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Acid Deposition Monitoring Network in East Asia (EANET)

EANET Science Bulletin

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Acid Deposition Monitoring Network in East Asia

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Objectives

- To create a common understanding of the state of acid deposition in East Asia;
- To provide useful inputs for decision making at local, national and regional levels aimed at preventing or reducing adverse impacts on the environment caused by acid deposition; and
- To contribute to cooperation on the issues related to acid deposition among the participating countries.

Photo of the cover by Akihiko Sato.

EANET Science Bulletin

Volume 4

The EANET Science Bulletin is published by the Network Center for the Acid Deposition Monitoring Network in East Asia (EANET) regularly. For the publication of the EANET Science Bulletin (Vol. 4), we established the Editorial Board as follows:

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Preface

The EANET Science Bulletin (Volume 4)

EANET Science Bulletin is published to share the scientific findings from the EANET research activities and to provide a platform for scientists from member countries to publish their scientific and technical research activities which are relevant to the EANET.

EANET Science Bulletin (Volume 4) is comprised of the reports of the EANET Fellowship Research Program (2013-2015), joints projects of EANET with participating countries, and scientific and technical research papers using EANET data from participating countries. This Science Bulletin is published as an activity in the Medium Term Plan (MTP) for the EANET (2016-2020) that approved at the Seventeenth Session of the Intergovernmental Meeting (IG17) on the EANET in 2015. The Bulletin would contribute to better understanding on the state of acid deposition and its impacts on the environment in East Asia and to promotion of the research activities as well as the improvement of capacity building on various air pollution issues such as Ozone/PM2.5 problems and atmospheric transport modeling for participating countries.

The IG17 in 2015 approved the budget for the publication of the Bulletin which is included in the Work Program and Budget of the EANET in 2016. Research results are very important and necessary for improving the situation of the scientific and technical matters of our EANET community. In fact, the quality of the EANET monitoring data is getting better and better and the number of scientific research papers using EANET data published in international scientific journal is increasing.

Finally, I'd like to express my sincere appreciation to all authors for the research papers and the members of Editorial Board for this EANET Science Bulletin. I hope your continuous cooperation and concern for the EANET activities.

Dr. Sakamoto Kazuhiko
Director General
Asia Center for Air Pollution (ACAP)
Network Center for the EANET

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Message from the Editor

Research activity is one of the important activities of the EANET as well as monitoring activity of acid deposition in order to understand the state of acid deposition and to improve air quality monitoring methodologies. The EANET has encouraged the implementation of the research work on acid deposition and published the Science Bulletin periodically. The research papers in the Bulletin are reflecting recent situation on acid deposition in East Asia and our effort to solve the emerging issues of the EANET through research activity will be continued.

Recent Scientific Activities of the Acid Deposition Monitoring Network in East Asia (EANET)

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1. Introduction

The Acid Deposition Monitoring Network in East Asia (EANET) is an intergovernmental regional network established for promoting cooperation among countries in East Asia to address acid deposition problems. The First Session of the Intergovernmental Meeting (IG1) on the EANET was held in March 1998 in Yokohama, Japan. Based on the agreement at the IG1, the EANET was started the preparatory phase activities in April 1998. On an interim basis, there were useful inputs to provide to the Second Session of the Intergovernmental Meeting (IG2) on the EANET which was held in 2000 in Niigata, Japan with participation of ten countries in East Asia to cover both the atmospheric and ecological environments. These countries were China, Indonesia, Japan, Malaysia, Mongolia, Philippines, Republic of Korea, Russia, Thailand and Viet Nam for the formal establishment of the Network.

The IG2 concluded that the preparatory phase activities of the EANET had been successful and decided to start the EANET activities as a regular basis from January 2001 based on the "Joint Announcement (JA) on the Implementation of the EANET" and the "Tentative Design (TD) of the EANET". The Meeting also designated the United Nations Environment Programme (UNEP) as the Secretariat and the Acid Deposition and Oxidant Research Center (ADORC) in Japan as the Network Center for the EANET respectively. The ADORC was renamed to Asia Center for Air Pollution Research (ACAP) in June 2010 taking into account of future prioritized research activities and so on. The Third Session of the Intergovernmental Meeting (IG3) was held in November 2001 in Chiang Mai, Thailand and the “Rules of Procedure for the EANET” were adopted. Since then, Sessions of the Intergovernmental Meetings (IGs) have been taken place annually following the Rules of Procedures. In 2001, 2002 and 2005, Cambodia, Lao PDR, and Myanmar have joined the EANET respectively, which now covers the thirteen countries in East Asia.

The objectives of the EANET are as follow:

(1) To create a common understanding of the state of acid deposition problems in East Asia;
(2) To provide useful inputs for decision-making at the local, national, and regional levels aimed at preventing, or reducing adverse impacts on the environment caused by acid deposition; and

(3) To contribute to cooperation on the issues related to acid deposition among the participating countries.

In general, the scientific activities of the EANET are implemented by the participating countries and the Network Center (NC) under the guidance of the Scientific Advisory Committee (SAC), and stipulated in the Instrument for Strengthening the Acid Deposition Monitoring Network in East Asia (EANET) (hereinafter "Instrument") adopted at the Twelfth Session of the Intergovernmental Meeting (IG12) on the EANET held in November 2010 in Niigata, Japan. And implementation of the activities is also in accordance with the "Medium Term Plan (MTP) for the EANET" and "Work Program and Budget of the EANET" each year approved at the Session of the Intergovernmental Meeting (IG) on the EANET annually held.

The EANET addresses the acid deposition including ozone and particulate matter (PM) issues in an integrated approach embarking on the following major activities:

1) Acid deposition monitoring
   a. Review and revision/establishment where appropriate of the national monitoring plans by the participating countries
   b. Implementation of monitoring using common methodologies by the participating countries
      (a) wet deposition, (b) dry deposition (air concentration), (c) soil/vegetation
      (d) inland aquatic environment, (e) catchment
2) Compilation, evaluation, storage, analysis and provision of data
   a. Submission of monitoring data to the Network Center from the participating countries
   b. Issuing of Data Report
   c. Periodic assessment report on the state of acid deposition
   d. Dissemination of the data and relevant information through EANET web site
3) Promotion of quality assurance and quality control (QA/QC) activities
   a. Development and implementation of QA/QC programs
   b. Development of Standard Operational Procedures (SOPs) by the participating countries
   c. Inter-laboratory comparison projects
4) Implementation of technical support and capacity building activities
   a. Dispatch of technical missions
   b. Individual training at the Network Center
   c. National training by the participating countries
   d. Utilization of existing training programs (JICA training course and others)
5) Promotion of research and studies related to acid deposition and air pollution problems
   a. Joint studies on wet/dry deposition, soil/vegetation and inland aquatic environment
   b. Joint studies on catchment analysis
   c. Fellowship Research Program at the Network Center
6) Promotion of public awareness activities
   a. Joint project on the development of the national brochures and the environmental studies
   b. Workshops on public awareness
7) Promotion of future development of the EANET and enhancement of international cooperation
   a. Discussion on future development of the EANET including expansion of its scope review and development and review of the Medium Term Plan (MTP) for the EANET
   b. Arranging various EANET sessions
   c. Cooperation and exchange of information and experiences with other regional and global networks/initiatives

d. Others

2. Recent scientific activities of the EANET

2.1 Acid deposition monitoring including QA/QC activities

(1) Compilation, verification, evaluation, storage and provision of data and relevant Information (Data Report)

The EANET data is submitted from the participating countries to the Network Center by the end of June every year. The EANET data is compiled and stored by the Network Center for the EANET and verified and evaluated by the experts in East Asian region. After compilation and evaluation, the Network Center develops draft Data Report. The contents of the Draft Data Report are discussed at the Sessions of Senior Technical Managers' Meeting and Scientific Advisory Committee (SAC) every year. After adoption at the Session of the SAC, the Network Center publishes the Data Report every year.

Recently the Draft Data Report 2014 based on the data and related information submitted by the participating countries was discussed at the Sixteenth Senior Technical Managers' Meeting (STM16) held in 2015, submitted to the Fifteenth Session of the SAC (SAC15) held in 2015 and adopted. After that, Data Report 2014 was published and distributed to the participating countries including National Focal Points (NFPs), National Centers, SAC members and other relevant organizations in November 2015.

In accordance with the "Detailed Mechanism of Article 4 of the Procedures on Data and Information Disclosure for the EANET" endorsed at the Fifth Session of the Intergovernmental Meeting (IG5) on the EANET in 2003, all the EANET monitoring data (Data Report) from 2000 up to 2014 were disclosed outside of the EANET. The raw data (including hourly data of air concentration/dry deposition) of the EANET in 2000-2013 was already disclosed outside of the EANET in January 2016 in accordance with the official request form of the EANET from outside.

(2) Development/Review of monitoring guidelines, technical manuals and strategy papers

The development of the Draft of the Strategy Paper for Future Direction of Air Pollution Effects on Agricultural Crops, Forest and Inland Water of EANET (2015-2020) was discussed at the Fourth Meeting of the Task Force on Soil and Vegetation Monitoring (TFSV4) of the Scientific Advisory Committee (SAC) held in June 2014 in Niigata, Japan. The Final Draft was submitted to the Fourteenth Session of the SAC (SAC14) in 2014 and the SAC14 adopted the "Strategy Paper for Future Direction of EANET on Monitoring of Effects on Agricultural Crops, Forest and Inland Water by Acidifying Species and Related Chemical Substances" with necessary corrections. The new strategy paper was printed in 2014 and sent to the EANET members.

Regarding the Strategy Paper on Future Direction of Monitoring for Dry Deposition, development of the Draft Strategy Paper on Future Direction of Monitoring for Dry Deposition of EANET was discussed at the Fourth Meeting of the Task Force on Monitoring for Dry Deposition (TFMDD4) of the SAC held in August 2015 in Niigata, Japan. The Final Draft was submitted to the Fifteenth Session of the SAC (SAC15) held in September – October 2015 in Da Nang, Viet Nam and the Session adopted the "Strategy Paper on Future Direction of Monitoring for Dry Deposition of EANET (2016-2020)", the revised Terms of Reference of the Task Force, and establishment of two Expert Groups which are necessary to revise the Technical Manual on Dry Deposition Flux Estimation in East Asia and the Technical Manual for Air Concentration Monitoring in East Asia.

(3) Implementation and coordination of QA/QC activities

(EANET Inter-laboratory comparison projects)

In order to promote quality assurance and quality control (QA/QC) activities, the EANET Inter-laboratory Comparison Project was started in 1998 during the preparatory phase by the Network Center. The Network Center distributed samples for the Inter-laboratory comparison projects on the wet deposition, the dry deposition (filter pack method), the soil and the inland aquatic environment to the participating countries in October/November every year. The results of the projects are discussed at the Senior Technical Managers' Meeting (STM) held in summer/autumn every year and submit to the Session of the SAC for adoption. After adoption, the Network Center publishes the Report of Inter-laboratory Comparison Projects and distributes it to the participating laboratories by the end of every year for evaluation and review of the results by the Network Center and the participating laboratories.

(Other Inter-laboratory comparison projects)

Several EANET laboratories in Japan, Philippines, Russia, Thailand, Viet Nam, etc. also participate in the other international inter-comparison projects organized by the Convention on Long-range Transboundary Air Pollution (CLRTAP), the International Cooperative Programme on Assessment and Monitoring of Acidification in Rivers and Lakes (ICP-Waters), and so on.

2.2 Research activities

(1) Fellowship research in 2013-2015

In response to the invitation from the Network Center, 7 applications from 3 countries namely Mongolia, Myanmar and Thailand in 2013, 4 applications from 4 countries namely Cambodia, Mongolia, Russia and Thailand in 2014, and applications from 4 countries namely Indonesia, Malaysia, Mongolia and Thailand in 2015 were submitted respectively. Among the applications, 2 researchers from Mongolia and Thailand in 2013, 2 researchers from Mongolia and Russia in 2014, and 2 researchers from Mongolia and Thailand in 2015 were selected respectively. The fellowship research in 2013-2015 was finished successfully, and all of research fellows already submitted their research reports to the Network Center. Their reports were reviewed by the reviewers nominated by the Task Force on Research Coordination (TFRC).

(Research themes of the fellowship research)

Research themes of the fellowship in 2013-2015 are the following:

i) 2013

Mongolia: Long range transport of air pollutants (trace gases) related with meteorological process in Asia

Thailand: Temporal distribution of atmospheric nitrogen deposition and correlation with biomass burning, Chiang Mai Thailand during 2008-2012

ii) 2014

Mongolia: Meteorological condition during the highly polluted and clean days in Ulaanbaatar, Mongolia

Russia: Investigation of long-term changes of air pollutants concentrations in North-East Asia including Russia using EANET monitoring data

iii) 2015

Mongolia: Chemical Characterization of PM in Ulaanbaatar, Mongolia

Thailand: Influence of long-range transport on air quality in northern part of Southeast Asia during open burning season

(2) Joint research project on catchment analysis with Thailand, Malaysia and Japan

Catchment analysis is one of the issues described in the Strategy Paper for Future Direction of Soil, Vegetation and related Ecosystems Monitoring of the EANET (2009-2014) and also the new strategy paper, "Strategy Paper for Future Direction of EANET on Monitoring of Effects on Agricultural Crops, Forest and Inland Water by Acidifying Species and Related Chemical Substances". The Network Center has been conducting the joint research projects on the catchment analysis with the Royal Forest Department (RFD) and the Environmental Research and Training Center (ERTC) in Thailand, with the University Putra Malaysia (UPM) and the Malaysian Meteorological Department (MMD) in Malaysia, and with Niigata Prefecture and Niigata University in Japan. The project was supported by Asia-Pacific Network for Global Change Research (APN, ARCP2013-13CMY-Sase) until July 2015, dynamics of sulfur derived from atmospheric deposition was studied utilizing S isotopic analysis in forest catchments in the countries above.

(3) International workshop on atmospheric modeling research in East Asia

The International Workshop on Atmospheric Modelling Research in East Asia was held once a year during 2010-2015 in China. At the workshop, the Work Plan for Model Inter-Comparison Study for Asia (MICS-Asia) Phase III, settings of model simulations including input data such as meteorological fields, initial/boundary conditions and emission inventories, and submission list of variables required for participant models were discussed. Preliminary analysis of inter-comparison of first submitted results was reported. The outcomes of the MICS-Asia Phase III activities will be useful to policy makers of countries in East Asia to identify high pollutant concentration areas.

(4) Modelling and emission inventory training workshop

A Modelling and Emission Inventory Training Workshop was held from 30 January to 3 February 2012. Around 20 participants from 11 EANET participating countries attended the training workshop. In the Workshop, instruction materials were provided to the participants as reference documents.

2.3 Technical support and capacity building activities

(1) Training for EANET participating countries

(a) EANET individual training

The EANET individual training course at the Network Center has been implemented once or twice a year from the beginning of the EANET activities. The training courses consist of wet deposition, dry deposition, soil/vegetation and inland aquatic environment monitoring, and data management. Recently, the course includes hands-on trainings of automatic monitoring on air concentration, responding to the requirement from the participating countries. 7 trainees were taken the individual training during 2013-2015 at the Network Center from Cambodia, Malaysia, Myanmar and Thailand. In addition, The Network Center also implemented the annual questionnaire survey on training activities conducted by the participating countries to gather information on training requirements and suggestions on new training areas.

(b) JICA country focused training course (for China)

The 1st JICA country focused training course on "Capacity Building on Planning Mitigation Measures for Ozone and $PM_{2.5}$" was held in June – July 2014 in Tokyo, Tsukuba and Kawasaki, Japan. During 10 days period, 9 trainees from China received 18 lectures, 4 sites visits and one hands-on training. In 2015, the 2nd training course was held in December. Based on the requirement from the

Ministry of Environmental Protection (MEP), China, ACAP arranged the course curriculum.

(2) Support to the participating countries of the EANET

(a) Dispatch of Technical Missions to the Participating Countries of the EANET

Recently 3 or 4 times a year, the Network Center has dispatched technical missions to the participating countries of the EANET. The mission was dispatched to Malaysia, Russia, Mongolia and China in 2013, to Indonesia, Malaysia, Philippines, Mongolia and Republic of Korea in 2014, to Indonesia, Philippines and Russia in 2015 to exchange information, provide technical advice, calibrate ozone monitors in particular country and observe operation of EANET monitoring sites. Condition of the onsite and lab's instruments provided to countries were also checked at that time.

(b) Technical support to the participating countries of the EANET

Technical assistance was also provided by the Network Center to Cambodia, China, Indonesia, Mongolia, Myanmar, Russia and Viet Nam in 2014 and/or 2015 for their monitoring activities including dissemination of the latest information, provision of equipment and its spare parts and consumables such as filter pack kit, standard solutions and so on.

2.4 Dissemination of the latest information to the participating countries

The Network Center with the Secretariat maintained close communication with the participating countries in all related activities of the EANET as well as relevant international organizations, resource institutions and programs such as the United Nations Environment Programme (UNEP); United Nations Economic and Social Commission for the Asia and the Pacific (UNESCAP); United Nations Economic Commission for Europe(UNECE), Convention on Long-range Transboundary Air Pollution (UNECE CLRTAP) and its programme including the Cooperative Programme for Monitoring and Evaluation of the Long Range Transmission of Air Pollutant in Europe (EMEP); Stockholm Environment Institute (SEI); Climate and Clean Air Coalition to Reduce Short-Lived Climate Pollutants Initiative (CCAC); Clean Air Asia (CAA); Governmental Meeting on Urban Air Quality in Asia; Malé Declaration on Control and Prevention of Air Pollution and Its Likely Transboundary Effects for South Asia (Malé Declaration); Joint Forum on Clean Air for Asia and the Pacific (Joint Forum); Atmospheric Brown Cloud (ABC) Project; North-East Asian Sub-regional Programme for Environmental Cooperation (NEASPEC); the Long-range Transboundary Pollutants (LTP) in Northeast Asia; Asian Co-benefits Partnership (ACP); German International Cooperation (GIZ); Institute for Global Environmental Strategies (IGES); etc. The Network Center communicated with participating countries also regarding scientific and technical issues to improve monitoring and assessment of acid deposition and related air pollution in the region.

New information has been uploaded on the EANET website such as publication (e.g. Data Report and Inter-laboratory comparison projects), EANET Newsletters, EANET meeting reports, monitoring site information, and so on. The Third Report for Policy Makers (RPM3) "EANET and Clean Air for Sustainable Development" was finalized after 2 years discussion and published in December 2014. The RPM3 was uploaded to the EANET website and distributed to the participating countries and other relevant institutions.

2.5 Discussion on expansion of the scope of the EANET

After development of the "Instrument for Strengthening the Acid Deposition Monitoring Network in East Asia (EANET) (hereinafter "Instrument")", discussion on scope expansion of the EANET was started in 2011 at the EANET Sessions. During discussion, following views and comments

were expressed.

(a) Scientific evidence is regarded as an important basis to consider the future priorities. In order to discuss the expansion of the scope of the EANET, it is necessary to review the present status of air pollution in East Asia.
(b) Clean air technologies (CAT) should be considered as one of the important areas of cooperation using the EANET as a platform.
(c) The Convention on Long-range Transboundary Air Pollution (CLRTAP) and its eight associated protocols provide a useful example of science-based instruments.
(d) Although studies on modeling and emission inventories are included in the present scope of the EANET as research activities, further development on modeling and emission inventory is essential to move on to the next step for better assessment of regional air pollution and so on.

(1) Development and publishing Report of "Review on the State of Air Pollution in East Asia"

Taking into account of the view of (a) above, etc., the "Reviewing Committee on the Status of Air Pollution in East Asia (RC)"was established under the Task Force on Research Coordination (TFRC) in February 2013. After accumulation of the data in East Asia relevant to air pollution and impacts on ecology and human health, etc, and several times discussion at the Reviewing Committee was made, finally the Reviewing Committee developed "Review on the State of Air Pollution in East Asia" in February 2015 and reported to the Task Force Meeting and the Session of the SAC in 2014 (SAC14). Contents of the Chapter 9 "Conclusions and Recommendations" in the Report of "Review on the State of Air Pollution in East Asia" officially developed by the TFRC is attached as Annex 1. Development of this report was also described in "2.6 (3)(d) Task Force on Research Coordination".

(2) (Pre-)Feasibility on the Expansion of the Scope of the EANET

Based on the discussion on EANET scope expansion at the EANET Sessions in 2011-2013 and reviews on the state of air pollution in East Asia, and also in accordance with the discussion at the Fifteenth Session of the Intergovernmental Meeting (IG15) in December 2013, Report of the Prefeasibility Study on the Expansion of the Scope of the EANET was developed in 2014 by the Network Center.

Based on the discussion at the Sixteenth Intergovernmental Meeting (IG16) in November 2014, Draft Report of the Feasibility Study on the Expansion of the Scope of the EANET was developed by the Network Center and discussion was made at the Fourteenth Session of the Working Group on Future Development of the EANET (WGFD14) held in June 2015 in Thailand. The Draft Report included 3 Categories on the expansion of the scope of the EANET shown below and the Session provided guidance on the Report to be used as a basis for the development of the MTP on the EANET (2016-2020).

<u>Category 1: Strengthening current monitoring activities</u>

1) Promotion of the monitoring of ozone and $PM_{2.5}$ including research cooperation and communication on the results as appropriate;
2) Capacity building activities especially for ozone and $PM_{2.5}$ monitoring;
3) Increasing number of EANET monitoring sites; and
4) Strengthening understanding and capacity for monitoring of meteorological parameters and reporting on these as appropriate.

<u>Category 2: Promotion of activities other than monitoring</u>

1) Promotion of information exchange: Information exchange on clean air technologies and regional impact assessment through workshops, seminars, etc.;

2) Promotion of research and technical cooperation on emission inventory to support the capacity building: Research on technical guidelines on emission inventory to support the capacity building, and capacity building for emission inventories;
3) Promotion of research activities: Research activities on regional impact assessment to plants and ecosystems; and Research activities on measurement methodology for elemental carbon (EC) and organic carbon (OC).

Category 3: Long term future activities

1) Monitoring of EC, OC, VOC (volatile organic compounds) and other related items as appropriate;
2) Submission of the emission inventory developed by each participating country;
3) Transfer following present Additional budget activities to Core activities of the Network Center;
   - Technical support to the participating countries including capacity building activities.
4) Name change of the network from "Acid Deposition Monitoring Network in East Asia (EANET)" to "Air Pollution Monitoring Network in East Asia (EANET)".

Draft Medium Term Plan for the EANET (2016-2020) with the conclusion that the extended activities listed in Category 1 and 2 above to be incorporated into the revised draft MTP for the EANET (2016-2020).

The finalized document of the Feasibility on the Expansion of the Scope of the EANET was submitted to the Sessions of SAC in 2015 (SAC15) and the IG in 2015 (IG17). Major contents of the Feasibility Study on the Expansion of the Scope of the EANET is attached as Annex 2. Revised Medium Term for the EANET which includes the activities of Category 1 and 2 was approved at the IG17 in November 2015.

2.6 Discussion on scientific issues at the recent Sessions of the EANET

At the recent Sessions of the EANET, various issues which include scientific aspects have been discussed, such as promotion of ozone and PM monitoring, quality assurance and quality control (QA/QC) activities, data disclosure to outside of the EANET and data sharing with other organizations, revision of the national monitoring plans (NMPs), development of periodic report on the state of acid deposition in East Asia, development of Strategy paper for future direction of the EANET monitoring, ongoing EANET research activities, future high priority research projects, future expansion of the scope of the EANET and so on. Major scientific issues discussed at recent each EANET Session are the below.

(1) Senior Technical Managers' Meeting (STM)

The senior technical staff (senior technical managers) of the National Centers in the participating countries of the EANET discusses the EANET data and the relevant technical and scientific issues at the EANET Senior Technical Managers' Meeting (STM). The latest STM, Sixteenth Senior Technical Managers' Meeting (STM16) on the EANET was held in Ulaanbaatar, Mongolia in August 2015. At the STM16, major discussion items were the following:

(a) Report on the progress of the EANET since STM15;
(b) Overview of the preliminary draft Data Report 2014;
(c) Evaluation for the results of the Inter-laboratory Comparison Projects in 2014;
(d) Consideration of the National Monitoring Plans (NMPs) and current monitoring activities for the EANET monitoring of the Participating Countries; and
(e) Other issues.

At the STM16, current monitoring activities for the EANET of the participating countries was discussed. And it was emphasized the important consideration of PM and ozone monitoring within the present scope of the EANET to be included into the National Monitoring Plan (NMP), so the QA/QC process and capacity building program for these pollutants could be schedulable in collaboration with the Network Center.

The participants of STM16 shared the current status on air pollution monitoring in Mongolia presented by Mongolia. In addition, the Network Center presented proposal of modification of curriculum of training activities to include automatic Ozone/PM monitoring technique/knowledge.

(2) Scientific Advisory Committee (SAC)

In accordance with the "Instrument", the SAC, composed of two scientific and technical experts from each participating country, will advise and assist the IG with various scientific and technical matters related to the EANET activities as mandated to it by the IG. These matters include the following:

(a) scientific and technical aspects for the EANET;
(b) development and revision of the monitoring guidelines and technical manuals;
(c) matters related to the selection of monitoring sites, QA/QC programs, data reporting procedures and formats;
(d) matters related to collection, evaluation, assessment and analysis of monitoring data;
(e) preparation of periodic assessment reports on the state of acid deposition in East Asia, based on, but not limited to the data compiled by the Network Center;
(f) matters related to studies on acid deposition; and
(g) other scientific matters as requested by the IG.

In accordance with the Instrument, the Session of the SAC discusses various scientific and technical issues of the EANET. The latest Session of the SAC, the Fifteenth Session of the SAC (SAC15) held in Viet Nam in 2015 also discussed many important scientific and technical issues including the following:

- Review of the Report on the Progress of the EANET since the Fourteenth Session of the SAC (SAC14) and the Financial Report in 2014 from scientific and technical viewpoints.
- Adoption of the EANET Data Report 2014
- Adoption of the Report of the Inter-laboratory Comparison Projects 2014
- Overview of the Updated National Monitoring Plans (NMPs) of the Participating Countries
- Adoption of the "Strategy Paper on Future Direction of Monitoring for Dry Deposition of EANET (2016-2020)", the revised Terms of Reference of the Task Force, and establishment of two Expert Groups which will consider revision of the Technical Manual on Dry Deposition Flux Estimation in East Asia and the Technical Manual for Air Concentration Monitoring in East Asia.
- Review on the Report of the Feasibility Study on the Expansion of the Scope of the EANET.
- Consideration of the Report of the Third Periodic Report on the State of Acid Deposition in East Asia (PRSAD3)
- Consideration of the Draft Medium Term Plan (MTP) for the EANET (2016-2020) from scientific and technical viewpoints
- Consideration of the Relevant Scientific Activities including the Progress of the Joint Research Activities on Model Inter-comparison Study in Asia (MICS-Asia) and Joint Research Project on Sulfur Dynamics in Forest Ecosystems in Thailand, Malaysia and Japan.
- Consideration of the cooperation with WMO/GAW.

- Consideration of the Draft Work Program and Budget of the EANET in 2016 from Scientific and Technical Viewpoints
- Other Issues

During the SAC15, 1) It was informed that WMO has the standard document for meteorological monitoring and observation by meteorological agencies in their respective countries and the activities have been conducted according to the official procedures. 2) The Session took note of the document of the Feasibility on the Expansion of the Scope of the EANET. 3) It was recommended to consider including existing monitoring stations of $PM_{2.5}$ and ozone in the participating countries to the EANET network in order to conduct regional assessment. The Network Center will initiate discussion with the countries on this regard. 4) It was clarified that Executive Summary would be prepared for the Third Periodic Assessment Report in order to introduce major outcomes to policy makers. It was stressed that Report for Policy Makers (RPM) would be prepared for such specific purpose, as decided by the IG. 5) It is suggested to promote research activities of measurement methodology including elemental carbon (EC) and organic carbon (OC) because there are perceived discrepancies of measured values among different measurement protocols. It is suggested that it is important to take into consideration existing methodologies in scientific knowledge worldwide.

(3) Task Forces and Expert Groups established under the SAC

According to the "Procedures for establishing Task Forces and Expert Groups under the SAC of the EANET" in the "Guidelines on Administrative and Financial Management for the Secretariat and the Network Center (latest one is Annex 4 of the Report of the Session of the IG14 (EANET/IG 14/14), Task Forces and Expert Groups can be established in accordance with the Decision of the Intergovernmental Meeting (IG). As of 2016, following four Task Forces have been established:

- Task Force on Monitoring for Dry Deposition
- Task Force on Soil and Vegetation Monitoring
- Task Force on Monitoring Instrumentation
- Task Force on Research Coordination

In addition, under the Task Force on Monitoring for Dry Deposition, Expert Group on Preparation of Technical Manual for Air Concentration Monitoring was established. As of 2016, totally five Task Forces and Expert Group have been established.

(a) Task Force on Monitoring for Dry Deposition

The members of the Task Force on Monitoring for Dry Deposition were nominated by the National Focal Points (NFPs) of the EANET and appointed by the Chairperson, and the Task Force has been recently discussing the Terms of Reference of the Task Force, the Strategy paper on future direction of monitoring for dry deposition of the EANET (2016-2020), Technical manual for air concentration monitoring, Technical manual for dry deposition flux estimation, etc. All these documents were discussed carefully by the EANET experts and adopted at the Sessions of the SAC and endorsed by the Sessions of the IG.

As mentioned in the paragraph above, the Expert Group on Preparation of Technical Manual for Air Concentration Monitoring was established under this Task Force. The Expert Group discussed the draft Technical manual for air concentration monitoring for a few years, finally it was adopted in 2013 and published. However a few years have already passed after the publishing, the contents has to be revised e.g., because at the Session of the SAC it was suggested that the latest scientific and technical

information should be accumulated and included, etc. In this regard, it will be planned to revise this manual and also Technical manual for dry deposition flux estimation by this Expert Group or to establish other new Expert Group(s) under the Task Force in the near future.

(b) Task Force on Soil and Vegetation Monitoring

The members of the Task Force on Soil and Vegetation Monitoring were directly appointed by the Chairperson of the Task Force. Recently Task Force on Soil and Vegetation Monitoring has been discussing the "Strategy Paper of Future Direction of EANET on Monitoring of Effects on Agricultural Crops, Forest and Inland Water by Acidifying Species and Related Chemical Substances", including risk maps on acidification of soil or inland water as an example of "identification of the areas susceptible to acid deposition" as one of the specific activities in the strategy paper. The strategy paper was adopted at the Session of the SAC in 2014 (SAC14) and finally endorsed following Session of the IG in 2014 (IG16).

(c) Task Force on Monitoring Instrumentation

The members of the Task Force on Monitoring Instrumentation were nominated by the National Focal Points (NFPs) of the EANET and appointed by the Chairperson. The main issues of the Task Force are to discuss situation of monitoring equipment of the sites and labs and other relevant issues. Since recently these issues have been discussed at the STM Meetings, the Task Force Meeting has not been held for a few years.

(d) Task Force on Research Coordination

The members of the Task Force on Research Coordination were directly appointed by the Chairperson of the Task Force. Recently Task Force on Research Coordination has been discussing 1) Review of the fellowship research reports, 2) Future high priority research projects including establishing informal small group for creating research proposals of high priority themes to apply potential external funding sources or donor agency", 3) Discussion on review on the present status of air pollution in East Asia taking into account of the expansion of the scope of the EANET, this issue was discussed at the Session of the SAC. As the result of discussions, the SAC in 2011 (SAC11) agreed to refer the review of the status of air pollution in East Asia to the Task Force on Research Coordination (TFRC). Based on the discussion of this issue Reviewing Committee on the Status of Air Pollution in East Asia was established in February 2013.

(Development of draft research proposals to the external funding agencies)

Taking into account of acquisition of external fund from (international) funding agencies, importance of appropriate application on serious air quality issues such as ozone and PM to the funding agency was pointed out at the Session of the SAC in 2012 (SAC12). Based on the suggestion to establish informal small group to develop research proposals at the Sessions of the SAC in 2012 (SAC12) and SAC in 2013 (SAC13), the Network Center established the Research Planning Group (RPG) which consisted of 5 scientists from 4 countries, and held its First Meeting in Niigata in May 2014. After the meeting draft proposals on $PM_{2.5}$ and SLCPs (Short-Lived Climate Pollutants) were developed for future further consideration.

(Review on the State of Air Pollution in East Asia)

Taking into account of the provision of the Instrument, “the scope of this Instrument may be extended, as decided by the Intergovernmental Meeting (IG)”, “Discussion on the issues of expansion of the scope of the EANET” was implemented in 2011-2015 at the Sessions of the WGFD, SAC and IG. The Session of the SAC in 2011 (SAC11) agreed to refer the review of the status of air pollution in East Asia to the Task Force on Research Coordination (TFRC). The IG in 2011 (IG13) also took note that the TFRC would review the status of air pollution in East Asia. Based on the discussion at the SAC and IG described above, the TFRC in November 2012 agreed that the review of the status of air pollution in East Asia would be implemented by the “Reviewing Committee on the Status of Air Pollution in East Asia (RC)”. Because Scientific evidence is regarded as an important basis to consider the future priorities. Finally the Reviewing committee which consists of several experts on atmospheric environment and ecological impact assessment issues was established in Feb. 2013 under the Task Force. The Reviewing Committee developed “Review on the State of Air Pollution in East Asia” in Feb. 2015 and reported to the Task Force Meeting and the Session of the SAC in 2014 (SAC14).

(4) Drafting committee (DC) for the Third Periodic Report on the State of Acid Deposition in East Asia (PRSAD3)

The First Periodic Report on the State of Acid Deposition in East Asia (PRSAD1) was developed by the SAC in 2006 (SAC6), and the Second Periodic Report on the State of Acid Deposition in East Asia (PRSAD2) was developed by the SAC in 2011 (SAC11). In order to draft Third PRSAD, Drafting committee (DC) for the Third Periodic Report on the State of Acid Deposition in East Asia (PRSAD3) was established after adoption at the SAC and endorsement at the IG in 2014. All the members of the DC for the PRSAD3 were nominated by the National Focal Points of the EANET (NFPs). The PRSAD3 will consist of “Part I Regional Report”, “Part II National Reports” and “Part III Executive Summary”. The Table of the contents of the Part I Regional Report of the PRSAD3 is as follows:

- Chapter 1: Introduction - Chapter 2: Data quality
- Chapter 3: Precipitation chemistry - Chapter 4: Trace gas and aerosol concentration
- Chapter 5: Wet and dry deposition - Chapter 6: Impacts of atmospheric deposition on ecosystem
- Chapter 7: Summary and recommendations for future activities

(5) Working Group on Future Development of the EANET (WGFD))

Although the WGFD is not stipulated in the Instrument, it was established in accordance with the Decision of the Intergovernmental Meeting (IG) on the EANET. The tasks of the WGFD in 2013-2015 included scientific and technical issues are the following:

(1. General Tasks)

(a) Review the implementation of the Instrument for Strengthening the EANET;
(b) Review the Guidelines on Administrative and Financial Management for the Secretariat and the Network Center adopted at the IG14 in 2012;
(c) Review the Work Program and Budget, Progress Report, Financial Report and Audit Report for the IG’s adoption; and
(d) Guide the Secretariat and the Network Center on public awareness and capacity building activities.

(2. Specific Tasks)

(a) Review the draft Mid-Term Report on the implementation of the MTP (2011-2015);
(b) Review the draft Third Report for Policy Makers;
(c) Review of the Draft Report of the Review on the Status of Air Pollution in East Asia;
(d) Review the issues of the future development of the EANET, including the following;

- the institutional arrangement of the EANET Secretariat, and
- the future expansion of the scope of the EANET;

(e) Review the performance of the Secretariat and the Network Center for 2012-2013;
(f) Review the strategies and action plan in obtaining external funding from international organizations and other funding agencies;
(g) Review the draft Report on the Implementation of the MTP for the EANET (2011-2015); and
(h) Review the draft MTP for the EANET (2016-2020).
(3. Other tasks requested by the IG, as appropriate)

The latest WGFD Session, the Fourteenth Session of the WGFD (WGFD14) held in June 2015 in Thailand included the following issues:

- The Session discussed the Draft Financial Report of the Secretariat and the Network Center in 2014 to be submitted to the IG17;
- The Session also considered the Revision of the Revised Guidelines on the Administrative and Financial Management for the Secretariat and the Network Center including calculating voluntary financial contributions expected from the participating countries and making revision and improvement of the working procedure of the SAC as appropriate; and
- The Session provided guidance on the Report of the Feasibility Study on the Expansion of the Scope of the EANET and agreed that the extended activities listed below will be incorporated into the next MTP for the EANET (2016-2020).

