Furthermore, the difference between Japan and North America, Europe could be considered the difference of neutralization in rain water between them. It was suggested that the ratio of the neutralization of rain water in Japan was lower than that in North America and Europe.

Figure 3-4-1-2 shows the same relationships by adding the data of monitoring sites in East Asia. There were considerable variations of value between each site, and so mean concentration and mean precipitation amount were used for the plot in this figure with no statistical procedures. High concentrations of nss-SO_4^{2-} were observed in China with highest concentration more than 150μmol L^{-1}, which was three times higher than the median value in Japan. Precipitation amounts in China were relatively low, however deposition amounts were six times larger than the median value in Japan due to high concentrations in China.

Relatively large deposition amounts, which were upper range of deposition amounts in Japan, were also observed in Korea, Indonesia, Viet Nam. NO_3^- deposition amounts in China, Korea and Indonesia were over the range of those in Japan. These areas were relatively low precipitation amounts, however because of high concentration in China and Indonesia the deposition amounts were 2 times larger than the median value in Japan.

In spite of high deposition amounts of acidic substances in East Asia, the level of acidity in rain water and H^+ deposition amounts were below the range of those in Japan. It was suggested that the ratio of the neutralization of rain water in East Asia region was higher than that in Japan.

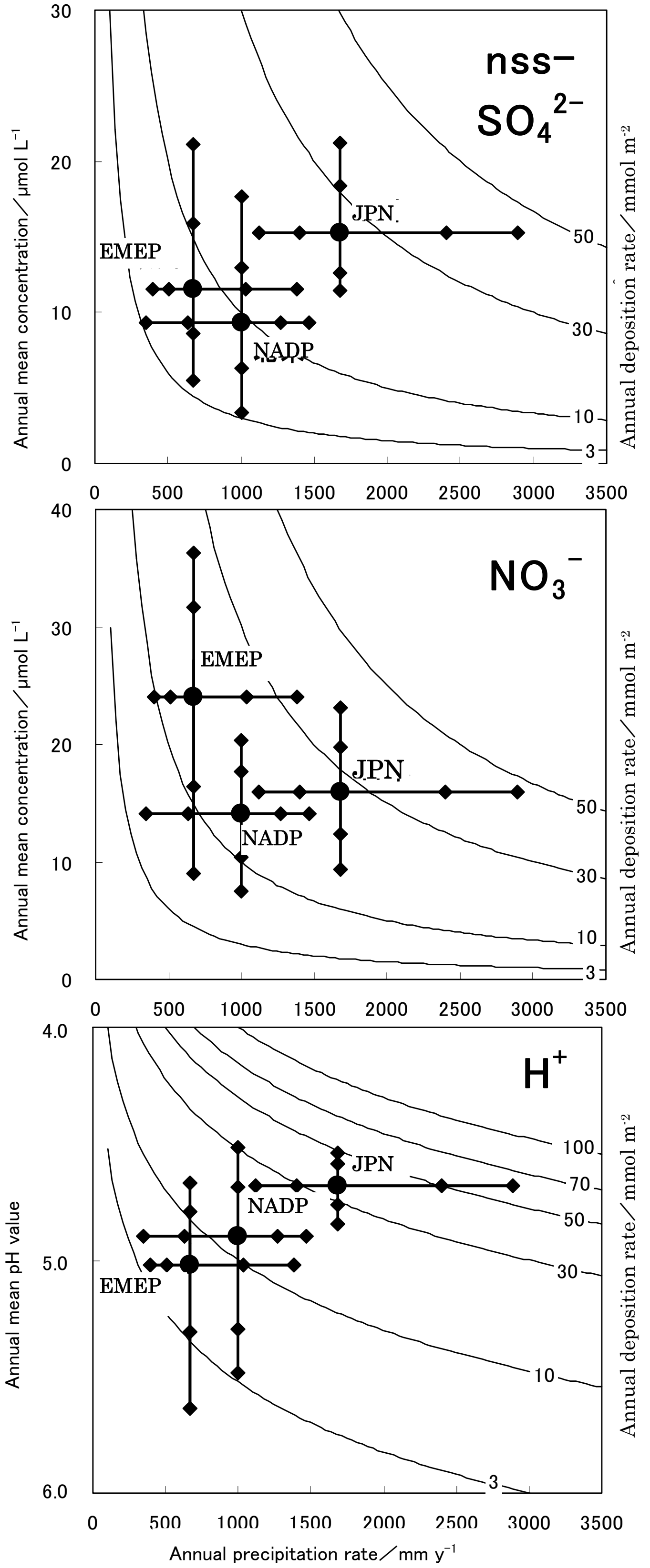


Fig.3-4-1-1 Wet deposition amounts in Japan and comparison with the data in Europe and North America(2003-2006)

1) Japan: JPN, n=68, North America: NADP, n=250, Europe: EMEP, n=97

2) Black points Dot point shows 10, 25, 50, 75, and 90 percentiles of ion concentration and precipitation amounts in Japan, North America, and Europe.

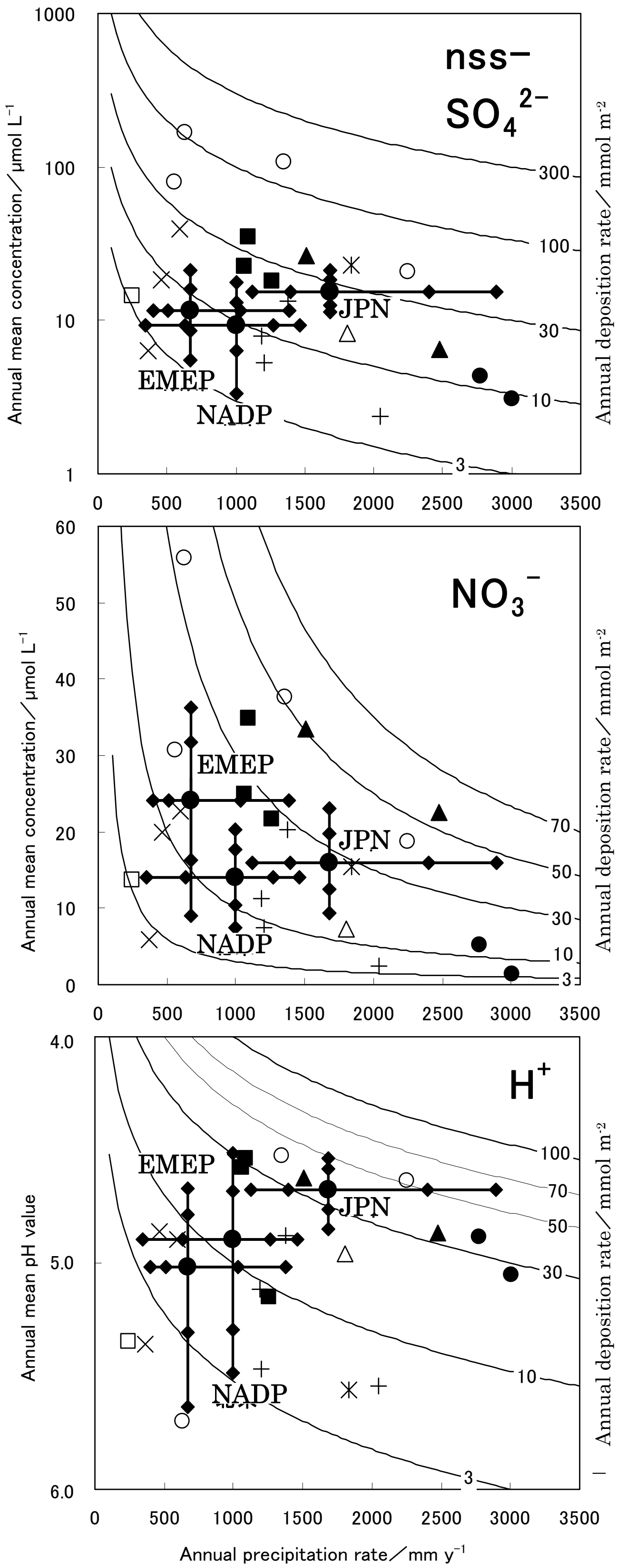


Fig.3-4-1-2　Wet deposition amounts in East Asia and comparison with those in Japan, Europe and North America(2003-2006)

1)　Japan:JPN, n=68, North America:NADP, n=250, Europe:EMEP, n=97

2)　East Asoa: ○:China, ▲:Indonesia, ●:Malaysia, □:Mongolia, △:Philippines, ■:Korea、 ×:Russia, ＋:Thailand, ＊:Viet Nam

(2) State of dry deposition

The concentrations of ionic compounds in gas and aerosols were compared with those in Japan, Europe and North America. Figure 3-4-1-3 shows the mean concentrations and 25, 50 and 75 percentiles and the minimum and maximum concentrations in JFY 2006(Japan) and 2006(Europe and North America). Mean concentration of ionic compounds in Japan was almost same levels with those in Europe and North America, however the ranges were narrow. Especially, not only ranges also mean values of O_3 concentrations were almost same levels with three regions.

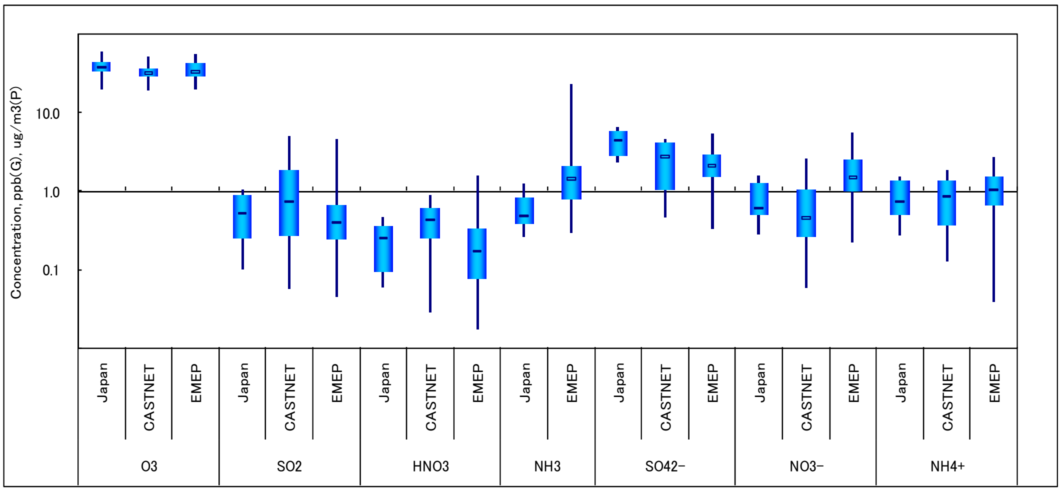


Fig.3-4-1-3 Comparison of ionic compounds in gas and aerosols in Japan, Europe and North America

Figure 3-4-1-4 shows the 5 years' mean concentrations of SO_2 from 2003 through 2007 in each EANET sites. The range of SO_2 concentrations were from 0.2 ppb to 15.8 ppb. Especially SO_2 concentrations in China were higher than other sites, and the concentrations of almost all the sites except Japan were over 1 ppb. However, the SO_2 concentrations at almost sites in Japan were below 1ppb, and SO_2 concentrations in Japan were low levels compared with other East Asia regions.

Figure 3-4-1-5 shows the mean concentrations (2003-2007) of S and N contained ionic compounds in gas and aerosol in remote area and urban area, respectively. The data of Japan is 5years' mean concentrations from 2003 to 2007, Urban site's data was Tokyo. It was clear that the concentration in urban sites were higher than that in remote sites.