Category 1: Strengthening current monitoring activities

1) Promotion of the monitoring of ozone and $PM_{2.5}$ including research cooperation and communication on the results as appropriate;
2) Capacity building activities especially for ozone and $PM_{2.5}$ monitoring;
3) Increasing number of EANET monitoring sites; and
4) Strengthening understanding and capacity for monitoring of meteorological parameters and reporting on these as appropriate.

Category 2: Promotion of activities other than monitoring

1) Promotion of information exchange: Information exchange on clean air technologies and regional impact assessment through workshops, seminars, etc.;
2) Promotion of research and technical cooperation on emission inventory to support the capacity building: Research on technical guidelines on emission inventory to support the capacity building, and capacity building for emission inventories;
3) Promotion of research activities: Research activities on regional impact assessment to plants and ecosystems; and Research activities on measurement methodology for elemental carbon (EC) and organic carbon (OC).

(6) Intergovernmental Meeting (IG)

Since the Session of the IG is mainly held for decision making, scientific issues have been fundamentally discussed at the Sessions of the SAC. However the discussion of the Sessions of the IG sometimes includes scientific issues, e.g., recently expansion of the scope of the EANET which includes scientific issues was also one of the important discussion issues of the Sessions of the IG. The major discussion items at the latest Session of the IG in 2015 (IG17) are the following:

(a) Consideration on the Report on the Progress of Acid Deposition Monitoring Network in East Asia (EANET) since the Sixteenth Session of the IG (IG16) and the Financial Report of the Secretariat and the Network Center in 2014;
(b) Consideration on the Report on the Outcomes of the Fifteenth Session of the Scientific Advisory Committee (SAC15);
(c) Consideration on the Framework Document on the Arrangements of the Secretariat for the Acid Deposition Monitoring Network in East Asia (EANET) between UNEP and the EANET;
(d) Consideration on cost saving measures for implementation of EANET activities including the future of the Working Group on Future Development (WGFD) of the EANET;
(e) Consideration on the Draft Medium Term Plan for the EANET (2016-2020);
(f) Consideration on the Revision of the Revised Guidelines on the Administrative and Financial Management for the Secretariat and the Network Center;
(g) Consideration on the Draft Work Program and Budget of the EANET in 2016; and
(h) Updates on Activities of Other International Programs related to Acid Deposition and Transboundary Air Pollution:
    - Information on activities under Convention on Long-range Transboundary Air Pollution/ Co-operative Programme for Monitoring and Evaluation of the Long Range Transmission of Air Pollutants in Europe (CLRTAP/EMEP) of relevance for the EANET
    - Asia Pacific Clean Air Partnership (APCAP)

3. Summary and recommendation

Although the EANET monitoring of acid deposition (including ozone and PM) has been implemented among the participating countries of the EANET since the commencement of the EANET regular phase activities with cooperative effort and produced brilliant achievements, as also described in the attached Feasibility Study Report, some serious issues in East Asia and the world still have been pointed out, e.g., air pollutants in East Asian region have already exceeded those of Europe and North America since the end of the last century, intercontinental transport of ozone and aerosols ($PM_{2.5}$ episodes in Northeast Asia and haze episodes in Southeast Asia) has been also recognized as important issues from the viewpoint of hemispherical air pollution as well as an outbreak of strong public concern on haze and $PM_{2.5}$ in East Asia as WHO already reported that around 7 million people died as a result of air pollution exposure in 2012 and so on.

It is important for the EANET in the future to enable continuation of benefits to the participating countries and to further contribute to national, regional and global efforts to improve air quality. In this respect, the activities consisted of a continuation of the long-term activities and included new projects that would take into consideration the changing situation and address emerging issues in East Asia. It also includes provisions for continuous assessments and reporting on the status of acidification to policy makers and implementation of appropriate studies to evaluate impacts of acid deposition and other priority chemical species.Taking into account of the current serious situation above, although the objectives of the present EANET have not targeted the mitigation of acid deposition in this moment, if the EANET could address these serious problems appropriately to mitigate air pollution, much progress would be expected in the future. In this regard, as a first step it is appropriate to make best effort to create an epistemic community with a common understanding on the state of air pollution in East Asia through promotion of information sharing, cooperation among the participating countries and so on for future expansion of the scope of the EANET and establishment of an appropriate

mechanism of mitigation of the air pollutants in East Asia.

Annex 1

Major Contents of the "Review on the State of Air Pollution in East Asia" developed in 2014 by the Task Force on Research Coordination under the SAC of the EANET

Chapter 9 Conclusions and Recommendations

For the past 50 years, Asia has been a home for the world's fastest economic growing countries, starting from Japan to Korea, Singapore, and now to China, and India, two of the fastest growing major economies. East Asian and Southeast Asian countries' economies heavily depend on manufacturing, emitting various air pollutants in a large quantity. This report reviews the present status of air pollution and their environmental impacts focused mainly on East Asia and Southeast Asia. Specifically, the present status of particulate matter (PM) and haze, oxidants, and acidification and eutrophication, including their environmental impacts, are reviewed. In addition, air toxics and climate-air pollution interaction are included as the more recent atmospheric environmental issues.

Particulate Matter (PM) and Haze

It was estimated that the PM level could be reduced to a safer level, and about 300,000-700,000 premature deaths could be prevented in developing countries. The seriousness of PM pollution in East and Southeast Asian countries has been unveiled by a series of international field campaigns, such as the Indian Ocean Experiment (INDOEX), Aerosol Characterization Experiment (ACE)-Asia, the East Asian Study of Tropospheric Aerosols and their Impact on Regional Climate (EAST - AIRC), and Atmospheric Brown Cloud (ABC). The East and Southeast are identified as one of the five major regional hot spots where anthropogenic aerosol optical depth exceeded 0.3 and the absorption AOD exceeded 0.03.

Atmospheric measurements have developed quickly in East Asia during the past decade. However, the number of monitoring sites is still limited in the East and to make things worse only part of the monitoring data is accessible. Additionally, the official monitoring mainly focuses on the routine measurement of regular pollutants but rarely focuses on the atmospheric chemistry. The long-term measurement data is also very limited in most countries of East Asia.

Currently, EANET set up their measurement sites mainly in Japan and Southeast Asia. There are only several sites in China, which contributed to the largest pollutant emission in East Asia, of which most of are background sites. It is suggested that additional sites will be set up in some proper locations in the North China Plain and East China, where the strongest human activities occur.

As for the aerosol measurement within the framework of EANET, carbonaceous aerosol measurement is suggested to be added as carbonaceous aerosol is playing a more dominant role in the formation of haze. The sampling resolution of filters at most EANET sites is from weekly to bi-weekly. The coarse time resolution could cause smoothing of typical short pollution episodes and difficulties of analyzing temporal variations, employment for model evaluation, and so on.

In East Asia, rapid industrialization and an increase in automotive vehicles on the continent have contributed to the recent increases of PM concentrations. This increase subsequently has increased the PM levels of downstream countries. The health effect attributable to transboundary PM pollution has not been well investigated.

The chemical composition of PM varies with areas and emission sources, hence the study on

the effect of PM chemical composition should be investigated for different areas. This type of investigation also makes it possible to distinguish local PM and transboundary PM, which leads to the study on transboundary PM attribution.

Studies on long term effects of PM in East Asia showed inconsistent results, partly because the studies were not designed for evaluating air pollution effects. Cohort studies specifically designed for air pollution effect evaluation with sufficient size are needed.

Tropospheric Ozone

Together with PM, ozone frequently violates the WHO guidelines causing a significant adverse impact on human health and vegetation. Regional-scale distributions and seasonal cycles along with controlling factors are well understood by enhanced observational evidence and model analysis in East and Southeast Asia. General consensus has been reached that long-term trends in East Asia are increasing. Despite the recent advances in ozone monitoring and modeling, there are still a number unresolved problems as listed below.

- The long term trends were not quantitatively reproduced by state-of-science models incorporating the latest emissions inventories.
- Intranasal variations have not been sufficiently explored.
- Monitoring of ozone precursors and tracers need to be enhanced, particularly free tropospheric data, which is very limited, needs to increase.
- The impacts of climate change on the source-receptor relationship needs to be explored more.

Multi-country collaboration for studies with consistent methodology are necessary. The study should include a sufficient number of cities, including those with negative or non-significant ozone effect.

The adverse impact of ozone on vegetation is deemed to be serious with possible increasing trends to make it necessary to take measures to minimize the ozone induced reduction in agricultural production in Asia. However, experimental and field studies are substantially lacking neither to ascertain the above assessment nor to contemplate efficient control measures. The species or cultivar difference in the sensitivity to the ozone should be considered to evaluate the present and future effects of the ozone on crops and the critical levels of ozone for protecting agricultural production in Asia. A similar study is also necessary to forest tree species with the additional study on the combined effects of the ozone and other environmental stresses on Asian forest tree species.

Acidification and Eutrophication

The acid deposition has been studied extensively for the characterization of precipitation and the impact assessment on soil and vegetation by field measurement and for the analysis of acid deposition phenomena and source-receptor relations by modeling since the 1980s. It has been reported that a significant decrease of soil pH in the last 20 years or so in China and Japan was probably caused by acid rain. Despite these studies, soil acidification manifested by field observations in East Asia are still very limited to draw a reliable scientific conclusion.

Based on input–output measurements conducted in Japan and China, a majority of ecosystems have a high nitrogen retention ability, whereas nitrogen saturation occurred in some particular forested ecosystems. Spatial distribution of nitrate concentrations in stream water in Japan showed that there were some areas with high concentrations in the vicinity of large cities and intensive agricultural areas, and the area with the highest concentrations were located on the air mass advection path from central Tokyo.

The field measurements and modeling in the 1990s and 2000s, and the recent studies in the 2010s all noted that the sulfur and nitrogen depositions have increased in China and Japan from the 1980s to 2000s most likely due to increased emissions of SO_2 and NO_x in China. Recently, some researchers suggested that sulfur deposition might start to decrease as early as after 2006.

The long-term data on inland water chemistry is limited in the Asian region. Re-measurement of previously surveyed points or assessment of the public data on water quality may be informative to evaluate the current situation on inland water acidification and nitrogen leaching. Regional scale evaluations of acidification and nitrogen leaching are necessary. It should be useful to analyze the causal relationship between regional deposition and impacts by the collaboration of atmospheric modeling and impact research.

A significant advance has been made in acid deposition monitoring and modeling in the past 20 years or so in East Asia. However, there are large differences among the reported source-receptor analysis results and the parameterizations of individual processes still heavily rely on the United States' and Europe's studies. On the other hand, there are only limited studies in literature for Southeast Asia. In addition, collecting data on biogeochemical cycles especially in Southeast Asia and analysis on interactions between nitrogen and carbon cycles are recommended as a future initiative.

Air Toxics

The POPs concentration in environmental samples have been evaluated under the Global Monitoring Plan of the Stockholm Convention (GMP). Japan organized the POPs Monitoring Project in East Asian Countries (POPSEA project) from 2002. Participating countries are Cambodia, Indonesia, Japan, Republic of Korea, Lao PDR, Malaysia, Mongolia, Philippines, Singapore, Thailand and Viet Nam. These studies demonstrated that the environmental POPs level and emission strength are assessed to not be significant, which may cause only a moderate environmental impact.

The monitoring methodologies have already been established in advanced countries (Japan, South Korea, Singapore and China). However, the amount of POPs monitoring data in the East Asia sub-region is still low and the preparation of the emission inventory is not enough. The preparation of detailed emission inventory should be expected to consider trans-boundary pollution and establishing the measure against POPs issues.

Emission Inventories

East and Southeast Asia became the regions of the world with the largest air pollutant emissions surpassing those in Europe and North America. The natural and anthropogenic emissions in this important region are only estimated with large uncertainties despite enormous international and national efforts.

The uncertainties of biogenic volatile organic compounds (VOC) emission inventories are very high in the Asian region. More and continuous studies to understand and evaluate the fundamental model and key parameters of biogenic emission estimations are required at many places. The Asian dust emission could be varied significantly depending on the used model and meteorological conditions. It is desirable for more reasonable estimations of emission amounts to use an ensemble average of several different dust emission models for several years.

In order to better understand the state of biomass burning emissions in East Asia, especially in Southeast Asia, field campaigns/experiments for measurement of regional specific parameters of fires are needed. Additionally, Asian databases of biomass burning emissions as well as their fundamental information, such as type of biomes and fires, combustion completeness, and environmental conditions,

should be set up.

The quality of anthropogenic emission inventory strongly depends on the accuracy of activity data and emission factors. However, studies for emissions, especially in Southeast Asia and other developing countries, are still limited and thus, uncertainties of their emissions are large. Therefore, it is recommended that research activities (e.g., survey of detailed activity data and region-specific emission factors) and capacity building for their activities are required in many countries, especially in Southeast Asia.

Verification, improvement and updates of anthropogenic and natural emission inventories based on ground and satellite observations, chemical transport modeling and inverse modeling are essential in reducing uncertainties. This work requires strong collaborative and integrated research among emission inventory developing, monitoring, chemical transport modeling, and inverse modeling groups.

Mitigation Measures of air pollutant emissions in East Asia

The major countries and regions in East Asia (e.g., China, Japan, and South Korea) have implemented considerable policies for energy conservation and air pollution mitigation. Particularly, the Chinese government sets targets to reduce energy consumption per unit GDP and total SO_2 emissions by 20% and 10%, respectively, measured in 2010 against 2005 levels. In the long term, China aims to reduce CO_2 emissions per unit GDP by 40-45% in 2020 compared with 2005 levels. Japan has taken substantial actions to achieve the target of a 6% reduction in greenhouse gas emissions from 1990 to 2008-2012, required by the Kyoto Protocol.

During 2005-2010, the emissions of SO_2 and $PM_{2.5}$ in East Asia decreased by 15% and 11%, respectively, mainly attributable to the large scale deployment of FGD for China's power plants, and the promotion of high-efficient PM removal technologies in China's power plants and cement industry. During this period, the emissions of NO_X and NMVOC increased by 25% and 15%, driven by the rapid increase in the emissions from China owing to inadequate control strategies. In contrast, the NO_X and NMVOC emissions in East Asia except China decreased by 13-17% mainly due to the implementation of tight vehicle emission standards in Japan and South Korea.

The successful implementation of the control policies set in China's 12^{th} Five Year Plan, the recently released emission standards for various industrial sources, and slowly strengthened control measures after 2015 (defined as a "progressive" end-of-pipe control strategy) could reduce China's emissions of NO_X, SO_2, and $PM_{2.5}$ significantly. The resulted NO_X, SO_2, and $PM_{2.5}$ emissions would be 16-26% lower than the 2010 levels by 2020, and even lower by 2030. Therefore, it is essential to support and monitor the progress of the implementation of these legislations.

The contributions of advanced energy saving measures to the reduction of SO_2 and $PM_{2.5}$ emissions exceed those of progressive end-of-pipe control measures by 2030. The energy saving measures would play an irreplaceable role for further reduction of air pollutant emissions.

The full application of best available technologies could reduce the emissions of NO_X, SO_2, and $PM_{2.5}$ in East Asia to only about one-quarter and NMVOC to one-third of the levels of the baseline projection. There are still large reduction potentials for major air pollutants using currently known control technologies.

Annex 2

Major Contents of the "Feasibility Study on the Expansion of the Scope of the EANET" (EANET/SAC 15/11)

II. Important issues for further improvement of atmospheric environment in East Asia

18. The First Session of the United Nations Environment Assembly (UNEA) of the United Nations Environment Programme (UNEP) held on 23-27 June 2014 in Nairobi Kenya adopted a resolution on strengthening the role of UNEP in promoting air quality issues. It recognizes that poor air quality is a growing challenge in the context of sustainable development in particular related to health in cities and urban areas, and that efforts across sectors to improve air quality are needed, and encourages Governments to take action across sectors to improve air quality to protect human health and the environment, reduce negative impacts including the economy, and promote sustainable development.

19. Although the monitoring of acid deposition (including ozone and PM) has been implemented among the participating countries of the EANET since the commencement of the EANET activities with cooperative effort and produced a brilliant achievements attached as Appendix 2, some important issues in East Asia for the mitigation of air pollution shown in paragraph 20 are pointed out.

20. Overview of relevant initiatives on air quality management in the world is attached as Appendix 3 and in the relevant website shown in the References of this document for information. Since the ASEAN Agreement on Transboundary Haze Pollution (ASEAN Haze Agreement) specifically targets haze pollution with the objectives to monitor and prevent transboundary haze pollution as a result of land and/or forest fires that should be mitigated through concerted national efforts and intensified regional and international co-operation, the EANET expansion to the integration of acid deposition, $PM_{2.5}$ and ozone does not overlap with the ASEAN Haze Agreement.

(1) Large amount of air pollutants in East Asian region

It was pointed out that several scientific studies show that air pollutants in East Asian region have already exceeded those of Europe and North America since the end of the last century, and further development of East Asian countries will be expected. As also shown in the "Review on the State of Air Pollution in East Asia (2014)", it was revealed that anthropogenic NO_X emissions from East Asia in the year 2008 were recorded much more than Europe and North America. (Figure 1)

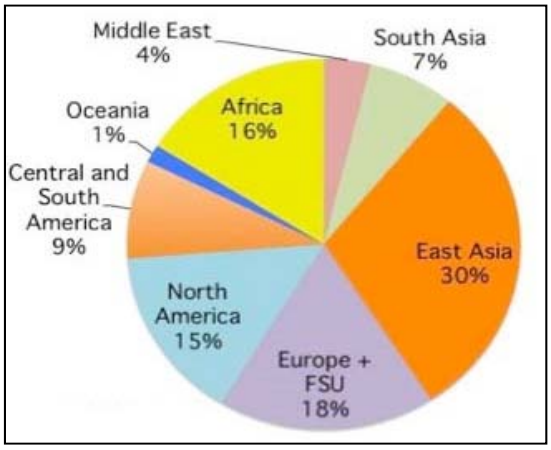

Figure 1. Shares of 2008 global anthropogenic emissions of NO_x by continent based on EDGAR 4.2

(2) Serious episode of ozone, $PM_{2.5}$ and haze in East Asia

Some air pollution problems have been highlighted as either a domestic, regional or hemispherical problem. For example, ozone and $PM_{2.5}$ are now emphasized as high priority air pollution species in some countries. Intercontinental transport of ozone and aerosols ($PM_{2.5}$ episodes in Northeast Asia and haze episodes in Southeast Asia) has been also recognized as important issues from the viewpoint of hemispherical air pollution in the Task Force on Hemispheric Transport of Air Pollution (TF HTAP) under the CLRTAP. In East Asia, strong public concern on the atmospheric concentrations of these species has been shown. It is also logical to use the EANET as a platform for getting information on them to meet the public request.

In March 2014 the World Health Organization (WHO) reported that around 7 million people died as a result of air pollution exposure in 2012 (WHO, 2014) that may now exceed the burdens of malaria, tuberculosis and AIDS combine. This finding is more than doubles of previous estimates and confirms that air pollution is now the world's largest single environmental health risk. The WHO also stated that the main culprits are small particulates—pollutants less than one tenth the thickness of a human hair that are produced by incomplete combustion of fossil fuels and biomass. The study also revealed a stronger link between both indoor and outdoor air pollution exposure and cardiovascular diseases, such as strokes and ischaemic heart disease, as well as between air pollution and cancer. This is in addition to air pollution's role in the development of respiratory diseases, including acute respiratory infections and chronic obstructive pulmonary diseases. In addition to the statement of WHO, the secondarily formatted particle/material from gases should not be disregarded in the process of their production.

(3) Not clear impacts of acid deposition to ecosystems and difficulties of obtaining financial support

From scientific viewpoint, acid deposition may have been marginalized these days because the impacts of acid deposition to ecosystems have not become so clear yet, for instance, in East Asia even after the accumulation of eleven years monitoring data in the EANET sites. Presently acid deposition is not necessarily recognized as an immediate or near future risk. As a consequence of the perception above, it is now very difficult to get support from financial authorities in some participating countries for only dealing with the acid deposition issues including monitoring, research activities and even for capacity buildings due to the little interest of policy makers. This may also apply to the cases of obtaining external funds. Financial support from international funding agencies is also very difficult.

(4) Inter-relationship between $PM_{2.5}$, ozone and acid deposition

As included in the Gothenburg Protocol (known also as the Multi-effect Protocol) to abate acidification, eutrophication and ground-level ozone developed in 1999 in the CLRTAP, one of recent concerned issues on the multi-pollutant, multi-effect for the air quality management is an effective method to reduce acidification, eutrophication and ground-level ozone simultaneously by setting emissions ceilings for sulphur dioxide, nitrogen oxides, volatile organic compounds and ammonia to be met by the new deadline year, 2020. As decided in the revised protocol adopted in 2012, acid deposition, $PM_{2.5}$ and ozone pollution should be treated simultaneously since the precursors of these air pollutants are in common (SO_2, NO_x and VOC) and the mitigation measures has to consider them together.

(5) Usefulness for the ultimate goal of air pollution mitigation including acid deposition, $PM_{2.5}$ and ozone

The EANET is useful as a platform for the ultimate goal of air pollution mitigation including

acid deposition, $PM_{2.5}$ and ozone in order to get public support. Although the objectives of the EANET have not targeted the mitigation of acid deposition in this moment, if the EANET expands its scope to mitigate air pollution, much progress will be expected, and the EANET community also understands usefulness of air pollution mitigation including acid deposition, $PM_{2.5}$ and ozone through its activities. As an example, the Malè Declaration on Control and Prevention of Air Pollution and Its Likely Transboundary Effects for South Asia (Malé Declaration) (Table 3 in Appendix 3) covers not only monitoring but also emission reduction to prevent and minimize transboundary air pollution in South Asia.

(6) Review on the State of Air Pollution in East Asia

Based on the discussion during the Sessions of the EANET in 2011-2013 on future expansion of the scope of the EANET as mentioned in i) of the paragraph 10, Review on the state of air pollution in East Asia (Northeast Asia and Southeast Asian) was implemented in 2013-2014 by the Reviewing Committee (RC) on the Status of Air Pollution in East Asia established under the Task Force on Research Coordination (TFRC). Detailed contents (Chap. 9 Conclusions and Recommendations) of the "Review on the State of Air Pollution in East Asia" are attached as Appendix 4. Major points of the "Review on the State of Air Pollution in East Asia" developed by the RC are shown as follows.

i) PM and Haze

It was estimated that about 300,000-700,000 persons could be prevented from premature death in the developing countries if the PM level could be reduced to the safety level. The seriousness of PM pollution in East Asian countries has been unveiled by a series of international field campaigns. East Asia is identified as one of the highlighted regions. The health effect attributable to transboundary PM pollution has not been well investigated.

ii) Tropospheric Ozone

Regional scale distributions and seasonal cycles along with and controlling factors are well understood by enhanced observational evidence and model analysis in East Asia. General consensus is reached that long-term trends in East Asia are increasing. The adverse impact of ozone on vegetation is deemed to serious with possible increasing trends to make it necessary to take measures to minimize the ozone induced reduction in agricultural production in Asia.

iii) Acidification and Eutrophication

It is reported that a significant decrease of soil pH in the last twenty years or so in China and Japan has been observed, which were probably caused by acid rain. The field measurement and modeling in 1990's and 2000's and the recent studies in 2010's all noted that the sulfur and nitrogen depositions has increased in China and Japan from 1980s to 2000s most likely due to increased emissions of SO_2 and NO_x in China.

iv) Emission inventories

The East Asia became the region of the world largest air pollutant emissions surpassing those in Europe and North America. The natural and anthropogenic emissions in this important region are estimated only with large uncertainties despite of the enormous international and national efforts. Verification, improvement and update of anthropogenic and natural emission inventories based on the ground and satellite observations, chemical transport modeling and inverse modeling are essential in reducing uncertainties.

III. Feasibility Study on expansion of the scope of the EANET

III-1 Possible expansion of the scope of the EANET

21. In accordance with the Prefeasibility Study in 2014 and based also on the previous discussion during the EANET Sessions in 2011-2013, the report of "Review on the State of Air Pollution in East Asia (2014)", difficult situation in East Asia to overcome the atmospheric environmental issues without expansion of the scope, capabilities of the NC(s) and so on, following possible activities for expansion of the scope of the EANET were proposed at the Fourteenth Session of the WGFD (WGFD14). The WGFD14 concluded on Category 1 and 2 as below and the revised Report of the Feasibility Study on the Expansion of the Scope of the EANET will be shared with the Fifteenth Session of the SAC (SAC15) of the EANET in 2015.

(1) Category 1: Strengthening current monitoring activities

[Expected activities]

i) Promotion of the monitoring of ozone and $PM_{2.5}$ including research cooperation and communication on the results as appropriate;
ii) Capacity building activities especially for ozone and $PM_{2.5}$ monitoring;
iii) Increasing number of EANET monitoring sites; and
iv) Strengthening understanding and capacity for monitoring of meteorological parameters and reporting on these as appropriate

[Rationale]

i) In order to respond to public concern to solve emerging $PM_{2.5}$ and ozone problems and to promote management of atmospheric environment so as to protect human health, plants and ecosystems, it is necessary to strengthen current EANET monitoring activities especially for $PM_{2.5}$ and ozone to better understand the problem first.

(2) Category 2: Promotion of activities other than monitoring

[Expected activities]

i) Promotion of information exchange;
- Information exchange on clean air technologies and regional impact assessment through workshops, seminars, etc.

ii) Promotion of research and technical cooperation on emission inventory to support the capacity building
- Research on technical guidelines on emission inventory to support the capacity building; and.
- Capacity building for emission inventories.

iii) Promotion of research activities.
- Research activities on regional impact assessment to plants and ecosystems; and
- Research activities on measurement methodology for elemental carbon (EC) and organic carbon (OC).

[Rationale]

i) Information exchange included in the Item 9 of the Instrument can provide lots of useful information and contribute to develop the mitigation policy of acid deposition problems of each participating country.

ii) Emission inventory already included as the EANET activities, e.g., in the MTP for the EANET (2011-2015) adopted at the IG12 in 2020 is one of the basic information to control the amount of air pollutants and develop national mitigation plan of each participating country. Therefore, as a first step, research on emission inventory guideline is essential.

iii) Promotion of research activities can provide useful information on potential risk of air pollution, identification of emission source as well as pollution mechanism of $PM_{2.5}$ and development of mitigation policy.

(3) Category 3: Long term future activities

[Expected activities]

It is recommended to consider following activities to include in the EANET activities in the future (after 2021) if necessary agreement is made among the participating countries:

i) Monitoring of EC, OC, VOC (volatile organic compounds) and other related items as appropriate;

ii) Submission of the emission inventory developed by each participating country;

iii) Transfer following present Additional budget activities to Core activities of the NC;
- Technical support to the participating countries including capacity building activities.

iv) Name change of the network from “Acid Deposition Monitoring Network in East Asia (EANET)” to “Air Pollution Monitoring Network in East Asia (EANET)”.

[Rationale]

i) In order to promote management of atmospheric environment so as to protect human health, plants and ecosystems, it is essential to implement air pollution monitoring including important components such as EC/OC using the name of Air Pollution Monitoring Network, and support to develop mitigation policy by each participating country.

ii) After development of emission inventory guideline, it is necessary to move on to the activities for developing emission inventory by each participating country for submission to the EANET.

iii) Since activities for technical support to the participating countries are essential for the EANET, it is necessary to include these activities in the Core activities of the NC if participating countries agree.

Reports of the EANET Research Fellowship Program (2013-2015)

The EANET Science Bulletin (Volume 4) is issued once every three years according to the Medium Term Plan (2016-2020) approved at the Seventeenth Session of the Inter-governmental Meeting (IG17) on the EANET held in Bangkok, Thailand, 2015.

Therefore, the research reports of the EANET Fellowship Research Program for 2013-2015 are included in this Bulletin.

Temporal distribution of atmospheric nitrogen deposition and correlation with biomass burning, Chiang Mai (Thailand) during 2008-2012

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Abstract

Biomass burning is the main source of air pollution in the dry season in the Southeast Asian region including Chiang Mai, Thailand. The research aims to assess the spatial and temporal distribution of N deposition and to find out the source of N deposition link with biomass burning in Chiang Mai Province during 2008 - 2012. The main concentrations of N species concentration were NO_2 and NO_3^-. The deposition velocity (V_d) is important for calculation deposition flux. The average V_d values of N_g in descending order were HNO_3 > NH_3 > NO_2. While the V_d values of NO_3^- and NH_4^+ in forest area were higher than grass area in particulate phase. The average V_d values during 5 years (2008 - 2012), it can be set for other study to calculate deposition flux of N species. The N deposition trend in gas phase and particulate phase were measured for 5 years. The NH_3 and NH_4^+ were estimated the main deposition for gas and particulate phase, respectively. The good correlations between N_t deposition including N_g and N_p and PM_{10} and hotspot number were found in whole periods. High N deposition was originated from west direction of Chiang Mai Province. Therefore it was supported by correlation and Principal component analysis (PCA) between N species deposition and the number of hotspots. Moreover, the main source of nitrogen deposition was accepted as biomass burning for this area in term both of local source and long rage transport in dry season periods.

Key words**:** Nitrogen deposition, Biomass burning, Backward trajectory, Wind rose deposition, hotspot

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1. Introduction

The global increase in nitrogen (N) emission has lead to concerns that chronic elevated atmospheric concentration which was many cause negative effects (e.g., eutrophication, losing biodiversity and acidification) (Cornelissen *et al.*, 2001; Krupa, 2003; Prenni *et al.*, 2014; Karlsson *et al.*, 2013). However, there are few dry N deposition studies in Asia which were many sources of N deposition (agricultural activities, fertilizer production, combustion of fuels, and biomass burning). There are not many studies of linking pollution of reduced nitrogen with biomass burning which are increased for many countries.

Biomass burning was a big problem for Asia especially South East Asia. Thailand, Myanmar, Lao PDR and Indonesia have been annually facing air pollution (biomass burning) during the dry season (Kim Oanh *et al.*, 2011). There have been several large-scale for biomass burning in Northern of Thailand which was many areas (Kim Oanh and Leelasakultum, 2011).This pollution has been recorded as a serious problem for well over 10 years. The burning area can represents by hotspots which can detect by remote sensing and GIS technologies to deliver global MODIS (Moderate Resolution Imaging Spectroradiometer). The number of hotspot in Chiang Mai Province presented in Figure 1, based on the data from FIRMS（Fire Information for Resource Management System) during 2008-2012. Recently, the problem has become worse than ever in dry season period (January - April). Chantara *et al.* (2009) were determinate particulate ions of airborne in Chiang Mai and Lamphun, Thailand from June 2005 to June 2006. Both anions (SO_4^{2-}, NO_3^-, and Cl^-) and cations (NH_4^+, Na^+, K^+, Mg^{2+} and Ca^{2+}) were significantly higher in dry period (December - March) and transition period I (October - November) than those in other seasons. The dominant anion and cation were SO_4^{2-} and NH_4^+, respectively. Moreover NO_3^- and NH_4^+ were high in dry season in Northern Thailand which has been large scale of biomass burning.

Therefore the objectives of the research are: To assess the spatial and temporal distribution of N species in dry deposition (NO_2, HNO_3, NH_3 in gas phase and NO_3^-, NH_4^+ in particulate phase) and to correlate biomass burning and nitrogen species concentrations in Chiang Mai Province. In this study, sampling of dry deposition has been carried out using the filter pack method at rural site in Chiang Mai Province.

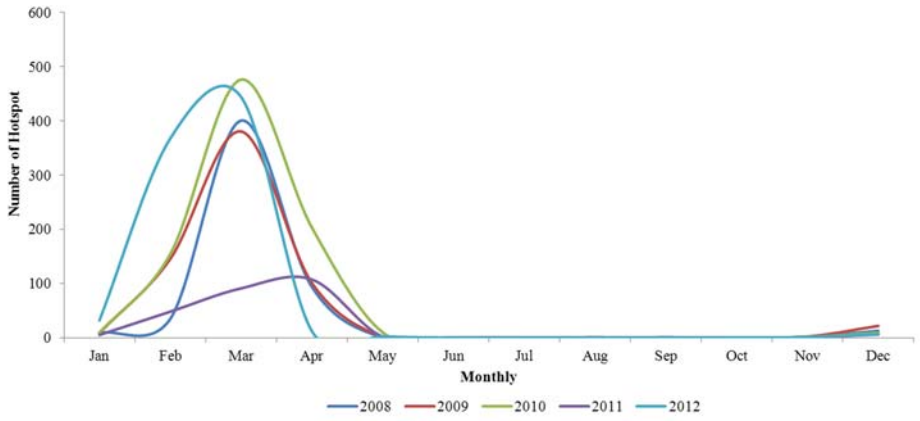


Figure 1. The Monthly hotspot numbers in Northern Thailand (Chiang Mai Province) during 2008-2012.

2. Methodology

2.1 Sampling site

The sampling site was at the meteorological station in the area of Mae Hia Research Center, Chiang Mai University, Muang District, Chiang Mai Province (Figure 2). This site was classified as rural site based on Acid Deposition Monitoring Network in East Asia (EANET) criteria since 2000. It is located at latitude 18° 45' 40.3" N and longitude 98° 55' 54.3" E. The sampling had been done within 5 year from 1st January, 2008 to 31st December, 2012.

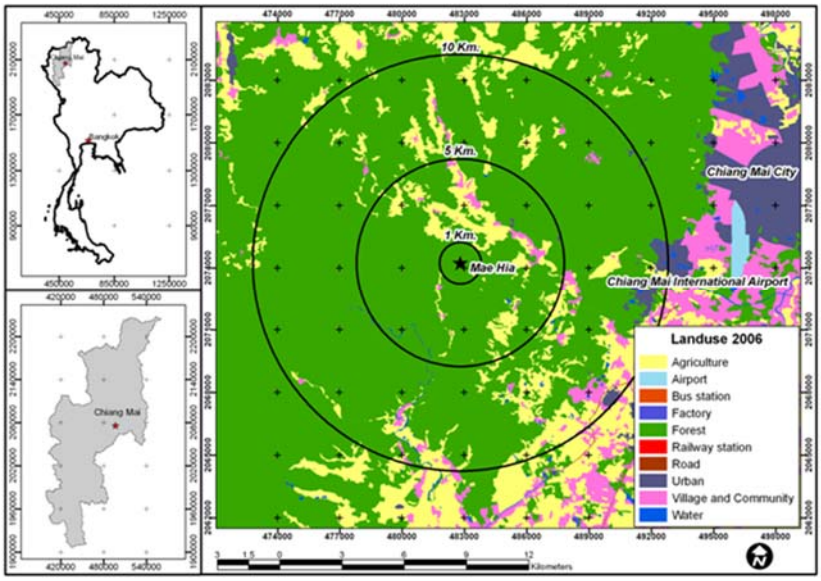


Figure 2. Location of the meteorological station in the area of Mae Hia Research Center, Chiang Mai University.

2.2 Sample collection

N species sample in gas phase such as HNO_3, NH_3 and in aerosol (NO_3^-, NH_4^+) were collected by a four-stage filter pack. The first two layers are polytetrafluoro ethylene filter; F0 and polyamine filter; F1. The other two layer filters are cellulose acetate filter (F2, F3), which impregnated with potassium carbonate and phosphoric acid and glycerin solution. Prior to sampling, a filters pack was connected to a sampling set at the monitoring site. Flow rate of air input was 1 L/min. Sampling was operated for 10 days long, 3 times a month during 2008 - 2012.

2.3 Samples Analysis

The filters of a four-stage filter pack, F0, F2 and F3 filter papers were extracted with 20 mL Milli Q water, while F1 was extracted with 20 mL 0.05% H_2O_2 by ultrasonication for 30 minutes. After extraction process, insoluble matters were filtered by cellulose acetate membrane (EANET. 2002). Ion chromatograph (Metrohm, Switzerland) was used for determination of major ions (NH_4^+ and NO_3^-). Analytical columns for anions and cations were Metrosep A Supp 5 (5×250 mm) and Metrosep C2 150 (4×150 mm), respectively.

2.4 Hotspot data

The hotspot data are provided by FIRMS that integrates remote sensing and GIS（Geographic Information System） technologies to deliver global MODIS. FIRMS was developed by the University of

Maryland with funds from NASA (National Aeronautics and Space Administration). The hotspots are detected using data from the MODIS instrument, on board NASA's Aqua and Terra satellites. The hotspots represented fire location the center of a 1 km pixel (approximately) flagged as containing one or more actively burning hotspot/fires within that pixel (NASA/University of Maryland, 2002).

2.5 Meteorological data

Thai Meteorological Department (TMD) provided data on temperature, relative humidity, radiation, rain precipitation, wind speed and cloud cover, which was used for this study. Most of the data were hourly data set. The data were used for calculation of velocity deposition (V_d). There were calculated follow method of the technical manual on dry deposition flux estimation in East Asia which was run by EANET (EANET. 2010).

2.6 Trajectory analysis

Backward trajectories arriving at the receptor (sampling site) were calculated using the HYSPLIT (Hybrid Single Particle Langrangian Integrated Trajectory) model. The 3 days backward trajectories are available online at http://ready.arl.noaa.gov/HYSPLIT.php. Air mass trajectories for each individual day were calculated only the highest N deposition period (10 days) for each year during 2008 - 2012.