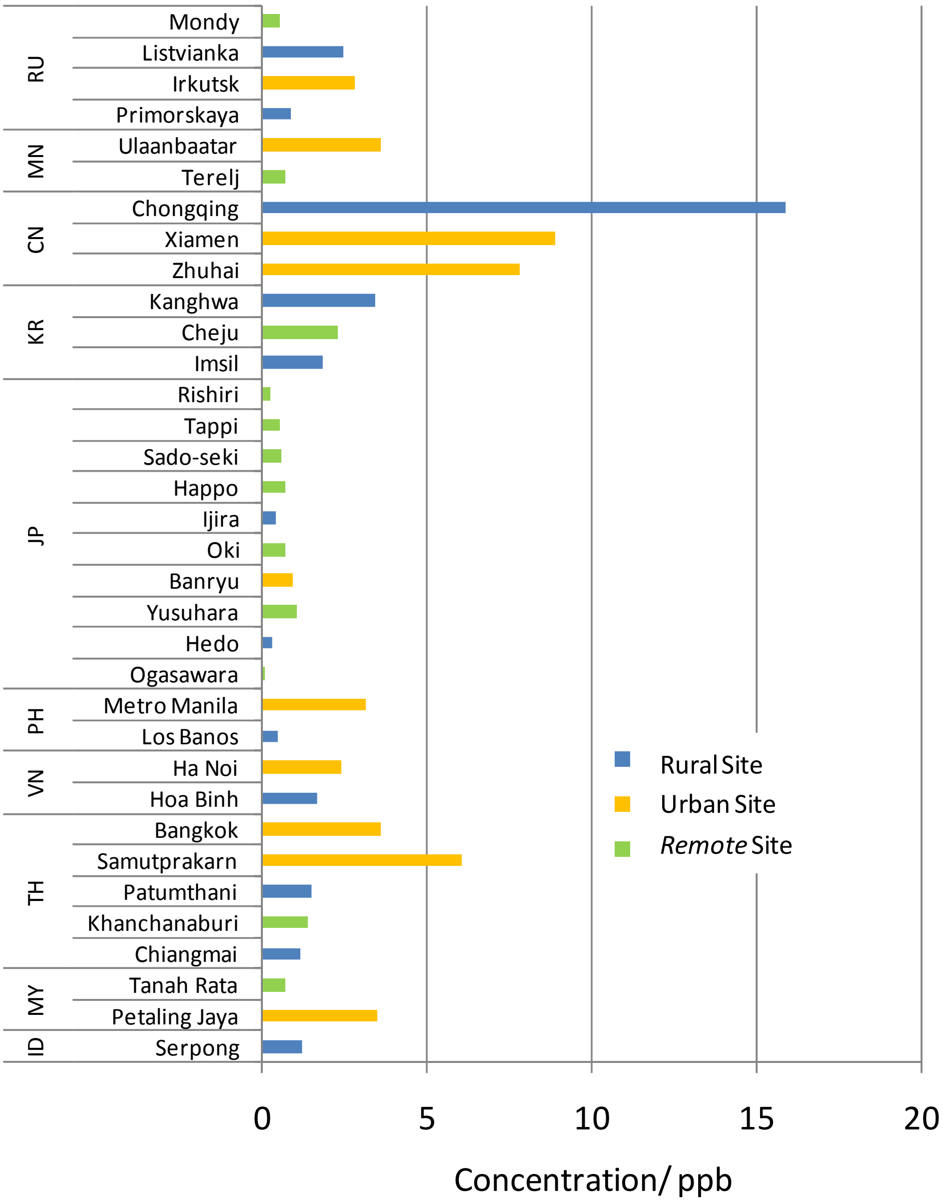
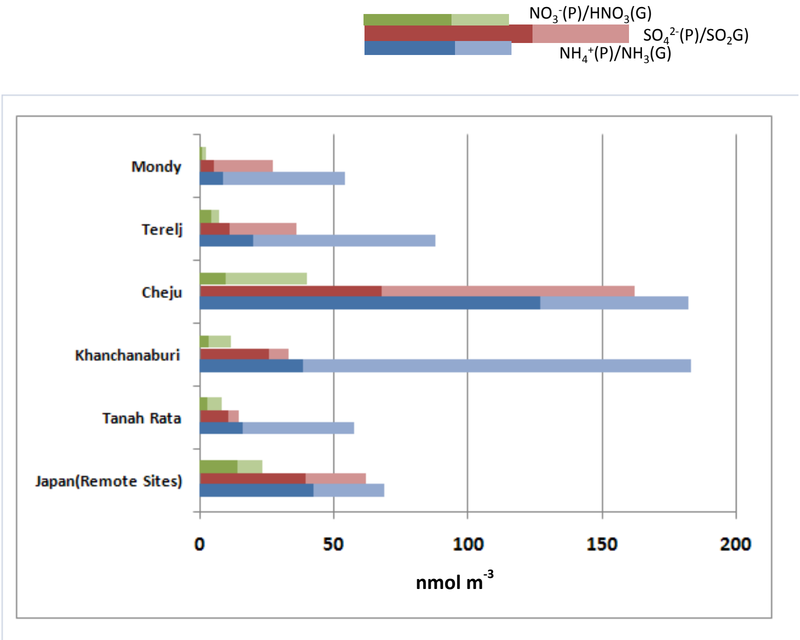


図3-4-1-4　SO2 concentrations in EANET sites (2003-2007)

Monitoring were carried out by automatic monitors and filter-pack method

(i) Remote Sites



(ii) Urban Sites

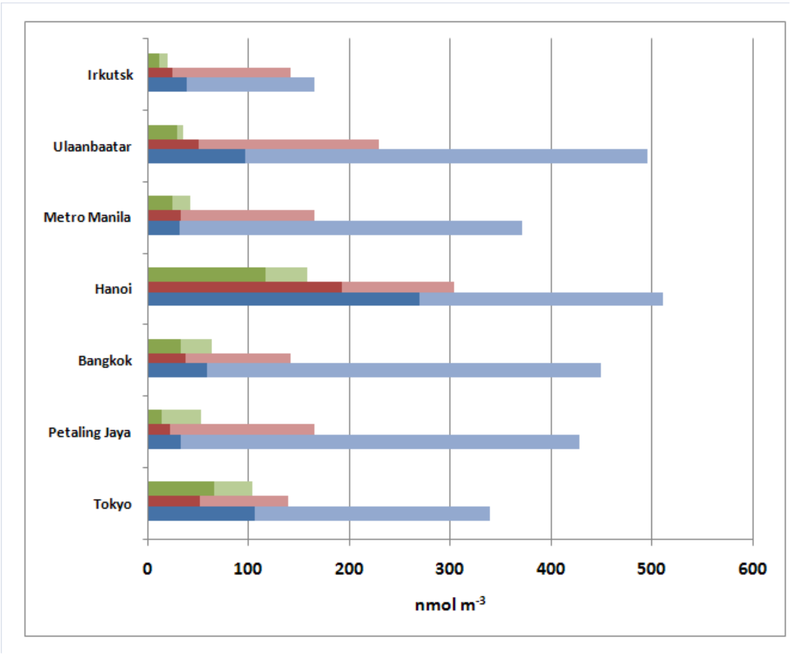


図 3-4-1-5　Air concentrations of S and N contained compounds in gas and aerosols in Japan and East Asia (Unit: nmol m^{-3})

3.4.2 Impacts on ecosystems

(1) Soil acidification

Soil has acid buffering capacity and acidification proceeds gradually with several steps. Therefore, short-term monitoring or temporary surveys mostly cannot detect small changes in soil chemical properties.

In EANET, the soil surveys have been done only once or twice in most sites, and it is impossible to evaluate soil acidification status only by the EANET data. However, in case of Lake Ijira catchment, one of the EANET monitoring sites, soil surveys have been conducted continuously by the Japanese Acid Deposition Surveys since 1988, and a soil acidification process was able to be detected there. At present, the soil acidification in Lake Ijira catchment is only one case, which may be attributed to acid deposition.

In Europe, the International Co-operative Programme on Assessment and Monitoring of Air Pollution Effects on Forests (ICP Forests) under the UNECE Convention on Long-range Transboundary Air Pollution started large-scale soil surveys at 5,300 plots since 1994. The second surveys are now being conducted in the respective countries. Therefore, chronological changes in soil chemical properties in European scale will be reported in near future. However, in Europe, soil acidification has been reported in several forest areas. A case in Southeast Sweden may be the most famous as a long-term acidification of soil. In this case, the area, which was originally surveyed in 1927, was surveyed again from 1982 to 1984, after 55 years from the first survey, and then, the soil acidification phenomena were obviously detected there.

(2) Tree decline

The current situation on tree decline symptoms in the EANET monitoring sites was shown in Fig. 3-4-2-1. Tree decline symptoms were reported in most monitoring sites. Especially in Dabagou of China, and Bolshie Koty and Irkutsk of Russia, all the trees showed some decline symptoms. Possible causes of the tree decline were presumed to be disease or insects in most sites and meteorological or topographical conditions in some sites. However, the cause was also not identified clearly in many sites. In Irkutsk, where the cause was not identified, local scientists suggested that effects of air pollution derived from industrial area around Baikal Lake.

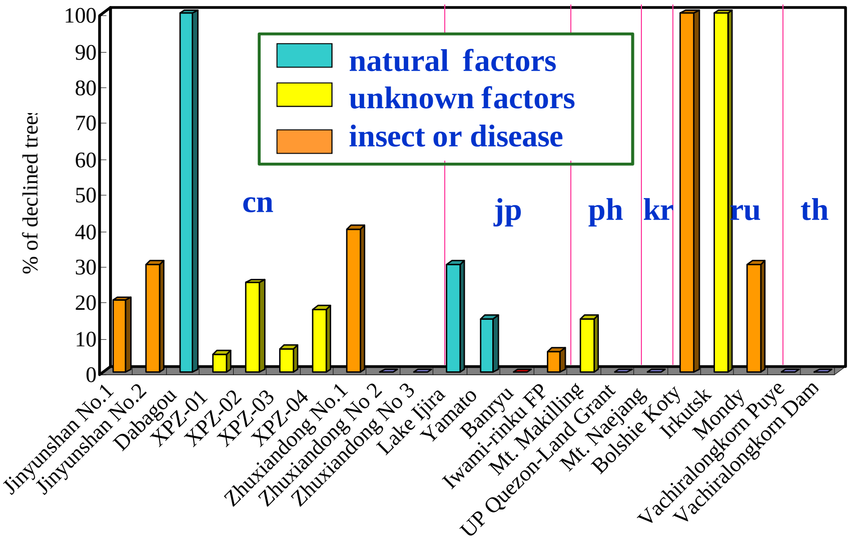


Fig. 3-4-2-1 Tree decline symptoms observed in the EANET sites

Based on the observation from 2000 to 2004; number of trees: 20 (at least 12); cn, China; jp, Japan; ph, Philioppines; kr, Republic of Korea; ru, Russia, th, Thailand (EANET, 2006)

In addition to the monitoring sites above, decline of larch (*Larix sibirica*) trees was reported in Bogdkhan Mountain near Ulaanbaatar City, the capital of Mongolia (Fig. 3-4-2-2). Effects of SO2 and other acidic pollutants derived from coal combustion in thermal power plants or mobile houses (Ger) were suggested as one of possible cause of the tree decline. Since the city of Ulaanbaatar is located in a basin surrounded by mountains, air pollutants will be still in the basin especially in winter. Airborne particulate matters could be detected visually in the city area and the slopes of Bogdkhan Mountain were smoggy in winter. Sulfur concentrations of larch needles were twice in the

Fig. 3-4-2-2 Decline of larch trees in Mt. Bogdkhan near Ulaanbaatar City

The photo was taken on the slope facing the Thermal Power Plant NO.3.

slope facing a thermal power plant than in reference forests (approx. 2mg-S g^{-1}).

As described above, tree declines reported in the EANET countries were mainly caused by relatively local air pollution. In many cases of Japan also, the tree decline symptoms were reported around big cities, such as decline of *Cryptomeria japonica* in Kanto Plain (mainly in Saitama next Tokyo) and tree decline in Nikko or Akagi (Tochigi or Gunma, downwind direction of Tokyo). So far, effects of transboundary air pollution on tree decline have not been reported.

On the other hand, recently, ozone concentrations have increased gradually, and the Photochemical Oxidants Warning has been issued in wider areas. As one of the possible causes of the high ozone concentrations, transboundary air pollution from the Asian Continent is pointed out. The annual mean ozone concentrations were higher than 60 ppb in higher altitudes, such as Akagi and Happo-One, and higher than 40 ppb in islands or coastal area along the Sea of Japan, such as Tappi Point, Sado-Seki Point, Oki Island, and Tsushima Island (see the section 3.1.2). In other East Asian countries, high ozone concentrations have been reported. The mean ozone concentrations of wet and dry seasons in a tropical seasonal forest in Thailand were shown in Table 3-4-2-1. The values shown here were 15-day mean values obtained by the Ogawa passive sampler. The maximum concentration was higher than 60 ppb and the mean concentration in dry seasons were higher than 40 ppb. Accumulated exposure over a threshold of 40 ppb (AOT40) is used as an indicator of plant growth in Europe. High ozone concentrations in forest area and their effects on plant should be monitored in detail.

Table 3-4-2-1 Ozone concentrations in a tropical seasonal forest, in Sakaerat Silvicultural Research Station, Thailand

Year	Season	Mean	Median	Minimum	Maximum
2005-2006	Dry (Dec - Mar)	43.0	40.6	29.7	64.2
2006	Wet (Apr - Nov)	29.2	25.6	19.8	66.7
2006-2007	Dry (Dec - Mar)	45.5	43.0	34.0	58.1

Note. The ozone concentrations above show the 15-day mean values obtained by the Ogawa passive sampler.

In Europe, ICP Forests started the surveys on ozone effects, including the measurement of ozone concentrations in forest area by passive samplers and observation of ozone visible injury. According to test phase of the passive sampling, the mean concentrations were higher than 45 ppb in many Mediterranean countries such as Italy and Spain.

(3) Inland water acidification

In EANET, acidification trends of inland water have not been reported. Acid-sensitive lakes, whose alkalinity is lower than 0.200 $\mathrm{mmol_c\ L^{-1}}$ and EC is lower than 10mS $\mathrm{m^{-1}}$, were found in China, Japan, and Russia. However, most monitoring sites in Southeast Asian countries did not meet the criteria above. It is difficult to find appropriate lakes in East Asian countries and acid buffering mechanisms of lakes are quite complicated. Rivers, which may directly reflect material cycles in the catchments, will be selected as the monitoring site in near future in EANET.

In Europe, the International Cooperative Programme on Assessment and Monitoring of Rivers and Lakes (ICP Waters) has promoted wide-range monitoring of inland waters. As the emission was reduced in Europe, SO_4^{2-} concentrations in rivers and lakes have been reduced year by year, and the reduction rate was the largest in 1990s. While SO_4^{2-} leaching was regulated by S deposition, NO_3^- concentrations in the inland waters did not show any clear trends and it was suggested that leaching processes of N may be more complicated. Alkalinity did not change at two-third of the sites but increased at one-third of the sites. From 1990 to 2001, acidification still proceeded at 14% of the sites. However, it was reported that acidification may not proceed in Europe from now on. Thus, inland waters in Europe may be in a process of recovery from acidification.

On the other hand, in Japan, the Synthesis Report of Japan Acid Deposition Surveys reported acidification trends in rivers and lakes in mountainous areas with low acid buffering capacity and acid shock phenomena in snow-melting seasons. Moreover, in this report, not only in Lake Ijira, but also in widespread area of Niigata Prefecture and Gifu Prefecture, acidification of rivers was reported. Therefore, in East Asia including Japan, effects of acid deposition on inland waters should be monitored continuously.

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4. Long-Range Trans-boundary Air Pollution

The term "Trans-boundary Air Pollution" can be traced back to the 1960s, when acidification phenomenon became serious environmental issues in Europe. When the interrelationship between sulfur emissions in continental Europe and the acidification in Scandinavian lakes were recognized as serious environmental problems, it was pointed out by Swedish meteorologist that air pollutants including acidic substances could be transported over long-range from Europe continent and UK1). After that, in accordance with worse condition on acidification phenomenon in Europe and boundary area between UAS and Canada, research on long-range transportation of acidic substances developed dynamically from 1970s to 1980s in Europe and North America2). During these decades, the Convention on Long-Range Trans-boundary Air Pollution (CLRTAP) was adopted in 1979 with the participation of European countries including USSR(Russia), USA and Canada. Since then, control measures to reduce emissions of sulfur and nitrogen oxides were enforced, and acidification phenomenon did not become so serious problems in 1990s.