2.7 Data analysis

PCA and Pearson correlation (r) was implemented to identify the relationships between number hotspot, PM_{10} and N deposition species in Chiang Mai Province. SPSS program was used for determination the factors underlying the inter-correlations between the measured species.

3. Results and discussion

3.1 Seasonal variability of N species concentrations

The concentrations of atmospheric N species had been done from 2008 - 2012. Total number of samples was 36 samples per year. The N species in gas phase (N_g), it was consist of NO_2, HNO_3 and NH_3 while N species in particulate phase (N_p) was NH_4^+ and NO_3^-. The total nitrogen species (N_t) was included all of N_g and N_p. All nitrogen species was analysed by IC except NO_2 was used chemiluminescence. The NO_2 data was provided by Pollution Control Department, Thailand (PCD). The 5 years average N_g concentration of NO_2, HNO_3 and NH_3 were 21.08 ± 11.52 μg/m^3, 3.12 ± 1.48 μg/m^3 and 3.25 ± 3.87 μg/m^3, respectively. While N_p concentration NH_4^+ and NO_3^- were 1.72 ± 0.67 μg/m^3 and 4.17 ± 1.05 μg/m^3, respectively. The main concentration of N_g was NO_2 and N_p was NO_3^-.

Figure 3 illustrated mean concentrations of hotspot number PM_{10} and N species (N_g, N_p and N_t) in periods during 2008 - 2012. Seasonal variations of hotspot number, PM_{10} and N species concentrations were almost the same. Their concentrations were highest in dry season decreasing in wet season. The important source was open burning including forest fire, agricultural waste and garbage burning in communities in dry season due to PM_{10} was very high concentration in the same period. They were assumed to be the main sources N species in this period. In addition, high frequency of forest fire was observed via number of hotspots detected by FIRM. Therefore May 2010, the concentration of N_p was high in rainy period which was abnormal situation. Moreover, in year 2011 was high amount of rain in the sampling area, which was diluted to low N_t concentration in dry season period (low number of hot spots and PM_{10} concentration).

3.2 N species deposition (2008 - 2012)

The amount of acid deposition per unit area per time or we know in term of deposition flux which is importance data to assess the impact by acid deposition. The products of air concentration and deposition velocity are necessary to be known. This study was followed the process from EANET method (EANET. 2010). The deposition velocities depend on many factors such as wind speed, humidity, temperature, net radiation etc.

Deposition velocity (V_d) of gases and particulates phase N species in forest and grass area in Chiang Mai Province are shown in Table 1. The deposition velocities are presented in the unit of cm^2/s. V_d of hourly data were calculated and then the V_d were averaged in the time resolution of N species concentration. The average values of V_d were estimated for gas phase (NO_2, HNO_3 and NH_3) and particulate phase (NO_3^-, NH_4^+) in forest and grass area during 2008 - 2012. The V_d values were high for HNO_3 in forest area (2.26 - 3.09 cm^2/s) while the V_d values of NO_2 (0.08 - 0.10 cm^2/s) and NH_3 (0.32 - 0.20 cm^2/s) were small variance in both of forest and grass area. NO_3^- and NH_4^+ were 0.40 - 0.50 cm^2/s and 0.31 - 0.40 cm^2/s, respectively in forest. While the V_d values of NO_3^- and NH_4^+ were used the same values (0.12 - 0.15 cm^2/s).

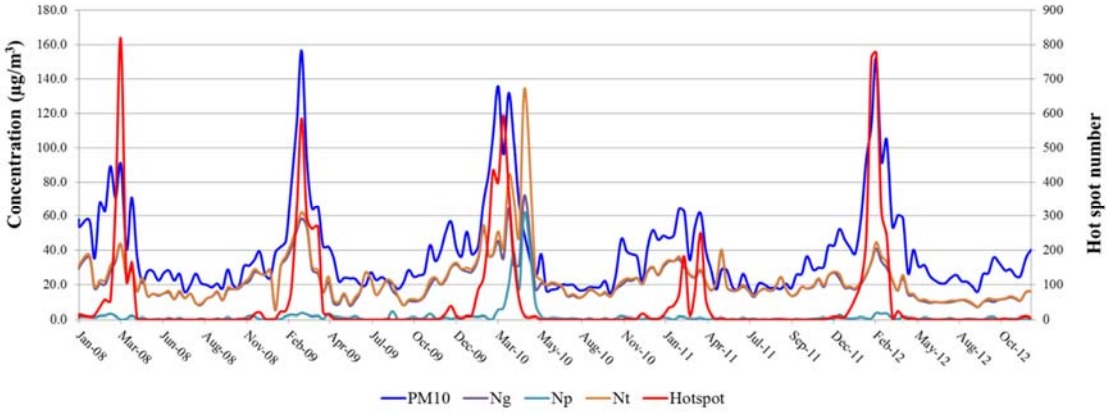


Figure 3. Ten days averaged concentration of N species, PM_{10} and hotspot number during 2008 - 2012.

Table 1. Deposition velocity of gases and particulates phase N species in forest and grass area in Chiang Mai (Unit: cm^2/s) .

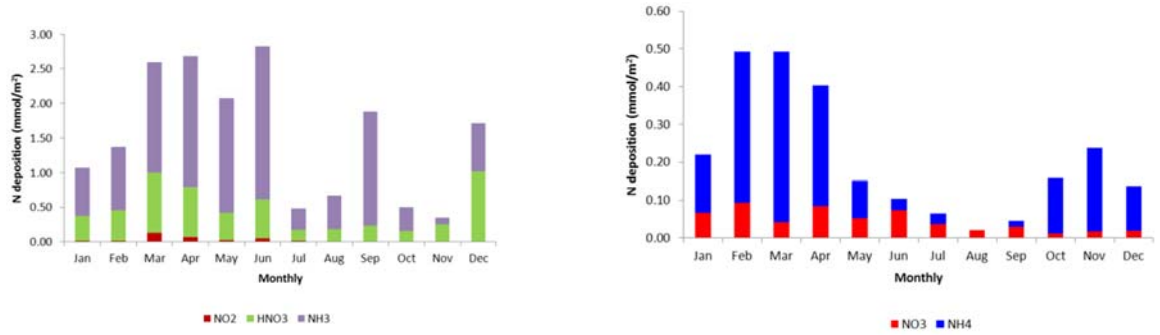
years	Gas Phase						Particulate Phase		
	forest			grass			forest		grass
	NO_2	HNO_3	NH_3	NO_2	HNO_3	NH_3	NO_3^-	NH_4^+	NO_3^-, NH_4^+
2008	0.09	2.26	0.32	0.08	0.76	0.22	0.40	0.31	0.12
2009	0.10	2.96	0.24	0.10	0.99	0.22	0.48	0.38	0.15
2010	0.09	3.09	0.22	0.10	1.04	0.21	0.50	0.40	0.15
2011	0.09	2.67	0.23	0.10	0.89	0.21	0.45	0.35	0.12
2012	0.09	2.66	0.21	0.10	0.89	0.20	0.44	0.34	0.13
Average	**0.09**	**2.73**	**0.24**	**0.10**	**0.91**	**0.21**	**0.45**	**0.36**	**0.13**

The dry deposition flux of N species were calculated from concentration of N species and V_d:

$$F_i = V_d^i \times C_i \qquad (1)$$

where F_i is flux of i species and C_i is concentration of i species. The N species deposition fluxes were calculated in gas and particulates phase in both of area (forest and grass). The percentage of forest area (85 %) and grass (15 %) were estimated by Arc-GIS land use program.

The profile of N species deposition in gas phase and particulate phase were illustrated in Figure 4 during 2008 - 2012. The main N species depositions were NH_3 and NH_4^+ for gas and particulate phase. Furthermore deposition in gas phase higher than in particulate phase. The particulate N depositions were high in the dry season and always highest in March - April. The most important factors are the amount of rain precipitation and scale and frequency of open burning. The gas N depositions were high in March - June while quite low for about 5 months (October - February). Then the concentrations increased again at in dry season (March) due to lower precipitation and higher burning activities in agricultural areas including forest area. Therefore in 2011 was low deposition for particulate phase due to the high amount of rain precipitation.



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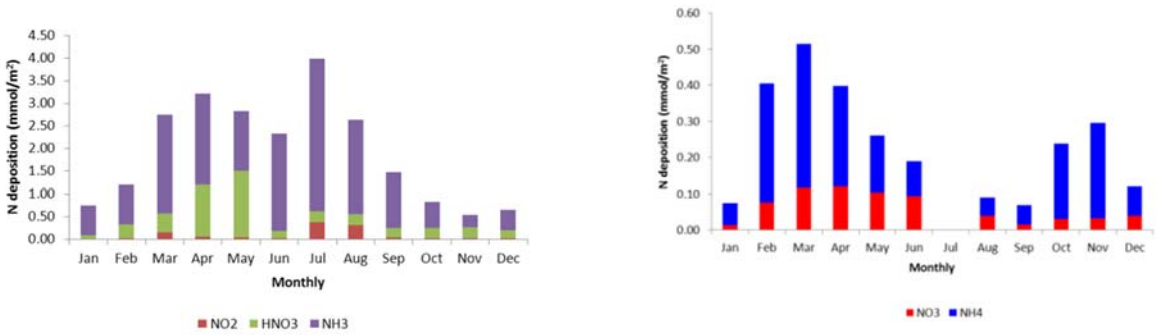

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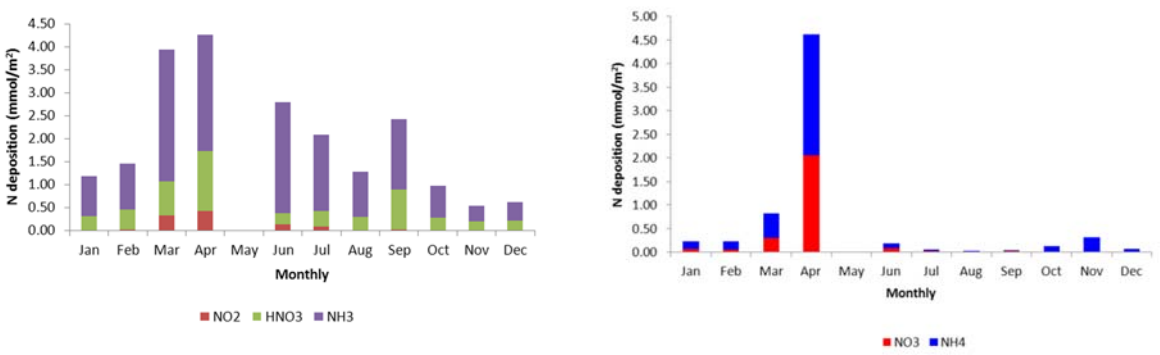

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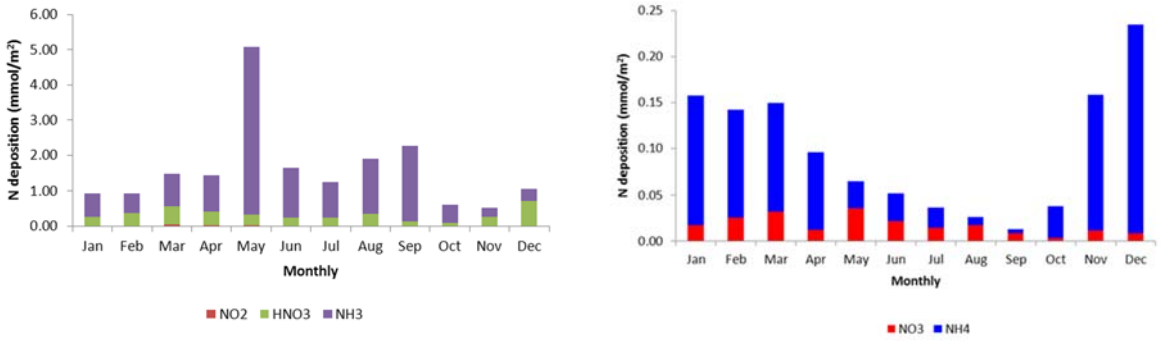

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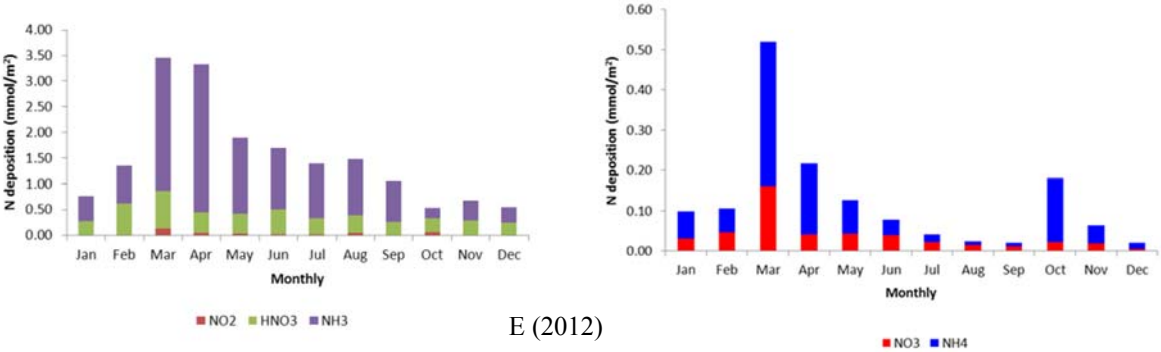


E (2012)

Figure 4. Monthly averaged N depositions in Chiang Mai during 2008 - 2012.

Figure 5 was illustrated the monthly averaged of N species deposition (N_g, N_p and N_t), PM_{10} and hotspot number during 2008 - 2012. The N_t were quite high in the dry season and getting lower in the wet season in every years. In May 2010, the concentration of N species was very high (2-3 times higher than normal) which was contaminated from the method. Therefore the data of May 2010 was deleted for data analysis. In 2011, it was started raining early than others year that was made lower PM_{10} concentrations and hotspot number.

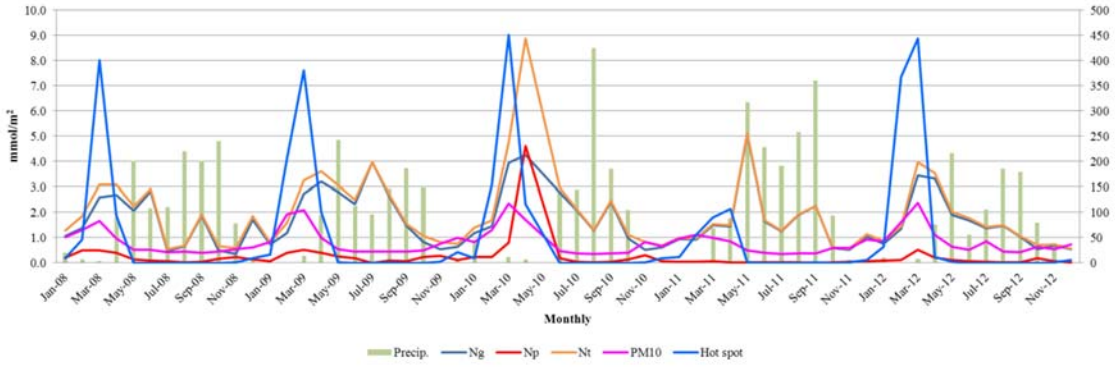


Figure 5. Monthly averaged trend of N species deposition, PM_{10} and hotspot number during 2008 - 2012.

The comparison annual average of N deposition in this study with others study was shown in Figure 6. The N_p deposition in Chiang Mai (CM) was lowest (5.68 mmol/(m^2 y)) while was highest deposition in EMEP (20.71 mmol/(m^2 y)). For Ng deposition, CM was highest deposition compare with other studies. The

Clean Air Status and Trends Network (CASTNET) in USA and The European Monitoring and Evaluation Programme (EMEP) were not reported N_g deposition. The N_t deposition in descending order were CM (26.10 mmol /(m^2 y)) > EANET (24.24 mmol/(m^2 y)) > EMEP (20.71 mmol / (m^2 y)) > CASTNET (9.09 mmol /(m^2 y)) .

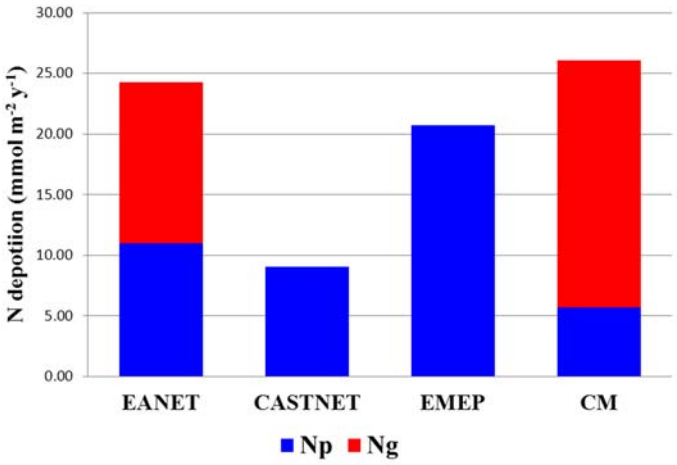


Figure 6. Comparison of N deposition per year with other studies.

3.3 Correlation between N species deposition and biomass burning factor

Correlations between N species deposition, PM_{10} and hotspot number were analysed by Pearson correlation during 2008 - 2012 in whole year (Table 2). The N_g and N_t (N_t+N_g) depositions were very strong correlated (r = 0.990). Furthermore, N_g deposition was correlated (r = 0.348) with hotspot number. While N_p deposition was moderately correlated with PM_{10} and hotspot number (r = 0.739 and r = 0.6565, respectively) with $p < 0.01$. Moreover, N_t deposition was weakly correlated (r = 0.372 and r = 0.432) with PM_{10} and hotspot number with $p < 0.05$.

Table 2. The correlation between N species deposition, PM_{10} and hotspot number during 2008 - 2012.

	N_g	N_p	N_t	PM_{10}	Hotspot	Precipitation
N_g	1.000					
N_p	0.349*	1.000				
N_t	0.990**	0.475**	1.000			
PM_{10}	0.280	0.739**	0.372*	1.000		
Hotspot	0.348*	0.656**	0.423*	0.891**	1.000	
Precipitation	0.077	-0.358	0.019	-0.621**	-0.444**	1.000

* Correlation is significant at the 0.05 level (2-tailed).

** Correlation is significant at the 0.01 level (2-tailed).

For assessment the impact of biomass burning was analysed data in dry season periods (January - April) which was demonstrated in Table 3. The N_t deposition was moderately with PM_{10} (r = 0.680) and hotspot number (r = 0.618) with $p < 0.01$. The strong correlations were found between deposition of N_g and N_t (r = 0.973) and N_p and N_t (r = 0.819). Moreover, N_g deposition was fairly correlated with PM_{10} (r = 0.603) and hotspot number (r = 0.9597) while N_p was good correlated with PM_{10} (r = 0.704) and hotspot number (r = 0.569). The results revealed that biomass burning emitted N species.

Table 3. The correlation between N species deposition, PM_{10} and hotspot number during 2008 - 2012 in dry season periods.

	N_g	N_p	N_t	PM10	Hotspot
N_g	1.000				
N_p	0.711**	1.000			
N_t	0.973**	0.819**	1.000		
PM_{10}	0.603**	0.704**	0.680**	1.000	
Hotspot	0.597**	0.567**	0.618**	0.802**	1.000

* Correlation is significant at the 0.05 level (2-tailed).

** Correlation is significant at the 0.01 level (2-tailed).

PCA has been applied to confirm the source from biomass burning. The factor analysis for N deposition and other factors were performed with two factors with an Eigen value >1, contributing ~85% of the variance. First factor contributes 50.85% of the total variance and high loading with N_p, PM_{10} and hotspot number (Figure 7). Therefore, loading scatter plots for secondary factors were grouped N_t and N_g which were the same coordinate. These results can be used for identification of the biomass burning source.

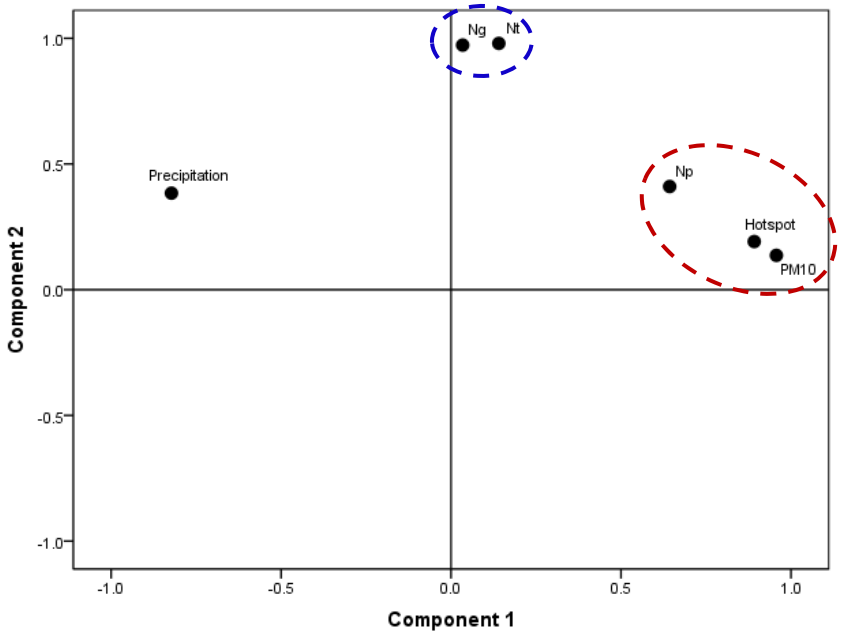
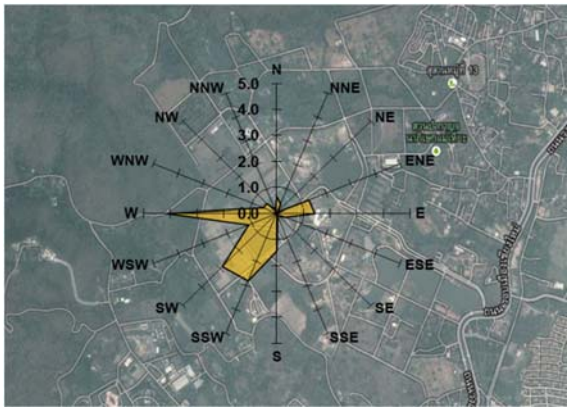


Figure 7. Loading scatter plot of N species deposition, PM10 and hotspot number during 2008 - 2012.

3.4 Wind rose deposition and back ward trajectory

In order to gain more information on N deposition, both in terms of type and position (local or long range transportation), wind rose deposition and three days backward trajectories of the air masses arriving at Chiang Mai were analysed by meteorological data and HYSPILT model.

The wind rose deposition was computed by wind direction and N_t deposition (Figure 8) during 2008 - 2012. All of them were recorded from the west direction, generated in western part of Chiang Mai (Mae Cham District). In dry season, high density of hotspots in the western part of Chiang Mai Province was found especially Mae Cham District and Mae Hong Son Province.



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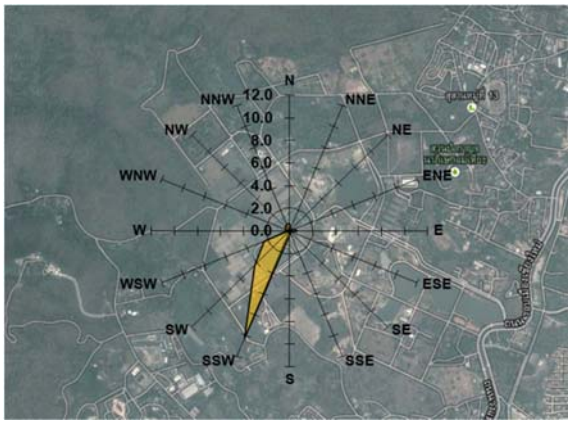

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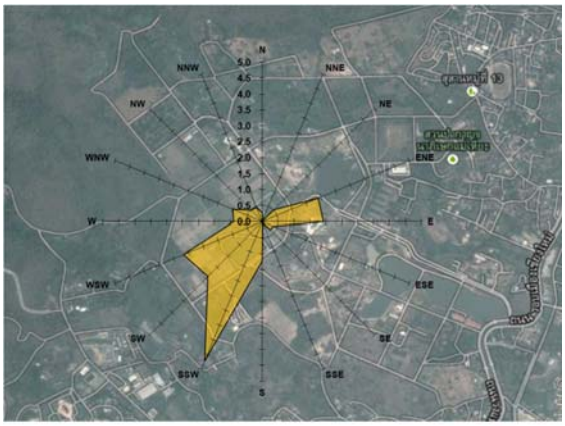

D(2011)

E(2012)

Figure 8. Wind rose N deposition during 2008 - 2012.

Therefore a movement of air mass from air mass west direction could bring pollutants generated from open burning to the receptor (sampling point).

Three days backward trajectories were run on the high Nt deposition one day per each year. The directions of trajectories were shown in Figure 9. Almost of direction came from the west direction, generated in the western region (Myanmar) and then travelling over western Thailand (Mae Hong Son Province), before arriving in Chiang Mai. The hotspots numbers were high in dry season periods which were occurred in the western part of Chiang Mai Province. Therefore a movement of air mass from air mass west direction could bring pollutants generated from open burning to the receptor (Chiang Mai Province). Air mass trajectories pattern were similar to wind rose deposition which were identified the biomass burning source.

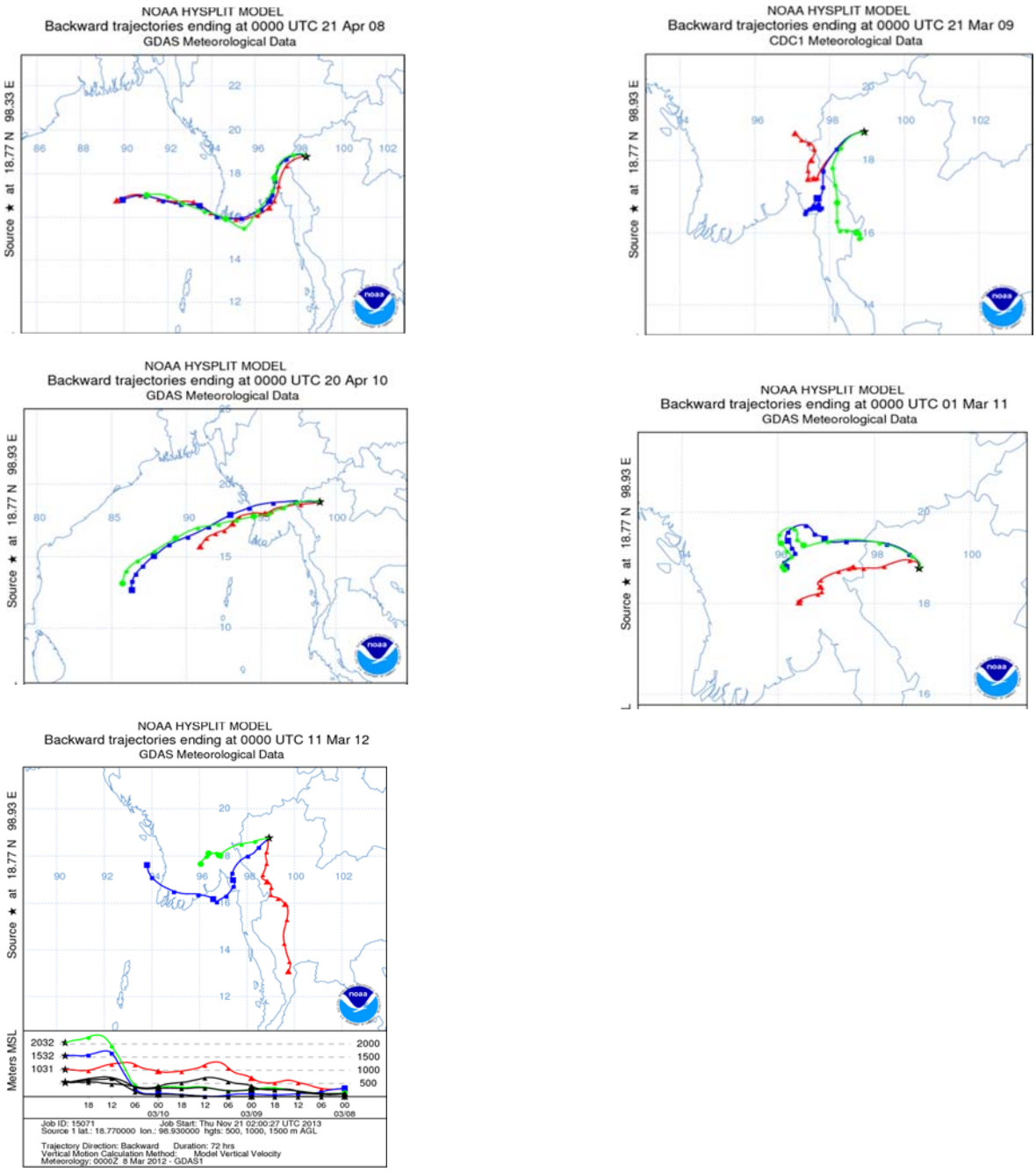

Figure 9. 3-day backward trajectories of the high N deposition periods during 2008 - 2012.

4. Conclusion

Chiang Mai and the northern part of Thailand have been faced with air pollution every dry season. The main air pollution source in this area is open burning especially forest fires. However, there were only few studies on N deposition from biomass burning in Thailand.

The concentrations of atmospheric N species were monitored in 2008 - 2012 in Chiang Mai Province (Chiang Mai University). The 5 years average N_g concentration in disorder were $NO_2 >> NH_3 > HNO_3$ while N_p were $NO_3^- > NH_4^+$. The dominant of N species concentration for N_g and N_p were NO_2 and NO_3^-, respectively. The importance data were deposition velocity are important for calculation deposition flux. The average V_d values were HNO_3 (2.73 cm^2/s) higher than NH_3 (0.24 cm^2/s) and NO_2 (0.10 cm^2/s) while NO_3^- and NH_4^+ were 0.40 - 0.50 cm^2/s and 0.31 - 0.40 cm^2/s, respectively in forest. While the V_d values of NO_3^- and NH_4^+ in forest were higher than (0.45 and 0.36 cm^2/s, respectively) in grass area (0.13 cm^2/s). The seasonal variation of N deposition in gas phase and particulate phase were conducted during 2008 - 2012. The NH_3 and NH_4^+ were estimated the main deposition for gas and particulate phase, respectively.

The good correlations between N_t deposition including N_g and N_p and PM_{10} and hotspot number were found in whole periods (2008 - 2012). Meteorological parameters (amount and frequency of precipitation) and number of hotspots were found to be the most influential factors affecting N deposition.

High N deposition during 2008 - 2012 originated from west direction of Chiang Mai Province in every year, which was confirmed by wind rose deposition and backward trajectory. In the dry season was large scale of biomass burning in this region. This was supported by correlation and PCA between N species deposition and the number of hotspots. In addition biomass burning was accepted as an important source of nitrogen for this area in term both of local source and long rage transport.

Acknowledgements

Financial supports from the Network Center for the Acid Deposition Monitoring Network in East Asia (EANET), Asia Center for Air Pollution Research (ACAP) are gratefully acknowledged. We would like to thank Mr. Shirai, Ms. Aoyagi and Ms. Nakamura (Data Management Department, ACAP) for the informative discussion on the data of research project. Moreover, we also thank for Pollution Control Department (PCD), Thailand, Fire Information for Resource Management System (FIRMS) and Thai Meteorological Department (TMD) for providing observation data and the NOAA Air Resources Laboratory (ARL) for the HYSPLIT dispersion model.

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Long range transport of air pollutants (trace gases) related with meteorological process in Asia (subregion)

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Abstract

This research focused on the air quality monitoring data analysis of Ulaanbaatar sites and some Asian sites which include EANET to determine seasonal diurnal variation and comparison of O_3, NO_2 and CO. Main purpose were an explanation of high concentration of trace gases related with local and regional air mass transport in the Asian region. Air quality monitoring data of Ulaanbaatar sites (July 2010 to June 2011) are indicated general air pollution trend of Ulaanbaatar city. There are big seasonal (high concentration in summer July, min-Jan) diurnal variation of ozone (maximum in summer), NO_2, CO (high concentration in winter Feb) and depending emission sources and meteorological condition. There is long range transport from neighbor countries (from Russia and China), but city pollution is dominated in Ulaanbaatar.

Key words: Trace gases, Air quality, Air pollutants, Transport

1. Introduction

Air pollutants are classified according to the manner in which they reached the atmosphere, namely primary pollutants, which emitted directly from sources and secondary pollutants which formed in the atmosphere by chemical interactions among primary pollutants and normal atmospheric constituents. The atmospheric distribution of nitrogen oxides is highly non-uniform. Although about ninety percent of nitrogen oxides in the earth's atmosphere are estimated to be produced by natural bacterial action, localized urban concentrations may far exceed clean air background concentrations. The oxides of nitrogen, NO, NO_2 play an important role in the air pollution chemistry. Most of the NOx formed in fossil fuel combustion. However, NO dominates the NOx. NO2 is formed to some extent from NO in combustion exhaust gases by $2NO + O_2 \rightarrow 2NO_2$. (1)

When NO and NO_2 are present in sunlight, ozone formation occurs as a result of the photolysis of NO_2.

$$NO_2 + hv \rightarrow NO + O \quad (2)$$
$$O + O_2 + M \rightarrow O_3 + M \quad (3)$$

Where M present N_2 or O_2 or another third molecule that absorb the excess vibrational energy and thereby stabilizes the O_3 molecule formed (John, 1986).

Ground-level ozone- what we breathe- can harm our health. Even relatively low levels of ozone can cause health effects. People with lung disease, children, older adults, and people who are active outdoors may be particularly sensitive to ozone. Children are at greatest risk from exposure to ozone because their lungs are still developing and they are more likely to be active outdoors when ozone levels are high, which increases their exposure. Children are also more likely than adults to have asthma. (http://www.epa.gov/air/ozonepollution/)

There is no significant source ozone in an atmosphere other than reaction →(3). Once formed O_3 react with NO to regenerate NO_2.

$$O_3 + NO \rightarrow NO_2 + O_2 \quad (4)$$

The photolysis of NO_2 is a key atmospheric reaction.

Also, ozone photolysis to produce both ground state (O) and excited single (O(‘D)) oxygen atoms. Oxygen atom just can react with O_2. However, O(‘D) collides with H_2O and produces two hydroxyl radicals.

$$O(‘D)+H_2O \rightarrow 2HO$$
$$CO+OH \rightarrow CO_2 + H \quad (5)$$

There is too complex reaction in the atmospheric system of NO, NO_2 and CO (John, 1986).

Meteorological conditions are a crucial factor contributing to poor air quality, in addition to the two other factors of emissions and atmospheric chemistry. Many studies are shown that very different local and regional meteorological influences can be dominant in various settings (Nelson and Sara, 1998).

In East Asia, especially China, emission of O_3 precursors have increased greatly since 2000 (Ohare *et al*. 2007) , (Zang *et al*., 2007). Tanimoto, Ohara and Uno (2009) noted that the anthropogenic emission became dominant contribution, showing Mt. Happo site of Japan to be an ideal location for detecting rapid changes in anthropogenic emissions in East Asia. This research focused on the air quality monitoring data analysis of Ulaanbaatar sites and some Asian sites which include EANET to determine seasonal diurnal variation and comparison of O_3, NO_2 and CO. Main purpose were an explanation of high concentration of trace gases related with local and regional air mass transport in the Asian region.

2. Method

2.1 Data and monitoring sites

Fifteen min averaged raw data of mixing ratios and concentrations of O_3, NO_x from June 2010 to July 2011 of Air quality monitoring sites of Mongolia and daily average data of EANET sites of Russia (Mondy), Japan (Ochiishi, Rishiri, Tappi, Sado-Seki) from ACAP used in this work. We converted 15 min concentration to 1-hour average and the same unit. A few days mixing ratio of the gases was extremely high from other days and we discharged such data for estimation diurnal and annual variations. But the extreme data’s used for trajectory analysis.

Table 1. Details of sites for used data.

	Data type	Sites	Parameters	Period	Data sources
1.	Air quality monitoring data	UB01(industrial) UB04 (urban) UB05 (urban) UB08 (background) Ulaanbaatar, Mongolia	O_3, NO_2, NO, NO_x, CO, temperature, wind direction, wind speed, pressure for every 15 min)	July 2010-June 2011	(ACAP from CLEM)
2	Air quality monitoring data	(UB02 (road-site)	(NO_2, CO, NO, NO_x, temperature, wind direction, wind speed, pressure),(15min)	July 2010-June 2011	ACAP from CLEM
3.	Air quality monitoring data	Mondy (remote-51.39N 100.55E), Russia	O_3 (daily average)	2010-2012	ACAP
4.	Air quality monitoring data	Ochiishi(43.09.43N 145.29.5E) Rishiri(45.07.11N,141. 12.33E) Tappi (41.15.06N, 140.20.59E) Sado-Seki(38.14.59 N, 138.24E)	NO, NO_2, NO_x, O_3	2010-2012	ACAP
5.	Short time observation	Sainshand, Mongolia	O_3	2010 (9-22Aug)	Russian-Mongolian expedition,

2.2 Location of sites

Ulaanbaatar city is located in the valley of the mountains. There are 6 automatic monitoring (Figure 1) sites based on influencing emission sources and activities: roadside (UB02), urban (UB04, UB05), industrial (UB01, UB07) and background (UB08). Remote sites were selected from EANET (Russia (Mondy)), (Japan (Ochiishi, Tappe, Sado-Seki, Rishiri)) in order to compare mixing ratios and determine long-range transport.

Figure 1. Location of Air quality monitoring sites in Ulaanbaatar Mongolia.

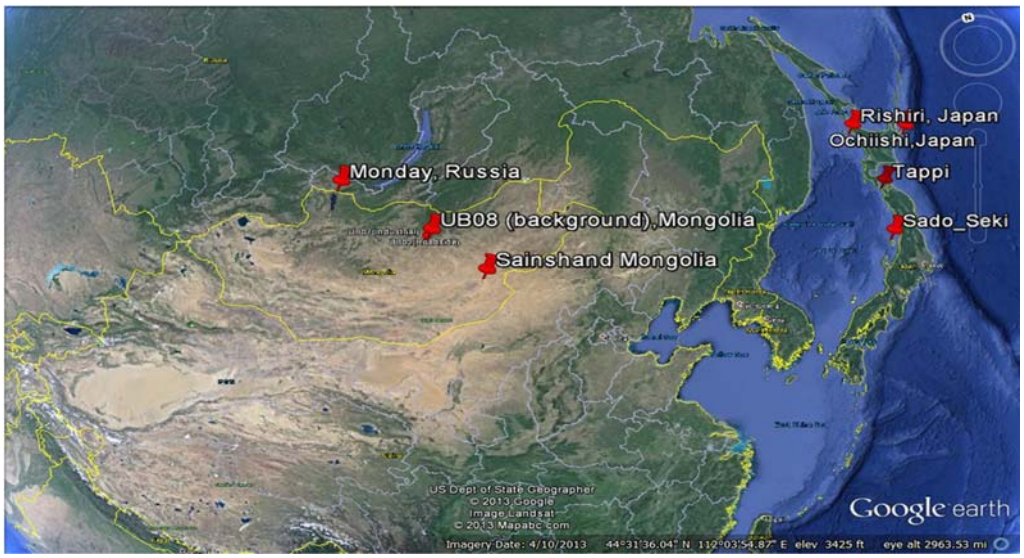


Figure 2. EANET sites (data used for the study).

2.3 Trajectory model

Hysplit trajectory model (http://ready.arl.noaa.gov/hypub-bin/trajtype.pl) has been used for identifying the origin of the air masses arriving at high concentration region in Asian subregion and long range transport, isobaric air mass trajectory used for high concentration O_3 days. Trajectories were calculated one a day with the initial level at 0, 500 and 1,500 m from ground level.

3. Results and discussion

3.1 Annual and diurnal variation of O_3 and NO_2

3.1.1 Data comparison of Ulaanbaatar city sites

The sources of tropospheric ozone are an influx from the stratosphere and generation by photochemical reactions in the troposphere. The influx of ozone from the stratosphere takes place mainly in

middle and high latitudes and is most active in early spring. The generation of ozone in the troposphere is most active in summer since it is caused by photochemical reactions involving nitrogen oxides (NO_x), carbon monoxide (CO) and volatile organic compounds (VOCs) (http://www.acap.asia/publication/pdf/ozone1.pdf). Therefore, we analyzed ground level O_3 data and its precursor's such NO_2 and CO. Before analysis, all measured 15 min data were converted to hourly averaged data. The comparing of daily averaged ozone and NO_2 data of Ulaanbaatar sites was shown on Figure 3, 4 and other Asian stations data's on Figure 5.

The daily averaged mixing ratios of O_3 were higher in summer lower in winter (Figure 3) and NO_2 lower in summer higher in winter (Figure 4) in Ulaanbaatar Mongolia. The highest mixing ratio of NO_2 measured at the road site (UB02).

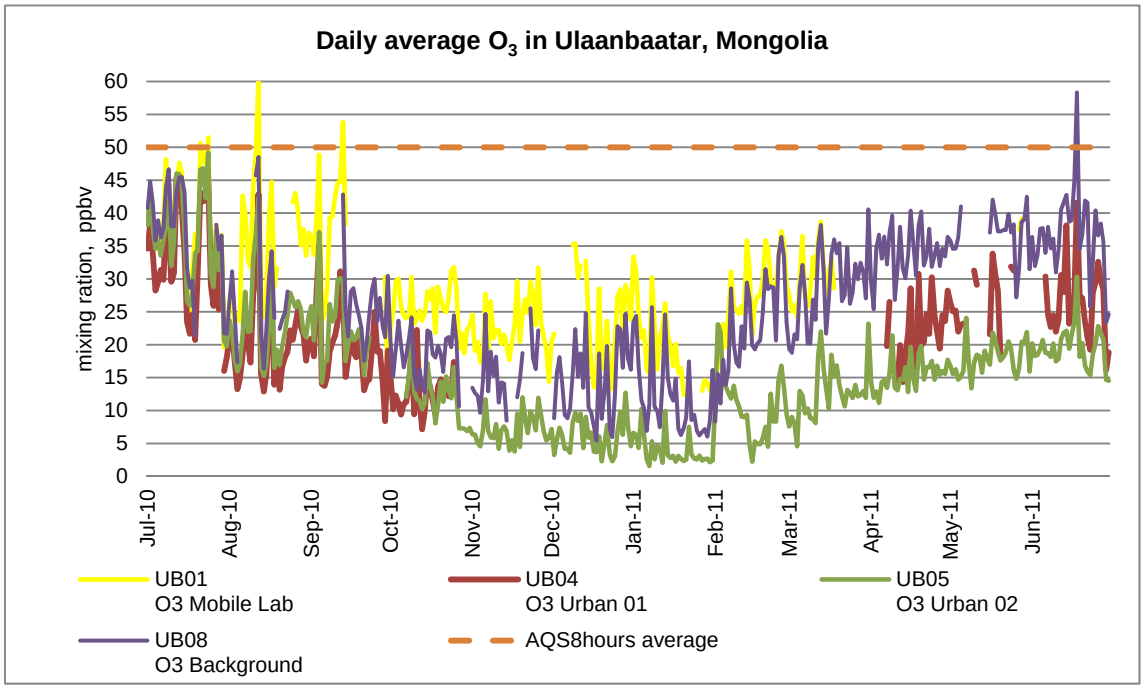


Figure 3. Annual variation of O_3 at Ulaanbaatar sites, Mongolia.

During summer, the daily averaged mixing ratios of ozone were exceeded the national air quality standard level (8 hour average) at UB01 site and UB08 on some days. In winter, the mixing ratios were under national air quality standard, lowest observed at Urban residential area (UB05 site) (Figure 3).

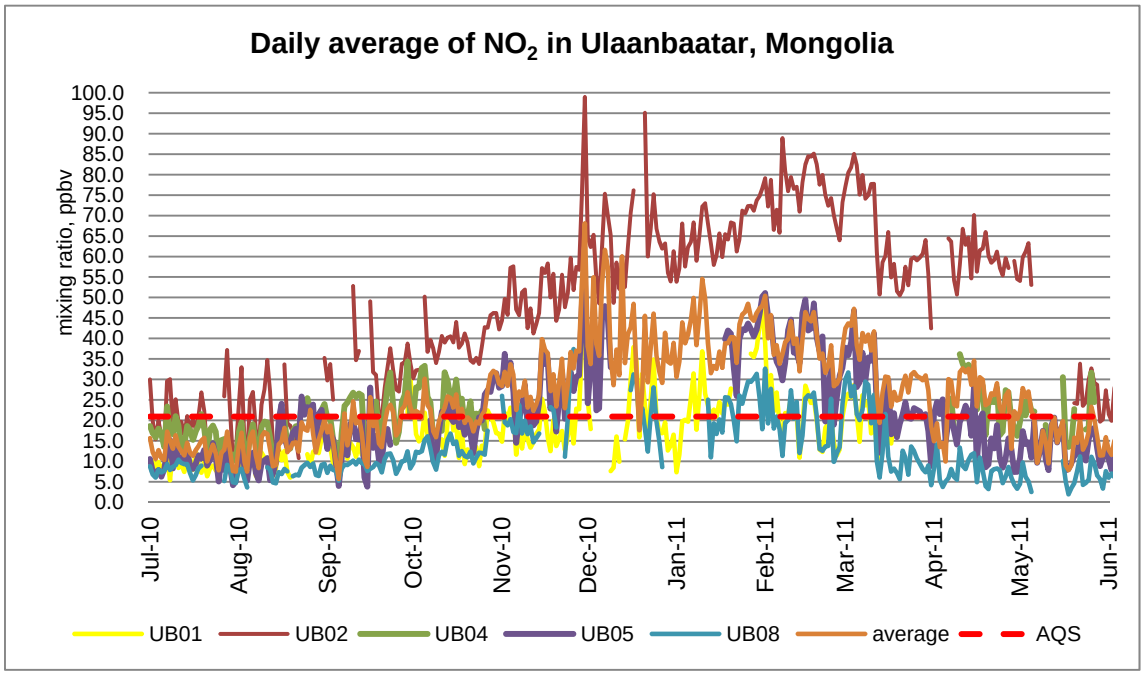


Figure 4. Annual variation of NO_2 at Ulaanbaatar sites, Mongolia.

The highest mixing ratio of NO_2 was measured at UB02 (central cross roadside), it was higher than air quality standard during the year. In the winter season, NO_2 was exceeded air quality standard at all sites of Ulaanbaatar. It emits from a residential area which is coal burning for heating.

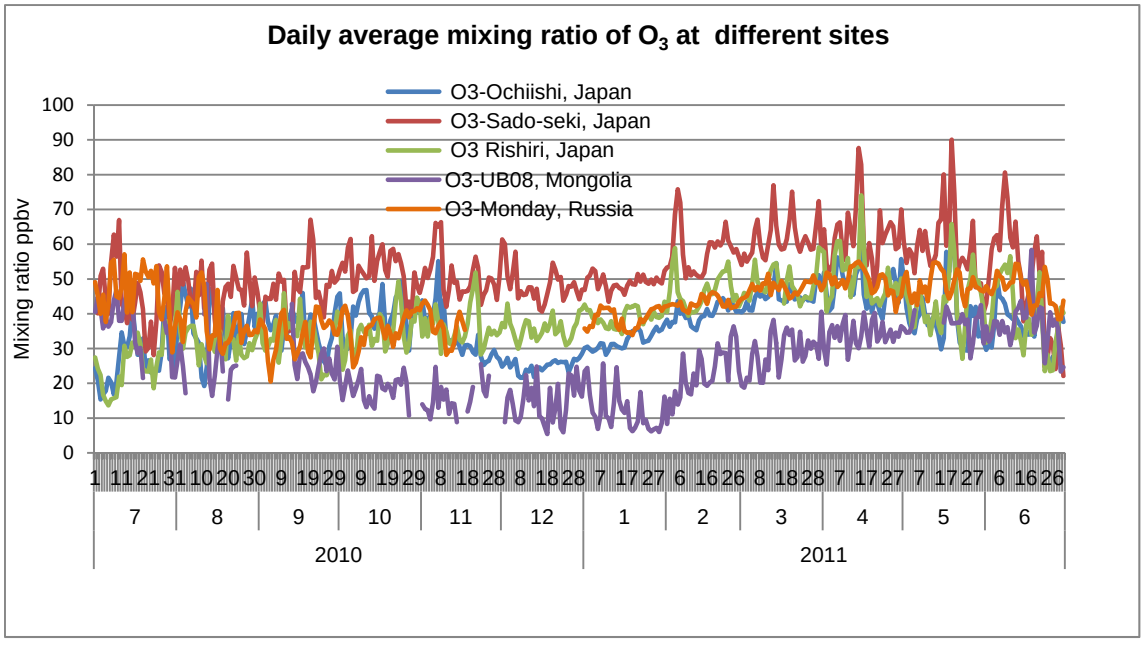
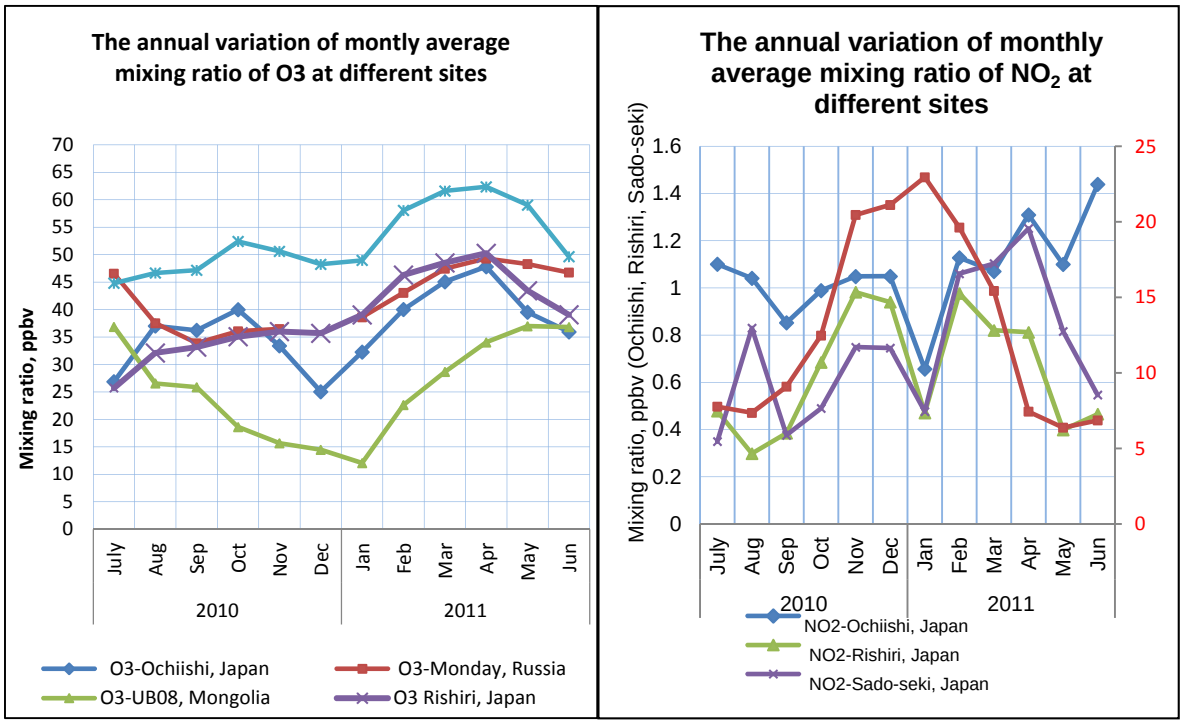


Figure 5. The daily average concentration of O_3 at different sites.

The daily averaged mixing ratio of O_3 was higher than other sites at Sado-Seki site of Japan during the year, lowest in Ulaanbaatar Mongolia every season from selected sites (Figure 5). The maximum mixing ratio was measured in July at Mondy of Russia and Ulaanbaatar (UB08) Mongolia. But it was observed in April and May in sites of Japan (Figure 5). In most of the Japan, the rainy season lasts from the beginning of

June to mid-July.



a) **b)**

Figure 6. Annual variation of O_3 (a.) and NO_2 at selected sites of Asia.

The lowest monthly average concentration of O_3 was observed in December at Ochiishi site and in January at Ulaanbaatar site. The highest monthly average concentration observed at Ulaanbaatar in January. NO_2 concentration was very low at sites of Japan.

3.2 Transport of ozone

3.2.1 Local transport of ozone

Ozone is likely to reach unhealthy levels on hot sunny days in urban environments. Ozone can also be transported long distances by the wind. For this reason, even rural areas can experience high ozone levels (http://www.epa.gov/air/ozonepollution/). The reason we have selected background site UB08 of Ulaanbaatar city. Average mixing ratios of gases were estimated by 16 wind direction at background site by hour and season.

At background site of Ulaanbaatar (UB08) has dominated 2 main wind direction Nigh time from East Southeast-East south (110-135), daytime West and West northwest (270-290). In winter time, highest concentration dominated when there wind direction from 270-340 and 110-135. In summer season higher concentration occurred with the (110-135) wind. It means NO_2 transported from residential area (270-340) and main central road 110-135) (Figure 7). The concentration of CO was shown similar. But it was higher when wind direction from near village such Khonhkhor and Nalaikh district (Figure 8).

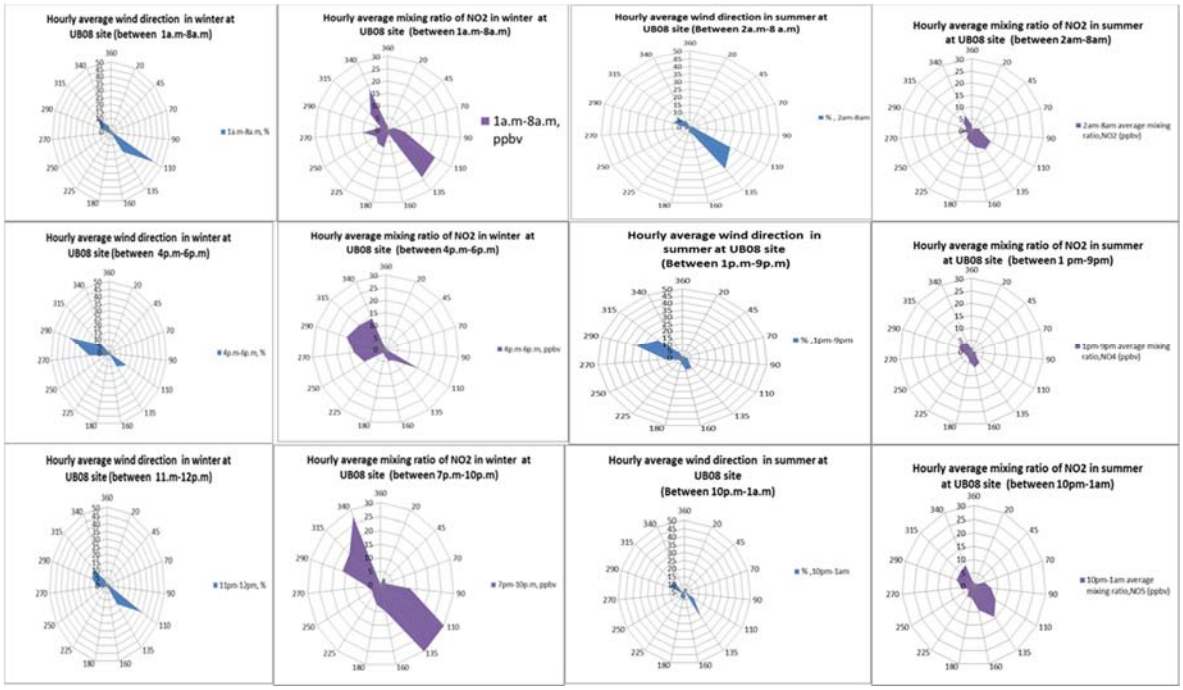


Figure 7. Frequency of wind direction (blue) and average concentration of NO_2 (purple) in winter and summer.

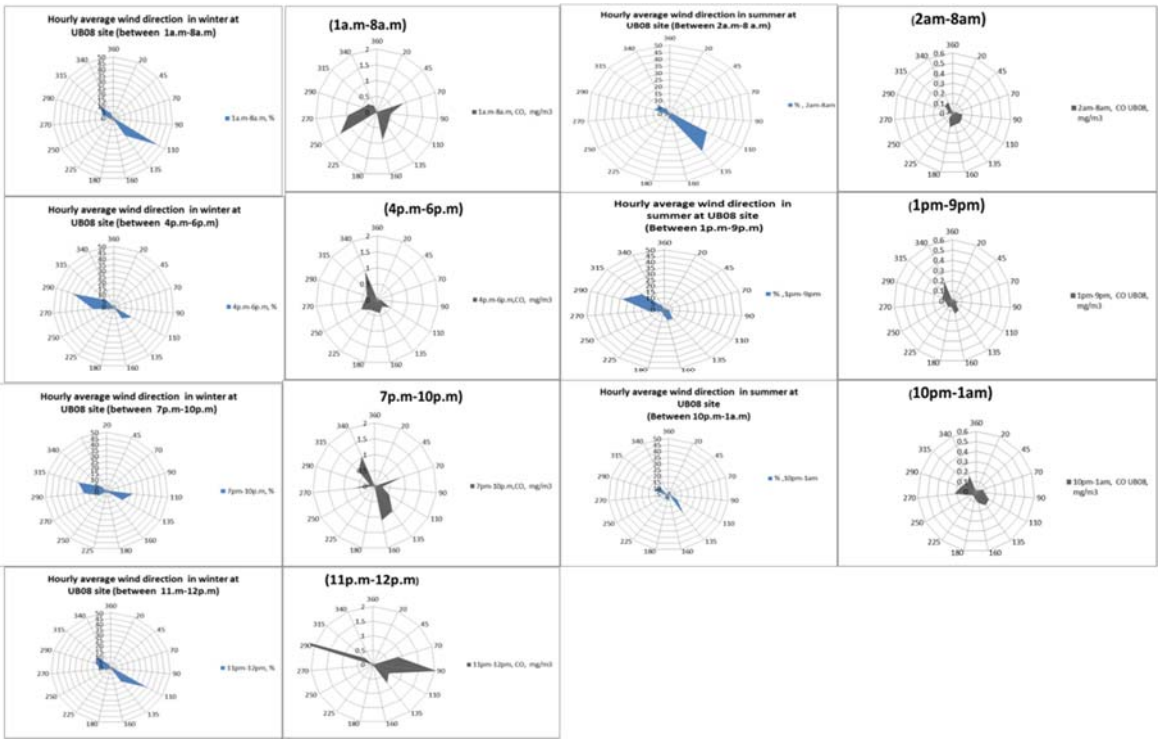


Figure 8. Frequency of wind direction (blue) and average concentration of CO (black) in winter and summer.

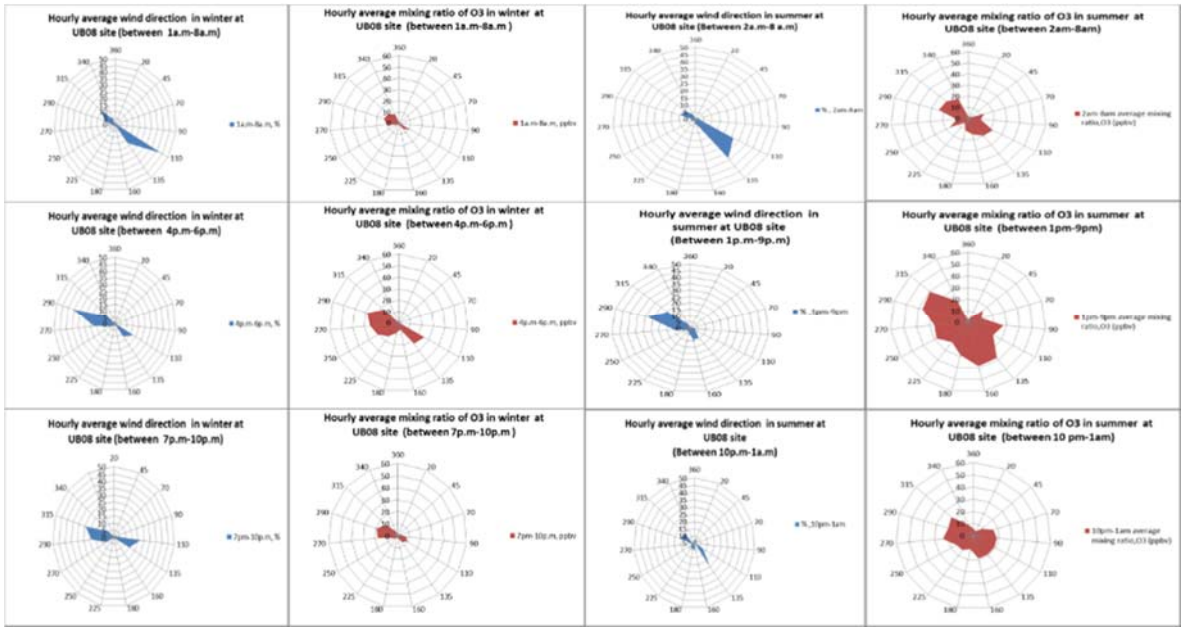


Figure 9. Frequency of wind direction (blue) and average concentration of O_3 (red) in winter and summer.

Higher concentrations of O_3 were the same direction with NO_2. But it was high in summer (Figure 9). It indicated that background site of Ulaanbaatar is located under the influence of city and major East road pollutants. There is local transport from the residential area of Ulaaanbaatar city usually daytime and main road nighttime.

3.2.2 Regional transport of ozone

We have compared daily average data of Mondy site and Ulaanbaatar sites. It was shown a higher concentration of O_3 measured at sites of Ulaanbaatar after 2-3 days Monday (Figure 10).

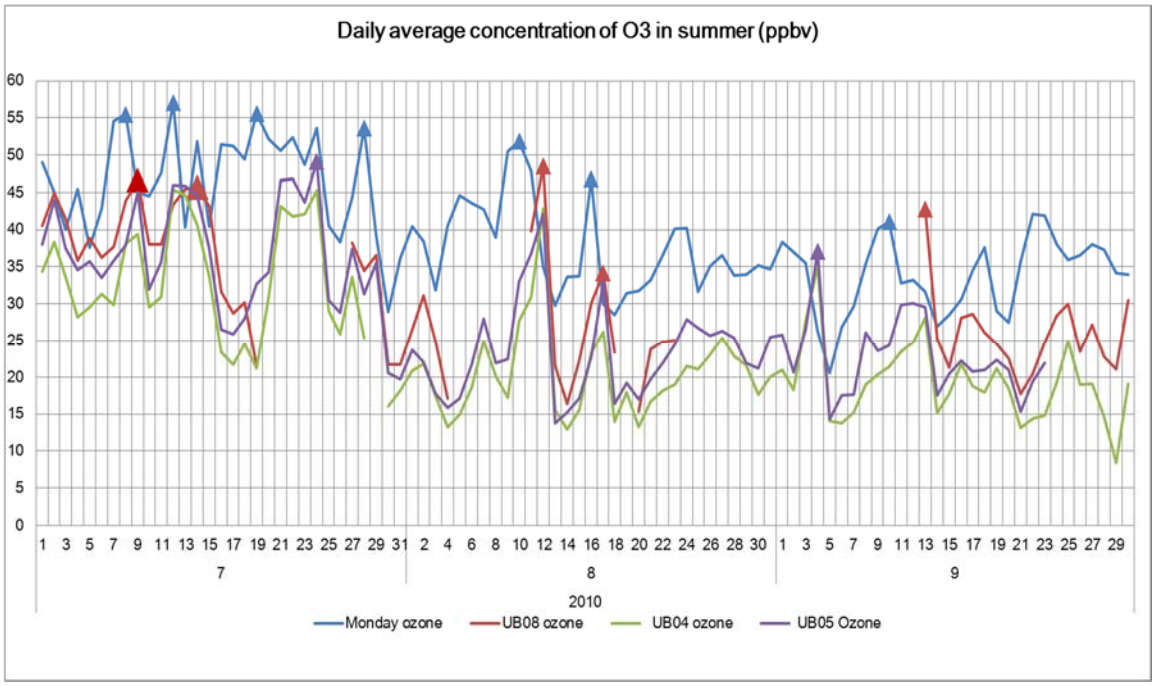


Figure 10. Summer mixing ratio of O_3 at Monday and Ulaanbaatar sites.

We have selected high mixing ratio of O_3 observed days and used 24 hours continuously meteorological parameters such temperature pressure, wind direction and speed and weather maps.

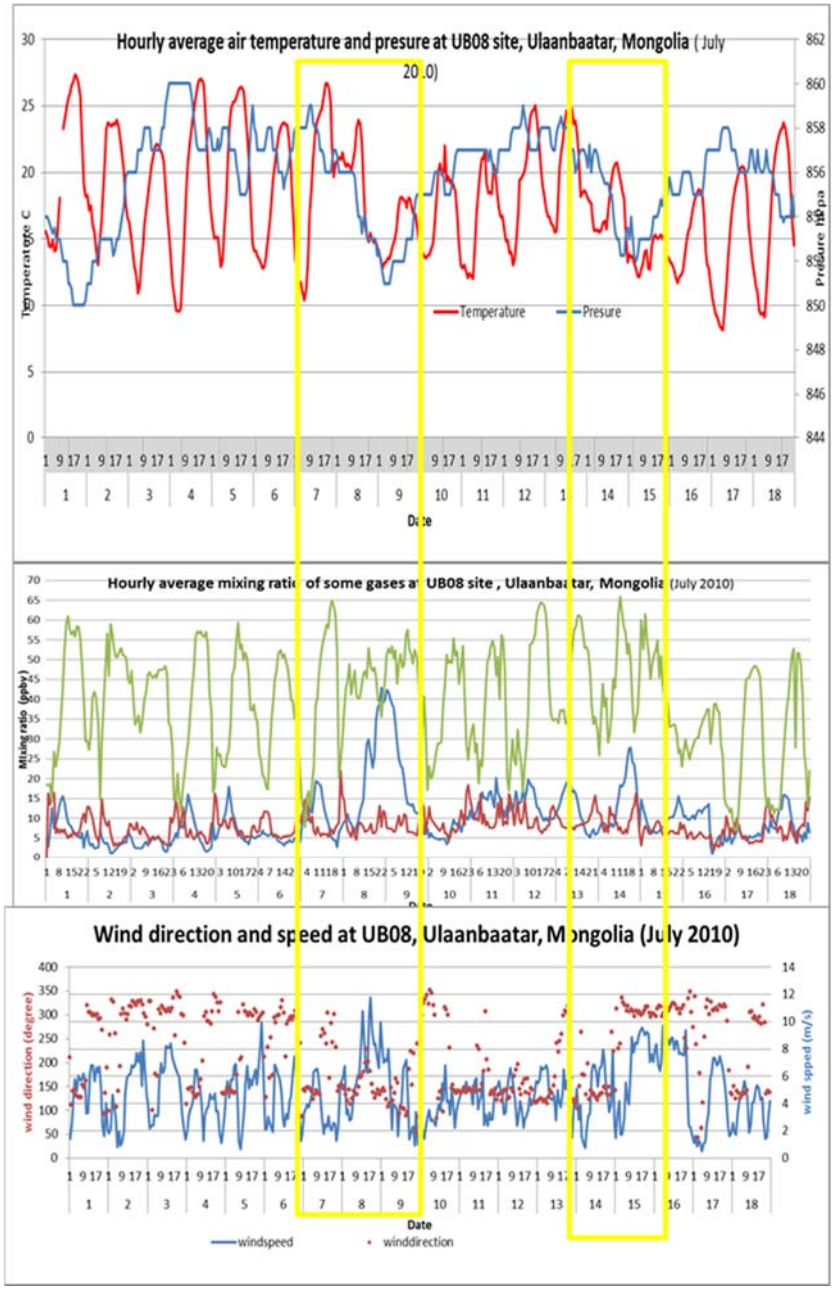


Figure 11. The time series of meteorological parameters during high O_3 episode days (7-9 and 14-15 July).

We selected 2 cases for 7-9 July and 14-15 July. The 2 cases, air temperature and pressure going down and up, the wind speed was unstable. It means weather front was crossing.

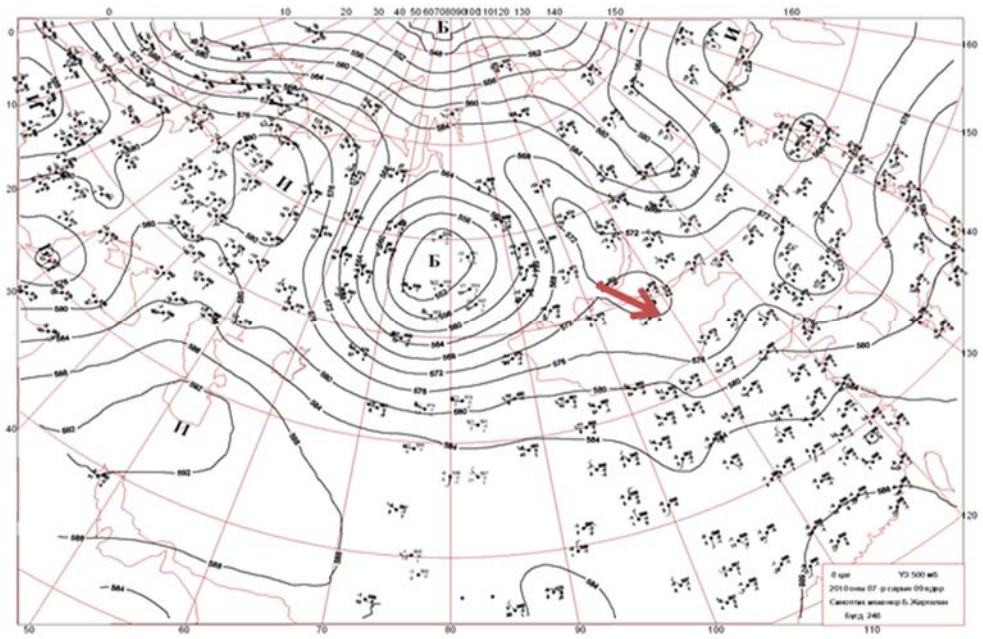


Figure 12. Weather map AT500 hPa upper air on 9 July 2010.

On 9 July 2010, the weak cyclonic system was found northern Mongolia. The same time deep strong cyclone was located on the Eurasian continent. Pakpong Pochard and Hajime Akimoto (2003) mentioned EU air masses are those originating in or passing through Europe before arriving on Monday. We would like to explain similar processing (Figures 12, 13) the high concentration of O_3 at Monday before 1-2 days Ulaanbaatar.

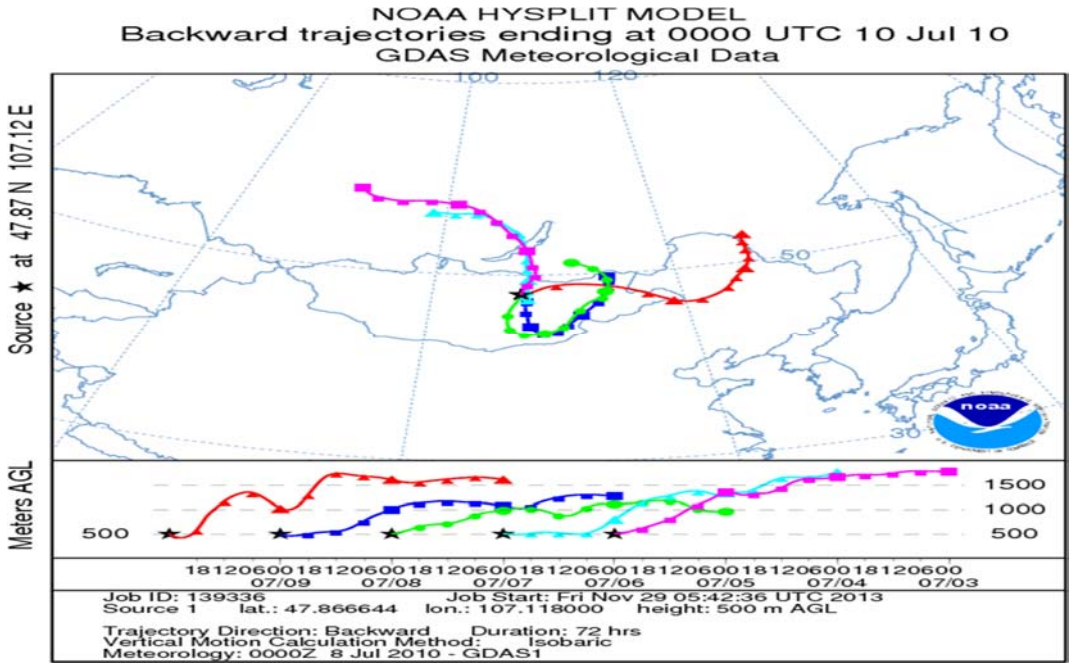


Figure 13. Hysplit model result.