Instead of acidification phenomenon, increasing of ozone concentrations was beginning to appear as environmental issues. In Europe and North America, counter measures for nitrogen oxide and VOC conducted properly, however ozone concentrations did not achieve the environmental standards levels. Possible causes of these results included the increasing of the back-ground ozone concentrations in northern hemisphere, and to achieve the environmental standards, it was necessary to address the ozone issue as a hemispheric issue and to discuss internationally. Taking above mentioned matter into account, the Task Force on Hemispheric Transport of Air Pollution(TF-HTAP) was established(ref. 4.4.1) under CLRTAP.

Meanwhile, in East Asia concern about acidification phenomenon arose after 1990s, and the Acid Deposition Monitoring Network in East Asia(EANET) was established. Same time, transportation of acidic substances in East Asia became a matter of concern, and the contribution of sulfur oxide in Japan from China and Korea etc. were examined and reported3). However, the results of the calculation varied widely and impact caused by acidification phenomena was considered as the issues of long-term risks.

Same as Europe, ozone issue became of interest lately in East Asia. In Japan, in spite of the decrease of the atmospheric concentration of precursor pollutants, VOC and NOx, long–term increasing trend of ozone concentrations has been shown. Then increasing ozone concentrations in China accompanied with rapid economic growth and the effect of trans-boundary air pollutants to Japan began to investigate. On the one hand, it became clear that hemispherical air pollution of ozone caused by the inter-continental transportation affected the increasing of back-ground ozone concentrations and related deeply with the difficulty of the achievement of environmental standard. Therefore, it is necessary to discuss the ozone control measures of Japan in future taking international framework into account, which includes the transportation issues in East Asia region and the inter-continental and hemispherical transport of air pollution.

4.2 The Increasing Emissions of Air Pollutants in East Asia

Using the emission inventory for Asia(Regional Emission inventory in Asia, REAS)$^{4)}$ emission trends between 1980 and 2003, and projections of future emissions of air pollutants(NOx, SO_2, NMVOC) in East Asia are presented here.

(i) Yearly emission changes of air pollutants

Figure 4-1 shows the yearly changes of regional emissions in the period 1980-2003. Also shown in table 4-1 are emission amounts in 1980, 1990 and 2003 in East Asia. Total emissions of NOx in Asia were 25.11 million tons in 2000 and China and India occupied 45% and 19% respectively. In China, the country with the largest emissions, coal-burning power plants were the largest emitters(34%), followed by industrial coal use and transport oil use(both at 25%). From 1980 to 2003, fuel consumption increased 2.3 times and NOx emissions also increased 2.8 times in Asia. However in China NOx emissions increased 3.8 times, yearly average 6%, during same period, and recently increased 3 times in 3 years after 2000, which were the highest record in China.

Total Asian emissions of SO_2 were 41.49 Mt in 2000, and 64% of them were discharged from coal combustion facilities in factories and power plants etc. From 1980 to 2003, SO_2 emissions increased 2.2 times in Asia, 2.1 times in East Asia, and 2.5 times in China. Then, emissions of NMVOC in Asia were 40.24 million tons, in which combustion facilities of factories and power plants occupied 38% and transportation, automobiles etc.,33%. From 1980 to 2003, emissions of NMVOC increased 2.1 times in Asia, 2.4 times in East Asia, and 2.5 times in China.

(ii) Future emission trends of air pollutants

Future projection of Asian emission was performed on the basis of emission scenarios and emissions for 2020 shown in table 4-2. Here, three emission scenarios for China have been developed taking into account of the trend of future energy consumption and environmental counter measures as follows; Policy Failure Case scenario(pessimistic scenario with high emission rates caused by continuation of the current energy structure, increased energy consumption, and the slow deployment of new energy technologies and new emission control technologies: PFC), Reference Case scenario (sustainable scenario with moderate emission rates caused by the suppression of energy consumption through energy conservation, a change to clean energy, and the moderate deployment of new energy technologies and new emission control technologies: REF), and Policy Succeed Case scenario (optimistic case with low emission rates owing to the implementation of strong energy and environmental policies and the fast deployment of new energy technologies and new emission control technologies: PSC). Each concept – PSC, REF, and PFC – resembled that of the B1 scenario, the B2 scenario, and the A2 scenario of IPCC, respectively5). For other countries other than China, fuel consumption was calculated on the basis of Reference Scenario Projections in the “World Energy Outlook”(IEA).

In China, NOx emissions under the REF scenario and PFC scenario were projected to increase by 39% and 128% respectively in 2020 compared with 2000 emissions, showing only a small decrease under the PSC scenario. However, considering the increasing trends of emission and fuel-consumptions from 2000, and recent satellite observations, present NOx emissions have accelerated impressively more than the predicted values of the PFC scenario in 2020(ref. figure 4-2a).

SO_2 emissions in Asian countries were projected to increase from 2000 to 2020. Emission in China showed 28% decrease in the PSC scenario, 3% decrease in the REF scenario, and 163% increase in the PFC scenario for 2020 compared 2000. 2020 NMVOC emissions in China would rapidly increase by 97%, 139%, 163% from 2000 in the PSC, REF and PFC scenario, respectively.

Table 4-1 Comparison of estimates emissions in East Asia, 1980-2003,
and increasing ratio from 1980 in parentheses.

Region	1980	1990	2003
NOx			
China	3.8	6.6 (1.7)	14.5 (3.8)
Other East Asia	3.6	3.8 (1.1)	4.7 (1.3)
East Asia Total	7.4	10.4 (1.4)	19.2 (2.6)
SO_2			
China	14.9	21.6 (1.4)	36.6 (2.5)
Other East Asia	3.9	3.8 (1.0)	2.4 (0.6)
East Asia Total	18.8	25.3 (1.3)	39.0 (2.1)
NMVOC			
China	6.8	9.7 (1.4)	17.2 (2.5)
Other East Asia	2.0	3.1 (1.6)	3.9 (1.9)
East Asia Total	8.8	12.8 (1.5)	21.1 (2.4)

Unit : Mt yr^{-1}

Table 4-2 Comparison of East Asian emissions in 2000, 2020 and increasing ratio from 2000 in parentheses.

Region	2000	2020		
		PSC	REF	PFC
NOx				
China	11.2	11.0 (1.0)	15.6 (1.4)	25.5 (2.3)
Other East Asia	4.4	5.5 (1.3)	5.5 (1.3)	5.5 (1.3)
East Asia Total	15.6	16.5 (1.1)	21.1 (1.4)	31.0 (2.0)
SO_2				
China	27.6	19.9 (0.7)	26.8 (1.0)	40.9 (1.5)
Other East Asia	2.6	3.6 (1.4)	3.6 (1.4)	3.6 (1.4)
East Asia Total	30.2	23.5 (0.8)	30.4 (1.0)	44.5 (1.5)
NMVOC				
China	14.7	29.0 (2.0)	35.1 (2.4)	38.6 (2.6)
Other East Asia	3.7	6.3 (1.7)	6.3 (1.7)	6.3 (1.7)
East Asia Total	18.4	35.3 (1.9)	41.4 (2.3)	44.9 (2.4)

Unit: Mt yr^{-1}

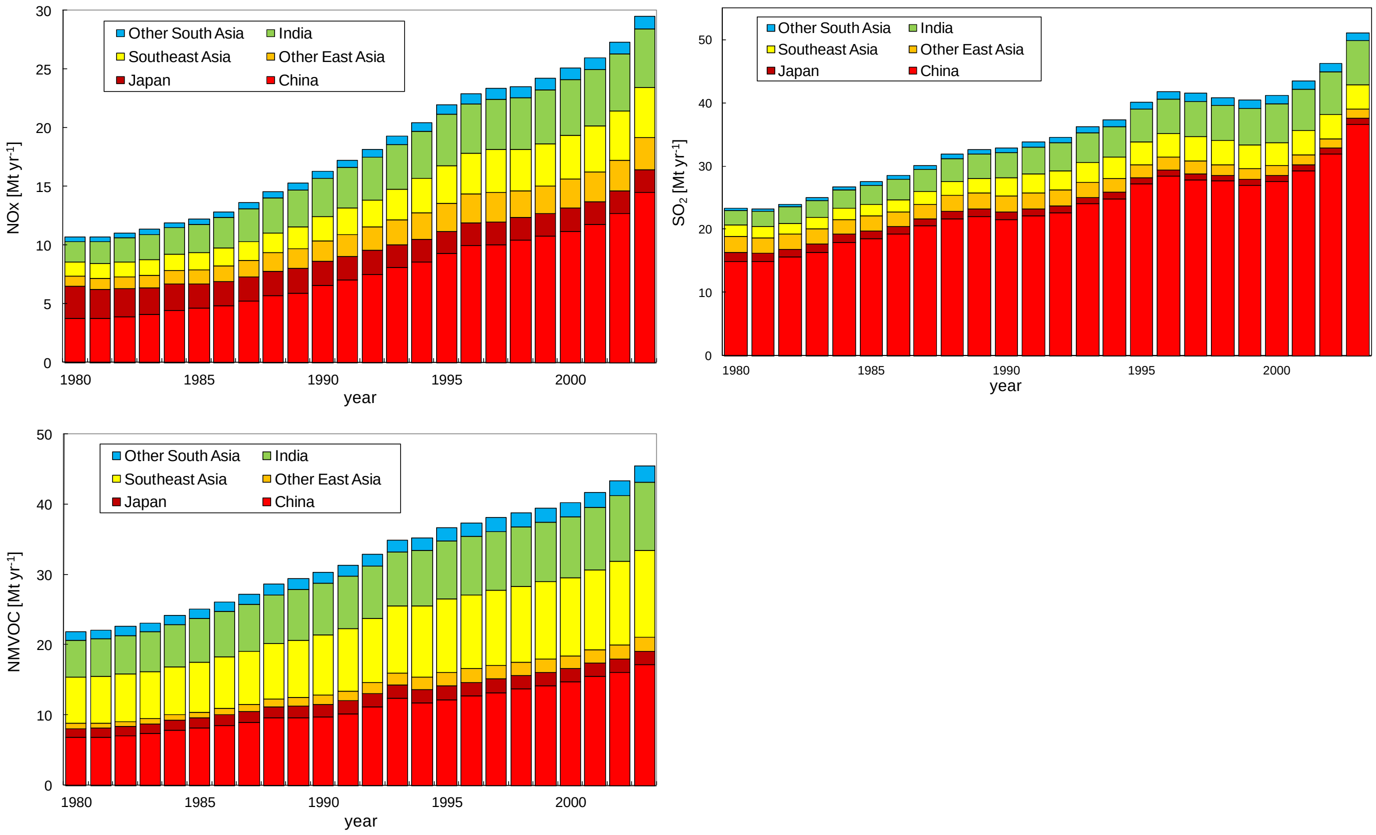

Fig.4-1 Temporal evolution of emissions for NOx, SO_2, NMVOC in Asia

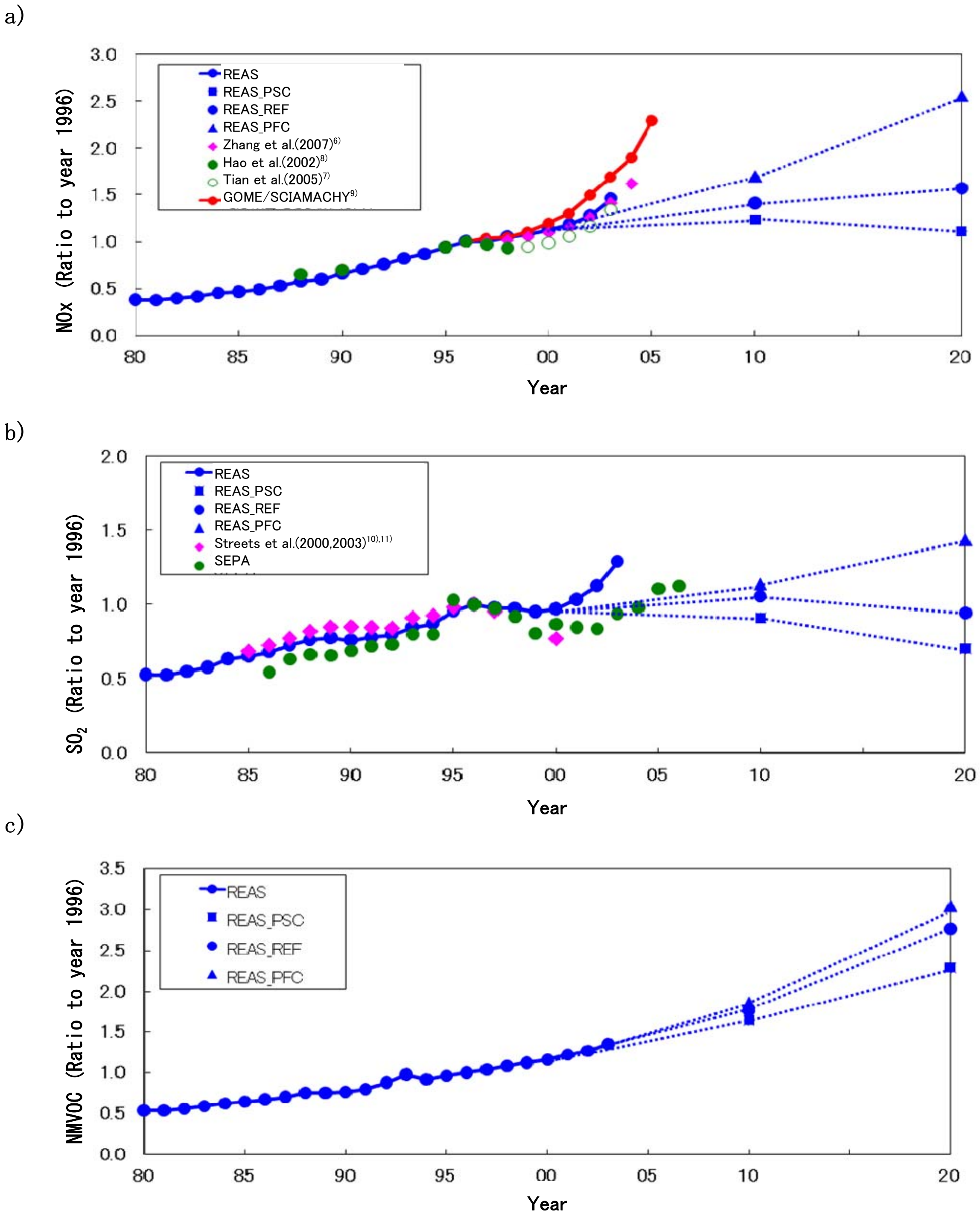

Fig. 4-2 Time series of NOx, SO_2, NMVOC in China.
Y axis shows the emission ratio of air pollutants to year 1996

REAS : Regional Emission inventory in Asia
REAS_PSC : Policy Succeed Case scenario
REAS_REF : Reference Case scenario
REAS_PFC : Policy Failure Case scenario
GOME/SCIAMACHY : NO_2 column concentration measured by GOME/SCIAMACHY [9)]
SEPA : Prediction calculated by the State Environmental Protection Administration, China (now Ministry of Environmental Protection)

4.3 Effect on trans-boundary air pollution in Japan

4.3.1 State of trans-boundary air pollution in Japan

(1) Ozone

(i) Air pollution of ozone in East Asia

O_3 concentration in East Asia were simulated using the Community Multi-scale Air Quality(CMAQ) model[14)] and the emission inventory for Asia(Regional Emission inventory in Asia, REAS)[13)] for considering the spatial and seasonal variation and the contribution of regional emissions[15)]. Figure 4-3 compares daily averaged simulated O_3 concentrations with daily averaged observed O_3 concentrations at Tsushima and Oki monitoring sites in 2000. O_3 concentrations over Japan are controlled by two meteorological factors, the regional-scale airflow and the solar radiation, and the characteristics of ozone concentrations in this area are as follows;

(i)During spring to early summer, O_3 concentrations increase caused by the increase in photochemical O_3 production following the increase in solar radiation and temperature.

(ii)During summer season, O_3 concentrations are lowest over year because of the penetration of clean marine air mass.

(iii)In autumn, O_3 concentrations increase again because of the weakness of clean marine air mass.

(iv)In winter, O_3 production is suppressed by weak solar radiation and low temperature

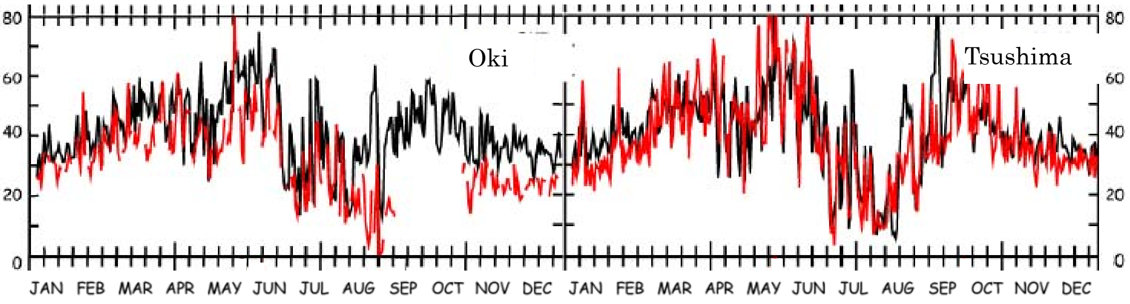

Fig.4-3 Observed (black lines) and simulated (gray lines) daily ozone concentrations at Tsushima and Oki monitoring sites in Japan[15)]. "R=" means each correlation coefficient between observed and simulated concentrations.

The solar radiation and the temperature control photochemical O_3 production, and the changes of airflow controls whether the polluted continental or cleaner marine air masses cover this area[15)], which phenomena can be confirmed using model analysis. Figure 4-4 illustrates 2-month, from May to July and June to

August, averaged O_3 concentration using CMAQ model. In May to July, high O_3 concentrations cover China, the Korean Peninsula and Japan at around 35-40N, and the concentrations of surface O_3 in the North China Plane are 65-70ppbv, and that in Northeast China, the Korean Peninsula, and a part of Japan reach 60-55ppbv. Then in summer, from July to August, the southern parts of China, the Korean Peninsula and Japan are under an effect of marine air masses, and thus O_3 concentration decrease. However, the northeastern parts of China, especially North China Plane, are covered with high O_3 concentrations of more than 60 ppbv.

To determine the contribution of trans-boundary transported O_3 to total O_3, a model run which all emissions in the model domain were turned off, zero emissions, was carried out and the dominant factors controlling the O_3 concentrations over East Asia was investigated by comparing the chemically produced O_3 by regional emissions and the inflow from outside of this model domain. Figure 4-5 shows the surface O_3 concentrations averaged over 2 months and chemically produced by the regional emissions over East Asia including Japan, and its contribution to the total O_3 concentration averaged over 2 months. From May to June, the O_3 concentrations chemically produced by the regional emissions over East Asia including Japan reach 20-35 ppbv in the southern parts from Kanto area of Japan, and reach 10-25 ppbv in Honsyu island from June to August. During spring to early summer, O_3 concentrations increase caused by the increase in photochemical O_3 production following the increase in solar radiation and temperature. The contribution to the surface O_3 concentrations (figure 4-4) is less than the concentrations chemically produced by the regional emissions over East Asia because of the outflow from outside of East Asia..

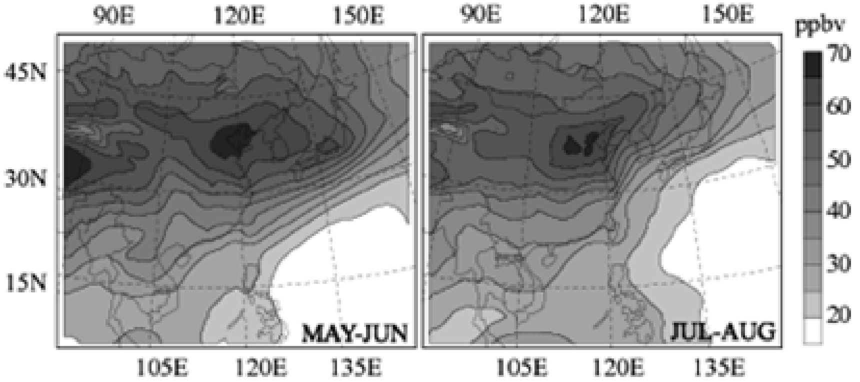

Fig. 4-4 Spatial distribution of O_3 concentrations (ppbv) in 2002[15)]

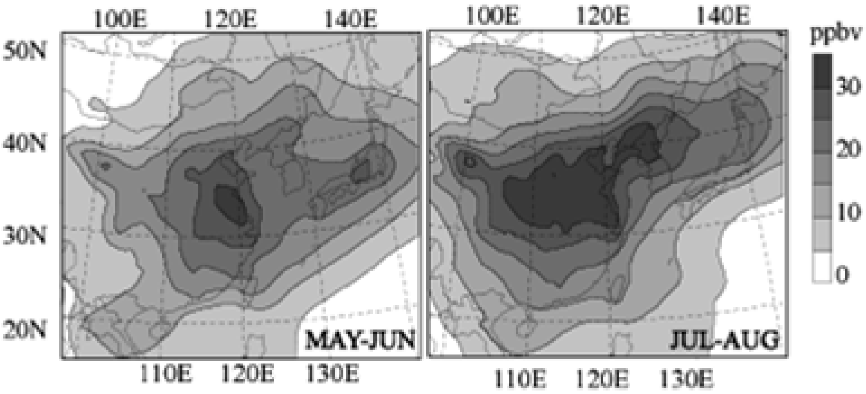

Fig. 4-5 Spatial distributions of chemically produced O_3 by East Asia emissions (ppbv) in 2000[15)]

(ii) Effect to Asia from other regions

Using global chemical transport model, the extent of impacts over Eurasia from Europe and North America on ozone concentration were investigated[16)]. The European and North American influence to Mondy in east Syberia, which represents the inflow to east Asia across Eurasia, were 0.5-3.5ppbv and 1.2-3.3 ppbv respectively. Their impacts increase in Winter and Spring, and after reaching maximum concentrations, they decrease in Summer. It was concluded that contribution ratio from Europe and North America to clean area in East Asia are totally 15%. We examined the contributions from various source regions to global distributions and budgets of tropospheric ozone (O_3) in the context of intercontinental transport, using tagged tracer simulation with a global chemical transport model[17)]. As for the impacts from Europe to East Asia, their contribution to Kagoshima was 2-2.5ppbv in April and below 1 ppbv in June, and in the case from North America to Japan, it was 1-2 ppbv in April and below 0.5 ppbv in June. Comparing the contribution of Europe and North America, there is a large contribution from Chinese planetary boundary layers (PBL) with a peak larger than 15 ppbv in spring. In summer O_3 from the Asian free troposphere increases and so total contribution also reachs 15 ppbv. Contributions of 10-15 ppbv from the stratosphere in winter and spring were calculated.

(iii) Transportation of the regional scale pollution in East Asia and their contributions

O_3 concentrations in the different air masses, regionally polluted continental air masses (RPC) and the background continental air mass (BK),were identified using trajectory analysis and O_3 data observed at Oki and Happo[18)]. Figure 4-6 shows seasonal variations of O_3 concentrations in the regionally polluted continental air masses, the background continental air mass, and the regional scale pollution in East Asia (EA), which were calculated by subtracting BK from RPC. O_3 concentration increases in the regionally polluted continental air mass increases in spring and summer, and average concentrations from April to September was about 55 ppbv. O_3 concentration in the regionally polluted continental air mass showed maximum concentrations in May, and average concentration is over 60 ppbv. These shows that monthly average O_3 concentrations in the air mass transferred from continent to Japan are almost 60ppbv, environmental standards in Japan, in terms of photochemical oxidants. Seasonal variations that O_3 concentrations in the regional pollution in East Asia showed maximum concentration in June –July, ranging 15-20 ppbv, and minimum in Winter, a few ppbv, were observed.

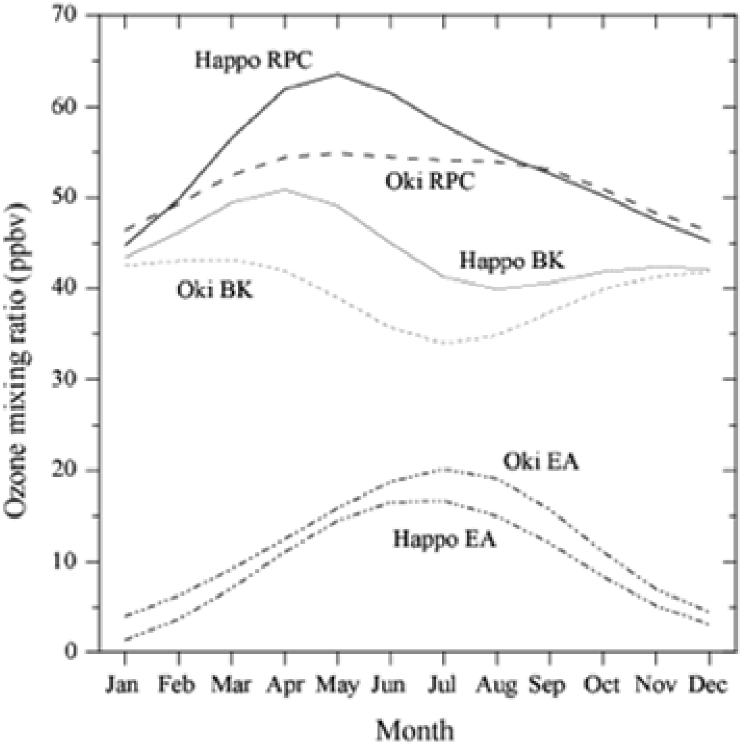

Fig.4-6 Seasonal variations of photochemical O_3 build up by East Asian pollutions(EA) at Oki and Happo calculated by RPC and BK[18)]

Figure 4-7 shows the monthly mean distributions of surface O_3 in April 2002 and regional build-up O_3 contribution from major anthropogenic sources in China and Korea. The monthly mean contributions of the regional build-up O_3 were 5-10ppb, and considering that O_3 concentrations in Honsyu Island were about 50-60ppbv[19)], contributions from East Asia to Japan were calculated about 10-20%

or more.

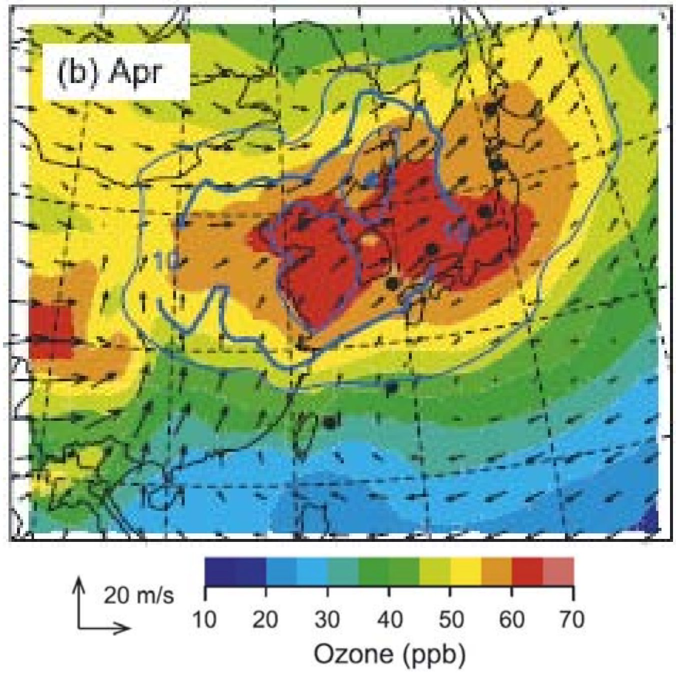

Fig.4-7 Monthly mean distributions of surface O_3 in the model for April 2002. Regional build-up O_3 due to major anthropogenic sources in China and Korea is also shown as blue contours. [19)]

(iv) Episodic pollution of photochemical ozone

The dynamics and formation mechanism of the episodic pollution of O_3 during 8-9 May 2007 were investigated using CMAQ model[20)]. (High O_3 concentrations were observed during 8-9 May, and on 9 May the high O_3 concentrations over 120 ppbv, which is national environmental standard, were observed at 22 prefectures in Japan) High O_3 concentrations over 120 ppb at monitoring stations in Japan were observed in North Kyushu area and the west part of Chugoku area 80 ppbv on 8 May. On next day (9 May) O_3 concentration over 100 ppbv were also observed over the wide area excluding Hokkaido and the north part of Tohoku area. Averaged contribution of Chinese emissions in the same period with high O_3 concentrations was estimated to be over 25% throughout Japan excluding the northern part of Japan, Aomori and Hokkaido, and to be 40-45% in Kyushu area (Figure 4-8).