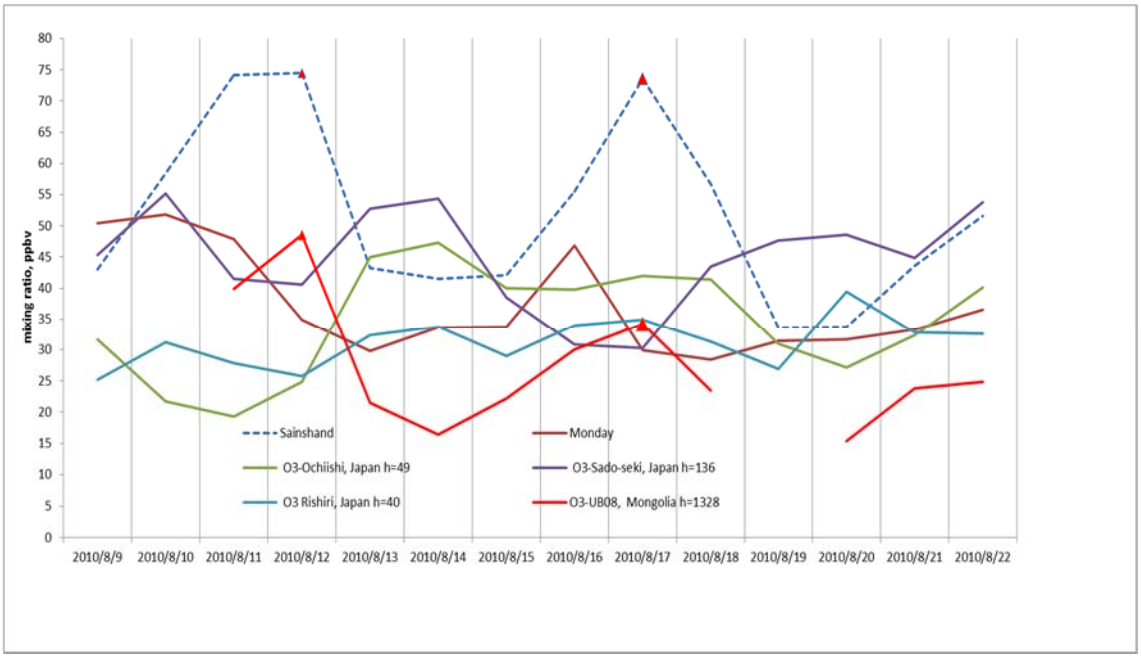


Figure 14. Ozone measurement on 9-22 August 2010.

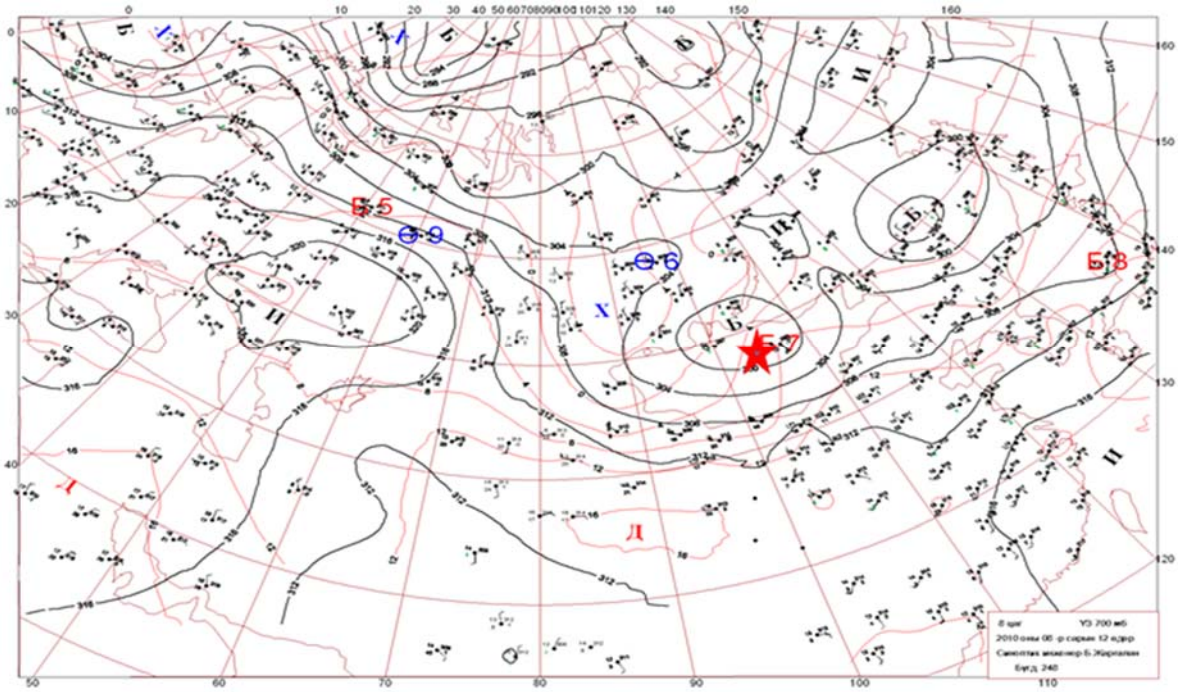


Figure 15. Weather map AT500 hPa upper air on 12 August 2010.

At the same day (12 and 17 August 2010) high concentration ozone was measured at Sainshand and Ulaanbaatar background station (Figure 14). We used weather maps and hysplit model to explain the cyclonic process related to the higher concentration. The day's cyclone was lying on the territory of Mongolia (Figure 15). It is possible to mix northern and southern air in the area. Hysplit model was shown the similar trajectory from northern and southern (Figure 16). The highest concentration of O_3 measured at Sado-Seki site of Japan on 14 and 19 April 2011.

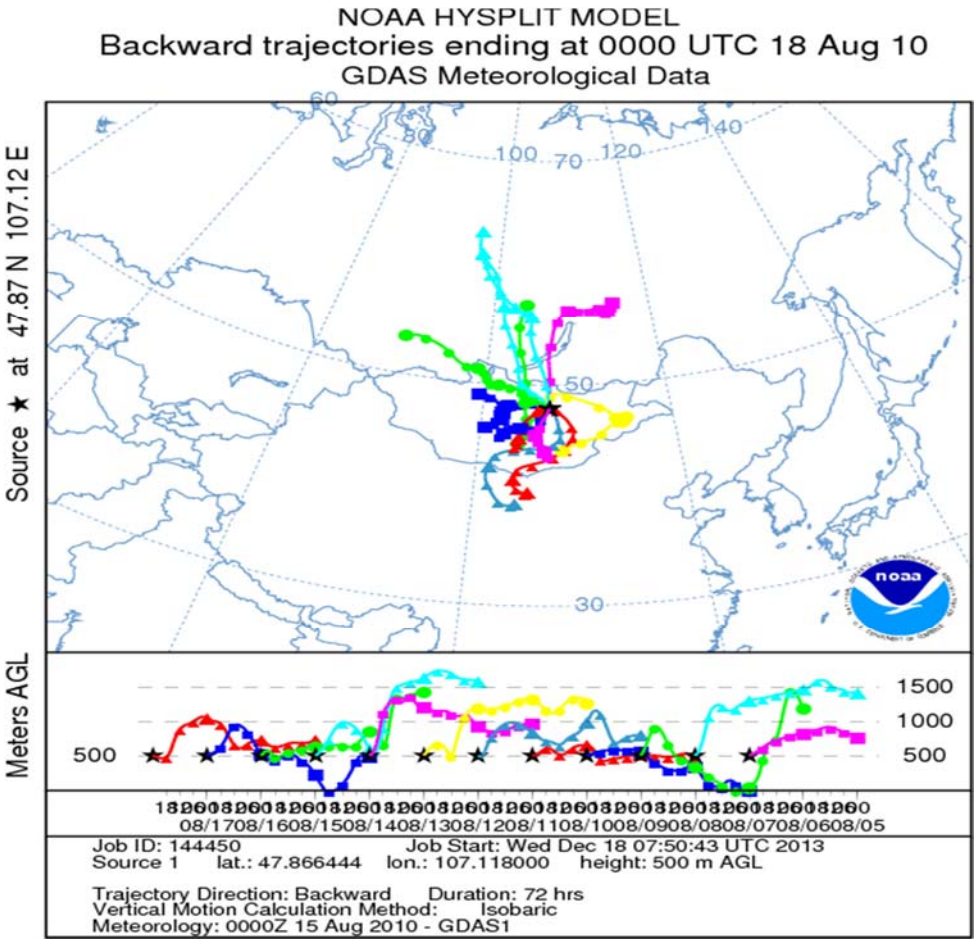


Figure 16. Air mass transport patterns arriving at Ulaanbaatar and Sainshand (Southeast Mongolia) by Hysplit model (NOAA) August 2010.

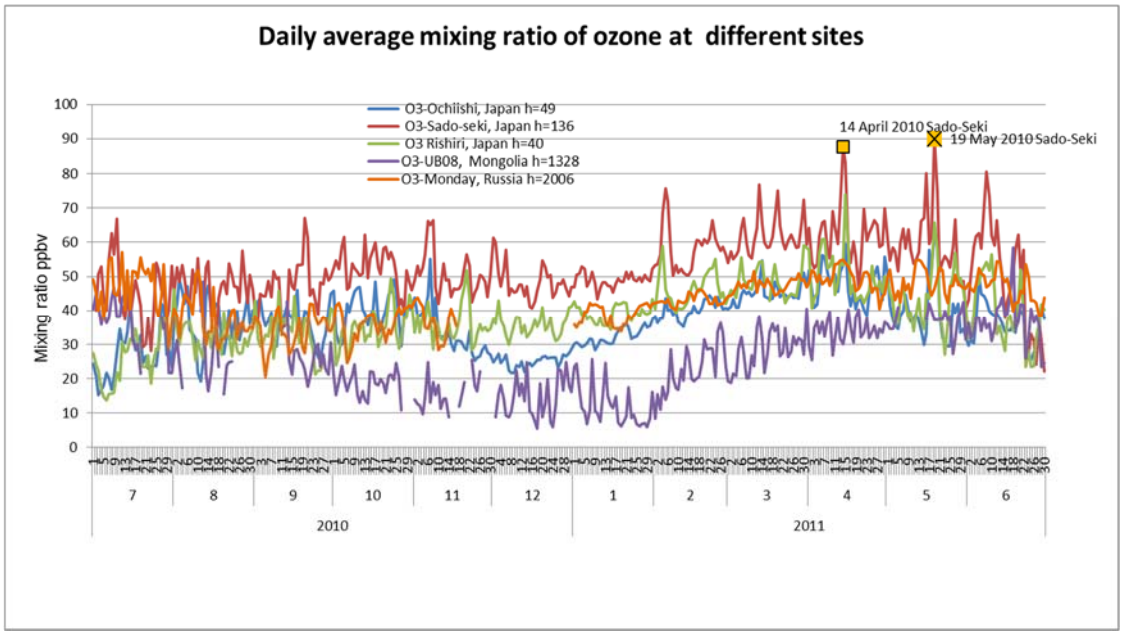


Figure 17. Daily average mixing ratio of O_3 at sites of Japan, Russia, Mongolia (July 2010-June 2011).

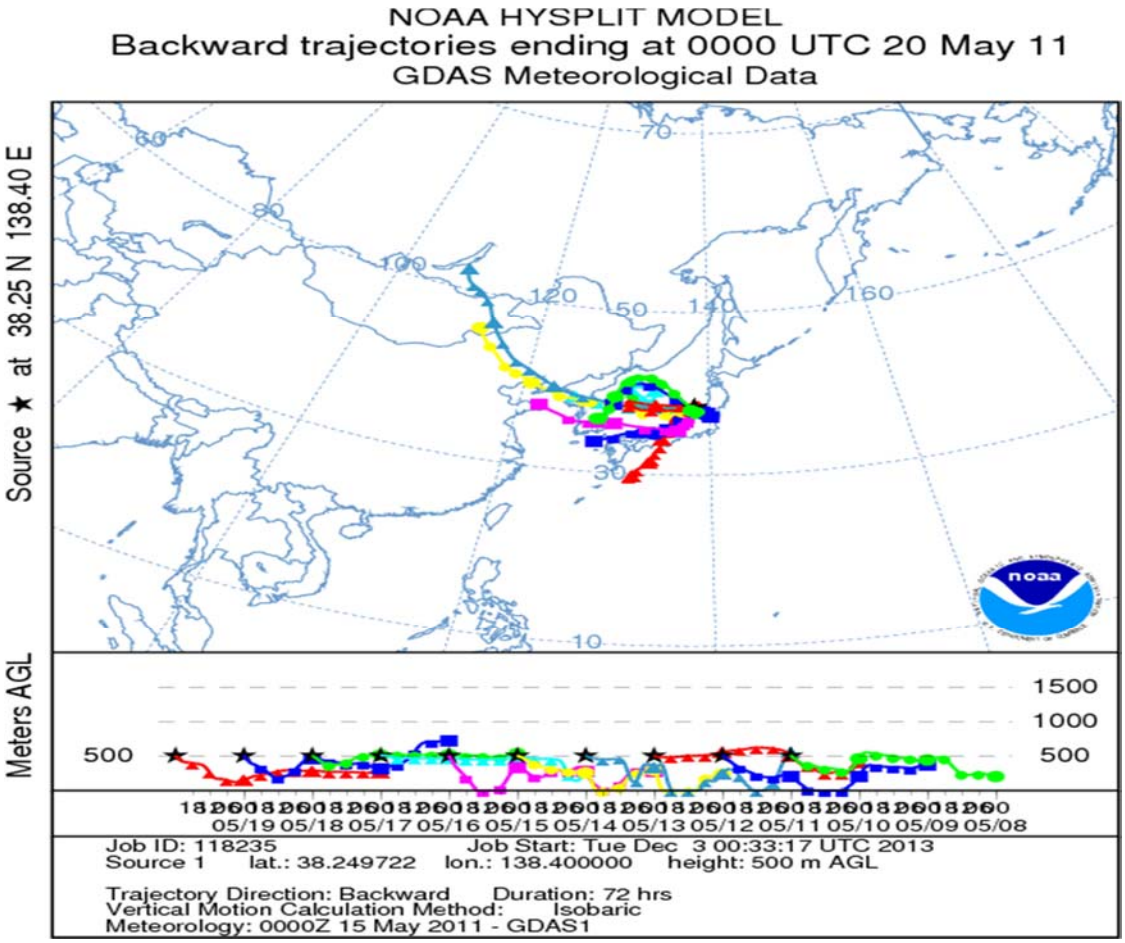


Figure 18. Air mass trajectories by Hysplit model (NOAA), May 2010.

The during days with high concentration ozone, the Hysplit model results are shown air mass transport from the west side such China and Korea. But main ozone precursor NO_2 was very low.

5. Conclusion

- Air quality monitoring data of Ulaanbaatar sites (July 2010 to June 2011) are indicated general air pollution trend of Ulaanbaatar city. There are big seasonal (high concentration in summer July, min-Jan) diurnal variation of ozone (maximum in summer), NO_2, CO (high concentration in winter Feb) and depending emission sources and meteorological condition.
- The UB08 site is not background station. It is under the influence of city pollution (270-320WNW) and main (ESE 110-135) road pollution (Maybe from Nalaikh district).
- Highest mixing ratio of O_3 observed at Sedo-Seki site, Japan (it may be transported from South East Asia because NO_2 is lower than 1ppbv in Japan remote sites)
- There is long range transport from neighbor countries (from Russia and China), but city pollution is dominated in Ulaanbaatar.

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Meteorological condition during the highly polluted and clean days in Ulaanbaatar, Mongolia

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Abstract

In order to investigate meteorological condition during highly polluted days in Ulaanbaatar, Mongolia, 1-h averaged PM_{10} (particulate matter with an aerodynamic diameter ≤ 10 μm), SO_2 (sulfur dioxide), NO_2 (nitrogen dioxide), wind speed and direction, air temperature, and relative humidity were analyzed at an urban (UB05), a roadside (UB02), an industrial (UB07) and a background (UB08) sites between January 1 and December 31, 2012. In general PM_{10} and SO_2 concentrations were the highest at UB05 followed by UB07 and UB02 and the lowest at UB08 for all seasons. Clear seasonal variation, winter high and summer low, was measured for PM_{10} and SO_2. During winter season, the highest PM_{10} was found to be 1112.0 ± 649.5 μg m^{-3} at the UB05 site whereas the lowest was found to be 175.1 ± 111.2 μg m^{-3} at the UB08 site. Strong diurnal variation, morning and evening high afternoon low, at the urban and industrial sites during winter months due to coal burning for space heating. Clear diurnal variation, evening and afternoon high, morning low was observed at the road site during winter months mainly from not only traffic but also fugitive dust. Pollution level in Ulaanbaatar during wintertime was highly dependent on temperature inversion, and wind speed and direction. Pollution was accumulated due to very thick and strong temperature inversion and low wind mostly from north and northeast. Pollution level during spring time was depending on synoptic condition. Low pressure system led unstable weather condition which brought high PM_{10}.

Key words: Best20%, Worst20%, Weather condition, Temperature inversion

1. Introduction

Poor air quality and pollution episodes at any location are highly depended on a wide range of factors that include pollutant emission sources and meteorological and topographical features of the region. Especially in cities, industrialization, population, traffic density and meteorological condition have significant effect on atmospheric air pollution. Measured urban PM_{10} quality monitoring sites which are operated by National Agency for Meteorology and Environmental

and $PM_{2.5}$ concentrations and their spatial and temporal variations for different locations have been discussed extensively (Malek *et al*., 2006), (Solberg *et al*., 2008). Godowska (2004) and Walczewski *et al* (1997) pointed out that black smog in winter and photochemical smog in summer were observed due to weather condition such as high pressure system, vertical distribution of temperature and air mass types. Stefan *et al.* (2013) concluded that the meteorological factors influence the pollutant mass concentrations by increasing the dispersion processes and also dry and wet depositions in near forest, lake and sub-urban area of Romania. The wind speed and wind direction showed deterministic impacts on trace gas pollution levels, providing information on regional pollution characteristics (Xu *et al*., 2011). The most significant variables for PM_{10} were temperature, wind, water vapor pressure and boundary layer height at Melbourne and the relative importance of local weather as driver of regional air pollution as well as the marginal effects of individual meteorological variables.

According to the WHO report (WHO. 2012), Ulaanbaatar, capital of Mongolia, is one of the most polluted city in the world. The Ulaanbaatar is located in a valley of the Tuul River surrounded by the Khentei Mountain area with elevations ranging from 1,652 m to 1,949 m above sea level. Its location causes Ulaanbaatar to experience frequent temperature inversion especially during cold months. Beside frequent temperature inversion, a stationary anticyclone which keeps stable weather condition, forms over north-western Mongolia mostly from October to April of following year (Natsagdorj, 1982), (Chung *et al*., 2004). There are several types of air pollution sources for instance; ger area, thermal power plants, vehicles heat only boilers, fugitive dust from paved and unpaved roads, petrol stations etc. In Ulaanbaatar City, wintertime air pollution mainly caused by coal combustion at 3 thermal power plants, 200 Heat Only Boilers (HOBs), 1000 Coal Fired Water Heaters (CFWHs) and 140,000 ger stoves. Several researches have been conducted on air pollution in Ulaanbaatar Mongolia, mainly focusing on chemical characteristics of fine particles which were measured at an urban site of Ulaanbaatar (Jung *et al.*, 2010), (Nishikawa *et al*., 2011), (Davy *et al*., 2011), (Batmunkh *et al*., 2013). However, few studies have been conducted on meteorological condition for different pollution level. Therefore, objective of the study is to investigate meteorological condition during clean and highly polluted days in Ulaanbaatar.

2. Method

2.1 Measurement site

The measurements were conducted from January 01, 2012 to December 02, 2012 at five different air quality monitoring sites: in an urban, a road, an industrial, and a background sites.

The UB02, representative of roadside, is surrounded by crossroads, and located 6-8 km away from two big power plants. There are two urban sites as UB04 and UB05. The UB05 is surrounded by ger area which is traditional dwelling of Mongolia whereas the UB04 is surrounded by residential area which is mostly apartments. The sampling site UB07 represents industrial area which is surrounded by small-scale industries including bricks and cements. The UB08 is background site, is located about 23 km away from downtown of the city.

2.2 Air quality and meteorological data

The 1-hour averaged air quality data (PM_{10}, SO_2, NO_2) and meteorological parameters (wind speed, direction, precipitation, air temperature, and relative humidity) were obtained from air

Monitoring. The $PM_{2.5}$ mass concentration was measured only at UB02 monitoring site. Radiosonde data were obtained from Tahilt meteorological station. Radiasonde was launched twice (8:00 a.m. and 8:00 p.m.) a day. Inversion intensity (dT) was determined as the temperature difference between the bottom and top of the inversion layer. Inversion depth (dH) was determined as the distance in meters from the surface to the top of the inversion. In order to characterize atmospheric condition, approximately 700 weather charts were examined for surface pressure characteristics.

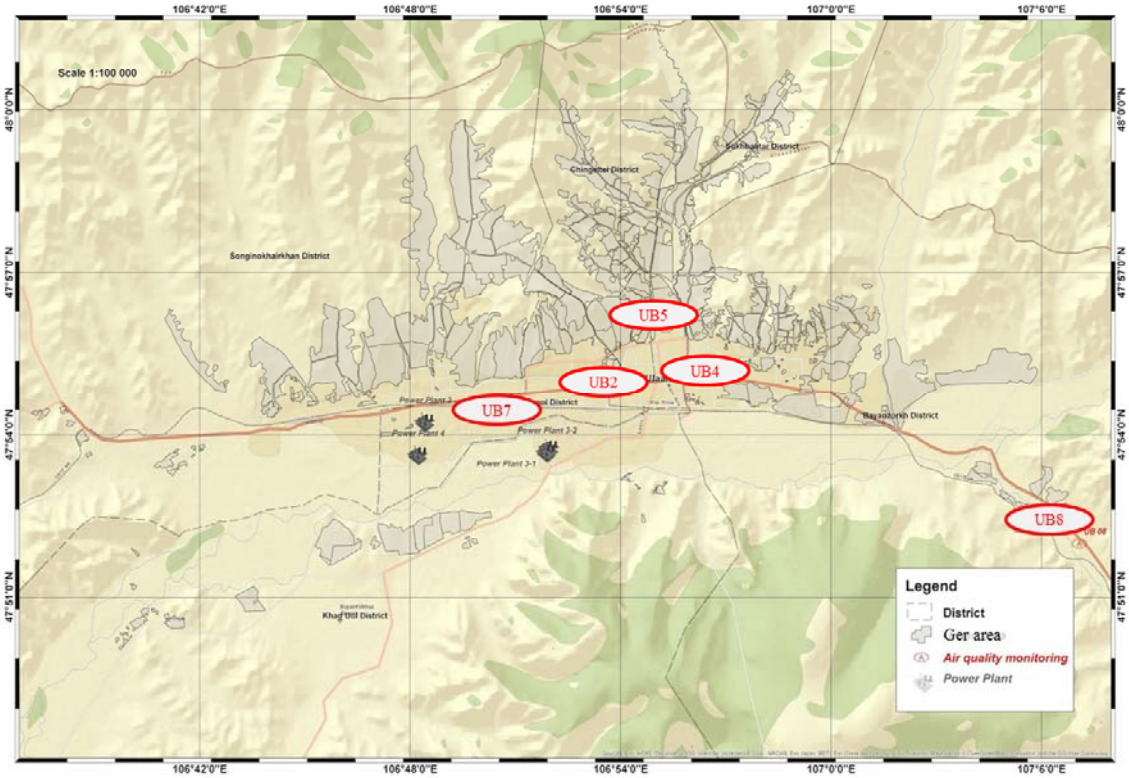

Figure 1. Location of air quality measurement sites.

2.3 Classification of pollution event

The pollution level was classified into two categories based on the daily average particulate mass concentration. Worst20% (daily average values for the 20% most impaired days) and Best20% (daily average values for the 20% least impaired days) (Kim *et al.*, 2006).

3. Results and discussion

3.1 General characteristics of atmospheric air pollution

In general PM_{10} and SO_2 concentrations were the highest at UB05 followed by UB07 and the lowest at UB08 for all seasons. During winter season, the highest PM_{10} was found to be 1112.0 ± 649.5 μg m^{-3} at the UB05 site whereas the lowest was found to be 175.1 ± 111.2 μg m^{-3} at the UB08 site, showing that air was highly polluted not only urban area but also background site. Clear seasonal variation, winter high and summer low, was measured for PM_{10} and SO_2. The mean of PM_{10} mass concentration at UB05 were measured to be 1112.0 ± 649.0 μg m^{-3} in winter, 447.2 ± 442.5 μg m^{-3} in spring, 125.9 ± 64.9 μg m^{-3} in summer and 233.6 ± 285.2 μg m^{-3} in fall.

Among winter months, January is the highest polluted month, 24-hour mean PM_{10} reached

2736.1 ± 2235.1 μg m^{-3} at the urban site of Ulaanbaatar. Diurnal variation was investigated for different sites and seasons. Diurnal variation was significantly depending on season and location (Figure 3). Strong diurnal variation, morning and nighttime high afternoon low, was observed for PM_{10} at UB05 (urban site) in winter and fall season. SO_2 had similar variation (now shown) with PM_{10} at same monitoring site and seasons indicating that morning and evening peaks were mainly from coal burning for space heating. In case of road (UB02) and industrial (UB07) sites, the first peak was observed between 2:00 a.m and 3:00 a.m (local time) then second peak was measured from 1:00 p.m. to 3:00 p.m. (local time). The first peak was associated with SO_2 peak showing that high PM level was mainly from coal burning. Since the UB02 and UB07 are roadside and industrial sites, respectively, the second peak was associated with fugitive dust from paved and unpaved roads.

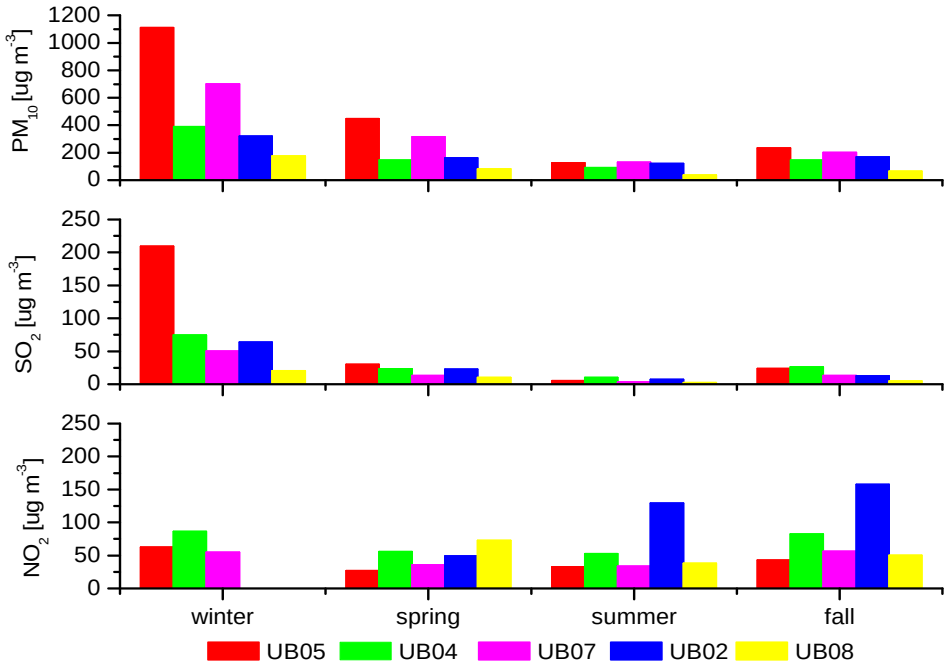

Figure 2. Seasonal variation of PM_{10} (upper), SO_2 (middle) and NO_2 (bottom) concentration at 5 monitoring sites in Ulaanbaatar.

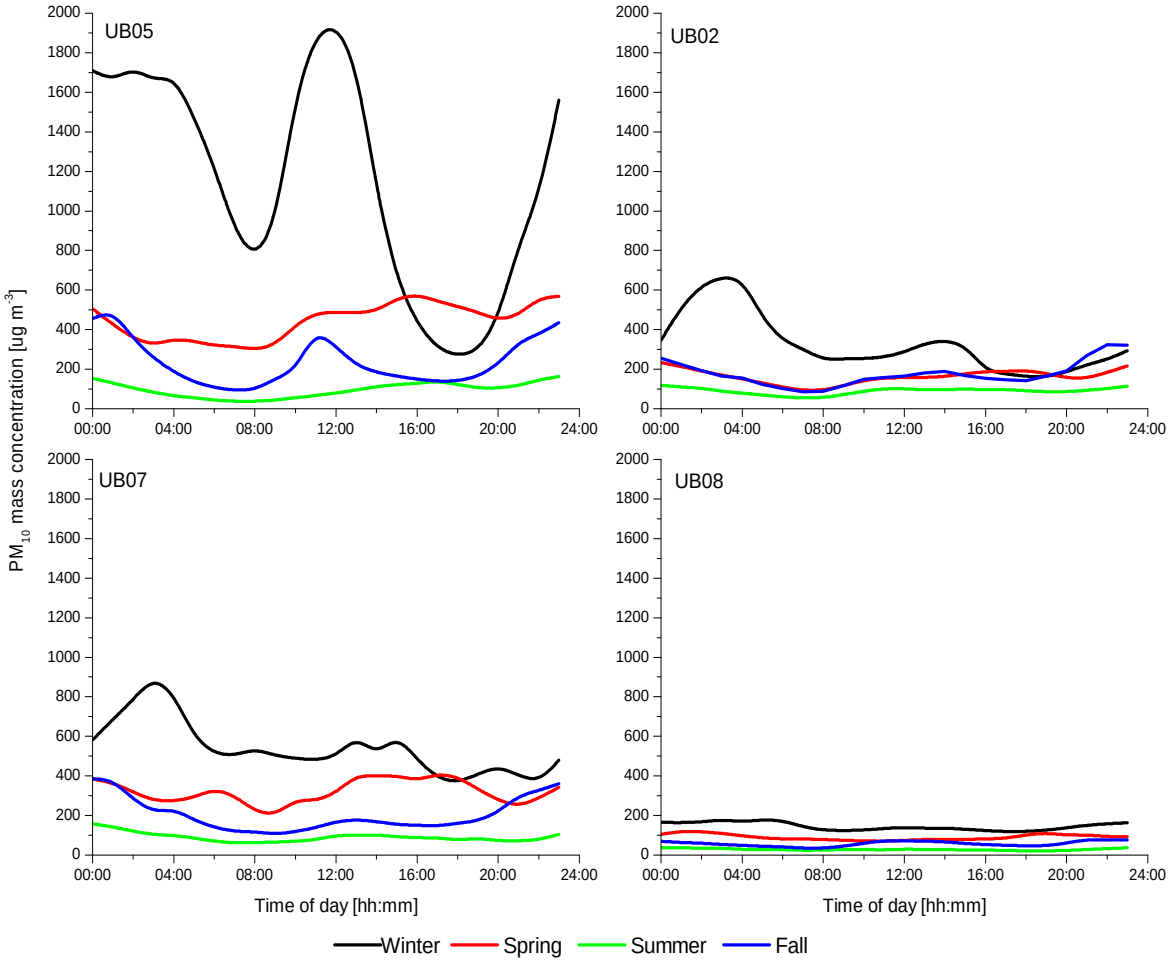

Figure 3. Diurnal variation of PM_{10} for different seasons and sites.

3.2 Analysis of severe and low pollution level

The pollution level was classified into two categories based on the daily average particulate mass concentration Best20% and Worst20% based on Kim's classification (Kim *et al.*, 2006). Table 1 shows mean concentration of PM_{10}, SO_2, and NO_2 for different events, sites and seasons. In winter season, the mean concentration of PM_{10} during the Worst20% case was approximately 5 to 11 times higher than that measured during the Best20% case for the different sites. The highest mean concentration of PM_{10} during the Worst20% condition was measured to be 2150.6 ± 256.3 μg m^{-3} at UB05 whereas the lowest was found to be 339.5 ± 142.3μg m^{-3} at the UB08 site. The mean concentration of SO_2 showed similar variation as PM_{10}, indicating that higher PM_{10} was mainly from coal combustion in Ulaanbaatar. In summertime, the mean concentration of PM_{10} during the Worst20% case was approximately 3 to 5 times higher than that measured during the Best20% case for the different sites. The SO_2 concentration was as low as, ranged between 3.7 μg m^{-3}-15.3 μg m^{-3} and 1.6 μg m^{-3}-7.8 μg m^{-3} during the Worst20% and Best20%, respectively.

Table 1. Mean of PM_{10}, SO_2, NO_2 for different events, sites and seasons.

		PM_{10} [ug m^{-3}]		SO_2 [ug m^{-3}]		NO_2 [ug m^{-3}]	
		Worst20%	Best20%	Worst20%	Best20%	Worst20%	Best20%
Winter	UB05	2150.6 ± 256.3	339.5 ± 80.3	350.4 ± 74.8	96.6 ± 30.5	85.8 ± 16.1	37.7 ± 4.1
	UB04	848.2 ± 126.7	101.4 ± 40.9	94.1 ± 31.8	37.8 ± 10.3	78.2 ± 19.3	73.2 ± 22.9
	UB07	1965.2 ± 512.2	166.1 ± 34.8	69.1 ± 11.7	41.5 ± 9.5	67.3 ± 11.3	51.2 ± 8.5
	UB02	693.5 ± 303.4	101.6 ± 22.5	71.3 ± 27.1	38.3 ± 17.1	-	-
	UB08	339.5 ± 142.3	67.6 ± 17.9	26.2 ± 4.6	11.4 ± 2.1	-	-
Summer	UB05	210.5 ± 29.9	42.7 ± 14.1	6.5 ± 3.0	6.9 ± 4.9	37.3 ± 4.8	31.0 ± 8.8
	UB04	165.8 ± 49.5	39.5 ± 10.4	15.3 ± 7.5	7.8 ± 4.6	59.2 ± 11.4	52.5 ± 8.6
	UB07	213.0 ± 38.7	37.4 ± 15.1	7.1 ± 4.8	3.5 ± 1.9	28.4 ± 19.9	31.7 ± 16.1
	UB02	186.0 ± 17.8	63.6 ± 20.1	6.6 ± 4.9	5.1 ± 5.1	131.5 ± 17.5	109.0 ± 37.4
	UB08	66.1 ± 19.3	18.9 ± 4.5	3.7 ± 1.3	1.6 ± 0.5	52.6 ± 25.6	35.7 ± 26.1

The highest NO_2 concentration was found to be 131.5 ± 17.5 µg m^{-3} and 109.0 ± 37.4µg m^{-3} at UB02 during the Worst20% and Best20%, respectively, in summertime showing combustion from transportation is dominant near the site. Moreover, high NO_2 level might be due to photochemical processes.

In order to define daily characteristics of PM level for different pollution level diurnal variation was plotted for different sites and seasons. Figure 4 shows diurnal variation of PM_{10} during the Worst20% and Best20% in winter, spring, and summer. During the winter time, PM_{10} has showed strong diurnal variation (SO_2 has similar diurnal variation—not shown here), evening & morning high, afternoon low, especially at the UB05 site which is surrounded by ger area indicating that the peaks are mainly from coal burning for space heating. PM_{10} has a similar behavior though magnitude is lower at the UB04 site. Diurnal variation of PM_{10} at UB02 has different trend such as nighttime and daytime high and morning low. Since UB02 is located at roadside, afternoon peak is mainly from not only traffic but also fugitive dust. Diurnal variation of PM_{10} at the UB07 was similar to the UB02, showing the afternoon peak was mainly from fugitive dust.