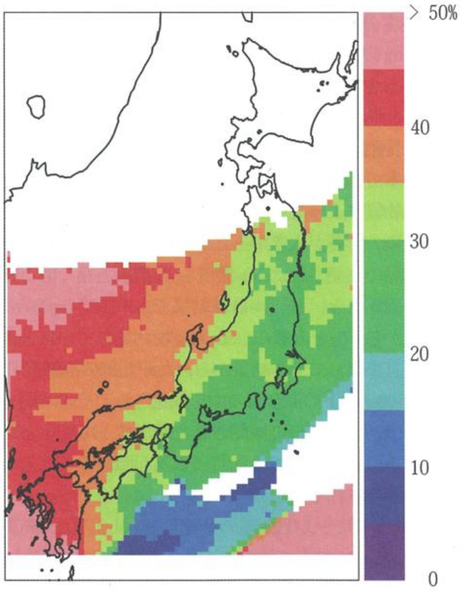

Fig.4-8 Chinese contribution to average O_3 concentration at surface around Japan during the high O_3 concentration in the period 8-9 May of 2007[20)]

（２） Aerosols

Sulfate, nitrate and their salts are long-range transported, and finally deposit with precipitation to the Earth's surface. Then evaluation of the chemical components in precipitation has been investigated taking acid rain into account. Atmospheric Brown Cloud-Asia (ABC) has been conducted under the United Nations Environment Programs (UNEP), which included the investigation on transportation, distribution and impact on climate of aerosols. Recently chemical analysis in aerosols has developed using the instruments with high time resolutions, therefore it has been possible to analyze the phenomena same as gaseous components. In future, it is expected that the discussion on long-range transport of air pollution will include the chemical components in aerosols.

(i) Observation of long-range transported aerosols

Kosa (dust) and anthropogenic aerosols transported from continent were observed in Hedo[21], Okinawa, which is introduced in the report on TF-HTAP[22] (ref. 4.4.1).

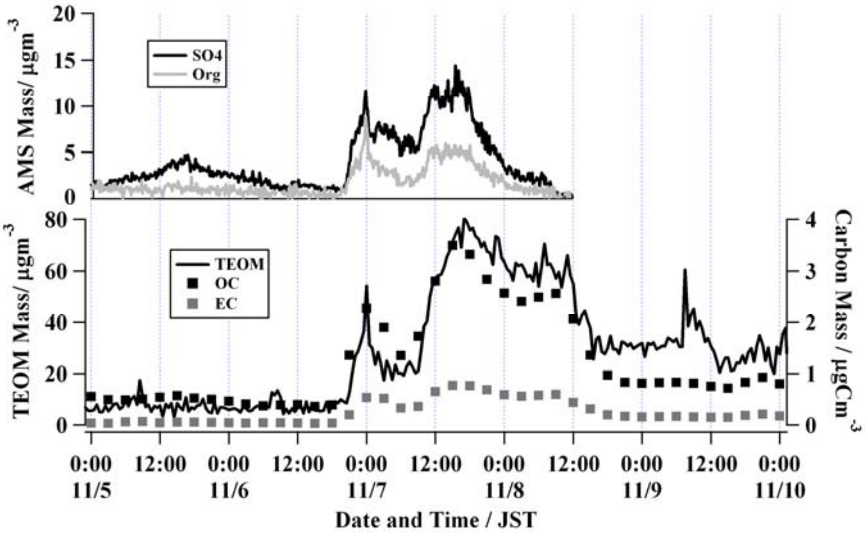

Fig.4-9 Mass concentration of SO_4^{2-}, OC, EC and PM2.5 measured by AMS, Carbon monitor and TEOM
(Org.:Organics, TEOM:PM2.5, OC: Organic carbon, EC: Elemental Carbon)

As shown in Figure 4-9, increasing the concentrations of sulfate and organic compounds were measured at 0:00 on November 7 with the cold front passing Cape Hedo, Okinawa. After the first peak, once these concentrations were decreased. However around 12:00 on November 7, same increasing was measured.

Considering from the back trajectory analysis, the first peak was originated from Taiwan, and the second peak from the central China. After that the concentrations of sulfate and organic compounds were decreased around 12:00 on November 8. Kosa appeared around 12:00 on November 7, and Kosa event dominated after 12:00 on November 8, which were measured by Lidar system. The EC/OC ratio was constant at 0.2 during the whole event. The anthropogenic aerosol originated from Taiwan was transported associating with the cold front passing the southern coast in Japan, then under the high pressure system moving from continent, both anthropogenic aerosol and Kosa were transported together from continent. Kosa was continuously transported under the second high pressure system. As a result, Kosa dominated the later period of this event.

Anthropogenic aerosols were measured in front of Kosa. It suggests that the trans-boundary air pollution occurs widely and its details is very different by meteorological conditions, which were already reported though[23,24)].

Continuous measurements of SO_2 and CO were carried out on the top of Mt. Fuji during the period January to July, 2004 for considering the influence of trans-boundary air pollution in remote area[25)]. These data displayed clear sporadic or seasonal signatures of Asian outflow over Japan to the North Pacific. Elevated SO_2 concentrations were always accompanied by elevated CO and the observed seasonal variation of SO_2 events had nothing to do with seasonality in SO_2 emissions from the Asian continent. These seasonal variations are attributed by cloud process. Little SO_2 is removed by cloud processes in winter, and increasing humidity levels in summer lead to the increased removal of SO_2 by cloud processes.

(ii) Model Analysis

Model analysis is also used in analyzing the long-range transport of aerosols. Regional Chemical transport model(RAMS/CFORS)[26)] is shown to accurately reproduce the features of transport of about 20 air-pollutants, both anthropogenic pollutants (CO, SO_2, sulfate, black carbon) and natural constituents(dust, sea-salt aerosol, volcano gas) etc., in East Asia region. Using these models, examples of the model analysis of trans-boundary air pollutions in April 2001 are shown below. Figure 4-10 shows the horizontal distribution of trans-boundary transportation during 9 to 12 April 2001. Huge air-mass of dust was distributed widely on the back of cold fronts and outflow from continent and Korea Peninsula to Japan could be seen clearly with the low pressure system and the front moving toward East. Moving of dust air-mass toward East in higher altitude was faster than in lower altitude, and air-mass reached to eastern part of Japan 1 or 2 days later than Rishiri. Confirming the existence of high concentration of sulfate in front of high concentration of dust and analyzing the results of measurements and model, it was clarified that Kosa arrives Japan a few hours later than SO_4^{2-} aerosol and that natural constituents and anthropogenic pollutants were trans-boundary transported almost same times accompanied with the moving of high and low pressure systems. During this periods, transportation of black carbon(BC) was very similar with that of SO_4^{2-} aerosol, and it was confirmed that the moving of low pressure systems and cold fronts toward East was the origin of large scale trans-boundary air pollutions.

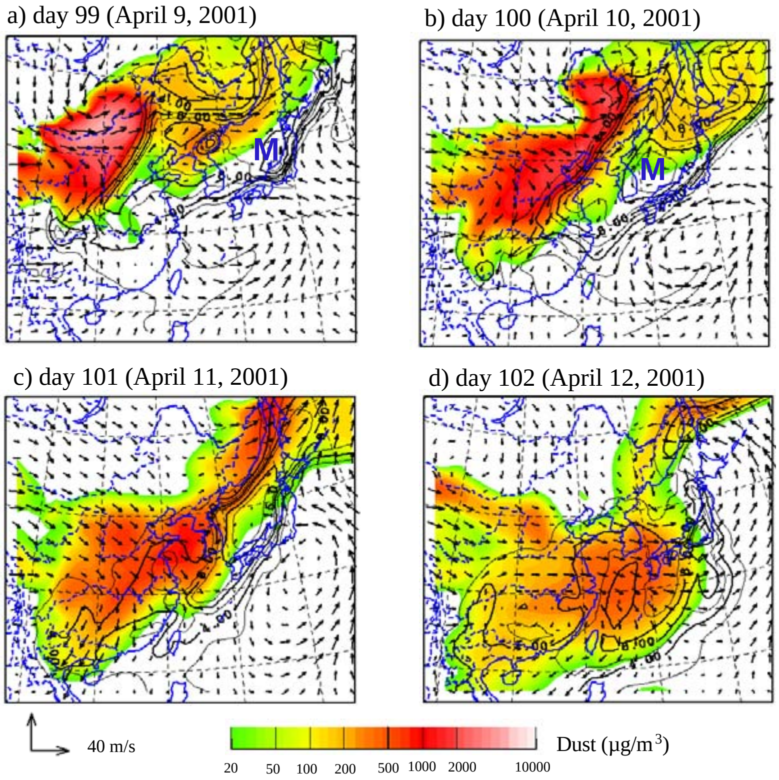

Fig.4-10 Spatial distribution of dust and SO_4^{2-} concentration from 9 to 12 April 2001
Contour plots means the mean dust concentration in boundary area, lines indicate the mean SO_4^{2-} concentration in aerosols, arrows denote the wind vectors above 500m, and M means large area with large contribution of SO_4^{2-} aerosols from Miyake volcano.

(iii) Changes of chemical components in the long-range transported aerosols

Research on chemical compositions and their changes in aerosol long-range transported aerosols have been carried out.

SO_4^{2-} is a dominant acidic substance over the East China Sea with outflow of the air-mass from North East Asia[28], and the ratio of SO_4^{2-}/ NH_4^+ is one or a little higher ratio. However under Kosa phenomenon, that ratio became larger[29](Figure 4-11, upper diagram). Generally it shows also not good correlations with concentrations of NO_3^- and NH_4^+. However under the conditions of transportation with air mass containing high concentrations of pollutants ($NO_3^->100neqm^{-3}$), it showed good correlation (Figure 4-11, lower diagram). In that case distributions of chemical components agreed well with that in coastal area in China with good correlation with SO_4^{2-}, NO_3^- and NH_4^+[30].

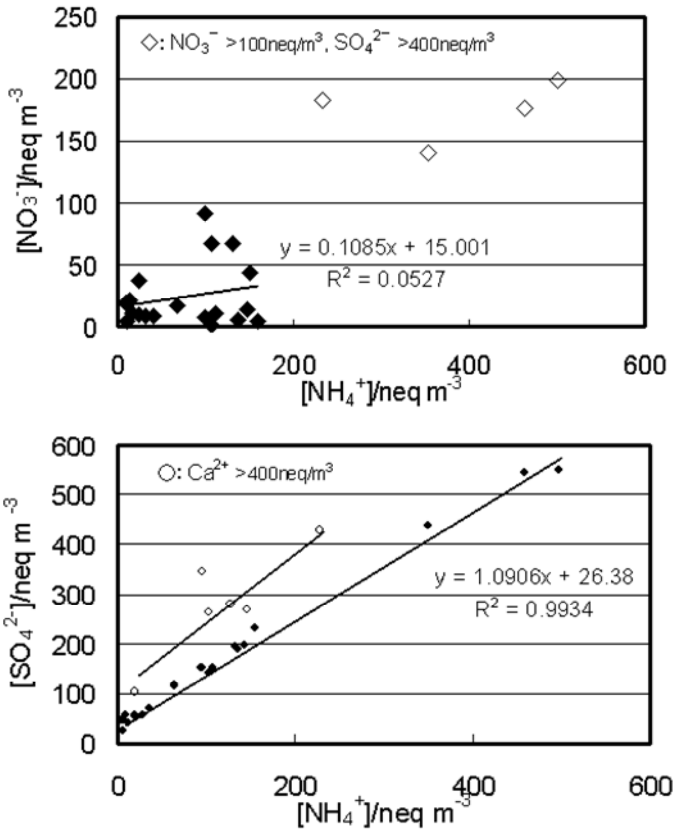

Fig.4-11 Correlations among ion species(NO_3^-, SO_4^{2-} and NH_4^+) in aerosols over the East China Sea

According to the statistics 2000, NH3 emissions in China were 13.6 Tg($8.1x10^{11}$mol), which were larger than SO_2 emissions, 20.4 Tg($8.0x10^{11}$mol), in mole unit[31]. Acidic substances produced in the atmosphere in China were neutralized rapidly with NH_3, and neutralized substances were measured over the East China Sea.

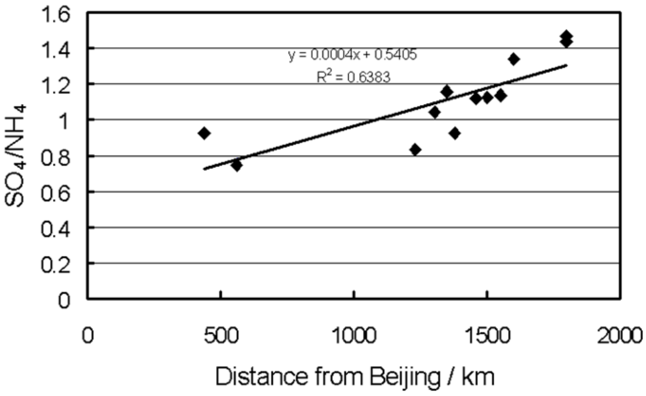

Fig.4-12 Correlation with the distance from Beijing and SO_4^{2-}/NH_4^+ equivalent ratio

The pH values in the rain and aerosols were low in Japan. It suggests that the acidification process goes on with long-range transportation. It was already reported more than 20 years ago that S compounds in aerosols over Pacific Ocean changed as follows in rain water with the distance from the continent;

$(NH_4)_2 SO_4 \rightarrow NH_4HSO_4 \rightarrow H_2SO_4$

It is suggested that same reaction occurrs in the aerosols transported from continent.

Figure 4-12[32)] shows the correlation with the distance from Beijing and SO_4^{2-}/NH_4^+ equivalent ratio in aerosols, which were measured in Dalian and Qingdao in March 2002 and in Japan in late 1990s. NH_4^+ to SO_4^{2-} ratio became lower when the air mass was transported long from the continent. This indicate that acidification process was promoted

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(3) Acid Deposition

Results of source-receptor analysis[1] studies for sulfur and nitrogen oxides obtained by numerical simulation models covering East Asia region were summarized, and effects of transboundary air pollution on acid deposition in Japan are considered.

① Evaluation of effects of transboundary air pollution by numerical simulation models

Table 4-3 shows source contributions to acid deposition amount in Japan from various source regions, which were presented in research papers of numerical model studies 33~42). According to the studies performed after 2000, contributions of transboundary air pollution were estimated to be 32-66% for non seasalt (nss)-SO_4^{2-} and 35-61% for NO_3^-. The reasons of variation in the estimated contributions are considered to be differences in structures of the simulation models, the used parameters, meteorological conditions in the base years, and emission source data, and so on 43,44). Especially, it should be noted that the contribution of volcano eruption to nss-SO_4^{2-} deposition in 2001 was significant because of the eruption in Miyake Island.

Table 4-3 Comparison of contribution evaluations to acid deposition in Japan from various source regions

	Implemented Organization	Published Year	Base Year	Model Type	Contribution (%)				
					Japan	Volcano	China	Korea	Others
nss-SO_4^{2-}	ADORC 33)	2008	2002	Grid	54		28	4	13
	U. of Tokyo 34)	2008	2001	Grid	18	50	15	4	13
	NIES 35)	2005	1995	Grid	21	13	49	12	5
	CRIEPI 36)	2001	1995 1990	Trajectory	26-29 27-30	24-32 25-31	29-32 24-27	12-13 17-19	1-2 1-2
	RAINS-Asia 37)	1995	1990	Trajectory	38	45	10	7	0
	CRIEPI 38)	1998	1988-1989	Hybrid	40	18	25	16	1
	OPU 39)	1997	1988	Grid	37	28	25	10	0
NO_3^-	U. of Tokyo 40)	2008	2001	Grid	56		21	15	8
	NIES 41)	2005	1995	Grid	39		34	18	9
	Columbia U. 42)	2002	1990	Grid	65		18	15	2
	OPU 39)	1997	1990	Grid	76		13	11	1

ADORC: Acid Deposition and Oxidant Research Center, NIES: National Institute of Environmental Studies, CRIEPI: Central Research Institute of Electric Power Industry, RAINS-Asia: Regional Air Pollution Information and Simulation-Asia developed by the World Bank, OPU: Osaka Prefecture University

As an example, Fig.4-13 illustrates contributions of nss-SO_4^{2-} annual deposition for classified regions in Japan 35). In Hokkaido and Tohoku district, contribution from China accounted for more than half of total deposition, and contribution from Japan was higher in Kanto, Chubu, Kinki, Chugoku and Shikoku districts because there are major SO_2 emission sources. In Kinki, Chugoku, Shikoku and Kyusyu districts, contribution from volcano was up to 20% because it was affected by contribution from active volcanoes such

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[1] Relationship between emission sources and receptors is quantitatively analyzed.

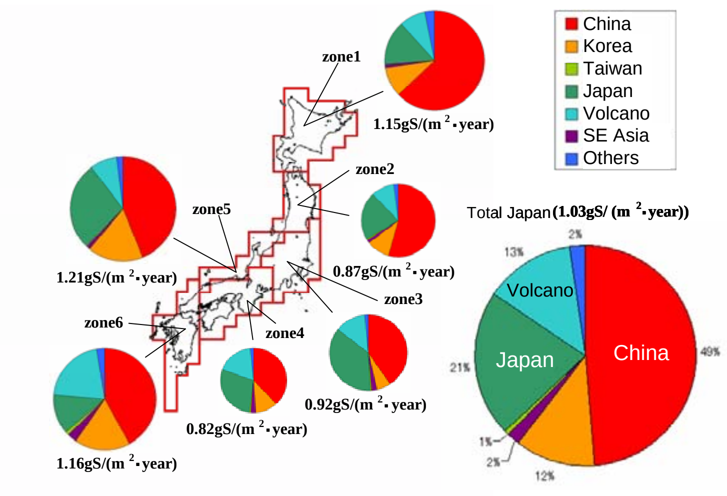

Fig.4-13 Estimated contributions of nss-SO$_4^{2-}$ annual deposition (gS m^{-2} yr^{-1}) for classified regions in Japan 1995.[35]

as Mt. Sakurajima. The reason why the contribution from China in the results of Fig.4-13 was higher is not clear at present. When estimated source contributions among different numerical modeling results were compared, the differences in domain of assessment, base year and absolute deposition amounts should be considered.

Table 4-4 shows estimated monthly source contributions of total sulfur deposition in Japan 2002 evaluated by Regional Air Quality Model (RAQM)[33]. On the basis of the annual average, contribution of transboundary air pollution accounted for approximately 35% (approximately 30% from China). From the point of view of seasonal variation, no clear variation of contribution from Korea was not seen, whereas contributions from central and southern China were high during autumn and spring and low in summer. The probable reasons of this seasonal trend are that consumption of coal which contains high amounts of sulfur compounds in China is highest in winter and that air pollutants are transported from continent to Japan by prevailing northwestern monsoon in winter.

Table 4-4 Estimated monthly source contributions of total sulfur deposition in Japan 2002 (%)[33]

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Annual Ave.
North China	4	5	3	2	2	3	3	3	2	3	4	5	3.3
Central China	24	28	21	14	16	13	17	9	9	21	23	17	17.7
South China	10	11	10	10	7	8	5	3	3	6	8	6	7.3
Korea	3	4	4	5	4	4	5	5	5	5	4	4	4.3
Japan	38	36	47	57	64	61	61	73	75	55	38	45	54.2
Others	21	16	15	12	7	11	10	7	6	10	22	24	13.4

According to estimated Table 4-4 shows estimated monthly source contributions of total sulfur deposition in Japan 2002 evaluated by Regional Air Quality Model (RAQM)[33]. On the basis of the annual average, contribution of transboundary air pollution accounted for approximately 35% (approximately 30% from China). From the point of view of seasonal variation, no clear variation of contribution from Korea was not seen, whereas contributions from central and southern China were high during autumn and spring and low in summer. The probable reasons of this seasonal trend are that consumption of coal which contains high amounts of sulfur compounds in China is highest in winter and that air pollutants are transported from continent to Japan by prevailing northwestern monsoon in winter.

According to long-term variation of estimated nss-SO_4^{2-} annual deposition in Japan by using emission amount of SO_2 during 1980 and 2000 obtained by REAS (Regional Emission Inventory in Asia, ref. Section 4.2) and Source-receptor matrix calculated for 1995 41), contributions from Japan and China were the comparable level (35%) in 1980. However, the contribution from China increased to 43 % in 1990 and has been 45-50% since 1992 because SO_2 emission in China has been increased since the 1980s whereas that in Japan decreased.

According to deposition calculation in Japan from 1995 to 2003 by using the Community Multiscale Air Quality (CMAQ) Modeling System and REAS 45), Fig.4-14 shows the calculated results well agreed with annual and seasonal variations of wet deposition amounts of nss-SO_4^{2-} (averages at 18 monitoring sites of the Japan Acid Deposition Surveys (JADS)). Furthermore, Fig. 4-15 demonstrates that the deposition amount has rapidly increased since 2000 because of increase in SO_2 emission in East Asian region (except the effect of Miyakejima volcano eruption). These results infer that the effect of transboundary air pollution from China is relatively increasing in recent years because SO_2 emission in China has been increased whereas that in Japan and Korea decreased.

Meanwhile, contributions of transboundary air pollution to NO_3^- deposition in Japan were estimated to be different levels, as well as those to SO_4^{2-} deposition, by various literatures, but it is highly likely that the contribution is increasing with NO_3^- deposition considering recent trend of NOx emission in Japan and China (ref. Section 4.2) 45).

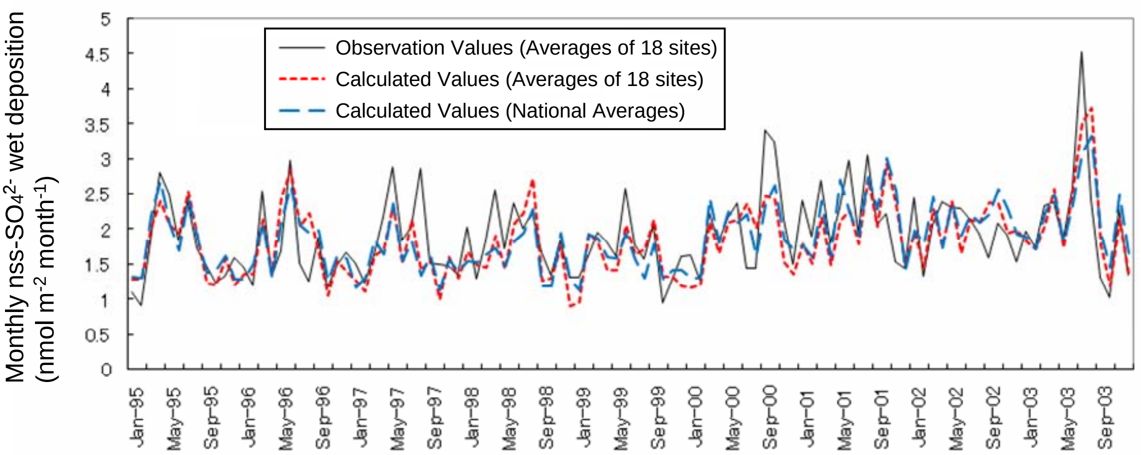

Fig 4-14 Comparison of observed values of nss-SO_4^{2-} wet deposition amounts (monthly values) and model calculated values 45).

The black solid line indicates the averaged value of 18 JADS monitoring sites, the red dotted line indicates the model calculated average values of 18 JADS monitoring sites, and the blue broken line indicates the model calculated average values of all monitoring sites in Japan.

In summary, model calculation results published after 2000 demonstrated that the recent contribution of transboundary air pollution to acid deposition amounts in Japan is estimated to be approximately 30-65% (approximately 15-50% from China) for nss-SO$_4^{2-}$ and approximately 35-60% (approximately 20-35% from China) for NO$_3^-$. It is considered that the contribution of transboundary air pollution is significant in winter-time northern Japan.

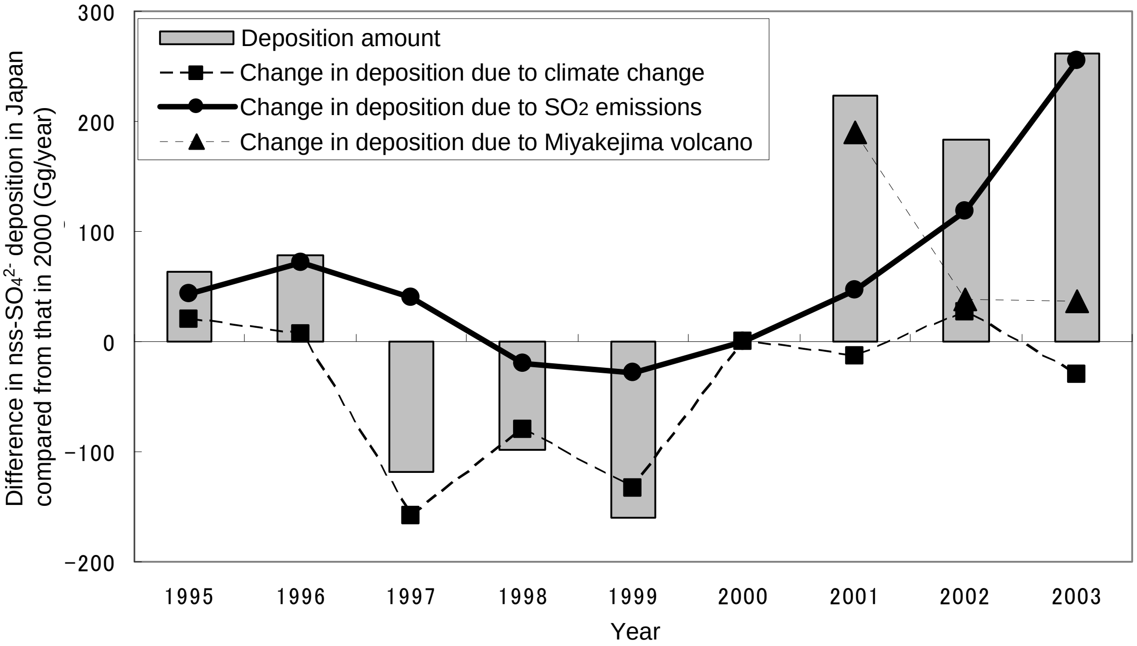

Fig 4-15 Estimated variations of nss-SO$_4^{2-}$ deposition in Japan from 1995 to 2003 45).

The fine broken line indicates the difference of nss-SO$_4^{2-}$ deposition due to climate change, the bold solid line indicates the difference due to SO$_2$ emissions other than Miyakejima volcano eruption, and the fine dotted line indicates the difference due to SO$_2$ emissions from Miyakejima volcano eruption. The base year for all differences is 2000.

② Evaluation of effects of transboundary air pollution by a sulfur isotope ratio[2]

It was reported that some researches evaluated effects of transboundary air pollution based on observation data of sulfur isotope ratio (δ^{34}S). δ^{34}S of wet deposition SO$_4^{2-}$ and nss-SO$_4^{2-}$ in Niigata prefecture was high in winter and low in summer. This seasonal trend is attributable to higher contribution of sulfur compounds (The contribution to SO$_4^{2-}$ was estimated to be approximately 20%) emitted from coal combustion origins in northeast Asia region during winter time 47). According to the ditto observation data at 19 Japan nationwide sites in winter, δ^{34}S of nss-SO$_4^{2-}$ was higher in northern Japan, and the δ^{34}S

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[2] Since a sulfur isotope ratio of ^{34}S to ^{32}S varies depending on emission origins, the isotope ratio provides information of sulfur cycle in the environment and emission sources of sulfur compounds, δ^{34}S indicates deviation from the isotope ratio of standard material in units of ‰.

value was close to that of coal mined in East Asian countries. This result implies that nss-SO_4^{2-} wetly deposited in northern Japan was influenced by long range transportation from the Asian continent.