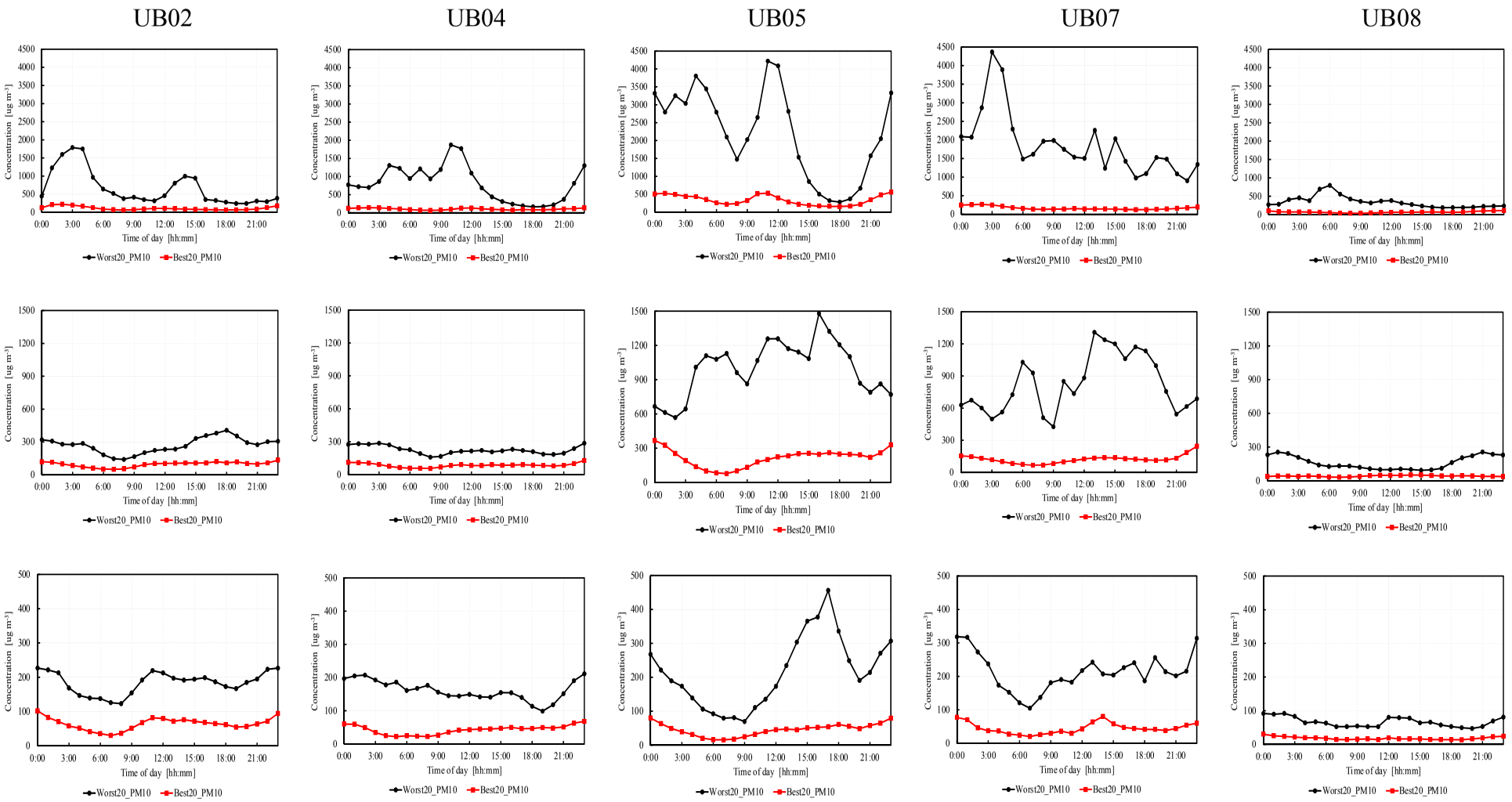

Figure 4. Diurnal variation of PM_{10} during the Worst20% (black line with circle) and Best20% (red line with square) in winter (upper panel), spring (middle panel) and summer (bottom panel).

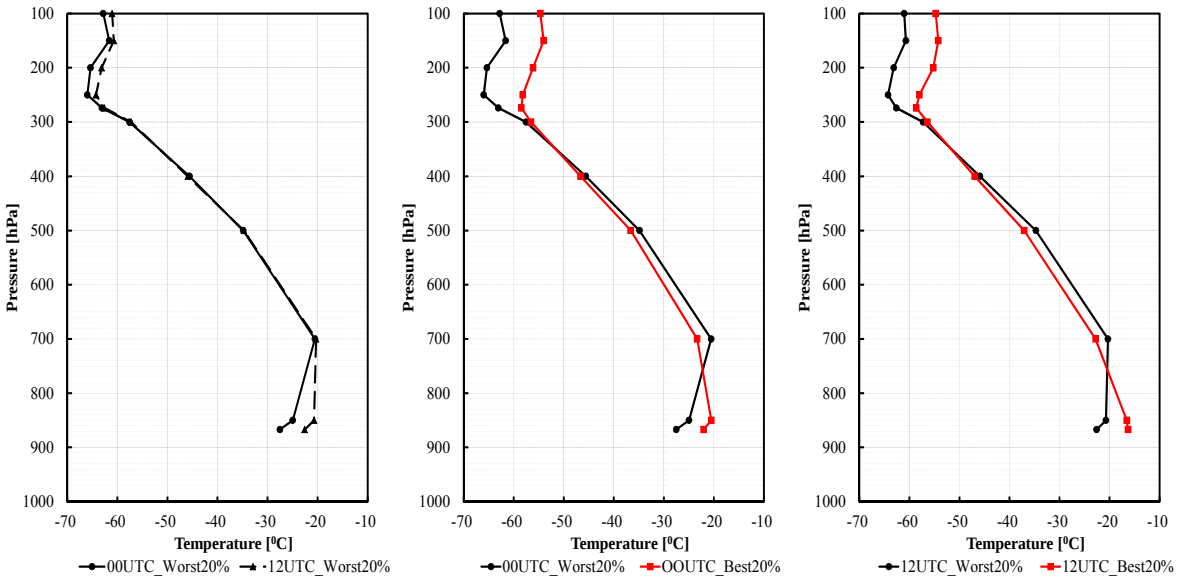

Figure 5. Temperature profile during the Worst20% and Best20% cases at morning and night time.

Temperature inversion intensity (dT) and thickness (dH) were calculated during the Worst20% and Best20% cases in winter season. The dT and dH at 8:00 a.m. (local time) and at 8:00 p.m. (local time) during the Worst20% were found to be 7.0 °C, 1640 m, and 2.2 °C, 1670 m, respectively. During the Best20% condition at 8:00 a.m. (local time), the dT was measured to be 1.5 °C and dH was 249 m which were much lower than that during the Worst20% condition. Temperature inversion disappeared at 8:00 p.m. during the Best20% condition. Hence, pollution level was highly depending on temperature inversion in winter season.

During the spring time, significant diurnal variation afternoon nighttime high morning low was observed at urban, industrial and roadside. In order to investigate pollution level and meteorological parameters, dT and dH were calculated for different pollution level. Temperature inversion was not occurred for the cases, Worst20% and Best20%, indicating pollution level was independent of temperature inversion. Therefore, surface weather chart and 4-day air mass back trajectories were examined during the Worst20% and Best20% conditions. According to surface weather analysis, low pressure system was placed over Mongolia and unstable weather condition was dominant during the Worst20% condition. As seen air mass back trajectories, the 4-day back trajectories were highly variable and originating from scattered area.

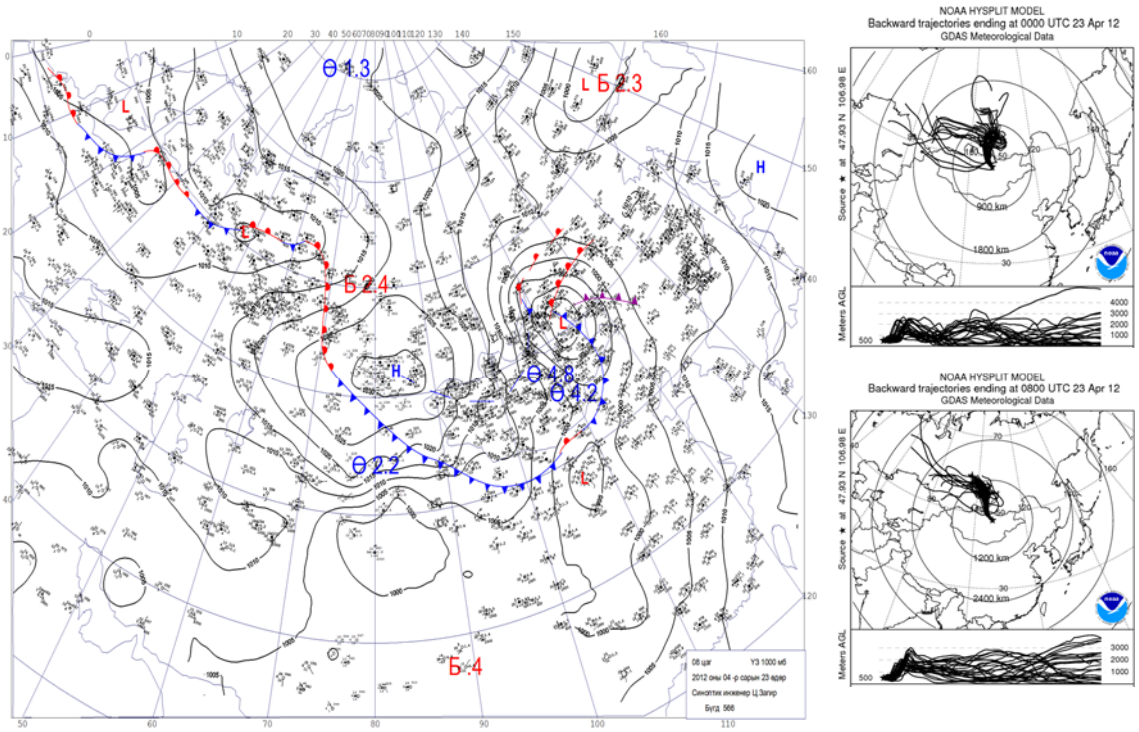

Figure 6. Surface weather chart at 8:00 (MST) and 4-day airmass backward trajectories on 23 April, 2012.

The relationship among the PM_{10} mass concentration and wind speed under Best20% and Worst20% conditions in winter and spring was illustrated in Figure 7. Wind speed is negatively correlated to PM_{10} in winter. Higher correlation is found to be 0.48 during the Best20% in winter. Wind speed is positively correlated to PM_{10} in spring. The highest correlation is obtained to be 0.74 during the Worst20% in spring, indicating that higher wind speed brings higher PM_{10}. It is indicating that wind speed was relatively important for particular mass concentration. Therefore, relationship between PM_{10} mass concentration and wind direction was examined under Best20% and Worst20% condition at UB05 monitoring site (Figure 8). Dominant wind direction was North and Northeast during the Worst20% case most of the time except spring. Wind direction was mostly from northwest for the Worst20% case in spring season. Wind direction was highly scattered during the Best20% condition in winter season. The ratio of $PM_{2.5}$ to PM_{10} was calculated at the UB02 monitoring site because $PM_{2.5}$ mass concentration data are available only that monitoring site. The mean $PM_{2.5}/PM_{10}$ was found to be 0.72 ± 0.19 in winter, 0.34 ± 0.23 in spring, 0.32 ± 0.19 in summer, and 0.46 ± 0.27 in fall, showing that fine particulate matter was dominant during the winter season whereas coarse particles are dominant other seasons. The obtained $PM_{2.5}/PM_{10}$ (0.72) in Ulaanbaatar during the winter season was comparable to that found in Beijing (0.71) from December 2002 to February 2003 (Zhang *et al.*, 2006).

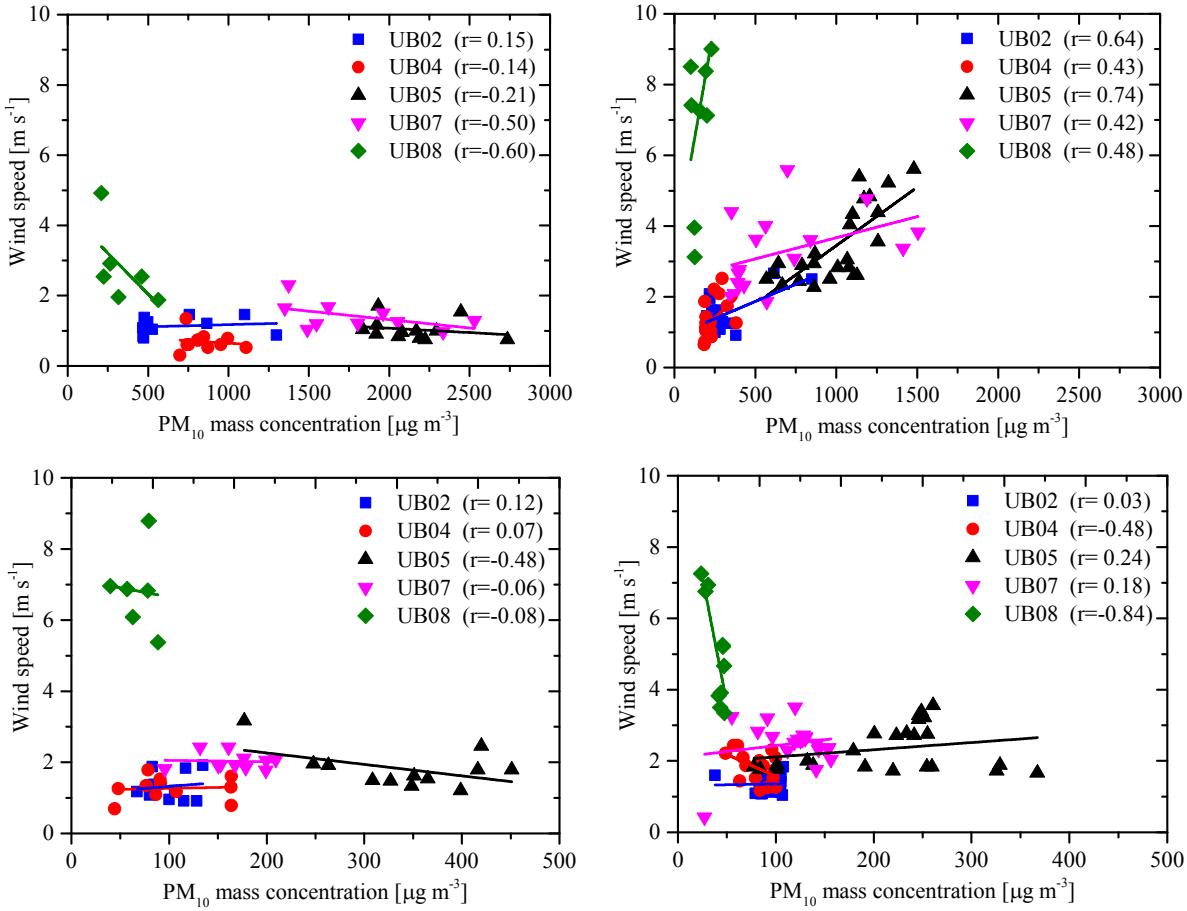

Figure 7. Relationship between PM_{10} and wind speed during the Worst20% (upper panel) and Best20% (bottom panel) in winter (left panel) and spring (right panel).

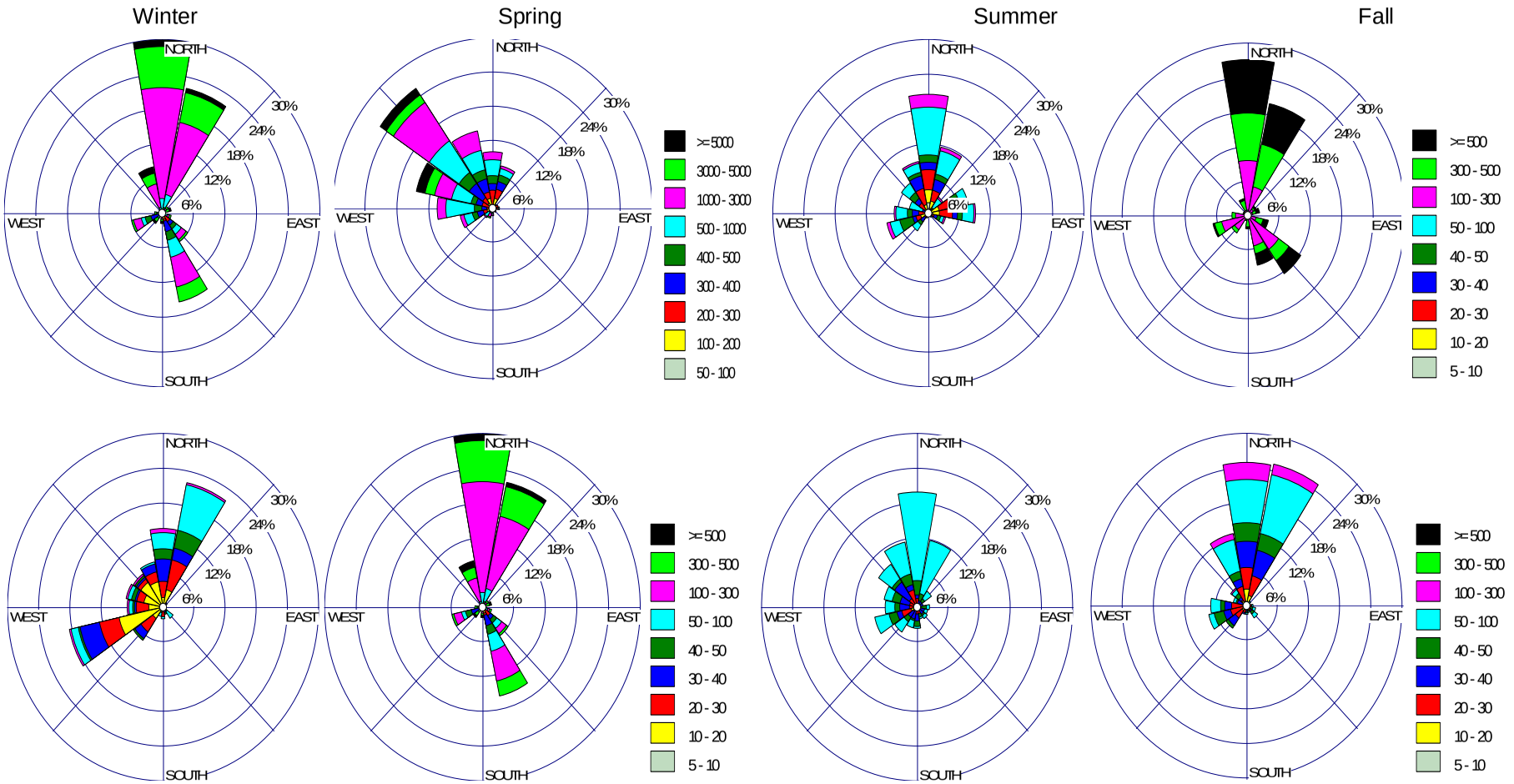

Figure 8. PM_{10} rose during the Worst20% (upper panel) and Best20% (bottom panel) conditions for different seasons at UB05 monitoring site.

4. Conclusion

In general PM_{10} and SO_2 concentrations were the highest at the urban site, especially at the UB05, followed by the industrial site, UB07, during the measurement period. The lowest PM_{10} and SO_2 concentrations were measured at the background site, UB08, for all seasons. Clear seasonal variation, winter high and summer low, was measured for PM_{10} and SO_2. Strong diurnal variation, morning and evening high afternoon low, at the urban and industrial sites during winter months due to coal burning for space heating. Clear diurnal variation, evening and afternoon high, morning low was observed at the road site during winter months mainly from not only traffic but also fugitive dust. Pollution level in Ulaanbaatar during wintertime was highly dependent on temperature inversion, wind speed and direction. Pollution was accumulated due to very thick and strong temperature inversion and low wind mostly from N, NE. Pollution level during spring time was depending on synoptic condition. Low pressure system led unstable weather condition which brought high PM_{10}. The mean PM_{10} mass concentration during the Worst20% condition was approximately 5 to 10 times higher than that in the Best20% condition. Coarse particles were dominant during most of the time except winter season, suggesting that air pollution in Ulaanbaatar was mainly from fugitive dust. Only during the winter season, contribution of $PM_{2.5}$ to PM_{10} was measured to be 0.72 indicating that fine particulate matter was dominant.

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Investigation of long-term changes of air pollutants concentrations in North-East Asia including Russia using EANET monitoring data

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Abstract

Permanent air quality monitoring networks provide valuable continuous measurement data. An interesting and important task is to analyze entire observational records revealing presence of monotonic long-term changes in air pollutant concentrations in the atmosphere. In this study we use EANET monitoring data on several major air pollutants for qualitative and quantitate assessment of trends at rural and remote stations in the North-East Asia region covered by EANET. Trends on the entire observational records as well as on selected seasonal subsets were estimated. For about a half of records considered we infer presence of statistically significant trends, whose spatial distribution we analyze further. Together with this trend assessment the comparison of mean air pollution levels at the North-East Asian (EANET) and European stations (from the European Monitoring and Evaluation Programme (EMEP)) is given. Statistical comparison of mean concentrations from the stations with similar environmental conditions was performed as well as comparison of seasonal trends.

Key words: EANET, Regional air pollution, Trend analysis

1. Introduction

One of the major aims of permanent monitoring networks is detection of long term changes and their similarity or difference over whole territory of interest. In addition, it is interesting to find interrelation between the changes in regions.

Long term changes of pollutants burden may be caused by different reasons. One of them is emission changes in the area affecting the station. This extent of this area can be large taking into account long range presence of one or many of them. Further investigations (e.g. modelling of long-range pollution transport) should be used to quantify these.

Trend assessment becomes more and more important taking into account changes in emissions and transport of pollutants in atmosphere. For example, such work was done by European Monitoring and Evaluation Programme (EMEP) (http://www.nilu.no/projects/ccc/).

To obtain a reliable trend assessment, long-term high quality monitoring data is needed. The minimum length of observational record required is determined by the specific questions and goals of the investigation. By now EANET provides fourteen years of continuous monitoring data that, as we show below, may be successfully used to obtain consistent trend estimates.

Annual and monthly means of pollutant concentrations observed during EANET monitoring period are available via in annual EANET Data Reports (EANET. 2015). This information lets one to grasp the overall variance in annual/monthly averages, however it is not well-suited for a consistent trend assessment. There are several reasons for that: (1) Statistically significant presence of monotonic increase or decrease of pollutant concentrations in the entire observational record may be very sensitive to statistical outliers, which detection is determined by the total sample size and data distribution. The latter substantially differ for the annual/monthly means and the source "raw" data. (2) Most of the pollutants have characteristic atmospheric lifetimes shorter than a few months, therefore their seasonal abundances are determined by different superposition of the relevant processes involved. Consequently, trends estimated for the seasonally selected data exhibit changes caused by these (dominating) processes, often in contrast to the entire observational record. In this study, we address the above mentioned problems by scrutinizing raw measurement data for the entire observational periods, establishing proper outlier detection criteria and dissecting the data according to each geographical region, season and even each month.

Another task set by Network Center was comparison of pollution levels at EANET stations and European stations of EMEP network.

2. Methods

In this paper non-urban sites in North-East part of EANET region. Due to lack of measurement data at rural and remote sites in China we used monitoring data from stations located in Japan, Korea, Mongolia and Russia only. Figure 1 illustrates stations whose monitoring data we considered.

We use filterpack measurement data of air concentrations of gaseous and aerosol species (SO_2, NH_3, HNO_3, HCl, SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, Na^+, K^+, Mg^{2+}, Ca^{2+}). A typical EANET biweekly sampling frequency yields approximately six points for each season and about two-dozen of points per year, respectively. Period of consideration was from 2000 to 2013, i.e. from the beginning of regular operation of the network.

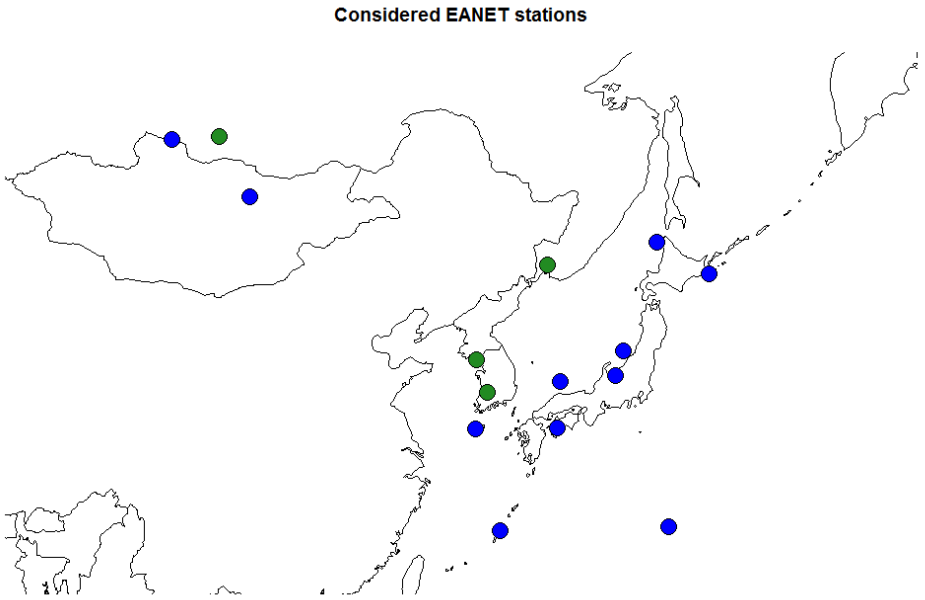


Figure 1. Rural/remote stations in North-East Asia in this work: Japan (Rishiri, Ochiichi, Tappi, Sado-Seki, Oki, Happo, Ijira, Yusuhara, Hedo, Ogasawara), Korea (Kanghwa, Imsil, Cheju), Mongolia (Terelj), Russia (Mondy, Listvyanka, Primorskaya). Rural stations a presented in green color, remote stations are presented in blue.

Time series were analyzed for presence of statistical outliers whose appearance is caused by measurement/data processing errors, exceptional local pollution events and meteorological conditions, or unidentified reasons. Filtered time series were analyzed for statistically significant presence of trends. We considered entire observational records and also divided them into four seasons and analyzed those seasons separately. To assess trends we used median regression (MR), which is essentially the linear trend separating the halves of the given distribution (Koenker and Bassett, 1978). Trend was considered as statistically significant if according confidence intervals didn't include zero and not significant otherwise trends.

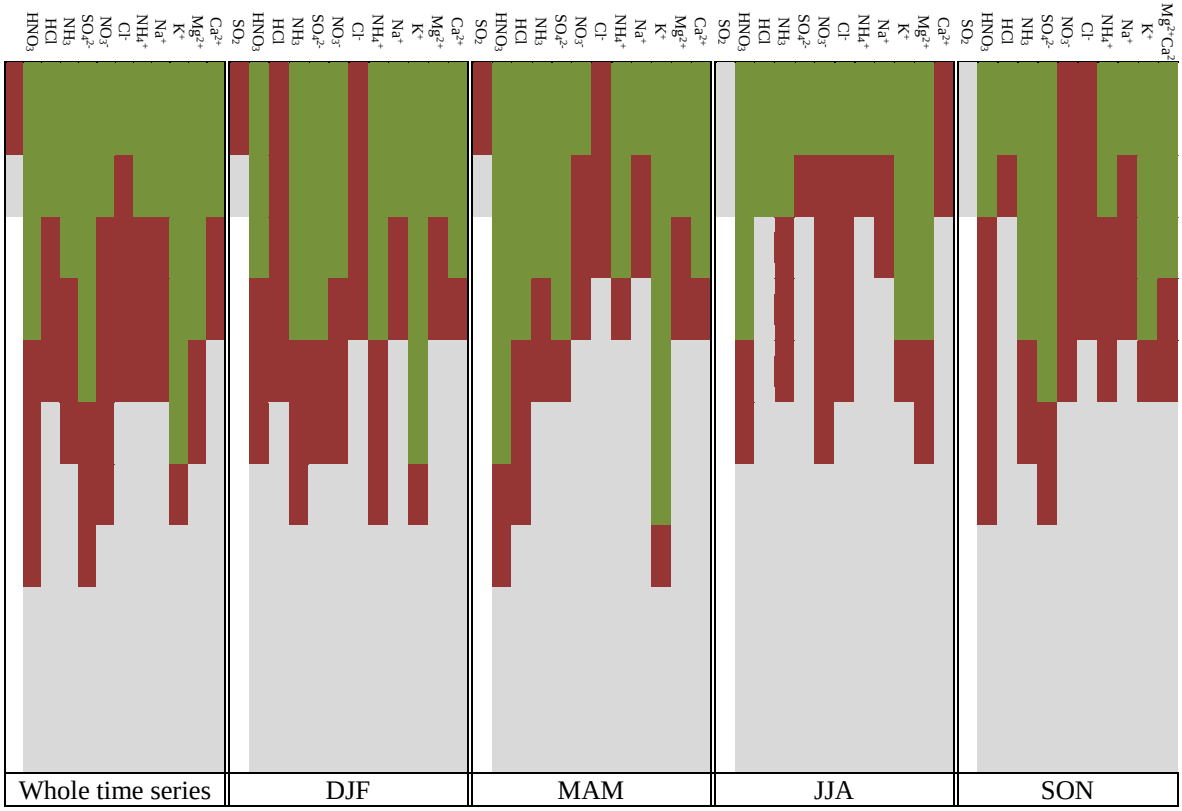
To compare monitoring data in EANET and EMEP networks the several EMEP stations were chosen to be similar rural and background sites in accordance with EANET classification (Technical Manual for Air Concentration Monitoring in East Asia). The selected stations for comparison are: Nord (Greenland), Bredkalen (Sweden), Oulanka (Finland), Svratouch (Czech Republic), Leba and Sniezka (Poland), Zingst (Germany). We used filterpack measurement data of these stations downloaded from Internet assessed data base of World Data Centre of Aerosols - EBAS (http://ebas.nilu.no/). In line with adopted sampling protocol EMEP sites provide one-day sampling filterpack data (EMEP Manual, http://www.nilu.no/projects/ccc/manual/index.html). Data used for comparison from mentioned stations cover mostly the same period of 2000 to 2012.

We used Student's two sample t-test to investigate whether mean concentrations are equal or not and after that which concentrations (EMEP or EANET) are statistically significant higher. It is necessary to note that for EANET stations we have two-week mean concentration for one sample while for EMEP it is one day mean concentration for one sample. Moreover the periods of data at EANET and EMEP stations for comparison are not fully equal but the first of them (EANET data) usually contents the second one. The comparison is the most interesting for the stations with similar site characteristics, i.e. rural Asian and rural European stations, remote Asian and remote European stations.

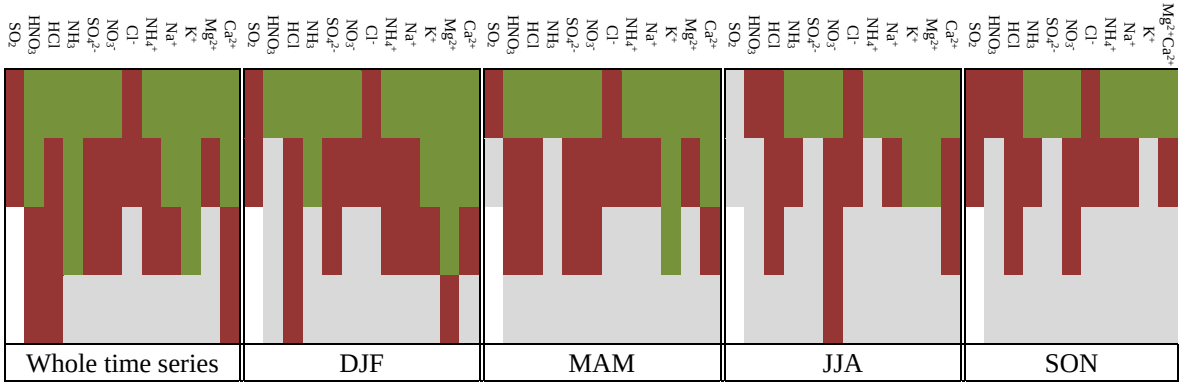
3. Results and discussion

3.1 Trend Assessment

There are statistically significant upward or downward trends found for about a half of considered stations. Numbers of both types of trend behavior are approximately equally. The sites with decreasing/increasing/absence trends for individual compounds as parts of total number of stations are presented at Figure 2 (a, b) for entire observational data and seasonal subsets with green/red/grey colors correspondently.



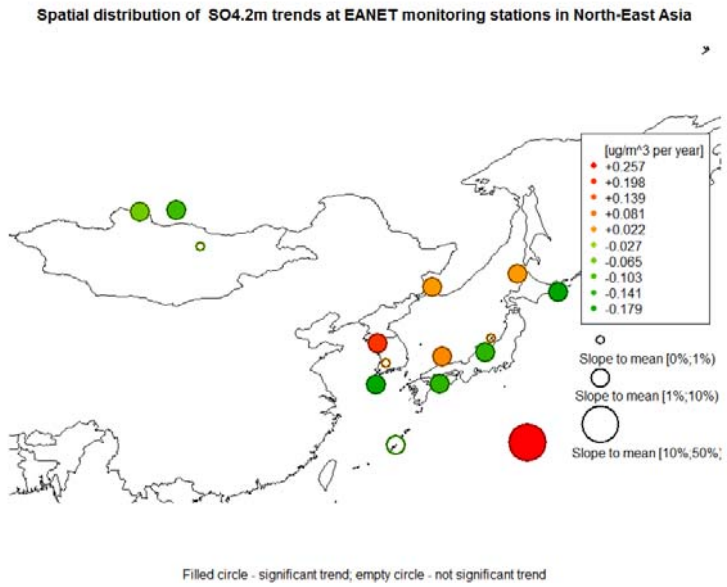
(a)



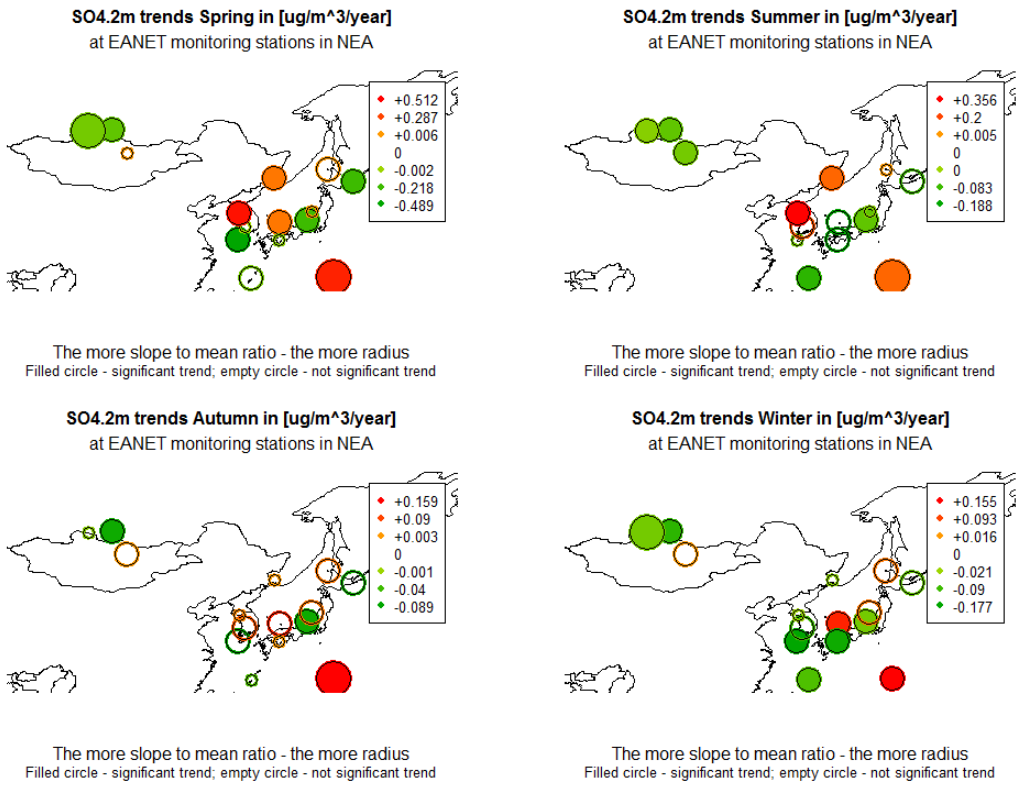
(b)

Figure 2. Parts of remote (a) and rural (b) stations with calculated decreasing (green), increasing (red) or insignificant (grey) trend for measured compounds to total number of stations.

Figure 3 (a, b) presents an example of illustrated results of trend assessment for sulfate been put on the map. Similar pictures were prepared and analyzed for all selected species. The description of results is given below.



(a)



(b)

Figure 3. Spatial distribution of entire observational record (a) and seasonal (b) trends of sulfate. Empty circles denote not significant trends. Color and its intensity represent direction and absolute value of the trend for the station: red tones indicate increasing trends, green indicate decreasing trends.

Aerosol sulfate trends illustrated at Figure 3 (a, b) indicate the increase of concentrations at central part of the region (Primorskaya, Kanghwa, Oki, Rishiri) while decreasing trends were evaluated for western part of the region as well as for eastern except Ogasawara. The same is for seasonal trends.

Filterpack measurements for sulfur dioxide were available only for four stations (three Russian and Mongolian). For all the stations there are increasing trends but seasonal trends are different. For example there are no statistically significant trends for autumn season. Highest increasing trend shows Mondy during winter season (increase about 0.5 ppb per year).

Whole situation for nitric acid is not so clear. For entire observational record trends one can see significant increase only for Primorskaya station. Other trends are decreasing or increasing but values of increase per year are less than one percent compared to mean value for whole time series at this station. Same nonhomogenity in trend behavior is true for seasonal trends. Only for summer we can see strongly increasing trend for Imsil.

Entire observational records of HCl show increasing trends for western part of region of interest. Same trends are true for seasonal trends. Also there is increasing trend at Japanese station Ogasawara for spring.

Entire observational records of ammonia show decreasing trends for central part of the region, whereas Cheju and Hedo show increasing trends. Spatial distribution for autumn and spring and summer trends is similar, however for winter the only increasing trends exist for western part of the region (Listvyanka, Mondy, Terelj).