4.3.2 Future prediction of impact of transboundary air pollution on Japan

In order to predict impact of transboundary air pollution on Japan, it is important that atmospheric concentrations of air pollutants are estimated by a chemical transport model and prospected emission amounts of air pollutants, as an input data, on the basis of future trend of socioeconomic activities. Research groups of Frontier Research Center for Global Change, Japan Agency for Marine-Earth Science and Technology (FRCGC/JAMSTEC) and National Institute for Environmental Studies (NIES), etc. promoted future prediction of atmospheric concentrations of air pollutants in East Asia by using the Regional Emission Inventory in Asia (REAS) 3),13) and the Community Multiscale Air Quality (CMAQ) Modeling System 49) as shown in Section 4.2.

Fig. 4-16 shows increase of tropospheric ozone concentration during 2000 and 2020 estimated by 3 kinds of scenarios of REAS and CMAQ 50). The REAS emission inventories employed the following 3 kinds of scenarios for China: Policy Successed Case (PSC), Reference Case (REF) and Policy Failed Case (PFC), and the standard emission scenario on the basis of energy demand prediction by International Energy Agency (IEA) was employed for other countries. It predicted that the annual average of tropospheric ozone concentration in Japan will increase by 0-4 ppbv per year during 2000 and 2020 if the moderate increase emission scenario (REF) was employed. If low emission increase scenario (PSC) was employed, the ozone concentration was estimated to increase by not more than 2 ppbv per year. If high emission increase scenario (PFC) was employed, the ozone concentration was estimated to increase by 2-6 ppbv per year on an annual average and by 2-14 ppbv per year on a monthly average of June when tropospheric ozone concentration becomes higher 50). Especially, ozone concentration in central Honshu was predicted to increase by approximately 6 ppbv on a three month average in summer (June - August) although NOx emission in Japan was predicted to decrease. Since there is no significant difference in emission amounts from other countries except China among different scenarios, future variation of emission from China may largely affect on tropospheric ozone concentration in Japan in the future. Moreover, it should be noted that the emission inventory scenarios of REAS considered 2000 as the base year, and that rapid increased emissions from China after 2000 4),52) were not sufficiently considered. If remarkable economical development in China continues in the future and the PFC scenario become a reality, the tropospheric ozone concentrations in Japan 2020 were predicted to be 45-60 ppbv on an annual average and 50-75 ppbv in June (Fig. 4-16). This prediction raises a concern that ozone pollution in Japan will reach a serious level 53). This prediction also demonstrates emission control such as NOx emission reduction is important to not only environmental protection in China but also countermeasure against photochemical pollution 51).

Medium-term prediction experiments aimed for contribution to the IPCC (Intergovernmental Panel on Climate Change) 4th Assessment Report was carried out by 26 kinds of global chemical transport models. The spate of models employed the 3 kinds of unified emission scenario as follows: Current Legislation (CLE), Maximum Feasible

Reduction (MFR) and IPCC Special Report on Emissions Scenarios A2 (IPCC-A2). When each scenario was incorporated for model simulation, the annual average of surface ozone concentration in East Asia (including Southeast Asia) was estimated to increase by 3.8 ± 0.7 ppbv per year (CLE) and 7.7 ± 1.8 ppbv per year (IPCC-A2) during 2000 and 2030. Only the estimations employed the MFR scenario exhibited decrease in annual ozone concentration (-3.6 ± 0.5 ppbv per year) during the period 54),55). Moreover, these model experiments demonstrated that nitrogen and sulfur deposition amounts were estimated to slightly change (NOy: -3.0 mgNm^{-2}, NHx: +47.0 mgNm^{-2}, SOx: -18.5 mgS m^{-2}) by employing the CLE scenario, largely decrease (NOy: -249.8 mgNm^{-2}, SOx: -404.3 mgS m^{-2}) by the MFR scenario and largely increase (NOy: +278.6 mgNm^{-2}, NHx: +135.0 mgNm^{-2}, SOx: -782.6 mgS m^{-2}) by the IPCC-A2 scenario 56).

It should be noted that these prediction results have large variation among the present global models and the predicted concentrations of air pollutants has larger differences among the different model calculations even if the same emission scenario is employed. Moreover, it should be also noticed that emission amounts in East Asia region will exceed the predicted amounts, and many emission scenarios employed by the above model prediction experiments may not consider this rapid increase of emission amounts. However, these experimental results implies that the atmospheric environment in East Asia region is expected to be in an alarming situation and transboundary air pollution may grow into a more serious problem unless emission regulation will be tightened in the future.

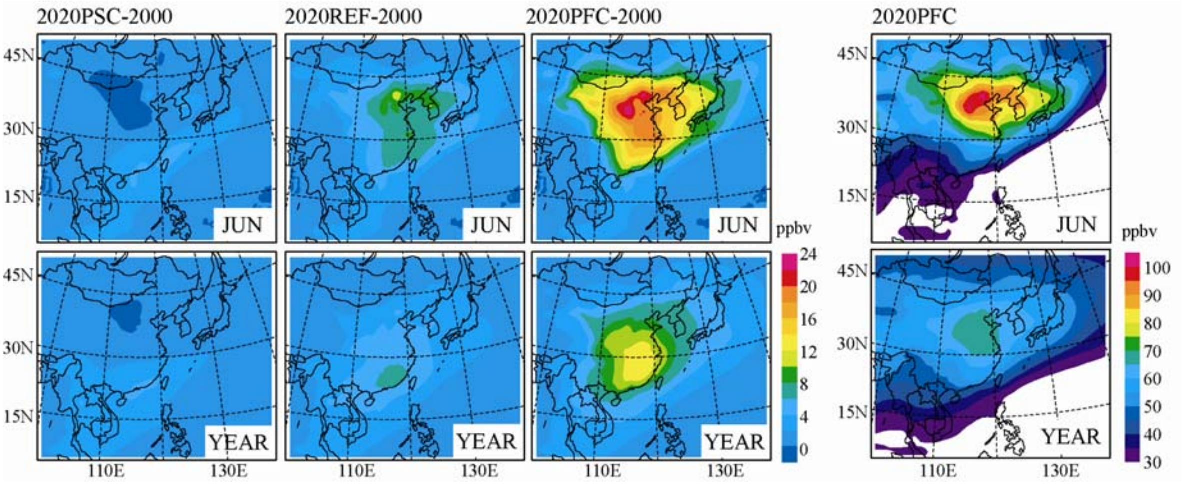


Fig 4-16 Predicted tropospheric ozone concentration by using 3 kinds of scenarios (PSC, REF and PFC) of REAS 50).

2020PSC-2000: increase in ozone concentration during 2000 and 2020 by using PSC, 2020REF-2000; increase in ozone concentration during 2000 and 2020 by using REF, 2020PFC-2000: increase in ozone concentration during 2000 and 2020 by using PFC, 2020PFC: predicted ozone concentration in 2020 by using PFC.

Overview of the model intercomparison study in Asia (MICS-Asia)

Despite the seriousness of transboundary air pollution issues in northern hemisphere, especially, East Asian countries have not built a consensus on common understandings of the issues. In order to build the consensus, numerical simulation by long-range transportation models will play an important role. However, there is significant difference in calculated results among the long-range transportation models targeting on air quality and acid deposition in East Asia developed by researchers in East Asian, American and European countries. Therefore, this discrepancy may be an impediment to the international consensus building. Against this background, the Model Inter-Comparison Study in Asia (MICS-Asia) was launched in 1998, and since 2003, precision comparison of model simulation targeting on SO_2, NOx and O_3 has been promoted as the MICS-Asia Phase 2 activity.

The MICS-Asia Phase 2 is a joint project between Acid Deposition and Oxidant Research Center and International Institute for Applied Systems Analysis in Austria. Approximately 20 research institutes participate the project, and intercomparisons of long-range transportation models and emission inventories have been promoted. The model calculation results were reported according to the provided data protocol, and finally intercomparison among 8 kinds of models was implemented. The target simulation period was March, July and December in 2002 and March in 2002. This study focused on yellow sand phenomena, high ozone concentration events in Northeast Asia, behaviors of air pollutants in summer and winter. The disclosed EANET (Acid Deposition Monitoring Network in East Asia) monitoring data was used for validation of models.

The projected members summarized the outcomes of the MICS-Asia Phase 2 in 8 research papers that was published in the special issue of "Atmospheric Environment". The MICS-Asia Phase 3 launched in 2008. The Phase 3 activities will promote analysis of emission contribution in East Asia, comparison with Northern Hemispheric models, analysis of impacts of mega cities in East Asia, relationships between air pollution and climate and development of new emission inventories.

References: MICS-Asia Phase 2 Web site
(http://www.adorc.gr.jp/adorc/mics.html)

4.4 Intercontinental transport of air pollutants and hemispheric air pollution

4.4.1 Task Force on Hemispheric Transport of Air Pollutants (CLRTAP/TF-HTAP)

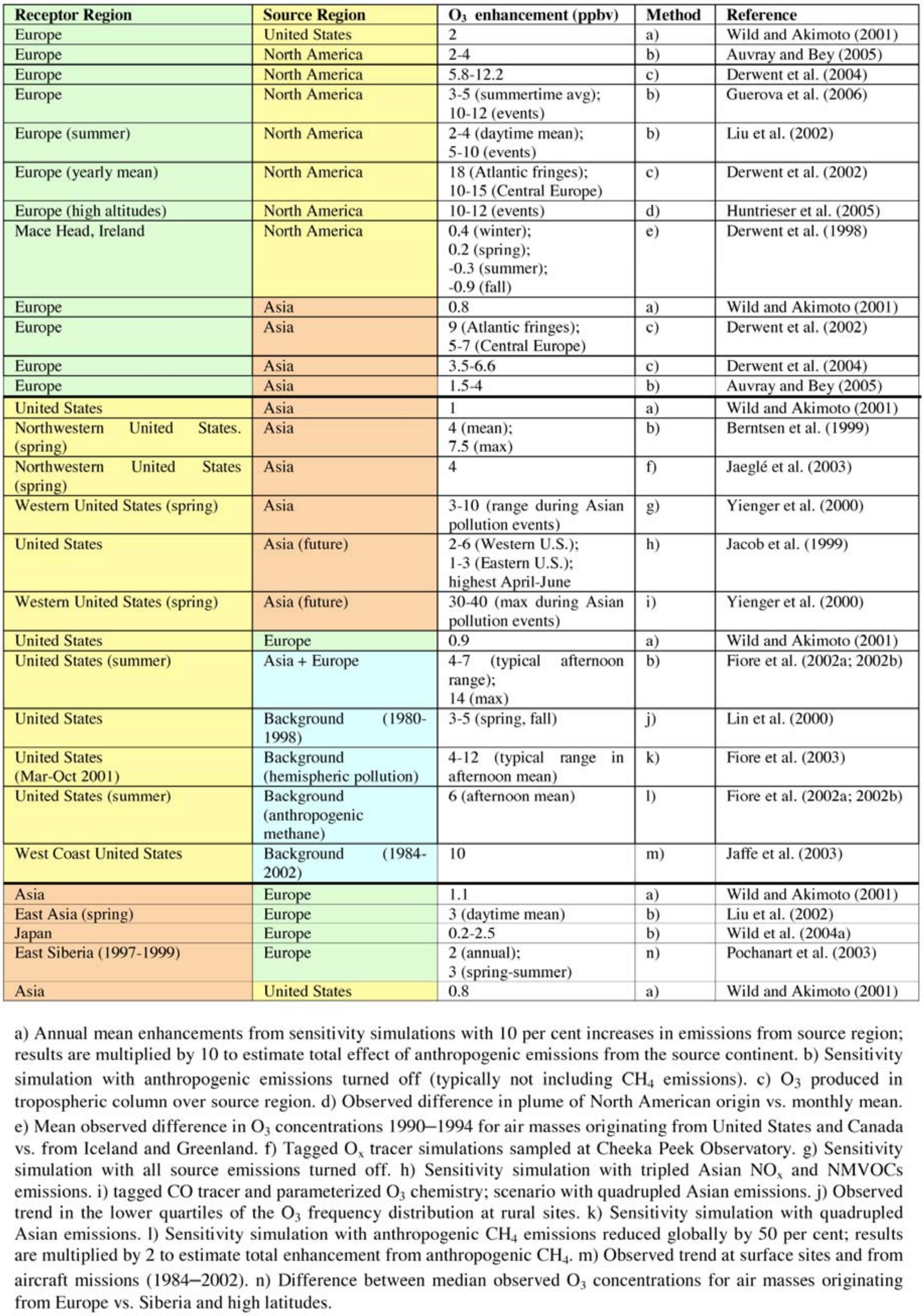
Scientific interest of intercontinental transport of air pollution increased in the latter half 1990s, and many research results were presented in the late 1990s 57). At the beginning, the intercontinental transport of yellow sand from the Asian continent to North America via the Pacific Ocean was paid attention 58),59),60),61),62). At that time, in the United States, the possibility that the effect of ozone derived from Asian region was not negligible for achievement of the air quality standards in the Unites States was discussed 63), 64), 65), 66), and, in Europe, the effect of ozone transported from the North America to Europe via the Atlantic Ocean was also discussed 67),68),69),70). The researches focusing on intercontinental transport of ozone from Europe to Asian region via the Eurasian continent demonstrated that effect of North America to Asia was significant as much as that of Europe and ozone originated from Europe and North America considerably impacted on ozone concentration in Asian region 71),72),73). As seen above, it came to light that air pollutants originated from each continent in Northern Hemisphere affected the other continents, and the concept of “hemispheric transport and hemispheric air pollution” was raised.

After the above scientific research outcomes, the Task Force on Hemispheric Transport of Air Pollutants under Convention on Long-range Transboundary Air Pollution (CLRTAP/TF-HTAP) was inaugurated in 2004 at the initiative of the United States and Europe. The secretariat was opened in the United States Environmental Protection Agency (US/EPA) and the United Nations Economic Commission for Europe (UN/ECE). The TF-HTAP is expected to cover technical issues necessary to understand transportation of the air pollutant in the northern hemisphere, and aims to make contributions to the protocol of CLRTAP with a support of a scientific support necessary for policy making. Ozone, aerosol, mercury and Persistent Organic Pollutants (POPs) are designated as target substances for hemispheric transport. The TF-HTAP is composed of experts in the participating countries of the CLRTAP, but it is encouraged to also invite experts in nonmember countries in the Northern Hemisphere. One task force meeting and a set of workshops have been held once a year, and researchers and administrative officers in Japan and other Asian countries have participated in these meetings. In 2006, a workshop on air pollutant emission was held in Beijing, China, which is the first time to be held in a nonmember country. The TF-HTAP prepared the interim assessment report in 2007 and will finalize the report in 2009.

A part of literature review published in the 2007 TF-HTAP interim assessment report is introduced in this section. Table 4-5 summarizes the model calculation results of enhancement of surface ozone in Europe, the United States and Asia as receptor regions affected by North America, Asia and Europe as source regions. For example, the enhancements of surface ozone concentration in Europe affected by North America were reported to be 2-4 ppb on an annual average and 10-15 ppb in some events, and those affected by Asia were 0.8-several ppb on an annual average. The enhancements in the

United States affected by Asia were reported to be 1-4 ppb on an annual average; however, those were 3-10 ppb in some events and up to 30 ppb in the highest event. and 10-15 ppb in some events, and those affected by Asia were 0.8-several ppb on an annual average. The enhancements in Asia affected by Europe and the United States were reported to be 2-3 ppb on an annual average 72),73). In summary, the enhancement of surface ozone concentration in Northern Hemisphere was estimated to be 1-5 ppb on an annual or seasonal average, but the preceding studies commonly pointed out that the enhancement was highest at the end of spring. Since the transport distance across the Atlantic Ocean is shorter than that across the Pacific Ocean or the Eurasian continent, the effect of intercontinental transport from North America to Europe is most significant.

Table 4-5 Estimated enhancements of surface ozone concentration in source regions affected by receptor regions according to existing literatures of intercontinental transport in mid-latitude Northern Hemisphere.

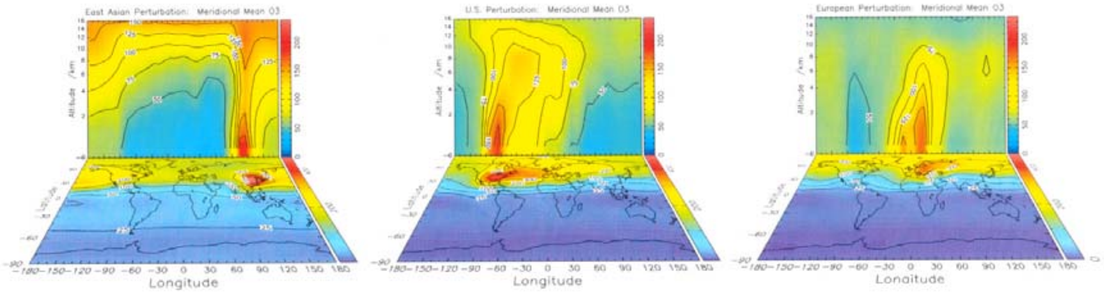


Receptor Region	Source Region	O_3 enhancement (ppbv)	Method	Reference
Europe	United States	2	a)	Wild and Akimoto (2001)
Europe	North America	2-4	b)	Auvray and Bey (2005)
Europe	North America	5.8-12.2	c)	Derwent et al. (2004)
Europe	North America	3-5 (summertime avg); 10-12 (events)	b)	Guerova et al. (2006)
Europe (summer)	North America	2-4 (daytime mean); 5-10 (events)	b)	Liu et al. (2002)
Europe (yearly mean)	North America	18 (Atlantic fringes); 10-15 (Central Europe)	c)	Derwent et al. (2002)
Europe (high altitudes)	North America	10-12 (events)	d)	Huntrieser et al. (2005)
Mace Head, Ireland	North America	0.4 (winter); 0.2 (spring); -0.3 (summer); -0.9 (fall)	e)	Derwent et al. (1998)
Europe	Asia	0.8	a)	Wild and Akimoto (2001)
Europe	Asia	9 (Atlantic fringes); 5-7 (Central Europe)	c)	Derwent et al. (2002)
Europe	Asia	3.5-6.6	c)	Derwent et al. (2004)
Europe	Asia	1.5-4	b)	Auvray and Bey (2005)
United States	Asia	1	a)	Wild and Akimoto (2001)
Northwestern United States. (spring)	Asia	4 (mean); 7.5 (max)	b)	Berntsen et al. (1999)
Northwestern United States (spring)	Asia	4	f)	Jaeglé et al. (2003)
Western United States (spring)	Asia	3-10 (range during Asian pollution events)	g)	Yienger et al. (2000)
United States	Asia (future)	2-6 (Western U.S.); 1-3 (Eastern U.S.); highest April-June	h)	Jacob et al. (1999)
Western United States (spring)	Asia (future)	30-40 (max during Asian pollution events)	i)	Yienger et al. (2000)
United States	Europe	0.9	a)	Wild and Akimoto (2001)
United States (summer)	Asia + Europe	4-7 (typical afternoon range); 14 (max)	b)	Fiore et al. (2002a; 2002b)
United States	Background (1980-1998)	3-5 (spring, fall)	j)	Lin et al. (2000)
United States (Mar-Oct 2001)	Background (hemispheric pollution)	4-12 (typical range in afternoon mean)	k)	Fiore et al. (2003)
United States (summer)	Background (anthropogenic methane)	6 (afternoon mean)	l)	Fiore et al. (2002a; 2002b)
West Coast United States	Background (1984-2002)	10	m)	Jaffe et al. (2003)
Asia	Europe	1.1	a)	Wild and Akimoto (2001)
East Asia (spring)	Europe	3 (daytime mean)	b)	Liu et al. (2002)
Japan	Europe	0.2-2.5	b)	Wild et al. (2004a)
East Siberia (1997-1999)	Europe	2 (annual); 3 (spring-summer)	n)	Pochanart et al. (2003)
Asia	United States	0.8	a)	Wild and Akimoto (2001)

a) Annual mean enhancements from sensitivity simulations with 10 per cent increases in emissions from source region; results are multiplied by 10 to estimate total effect of anthropogenic emissions from the source continent. b) Sensitivity simulation with anthropogenic emissions turned off (typically not including CH_4 emissions). c) O_3 produced in tropospheric column over source region. d) Observed difference in plume of North American origin vs. monthly mean. e) Mean observed difference in O_3 concentrations 1990–1994 for air masses originating from United States and Canada vs. from Iceland and Greenland. f) Tagged O_x tracer simulations sampled at Cheeka Peek Observatory. g) Sensitivity simulation with all source emissions turned off. h) Sensitivity simulation with tripled Asian NO_x and NMVOCs emissions. i) tagged CO tracer and parameterized O_3 chemistry; scenario with quadrupled Asian emissions. j) Observed trend in the lower quartiles of the O_3 frequency distribution at rural sites. k) Sensitivity simulation with quadrupled Asian emissions. l) Sensitivity simulation with anthropogenic CH_4 emissions reduced globally by 50 per cent; results are multiplied by 2 to estimate total enhancement from anthropogenic CH_4. m) Observed trend at surface sites and from aircraft missions (1984–2002). n) Difference between median observed O_3 concentrations for air masses originating from Europe vs. Siberia and high latitudes.

4.4.2 Analysis by intercontinental transportation model

As described above, since the late 1990s, some groups have presented the researches on intercontinental pollutant transport and transformation processes using the seasonal trend data of ozone, carbon monoxide, aerosol and other air pollutants with lifetimes longer than one month. Fig. 4-17 shows characteristics of intercontinental transport of tropospheric ozone via trans-Eurasia (from Europe to Asia), trans-Pacific (from Asia to North America) and trans-Atlantic (from North America to Europe) 72). This figure illustrates how the increased ozone will be transported in a hemispheric scale when emission amounts of anthropogenic NOx, VOC and CO from each continent are increased by 10%. As shown in the left of the figure, ozone generated in Asia is assumed to be transported to the upper troposphere by active vertical convection, which will affect on the hemispherical scale. On the other hand, ozone generated in Europe is assumed to be stay in the middle or lower troposphere because vertical convection is less active, and thus transport to Asia takes place in relatively lower altitude. Ozone generated in North America has the intermediate characteristics between Asia and Europe. These different characteristics are attributable to the difference in latitudes of major polluted areas in each region. The vertical convection in Asia tends to be active because of lower latitude, whereas air mass will stay in the lower troposphere at higher latitude of North America and Europe.



Increase in emission from Asia

Increase in emission from North America

Increase in emission from Europe

Fig. 4-17 Estimated vertical and planar distributions of ozone concentration when emission amounts of anthropogenic air pollutants from each continent are increased by 10% 72).

On the basis of the above shown results, effect on ozone and carbon monoxide concentrations in Happo One, Japan by emissions from North America or Europe is especially shown in Fig. 4-18, depicted as vertical and monthly distributions 73). The enhancement of carbon monoxide affected by Europe was highest in the atmospheric boundary layer during February and March. As is anticipated, the effect of Europe is larger than that of North America On the other hand, the enhancements of ozone affected by both Europe and North America are highest in the middle troposphere during April and May, and the effects of both regions are at the same level. As for surface ozone, the effect of

Europe is maximum during March and April, and that of North America is during February and March. The enhancements of surface ozone in both regions are at the almost same level (2-3 ppb on a monthly average). The reason why the effect of much farther North America is the same level as that of Europe is that, as shown in Fig. 4-17, ozone generated in North America will be transported into higher altitude than that generated in Europe, which will causes effective long range transport and deposit near Japan.

Fig. 4-19 shows the comparison between the observed surface ozone concentrations at Happo in April 1996, when the effect of intercontinental transport is considered to be relatively significant, and the coetaneous estimated ozone concentrations by a global model, which is the sum of contributed ozone concentrations from each region are shown in the bottom [73]. The observed ozone concentration often reached at 60-100 ppb during this period. The contribution from Japan was dominant after April 20, while the contribution of transboundary pollutant transport from East Asian region including China was comparable to that from Japan during April 5-10 and April 15-18. On the other hand, the advections from Europe, North America and the stratosphere significantly contributed to boost background ozone concentration throughout the period. It is a clear

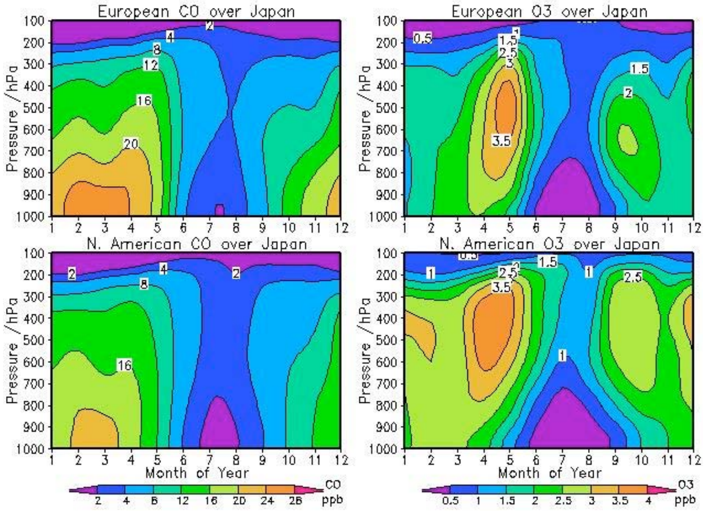


Fig. 4-18 Effects on CO (left) and O_3 (right) concentrations in Happo One, Japan by emissions from Europe (upper) and North America (lower), depicted as vertical and monthly distributions [73].

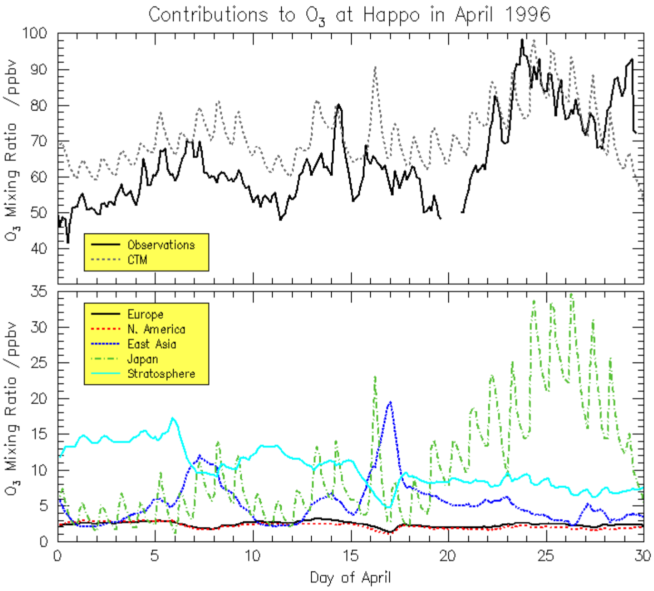


Fig. 4-19 Time profiles of surface O_3 (right) concentration in April 1996 at Happo One. The upper shows the observed and calculated concentrations, and the lower shows the contributed ozone concentrations from each region

characteristic that transboundary pollutant transport from East Asian region effects on high ozone concentrations in Japan, which apparently take place some days, and the effect of intercontinental transport has slight daily variation and continuously contributes to background ozone concentration in Japan at a constant level. The contributions from Europe and North America are estimated to be several ppb in total. This amount doesn't seem to be significant, but it is comparable to transboundary transport from East Asian region on a monthly average basis. This estimation will largely influence on the discussion on adverse effects of ozone on plants, which are generally evaluated by integrated exposed amounts such as AOT40.

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