At majority of stations we see increasing trends of NO_3^-, especially in central part of the region. Exceptions are Mondy, Listvyanka, Ochiishi that show decreasing trends. Seasonal trends are similar.

For chloride we can see increasing trends along the line from north-west to south-east (Terelj, three Korean stations, Oki, Ogasawara). Other stations show no significant trend or slightly decreasing. Seasonal trends are similar to entire observational record.

Situation for ammonium is similar to chloride in central part and Ogasawara there are increasing trends, but there are more stations showing decreasing trends. Seasonal trends are similar to entire observational record.

For Na^+ central part of the region and Ogasawara show increasing trends and northern part of stations show decreasing trends. Seasonal trends are similar to entire observational record trends.

K^+ entire observational record majority of stations show decreasing trends and only Ogasawara shows strongly increasing trend. Seasonal behavior is very similar except increasing trend at Primorskaya in spring and increasing trend at Oki in winter.

Group of three stations (Primorskaya, Oki, Ogasawara) show increasing trends for entire observational period as well as for seasonal trends for Mg^{2+}. Other stations show statistically significant decreasing trends or not significant trends.

Whole situation for Ca^{2+} is same as for Mg^{2+} except Kanghwa that also shows increasing trend.

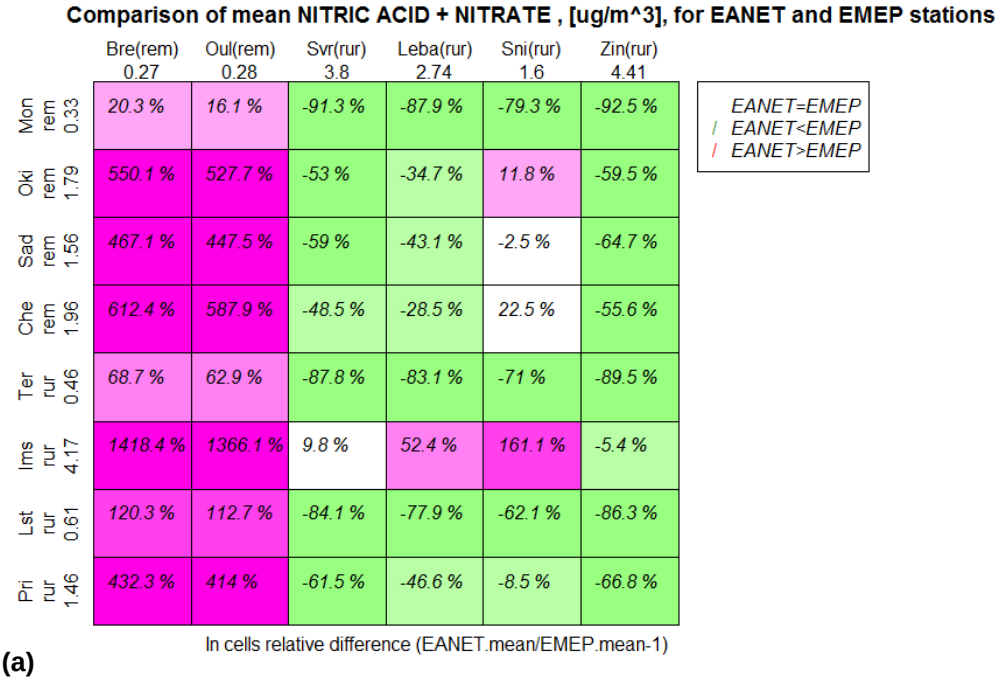
For majority of pairs station-species seasonal trends and entire observational record trends are similar. However there is a number of cases where there is no any statistically significant trend for entire observational record while there are two significant opposite trends for different seasons. An example of such situation is ammonia trends at remote Russian continental station Mondy. At Mondy there are statistically significant decreasing of ammonia concentrations in spring and summer significant increasing in winter and no significant trend for entire record.

Necessary to point out that Ogasawara shows strongly increasing trends for many species though it is very remote area. The reasons of such behavior should be considered.

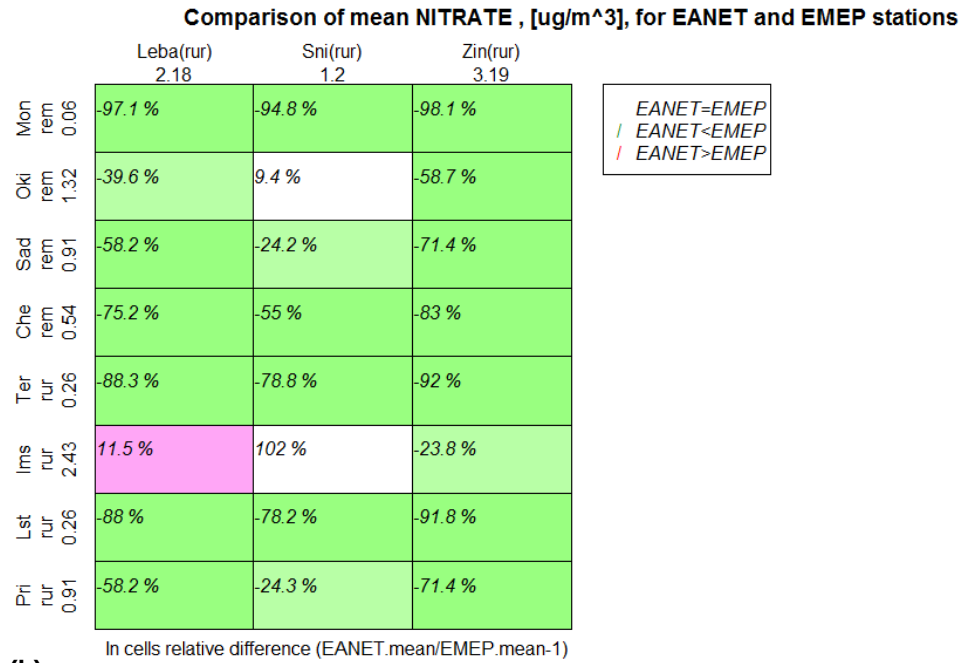
All results including spatial distributions of trends and tables with trend slopes values are available are available under request to correspondent author.

3.2 Comparison of the EANET and EMEP monitoring data (aerosols and gaseous species)

For EANET and EMEP stations results of comparison of mean concentrations are shown in Figure 4 (a, b) and Figure 5 (a, b). Green cells denote that mean concentration at EANET station is statistically significant lower than at EMEP station; red color – higher; white – no significant difference. Brief description of the results of comparison is given below.



(a)



(b)

Figure 4. Results of comparison of mean concentrations of Nitric acid+nitrate (a) and nitrate (b) at EANET and EMEP stations. Green cells denote the mean concentration at EANET station is statistically significant lower than at EMEP station; red – higher; white – no significant difference.

Remote EMEP stations have lower concentrations of the sum of nitric acid and nitrate comparing to EANET remote and rural stations. However, their concentrations at almost all rural EMEP stations considered here are higher than at rural EANET stations. The concentrations of nitrate are available only at three rural EMEP stations. Concentrations of nitrate at EANET stations are lower comparing to EMEP.

At the remote EANET stations average ammonium concentrations have higher values compared to remote EMEP stations. And its concentrations at remote Cheju station were even higher than those at rural EMEP stations. Rural EANET stations have lower concentrations except for station Imsil.

Comparison of mean SULFATE , [ug/m^3], for EANET and EMEP stations

	Gre(rem) 0.43	Bre(rem) 0.6	Oul(rem) 1.1	Svr(rur) 2.63	Leba(rur) 3.84	Sni(rur) 2.31	Zin(rur) 2.78
Mon rem 0.47	11.2 %	-20.4 %	-57.1 %	-81.9 %	-87.7 %	-79.5 %	-82.9 %
Oki rem 4.85	1036.9 %	714 %	339 %	84.6 %	26 %	109.6 %	74.3 %
Sad rem 3.55	732.5 %	496 %	221.5 %	35.1 %	-7.7 %	53.5 %	27.7 %
Che rem 5.67	1231.3 %	853.2 %	414.1 %	116.1 %	47.6 %	145.5 %	104.2 %
Ter rur 1.05	146.2 %	76.2 %	-4.9 %	-60 %	-72.7 %	-54.6 %	-62.3 %
Ims rur 6.41	1404.9 %	977.5 %	481.1 %	144.3 %	66.8 %	177.5 %	130.8 %
Lst rur 1.4	228.8 %	135.4 %	26.9 %	-46.6 %	-63.6 %	-39.4 %	-49.6 %
Pri rur 3.67	760.5 %	516.1 %	232.3 %	39.7 %	-4.6 %	58.7 %	32 %

EANET=EMEP / EANET<EMEP / EANET>EMEP

In cells relative difference (EANET.mean/EMEP.mean-1)

(a)

Comparison of mean SULFUR DIOXIDE , [ug/m^3], for EANET and EMEP stations

	Gre(rem) 0.14	Bre(rem) 0.12	Oul(rem) 0.79	Svr(rur) 2.79	Leba(rur) 2.51	Sni(rur) 2.1	Zin(rur) 1.39
Mon rem 1.04	623.8 %	798.5 %	32.5 %	-62.6 %	-58.4 %	-50.3 %	-25 %
Oki rem 1.73	1096.6 %	1385.5 %	119 %	-38.2 %	-31.2 %	-17.8 %	24 %
Sad rem 0.95	556.2 %	714.5 %	20.1 %	-66.1 %	-62.3 %	-54.9 %	-32 %
Ter rur 1.32	818.3 %	1039.9 %	68.1 %	-52.6 %	-47.2 %	-36.9 %	-4.9 %
Lst rur 7.56	5143.6 %	6409.4 %	859.7 %	170.7 %	201.4 %	260.1 %	443.2 %
Pri rur 2.46	1605.5 %	2017.2 %	212.1 %	-11.9 %	-2 %	17.1 %	76.7 %

EANET=EMEP / EANET<EMEP / EANET>EMEP

In cells relative difference (EANET.mean/EMEP.mean-1)

(b)

Figure 5. Results of comparison of mean concentrations of sulfate (a) and sulfur dioxide (b) at EANET and EMEP stations. Green cells denote mean concentration at EANET station is statistically significant lower than at EMEP station; red – higher; white – no significant difference.

The concentrations of sulfate were higher at almost all EANET stations compared to EMEP stations, except north-western continental EANET sites Mondy, Terelj and Listvyanka.

For sulfur dioxide rural continental station Listvyanka have higher concentrations than remote and rural EMEP stations as well as its concentrations at remote EANET stations are higher than at correspondent remote EMEP stations.

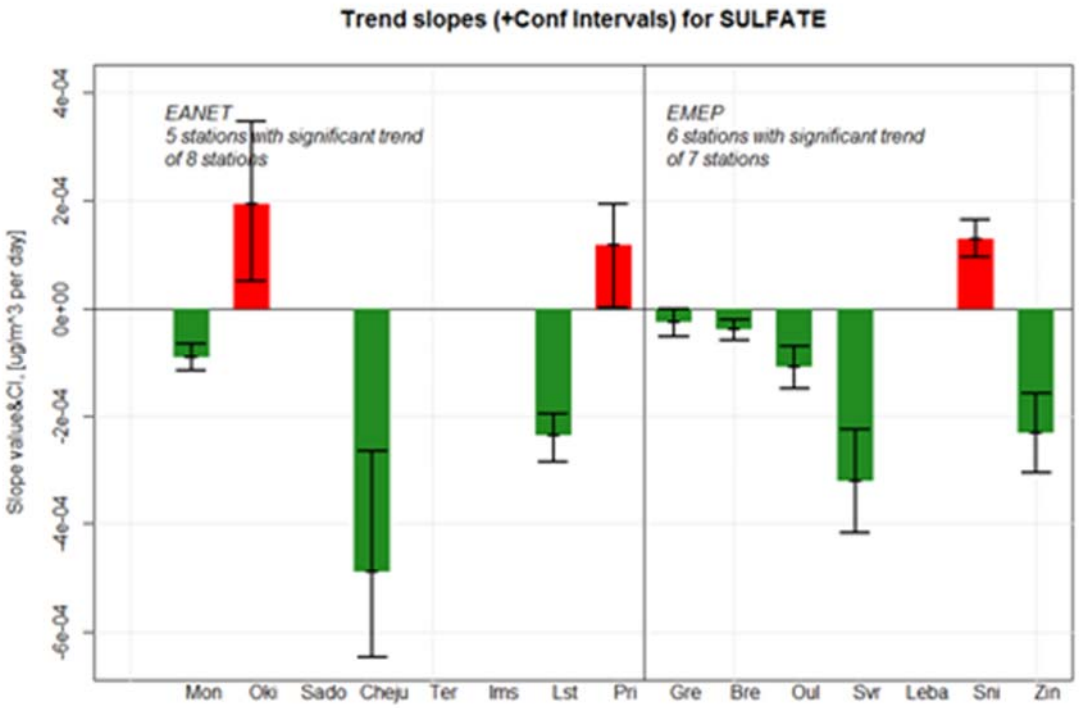
To summarize the evaluation results above we can make four groups of stations according to the site classification (remote or rural) and distance far from the sea. After that we compared the result outlines of correspondent stations from both EANET and EMEP monitoring networks. Groups of stations and results of comparison are presented in Table 1.

Table 1. Description of comparison results between EANET and EMEP stations of each of four (remote-seaside, remote-continental, rural-seaside, rural-continental) groups.

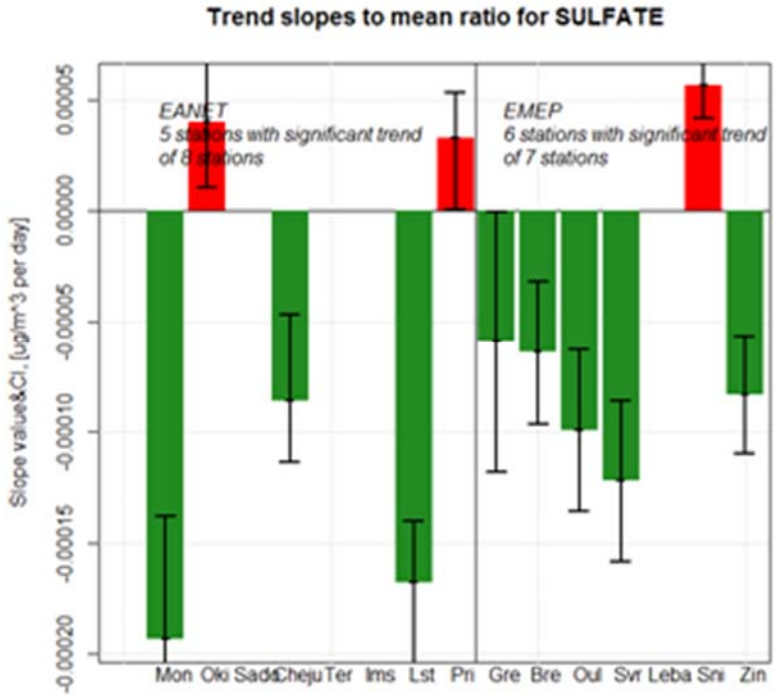
	Seaside	Continental
Remote	EANET stations: Oki, Sado, Cheju; EMEP stations: Bredkalen, Greenland. Concentrations at EANET stations are higher than at EMEP stations for all the species.	EANET stations: Mondy, Terelj EMEP stations: Oulanka Concentrations at EANET stations are higher compared to EMEP stations except for sulfate (both EMEP stations are higher).
Rural	EANET stations: Imsil, Primorskaya EMEP stations: Leba, Zingst Results are not so homogeneous For nitric acid+nitrate and nitrate Imsil has higher concentrations than Leba but lower than Zingst. Concentrations at Primorskaya for almost all cases for nitric compounds are lower than at EMEP stations. However, concentrations of sulfur compounds at Primorskaya and Imlsil are higer than at EMEP stations Leba and Zingst.	EANET stations: Listvyanka EMEP stations: Svratouch, Sniezka For almost all the species concentrations at EANET station Listvyanka are lower compared to EMEP except for sulfur dioxide.

To make the comparison of trends between stations is not so simple and easy due to presence of uncertainty in trend assessing itself. To assess this trend uncertainty we used confidence intervals (CI) in our study. Therefore to compare two trends the relevant CIs have to be taken into account. The conclusion on difference of the trends could be done only if correspondent CIs are not intercepted. But if CIs do intercept we cannot say anything about trend equality. Consequently, to compare long term changes at stations of the monitoring networks we compared the existence and order of statistically significant trends. Methods to assess trends at EMEP stations were the same used for trends assessment performed for EANET and described above. Based on available data sets we compared estimated trends at EMEP and EANET stations. The example of the results obtained is presented in Figure 6.

Trends at European stations is also not homogeneous similar to EANET stations. There are statistically significant increasing and decreasing trends for European stations. The orders of trend magnitude are the same for compared Asian and European stations. Moreover, when we compare the relative steepness of the trend slopes (i.e. slopes divided by the mean values) these values at Asian and European stations became even closer. There is no principal difference between trends estimated for entire observational data sets and seasonal subsets.



(a)



(b)

Figure 6. Comparison of slopes of trends at EANET and EMEP stations with correspondence confidence intervals for entire observational records (a); comparison of slope to mean ratios at EANET and EMEP stations with correspondence confidence intervals for entire observational records (b).

4. Conclusion

Multi-year general and seasonal longterm changes of main acidifying components were investigated in our study based on regular measurements of their atmospheric gaseous and aerosol concentrations at EANET stations over North-Eastern Asia. There are more than a half of all stations over the whole territory of interest which trends are found to be statistically significant. However, the ratio of increasing and decreasing trends is approximately equal. Trend analysis of separated seasonal datasets provided more detailed description of long term change processes at areas of stations. For some places the completely opposite direction trends had been recognized for different seasons while trend for common data of entire period was estimated to be not significant. Spatial peculiarities of trends are certainly different for the selected species but we recognized to be possible to distinguish two directions within region along each of those the trends at stations are mostly similar: from North-West to South-East, and from South-West to North-East.

Levels of pollutant concentrations at EANET stations in Asia are not homogeneous as well as ones at European stations. A comparison of pollution levels might allow us to conclude that mean air concentrations of main compounds at EANET stations are higher than at EMEP stations for majority of cases. However, a number of opposite cases was also found.

The statistically significant increasing and decreasing trends were found in monitoring data on airborne acidifying compounds at both North-East Asian and European stations. Absolute values of the trend linear slope vary among the stations and depend on pollution species at each region. Taking into account the confidence intervals for trend estimation the valid conclusion cannot be done whether absolute values of either increasing or decreasing trends are higher at EANET stations comparing to EMEP region or not. However, the orders of magnitude of trends are similar for whole sets of observational records as well as for seasonal subsets.

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Chemical Characterization of $PM_{2.5}$ particles in Ulaanbaatar, Mongolia

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Abstract

Ulaanbaatar (UB), the capital of Mongolia, has been suffered from severe air pollution because of large consumption of coal as power plant and household fuel. In addition to anthropogenic emission, topography and meteorological conditions of the city have brought worse air pollution particularly during the cold season. There have been a number of studies on air pollution sources and the adverse effect on mortality. However, very limited measurements on chemical characteristics of particulate matter have been carried out till now. For last several years, both the national and UB city governments routinely measure ambient concentrations of particulate matter and various gaseous pollutants at monitoring sites across the city. This study aims to conduct intensive monitoring of $PM_{2.5}$ at the selected sites in UB and chemical characteristics of $PM_{2.5}$ is clarified. This information is necessary to identify $PM_{2.5}$ emission sources as well as routine ambient monitoring in UB.

$PM_{2.5}$ particles were sampled with filters in the middle of Oct 2015 for 2 weeks at 2 sites in UB. Ionic components, metallic and metalloid components and carbonaceous components of $PM_{2.5}$ were analyzed. Arithmetic means of $PM_{2.5}$ at the UB03 and NAMEM sites were 152 and 68 $\mu g/m^3$, respectively. Carbonaceous components were most dominant contribution of $PM_{2.5}$ and accounted for approximately 40%. Contributions of metallic and metalloid components and ionic components were approximately 10% for both. Se, Cd, Sb at the both UB03 and NAMEM sites and Zn, As, Hg at the UB03 sites were highly enriched. These elements would be highly contaminated by local emission sources such as coal combustion, tire wear and brake wear dusts.

Key words: $PM_{2.5}$, Carbonaceous components, Coal combustion, Local meteorology, Enrichment factor

1. Introduction

Ulaanbaatar (UB) is the capital of Mongolia, with over 1.0 million people, approximately 35% of the total population in Mongolia. The city is located in a valley of the Tuul River, and surrounded by the Khentei Mountain area, with elevation ranging from 1,652 m to 1,949 m above sea level. Its location causes UB to experience frequent temperature inversions, particularly during the cold season. It was reported that the thickness and intensity of the inversion layer of air temperature over Ulaanbaatar during

the winter season were 629 m to 809 m, and 4.7 °C to 6.9 °C, respectively (Baasankhuu and Gomboluudev, 1996). The highest PM concentrations are often reported during stable meteorological conditions, such as inversion with low wind speeds (Pohjola *et al.*, 2004). A stationary anticyclone, named a Mongolian/Siberian or Asian anticyclone, forms over north-western Mongolia and the territory of Tuva, Russia, in the cold season, from October to April of the following year (Chung and Dulam, 2004). An Asian stationary anticyclone, placed over Mongolia, plays an important role, not only for the climate of Mongolia, but also for the level of air pollution. Meteorological condition is a noticeable determining factor for particular matter concentration level in UB (Batmunkh *et al.*, 2012). The winter season climate of UB is characterized by stable cold weather with low wind speed, which makes it the coldest capital in the world, with a winter mean air temperature of -23.5 °C. Coal is the main fossil fuel, and is a highly-utilized source of energy in UB. There are three large power plants in UB, which consume about 5 million tons of coal per year. About half of the total inhabitants of UB live in Gers, the traditional Mongolian dwellings, which use wood and coal for cooking and heating, without any pollution control devices. Coal consumption of Ger areas is 13 % of the total, and a large amount of lignite coal is used as household fuel in UB (ADB. 2006). There are approximately 0.35 million vehicles, approximately 61 % of the total number of vehicles in Mongolia, and most of these vehicles are second-hand. About 80 % of them do not meet emission standards, and about 54 % of the vehicle fleet is 11 years or older (ADB. 2006). Pollutant emissions, topography, and meteorological conditions of the city have brought severe air pollution problems in UB. There have been a number of studies conducted on air pollution and its sources, and on their adverse effect on mortality (Jung *et al.*, 2010, 2011) (Davy *et al.*, 2011) Allen *et al.*, 2011) (Batmunkh *et al.*, 2012). However, very limited measurements on chemical characteristics of particulate matter have been carried out. An air pollution episode can be defined as a situation during which air pollutant concentrations exceed a specified threshold value. The fine particles are the most significant factor that impacts visual air quality in many urban atmospheres.

For last several years, both the Mongolian national and the UB city governments routinely measure ambient concentrations of particulate matter and various gaseous pollutants at monitoring sites across the city. However, these measurements do not include the composition of ambient particles and this information is needed to apportion ambient particulate matter emissions to its sources. Even with the speciation data the apportionment is challenging due to meteorological variability. Therefore, objectives of this study are to determine chemical characteristics of filter-based $PM_{2.5}$ particles at 2 different sites in Ulaanbaatar during field study and provide useful dataset of $PM_{2.5}$ mass concentration and their chemical components (“speciation”) in order to investigate ambient fine particulate source apportionment study.

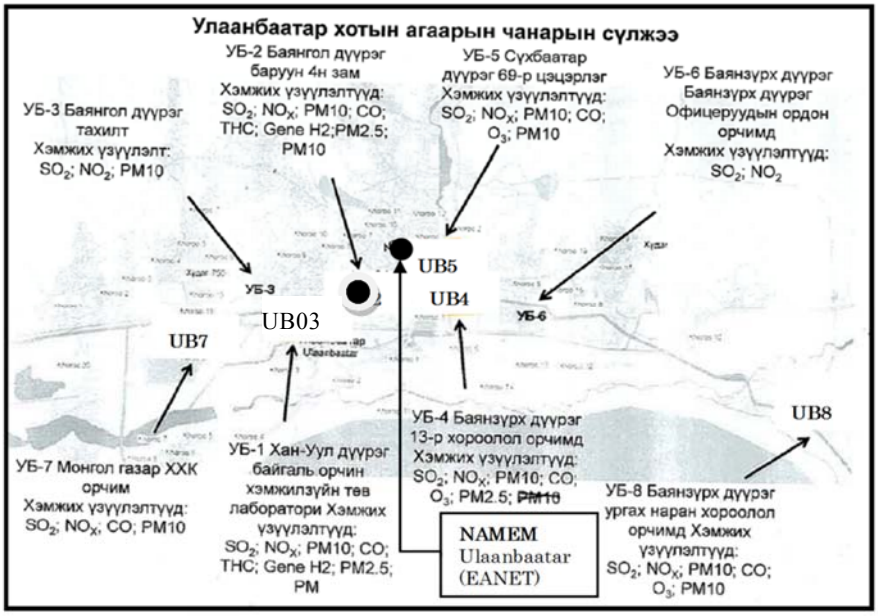

Figure 1. Map of measurement sites in Ulaanbaatar (The black dots indicate location of sampling sites).

2. Methodology

Chemical characterization of suspended particles is necessary, along with the application of source apportionment models, to attribute ambient concentrations to their sources for the development of emissions reduction strategies and to establish associations between particles and health. Particle sampling on filters is the most practical method currently available to characterize the sizes and chemical compositions of and $PM_{2.5}$. Ambient aerosol sampling systems consist of a combination of monitoring hardware, filter media, laboratory methods, and operating procedures which are specifically tailored to different monitoring objectives. No single sampling system can meet all needs, and it is often necessary to adapt existing sampling components to the specific situation being studied. Chemical analysis of filter deposits cannot be separated from the methods used to obtain the sample. Sampling for chemical analysis requires stringent attention to choice of filter media, sample handling, sample storage, and to the sampler used to obtain the filter deposits. Detailed descriptions of $PM_{2.5}$ sampling sites, system are given for following sub-chapters.

2.1 Sampling site and period

2.1.1 Sampling site

Locations of purposed sampling sites are shown in Figure 1 and brief descriptions are summarized in Table 1. Sampling sites located in downtown area and Ger area where people use much raw coal for heating. When we select the sampling sites we considered no local emission sources around the sites, the lowest distance between sampling sites and meteorological stations, and continuous and sufficient power supply in the stations.

Table 1. The description of the purposed measurement sites.

Name of sampling site	Site 1 (UB03)	Site 2 (NAMEM)
Location	UB03 national air quality monitoring station in Songinokhairkhan district	National Agency for Meteorology and Environment Monitoring (NAMEM) building in Chingeltei district
Geographical location and feature	(Latitude and Longitude) 47° 55' 07.2" N, 106° 50' 52.8" E UB03 is one of the air quality monitoring station in UB (sampler head at 2 m from ground level). The site is surrounded by Ger area where people use a raw coal for heating and land surface is degraded and contaminated by coal ash. The nearest meteorological station is located just behind the sampling site.	(Latitude and Longitude) 47° 55' 12.9" N, 106° 54' 42.0" E The site locate at roof top of NAMEM building (sampler head at around 15 m from ground level). It is down town area surrounded by many roads. By previous studies, it was learned that this site could be good representative for air quality study in city down town area. There is one of the EANET site is have been operating since 1998. The nearest meteorological station is located just behind the sampling site.
Point sources	Combined heat-and-power plants are located approximately 6 km to the Southwest of the site.	Combined heat-and-power plants are located approximately 4 km to the Southeast of the site.
Traffic source	Closest main road is located approximately 0.5 km to the south of the site.	NAMEM building is located just behind the city busiest road junction.

2.1.2 Sampling period

Ambient $PM_{2.5}$ particles were collected every day during field survey including a field blanks. Sampling duration usually was dependent on PM level i.e. 24 hours or 12 hours because of the sampling flow rate could be decreased due to aerosol loading on filter. We had been conducted the sampling for 2 weeks through middle of October 2015, and during that period, the PM level was not so high, so our proposed sampling time was noon to noon for 24 hours.

2.2 $PM_{2.5}$ sampling protocol

2.2.1 Sampling system

Particle filtration samplers consist of combinations of size-selective inlets, filter media, filter holders, and flow movers/controllers. Additionally, Denuder systems and absorbing materials that capture gases associated with volatile species such as ammonium nitrate and some organic compounds can be installed ahead the size-selective inlet and behind the particle collection filter. The dimensions, materials, and construction of these components affect the particles that are measured (Chow *et al.*, 1998).

As shown in Figure 2 and Figure 3, two different samplers are used for sampling. Patisol™ 2000 FRM sampler (Thermo Fisher scientific USA, Sampling flow rate is 16.7 L/min) used at the UB03 site and sampler has only one sampling line, therefore, quartz filter were used for $PM_{2.5}$ sampling. In case of another site, new Slit jet MCAS-SJ sampler (Murata Keisokuki Service Co.,Ltd, Japan, Sampling flow rate is 30 L/min for each line) was used for sampling. This sampler has a two sampling lines and each lines have a two stage filter holder. The "A" line that was used to collect samples for mass, ionic and elemental analysis while "B" line was used to collect samples for organic and elemental carbon analysis. Filter holder designated to collect $PM_{2.5}$ and $PM_{2.5\text{-}10}$ at same time. Coarser particles impacted on the first stage and fine particles collected on the second stage.

Figure 2. Left) Patisol™ 2000 FRM sampler; Right) 47 mm filter holder.

Figure 3. Left) Slit jet MCAS-SJ sampler; Right) 47 mm multi stage filter holders.

2.2.2 Filter media

A particle sampling filter consists of a tightly woven fibrous mat or of a plastic membrane that has been penetrated by microscopic pores. Several air sampling filter types are available. However, no single filter medium is appropriate for all desired chemical analyses, and it is often necessary to sample on multiple substrates when chemical characterization is desired. Several characteristics are important to the selection of filter media for compliance measurements 1) Particle sampling efficiency, 2) Mechanical stability, 3) Chemical stability, 4) Temperature stability, 5) Blank concentrations, and 6) Flow resistance and loading capacity, 7) Cost and availability (Chow and Watson, 1998). The filter medium for this study was ring supported membrane filter and quartz filters at the NAMEM site and

only quartz filter at the UB03 site due to samplers design. Summary of the purposed filter medium is shown in Table 2.

Table 2. Summary of the filter medium.

Filter Type	Filter Size	Physical Characteristics	Chemical Characteristics	Compatible Analysis Method
Ring supported membrane filter for fine particles at the NAMEM site (PALL PTFE)	47 mm	- White opaque surface diffuses transmitted light. - High particle collection efficiencies. - Melts at ~60EC. -High flow resistance. - 2 µm pore sizes. - Retains static charge.	- Usually low blank levels. -Inert to adsorption of gases. - Low hygroscopicity.	Gravimetry, IC, ICP/MS, ICP/OES
Quartz filter for OCEC analysis at the NAMEM and the UB3 sites (PALL 2500QAT-UP)	47 mm	- Mat of pure quartz fibers. - White opaque surface, diffuses transmitted light. - High particle collection efficiencies. - Melts at > 900°C. - Moderate flow resistance.	- Contains large and variable quantities of Al and Si. Some batches contain other metals. - Passively adsorbs organic vapors. Adsorbs little HNO_3, NO_2, and SO_2. - Low hygroscopicity	TOT, OA ICP/MS, IC

2.2.3 Flow measurement, control, and movement

Size-selective inlets require flow rates to be maintained within close tolerances to maintain the desired cut-point. There is several of the flow measurement and control devices in common use for particle sampling, for instance, Pitot tube, Bios defender, Dry Gas Meter, Critical Orifice or Device, Calibrated Rotameter, Calibrated Orifice, and Wet Test Meter. Bios Defender Dry cal used as primary standard in many laboratories and used to calibrate samplers.

2.3 Laboratory analysis

The most commonly applied aerosol analyses methods can be divided into the categories of mass, elements, ions, and carbon. The following subsections define filter preparation, handling, storage, and describe chemical analysis methods. Figure 4 illustrates a PM sampling and mass concentration analysis. After finish mass concentration analysis at Central Laboratory of Environment and Metrology (CLEM) in UB, all of samples had analyzed at Asian Center for Air Pollution Research under fellowship program.

2.3.1 Filter preparation, handling and storage

Teflon-membrane and quartz-fiber filters require pre-treatment prior to sampling, including:

- *Conditioning filters:* Gravimetric measures the net mass on a filter by weighing the filter before and after sampling with a balance in a temperature- and relative humidity-controlled environment. According to $PM_{2.5}$ reference methods to minimize particle volatilization and aerosol liquid water bias, filters must be conditioned for 24 hours at a constant relative humidity (30% – 40%) ± 5% and at a constant temperature between (20 °C - 23 °C) ± 2 °C in a temperature and humidity controlled chamber (Figure 5). These filter equilibrium conditions are intended to minimize the liquid water associated with soluble compounds and to minimize the loss of volatile species. New, clean unexposed filters placed in the chamber/conditioning

environment immediately upon arrival and stored there until the pre-sampling weighing. Filters conditioned for 24 hours to allow their weights to stabilize before being weighed. Within the filter conditioning environment, filters placed in a Petri-dish that allows air circulation over the filters while reducing the chance for airborne materials to settle on the filters. Lab blanks used to check for potential cross-contamination from airborne particulates. After sample receipt and filter inspection, the analyst stored filters/filter containers, exposed side up, in the lab refrigerator (< 4°C) until ready for filter post sampling equilibration. Prior to post sampling filter conditioning, the exposed filters were removed from the lab refrigerator and transferred to the chamber. Each filter/filter container kept for minutes to warm to room temperature before opening the filter Petri-dishes to preclude water condensation on a cold filter. After filters have warmed to room temperature, the filter Petri-dishes were opened and the filter equilibration process were began. During filter conditioning, lid was partially covered the open container.

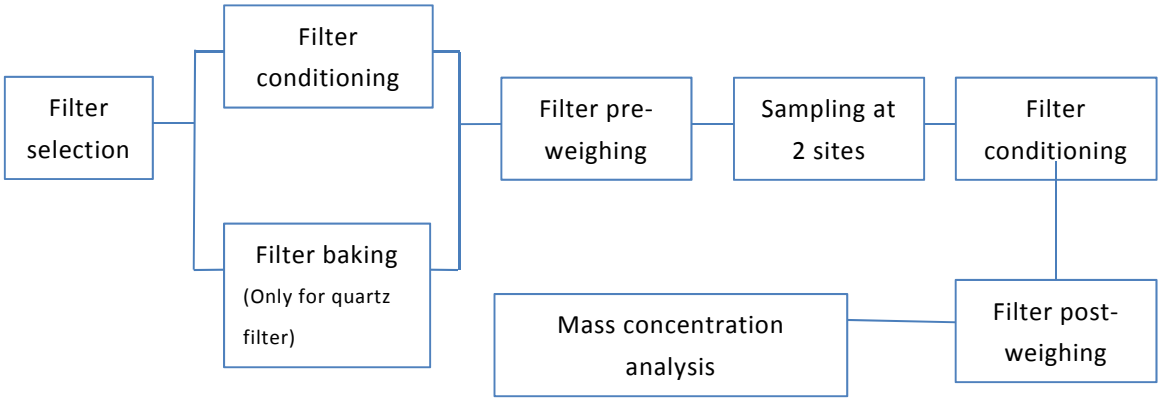

Figure 4. Flow chart for PM sampling and mass concentration analysis.

Figure 5. Orion PAP03B-KJ (temperature/RH controlled chamber).

- *Pre-baking of quartz filters:* Quartz-fiber filters adsorb organic vapors overtime. Blank quartz-fiber filters baked for at least four hours at 550 °C in CLEM. All pre-baked filters will be sealed with Teflon tape and aluminum foil and stored in a freezer prior to preparation for field sampling.
- After post-weighing, the filters kept in freezer until they were delivered to ACAP laboratory.

2.3.2 Mass measurement

Particulate mass concentration is the most commonly made measurement on aerosol samples. Gravimetric analysis is used almost exclusively to obtain mass measurements of filters in a laboratory environment. Accurate gravimetric analyses require the use of filters with low dielectric constants, high filter integrity, and inertness with respect to absorbing water vapor and other gases. Equilibration at low temperatures and relative humidity effectively removes liquid water associated with the particle deposit. Gravimetric analysis of the filters needs to be performed with a microbalance (Feeney *et. al*., 1984). Mettler Toledo Model XP6 whose readability is 1 µg used for mass measurement (Figure 6). Due to its sensitive, microbalances require isolation from vibration and air currents. Balances placed at in laminar flow hoods with filtered air minimize contamination of filters from particles and gases in laboratory air. The main interference in gravimetric analysis of filters results from electrostatic effects. Electrostatic charge eliminator (Product of HAUG GmbH & CO.KG) used to remove electrostatic charge. Balance calibration was done before and after each weighing session.

Figure 6. Orion PAP03B-KJ (temperature/RH controlled chamber).

Pre- and post-sampling filter weightings were performed on the same micro-balance. Though tolerances on re-weights of membrane filters are typically ±0.010 mg, these sensitive balances require isolation from vibration and air currents. Electrostatic effects contribute another main interference in gravimetric analysis of filters. It is established that residual charge on a filter could produce an electrostatic discharge between the filter on the pan and the metal casing of the electro balance, which induces non-gravimetric forces.

Electrostatic charge within the balance minimized by placing a static eliminator in the balance's weighing chamber. The balance operated according to the manufacturer's directions. Between each filter weighing, re-zeroed the balance. Remove the filter from the Petri dish by gently slipping the filter forceps under the outer. Each filter passed near a static eliminator for 30 - 60 seconds immediately before weighing and transferred to the microbalance's pan and closed the weighing chamber door.

2.3.3 Calculation of mass concentrations

The equation to calculate the mass of fine particulate matter collected on a filter is as below:

$$\Delta M = M_f - M_i$$

Where,

ΔM = total mass of particulate matter collected during sampling period (mg)

M_f = final mass of the conditioned filter after sample collection (mg)

M_i = initial mass of the conditioned filter before sample collection (mg)

Field records of $PM_{2.5}$ sampler are required to provide measurements of the total volume of ambient air passing through the sampler in cubic meters at the actual temperatures and pressures measured during sampling.

2.3.4 Chemical analysis

The samples collected on the membrane filter at the NAMEM sites and quartz filter at the UB3 sites used for the determination of the PM mass and water-soluble ions such as CH_3COO^-, F^-, Cl^-, NO_2^-, Br^-, SO_4^{2-}, NO_3^-, F^-, $HCOO^-$, PO_4^-, Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+} as well as following elements: Li, Be, Na, Mg, Al, K, V, Ca, Fe, Zn, Ba, Ni, Cu, Cr, Mn, Ga, As, Sr, Co, Se, Rb, Ag, Cd, In, Sb, Cs, V, Hg, Tl, Th, Mo, Pb, and U. The particles collected on the quartz filters at two sites used for the determination of the carbonaceous aerosol *i.e.*, concentration of organic carbon (OC) and elemental carbon (EC).

2.3.4.1 Elemental ~~a~~nalysis

The most common interest in elemental composition derives from concerns about health effects and the utility of these elements to trace the sources of suspended particles. Instrumental neutron activation analysis (INAA), atomic absorption spectrophotometer (AAS), inductively coupled plasma with atomic emission spectroscopy (ICP-AES) or with mass spectroscopy (ICP-MS), photon-induced X-ray fluorescence (XRF), and proton induced X-ray emission (PIXE) have all been applied to elemental measurements of aerosol samples for atomic numbers ranging from 11 (sodium) to 92 (uranium). XSeries2 ICP-MS (Thermo Fisher Scientific Inc.) was used to analyze elements. The ICP-MS can measure trace elements as low as one part per trillion (ppt) or quickly scan more than 70 elements to determine the composition of an unknown sample. ICP-MS was achieved by ionizing the sample with inductively coupled plasma and then using a mass spectrometer to separate and quantify those ions. External calibration using multi-element standard was performed at beginning and end of analysis. Sample concentrations were calculated using the slope of the calibration curve.

When other analyses are to be performed on the same filter, the filter is sectioned first using a precision positioning jig attached to a ceramic cutter to avoid metal contamination. The half of membrane filter and quarter of quartz filter were used for these analyses, so the results need to be multiplied by two and four, respectively to obtain the deposit on the entire filter. The cutting blade was cleaned between each filter cutting. To analyze the trace elements, procedure of filter extraction followed by the improved ACAP method illustrated in a simple flow chart of Figure 7 and described the procedure as follows.

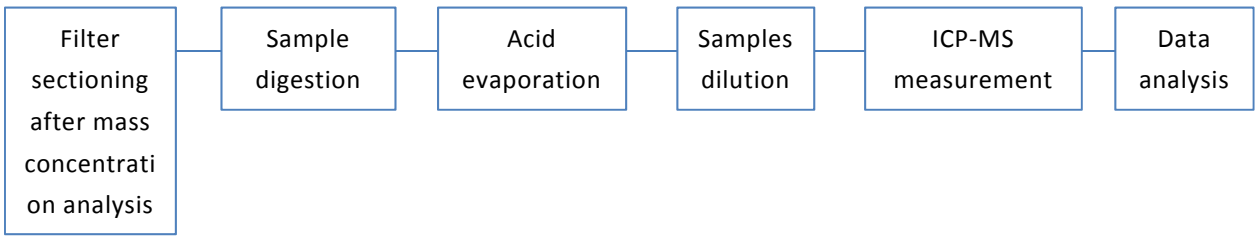

Figure 7. Simplified flow chart of elemental analysis.

1. Filter sectioning: Ceramic scissors were used to cut filters
2. Extraction: 3 ml of hydrofluoric acid (50 %), 6 ml of nitric acid (60 %) and 1 ml of hydrogen peroxide (30 %) were added to to the sectioned filter. Then, the samples were put into microwave digestion instrument (ETHOS 900, Milestone Inc.). The power of microwave was 250 Watt for 2 min., 0 Watt for 3 min., 250 Watt for 5 min., 400 Watt for 5 min., 500 Watt for 10 min. and 400 Watt for 20 min.
3. Acid evaporation: Hot plate was used for acid evaporation. The digested samples were heated at 200 °C until those were is reduced to about 0.1 ml.
4. Dilution: Diluted nitric acid (1 mol/l) was added to make 20 ml samples.

2.3.4.2 Water-soluble ion analysis

There are many different ways of determining ions qualitatively and quantitatively. One such technique that is widely used is ion chromatography. Ion chromatography is one member of the large family of chromatographic methods. Chromatography is a method for separating mixtures of substances using two phases, one of which is stationary and the other mobile moving in a particular direction. Ion chromatography includes all rapid liquid chromatography separations of ions in columns coupled online with detection and quantification in a flow-through detector. The sample is introduced onto the system *via* a sample loop on the injector. When in the inject position the sample is pumped onto the column by the eluent and the sample ions are then attracted to the charged stationary phase of the column. The charge diluent elute the retained ions which then go through the detector (which is most commonly conductivity) and are depicted as peaks on a chromatogram. Purposed instruments to ion analysis were DOINEX ICS 2100 for anions and DOINEX ICS 1100 for cations. External calibration had performed beginning of analysis. Sample concentration had calculated using the slope of the calibration curve and peaks of each ion from chromatography. Figure 8 illustrates flow chart of ionic species analysis. The brief procedure is shown as follows. Totally 44 samples including blanks have been analyzed using IC system.

- Filter sectioning: Ceramic scissors and other ceramic tools were used to cut filters. And then quarter of quartz filters and half of PTFE filters were used for ionic species analysis due to limited number of samples
- Sample extraction: Sample extraction was conducted by two steps according to the EANET manual (EANET. 2013). Each filter part was extracted with totally 40 mL Millipore water solution.
- Anions (F^-, $CH3COO^-$, $HCOO^-$, Cl^-, NO_2^-, Br^-, SO_4^{2-}, NO_3^-, $COOH^-$, PO_4^{3-}) and cations (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}) were analyzed by ion chromatography (ICS-2100 for anion and ICS-1100 for cation, Thermo Fisher Scientific Inc.).

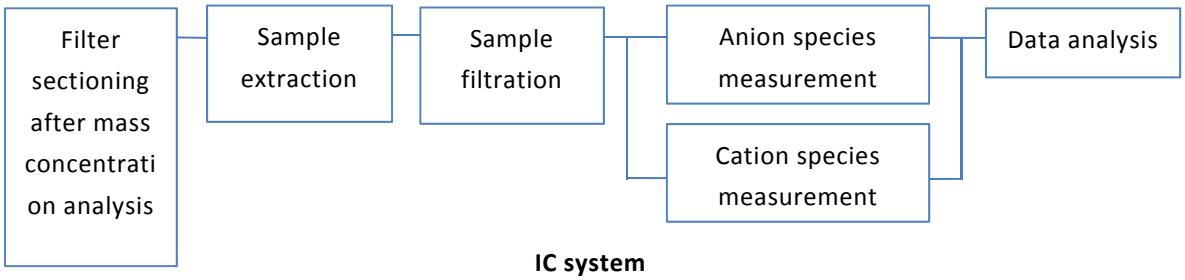

Figure 8. Simplified flow chart of ionic species analysis.

2.3.4.3 Thermal optical transmission method for carbon

Filter based particulate carbonaceous aerosol will be analyzed for elemental carbon (EC) and organic carbon (OC), with the thermal-optical transmittance (TOT) method used for pyrolysis correction (Birch and Cary, 1996). A 0.505 cm^2 filter punched from the sampled filter for OC and EC in PM analysis by the TOT method, based on NIOSH (National Institute for Occupational Safety and Health) 5040 temperature protocol (NIOSH. 1996). The analysis was conducted in a pure Helium atmosphere, and purged for 10 s, followed by two temperature steps at 600 °C and 840 °C, for 80 s and 90 s, respectively. The oven was briefly cooled for 35 s, before the gas valve was switched to a mixture of a 2 % O_2 and 98 % He atmosphere, followed by three steps of heating at 550 °C, 650 °C, 850 °C, for 30, 45 and 90 s, respectively. We identified organic carbon and elemental carbon as follows.

Organic Carbon (OC) - Optically transparent carbon removed (through thermal desorption or pyrolysis) and char deposited when heating a filter sample to a preset maximum (840 ºC) in a non-oxidizing (helium) carrier gas.

Elemental Carbon (EC) - Carbon (*e.g.*, in soot particle cores) that can only be removed from the filter under an oxidizing carrier gas (He/O_2). Optically absorbing carbon removed at high temperatures (*e.g.* 850 °C) in a non-oxidizing carrier gas when internal (sample matrix) oxidants are present.

IMPROVE thermal-optical reflectance (TOR) and TOT analyses are implemented on DRI Model 2001 thermal/optical carbon analyzers (Atmoslytic, Inc., Calabasas, CA, USA). Prior to analysis, a 0.5 cm^2 punch is removed from each quartz-fiber filter, using a calibrated punch, and placed into the quartz sample boat. This boat has a 4.56-mm diameter hole to minimize interference with the optical signal and to allow carbon from both sides to be entrained in the carrier gas. As temperatures are ramped from ambient (25 °C) to a preset plateau (140 °C to 840 °C), carbonaceous material in the sample is volatilized, and pyrolyzed to gas-phase compounds that are converted to CO_2 as they pass through a manganese oxide (MnO_2) oxidizer at 912 °C. The CO_2 is reduced to CH_4 as it passes through a granulated firebrick impregnated with a nickel catalyst at 420 °C, and the CH_4 is then quantified by a FID. A 5-mW He-Ne laser (λ = 632.8 nm, red light) is directed perpendicular to the deposit side of the sample punch. Reflectance (R) and transmittance (T) are monitored at angles of 180° and 0°, respectively. The laser signal is modulated with a chopper to separate R and T signals from stray light sources. The thermocouple extends under the sample boat so that it is as close to the filter as possible. Even so, calibration is required for the thermocouple reading to equal the filter temperature. A negative sign is assigned to pyrolyzed OC (*i.e.*, optical pyrolysis (OP)) if the laser split occurs in the He atmosphere prior to the introduction of O_2. This rarely occurs for the IMPROVE temperature protocols, but it is common for other temperature protocols due to the incorrect presumption that carbonate carbon evolves at 850°C in an inert analysis atmosphere, as real-world carbonate mixtures evolve across a wide range

of temperatures. Carbonate carbon typically accounts for < 5% of mass in $PM_{2.5}$ samples, and its analysis is only required for special studies. The procedure of EC/OC analysis is shown in Figure 9. To evaluate the potential for carbonate interference, an injection port on the Model 2001 analyzer allows 20 µL of 0.4 M hydrochloric acid (HCl) solution injected with a micro-syringe onto the filter to react with carbonates; the CO_2 evolving from this reaction can measured separately from other carbon fractions.

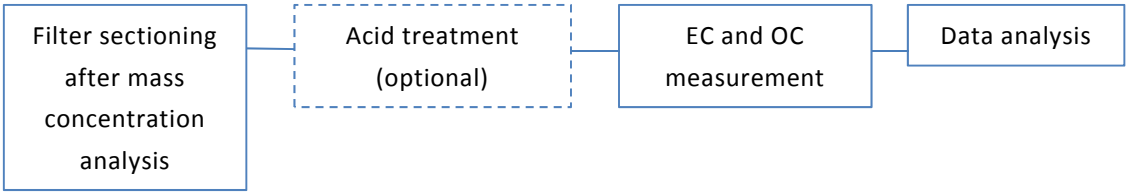

Figure 9. Simplified flow chart of EC/OC analysis.

3. Results and discussion

3.1 Time variations of $PM_{2.5}$ Mass concentration

The October is end of warm season of year in Mongolia. In this month, a house heating very depends on meteorological condition of days. During the field study period, ambient temperature was decreasing to below zero degrees Celsius at night time and some days it was windy. $PM_{2.5}$ mass concentration at the UB03 site was almost two times higher than that was at the NAMEM site for all the time of sampling period (Figure 10). $PM_{2.5}$ concentration at NAMEM site was highly correlated to automatic monitoring station measurement data of UB02 and UB04 while concentration at UB03 was much higher than other places.

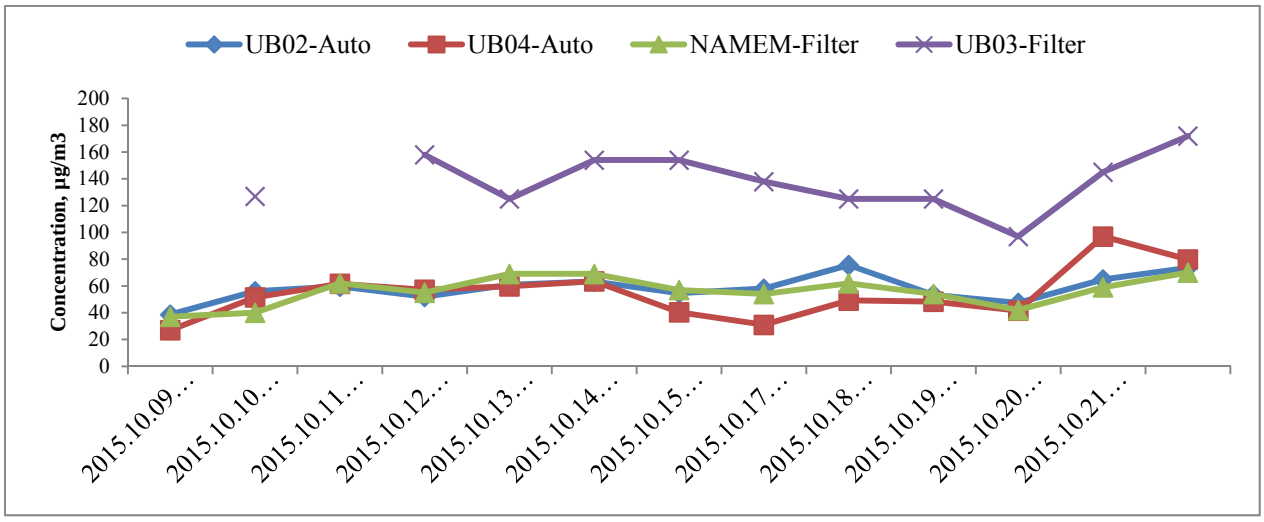

Figure 10. $PM_{2.5}$ concentration at air quality monitoring stations and selected sites.

3.2 Summarized data of metallic and metalloid components in $PM_{2.5}$

Table 3 shows the concentrations in ng/m^3 of the elements evaluated. A wide range of concentration can be observed with Ag (0.01 ng/m^3) being the element that exhibits the lowest concentration and Fe (12.8 µg/m^3) the highest. The guideline values recommended by the World Health Organization (WHO) for air pollutants of special environmental and health concern are also shown in Table 3. These pollutants include those substances with effects other than cancer, namely, Cd, Mn, Pb,

and V, and those with guideline values based on carcinogenic effects, As and Ni. Average Ni and As concentrations were above the reported concentration associated with an excess lifetime cancer risk of $1{:}10^{-6}$. Total $PM_{2.5}$ mass concentration was found to vary from 44 to 87 μg/m^3, with an average value of 68 μg/m^3 at the NAMEM site while it was vary from 105 to 189 μg/m^3, with an average value of 152 μg/m^3 at the UB03 site. They were much higher than 50 μg/m^3, the present national air quality standard for $PM_{2.5}$.

Table 3. Descriptive statistics of measured concentrations of $PM_{2.5}$ at two sites.

	UB03 site				NAMEM site				WHO guideline	
Element	Min	Geo-mean	Arithmetic mean	Max	Min	Geo-mean	Arithmetic mean	Max	Time-weighted average	Averaging time
$PM_{2.5}$	105	151	152	189	44	67	68	87	–	
^{23}Na	278	2704	3916	10659	190	550	596	963	–	
^{24}Mg	242	904	1083	3210	127	389	419	745	–	
^{27}Al	1623	4643	5328	12785	766	2356	2543	4295	–	
^{39}K	755	1564	1720	3812	301	707	771	1216	–	
^{44}Ca	698	2244	2606	6749	338	1195	1331	2381	–	
^{54}Fe	364	1770	2144	4945	391	1302	1447	2329	–	
^{56}Fe	499	2165	2565	5847	516	1390	1511	2408	–	
^{66}Zn	92.6	304.9	326.7	554.7	33.2	96.1	104.4	151.1	–	
^{137}Ba	11.4	65.4	127.4	300.2	8.5	22.6	24.3	38.7	–	
^{60}Ni	7.9	17.5	20.4	40.4	0.0	0.5	4.5	23.5	2.5	Lifetime[a]
^{65}Cu	3.0	10.1	11.6	24.4	5.2	9.8	10.0	13.6	–	
^{52}Cr	0.1	6.7	12.5	36.1	1.0	5.2	7.3	12.8	–	
^{55}Mn	11.6	47.7	56.3	124.1	12.2	36.4	41.8	67.6	150	Annual
^{69}Ga	2.6	23.7	30.2	70.4	1.2	2.4	2.6	3.8	–	
^{75}As	9.6	11.9	12.0	16.2	2.7	5.1	5.3	7.6	0.66	Lifetime[a]
^{88}Sr	4.2	19.2	22.9	52.2	3.9	10.5	11.6	19.5	–	
^{95}Mo	1.9	8.8	12.8	38.0	0.3	0.5	0.5	1.1	–	
^{206}Pb	19.2	26.9	27.8	40.8	8.0	14.0	14.7	24.3	–	
^{207}Pb	21.2	29.5	30.4	44.6	8.9	15.3	16.2	26.5	500	Annual
^{208}Pb	18.7	25.9	26.7	39.2	7.7	13.5	14.3	23.6	–	
^{7}Li	0.4	2.0	2.5	5.9	0.4	1.2	1.3	2.2	–	
^{9}Be	0.1	0.2	0.2	0.4	0.0	0.1	0.1	0.2	–	
^{45}Sc	0.3	0.7	0.8	1.8	0.0	–	0.2	0.5	–	
^{51}V	0.9	4.6	5.6	13.0	0.9	2.6	2.8	4.5	1000	24h
^{59}Co	0.3	0.9	1.1	2.4	0.2	0.6	0.7	3.0	–	
^{82}Se	0.5	1.0	1.0	1.6	0.3	0.4	0.4	0.6	–	
^{85}Rb	1.4	4.9	5.6	12.3	1.0	2.5	2.8	4.5	–	
^{107}Ag	0.0	0.1	0.1	0.3	0.1	0.1	0.1	0.3	–	
^{111}Cd	0.6	1.0	1.0	1.6	0.3	0.6	0.7	1.1	5	Annual
^{121}Sb	1.9	3.8	4.0	8.0	1.8	3.9	4.5	11.2	–	
^{133}Cs	0.1	0.4	0.4	0.8	0.0	0.1	0.1	0.2	–	
^{202}Hg	0.1	0.5	0.8	2.1	0.1	0.2	0.2	0.9	–	
^{205}Tl	0.1	0.2	0.2	0.3	0.1	0.1	0.1	0.1	–	
^{209}Bi	0.2	0.4	0.4	0.7	0.1	0.2	0.2	0.4	–	

^{232}Th	0.1	0.5	0.6	1.2	0.1	0.2	0.2	0.4	–
^{238}U	0.0	0.3	0.5	1.4	0.0	0.1	0.1	0.3	–

All element concentrations are in ng/m^3. $PM_{2.5}$ mass concentration is expressed in µg/m^3. Guidelines of the WHO for air pollutants of special environmental and health significance are informed as a reference.

[a] -Concentration of carcinogenic air pollutant associated with an excess lifetime cancer risk of 1 per 1,000,000.

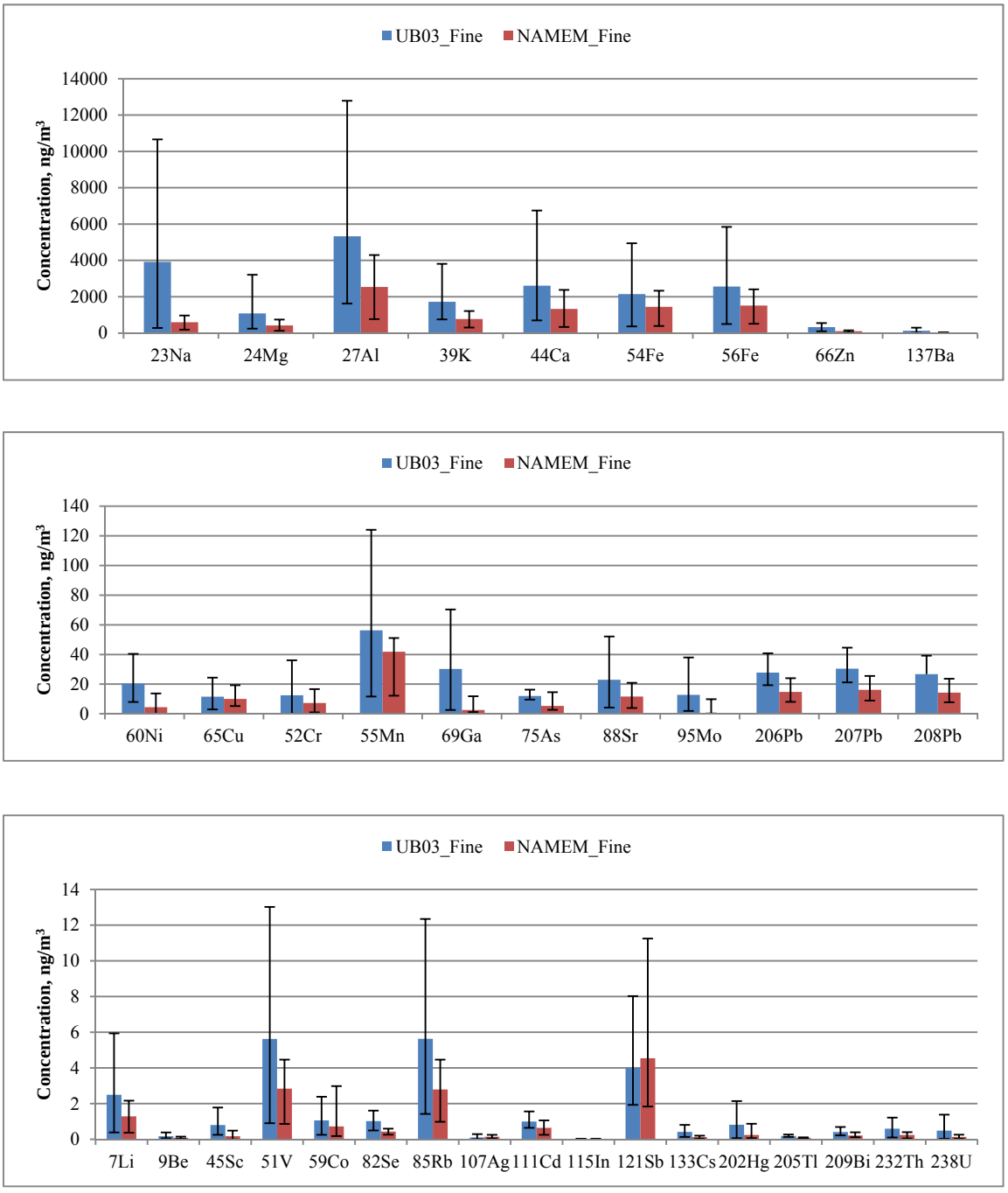

Figure 11. Ambient concentration of elements in fine particles at two sites in UB. Error bar shows the range of maximum and minimum values.

The element concentrations can be grouped in 3 ranges. The first group is composed by the geological elements, namely, Al and Fe that show the highest concentrations. Aluminum concentration ranges between 766 and 12,785 ng/m^3 and that of Fe ranges between 364 and 5,847 ng/m^3. The second group, Mn, Pb, As and Sr, exhibits relatively higher concentrations being As, the element that varies within the widest range, from 2.7 to 16.2 ng/m^3. The other trace elements in the order of V > Rb > Sb > Li > Hg conform the third group (Figure 11).

3.3 Summarized data ionic species in $PM_{2.5}$

Totally 44 samples including blanks have been analyzed using IC system. SO_4^{2-} was the most abundant ion followed by NH_4^+, Ca^{2+}, NO_3^-, Cl^-, CH_3COO^-, Na^+, K^+, Mg^{2+} and others at both two sites. Similarly to the mass concentration, most of ions concentration was higher at the UB03 site than the NAMEM site (Figure 12). Particularly, some organic ion concentration was much higher at the UB03 site.

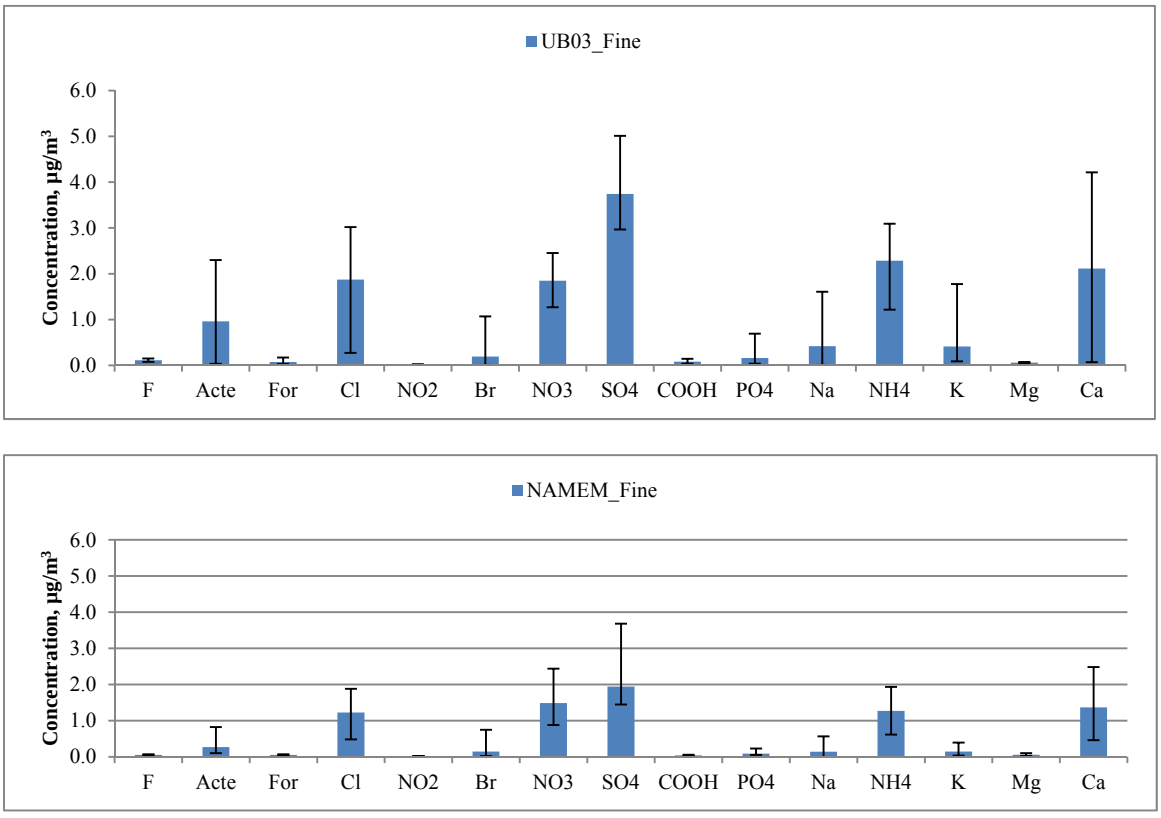

Figure 12. Ambient concentration of water soluble ions in fine particles at two sites in UB. Error bar shows the range of maximum and minimum values.

3.4 Summarized data of carbonaceous components in $PM_{2.5}$

The mean concentrations of the carbon fractions analyzed for each site are given in Table 4. The distribution of the OC and EC daily data is shown in Figure 13. According to these results, during the field sampling $PM_{2.5}$ mass mean concentrations were the highest in the UB03 site (152.4 µg/m^3) and the NAMEM site (68.4 µg/m^3). Total carbon contribution was 39.8 ± 9.4 % during field sampling.

Table 4. Carbonaceous species in $PM_{2.5}$ particles (All fraction concentrations are in $\mu g/m^3$).

Carbonaceous species	UB03 site				NAMEM site			
	Min	Geo-mean	Average	Max	Min	Geo-mean	Average	Max
$PM_{2.5}$ Concentration	105.5	150.6	152.4	189.0	44.2	67.0	68.4	87.1
VOC	6.8	13.0	14.2	26.4	1.8	4.6	4.9	8.1
OC high T	27.3	36.0	36.8	50.5	6.8	11.6	11.9	17.4
EC	9.8	13.5	13.8	19.7	3.0	6.5	6.8	10.2
EC high T	0.4	0.5	0.5	0.8	0.3	0.6	0.6	1.0
TC	46.5	63.9	65.7	96.8	12.8	23.9	24.8	37.4

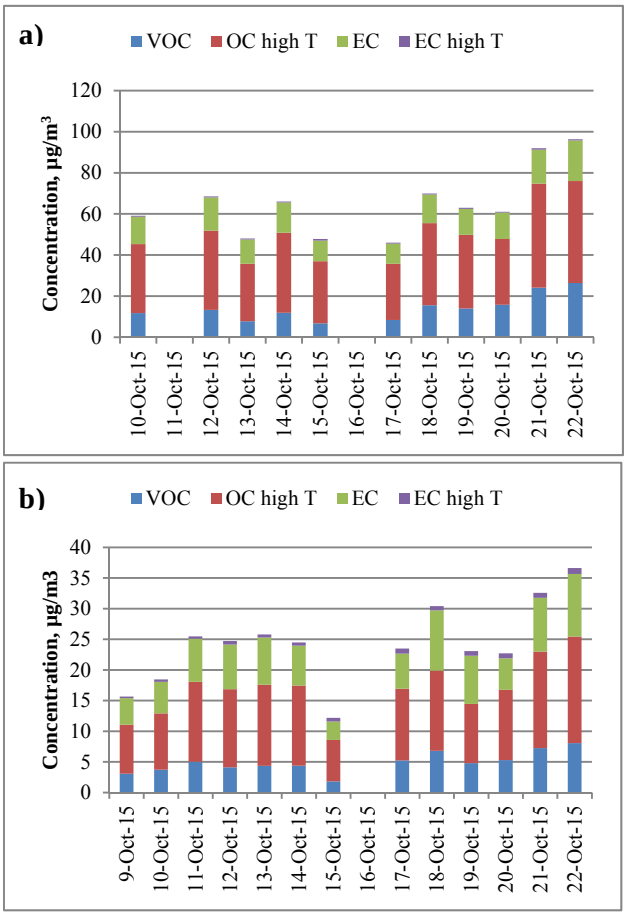

Figure 13. Carbonaceous species concentration. a) UB03 site; b) NAMEM site.

To eliminate carbonate interference 20 µl of 0.48 M hydrochloric acid (HCl) were deposited onto the filter punch just before analysis. But it was not considerable because high temperature EC contribution was too small it is around 1 % of total carbon in $PM_{2.5}$ samples.

3.5 Characteristics of $PM_{2.5}$ components in UB

The mass contributions of analyzed components to $PM_{2.5}$ were determined for each measurements in the UB03 and NAMEM sites. Table 5 shows mass contributions of respective components to $PM_{2.5}$ in UB. Carbonaceous components were most dominant contribution to $PM_{2.5}$ and accounted for approximately 40 % of $PM_{2.5}$ mass concentration. The contributions of metallic and metalloid components and ionic components were approximately 10% for both. It should be noted that the unknown contributions accounted for 39.1 ± 8.6% because it includes oxygen atom which were existed as hydrocarbons in carbonaceous components or oxides of metallic and metalloid components in $PM_{2.5}$. If constructed masses using finger print components of specific emission sources of $PM_{2.5}$ are considered (e.g. Frank, 2006), the percentage of "Unknown" must be decreased. We compared the mass contributions to $PM_{2.5}$ in UB with those in Japan as shown in Figure 14 (Ministry of the Environment in Japan. 2016). The contribution of OC in UB are remarkably higher than those at ambient and roadside stations in Japan. This implies higher emission originated from combustion process in coal power plant and domestic use in Gers.

Table 5. Mass contributions of respective components to $PM_{2.5}$ in UB.

$PM_{2.5}$ components	Contribution to $PM_{2.5}$, % (ave. ± stdev.)
Metals and metalloids	10.6 ± 4.3
Anions	7.0 ± 2.2
Cations	3.9 ± 0.8
EC	10.3 ± 2.3
OC	29.1 ± 8.4
Unknown	39.1 ± 8.6

Note: Arithmetic means of $PM_{2.5}$ at the UB03 and NAMEM sites are 152 and 68 μg/m³, respectively.

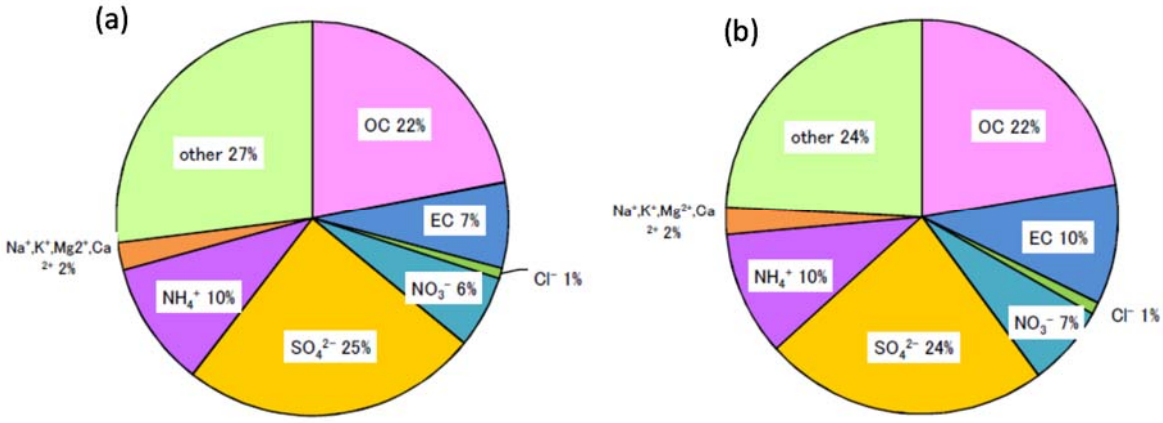

Figure 14. National averages of mass contributions of respective components to $PM_{2.5}$ in Japan.(Fiscal year of 2014) a) Average of 102 ambient stations ($PM_{2.5}$ = 14.3 μg/m³); b) Average of 34 roadside stations ($PM_{2.5}$ = 15.0 μg/m³).

Ambient concentrations of particulate matter and its chemical components depend strongly on wind dynamics in the research field. While average concentrations provide little information on the contributions of different emission sources and possible routes of transportation, concentrations measured during prevailing wind directions might be more useful. Calm conditions (wind velocity < 2 m/s) were observed around 90 % of sampling days. The highest $PM_{2.5}$ concentrations at two sites were associated with calm conditions suggesting influence of local sources rather than transport in relation to possible occurrence of temperature inversion and reduction of vertical diffusion.

The influence of wind speed on the elemental content of airborne particles varies among the sites as well as among different elements (Figure 15). High concentrations of geological elements concentration were observed at UB03 site when daily average wind speed was higher than 2 m/s. The strong wind generated soil dust at the UB03 site but at the NAMEM site there is no influence of soil generated dust in downtown area and PM mass concentration was decreased. The organic carbon exhibited higher concentrations during calm conditions suggesting emissions from local sources.

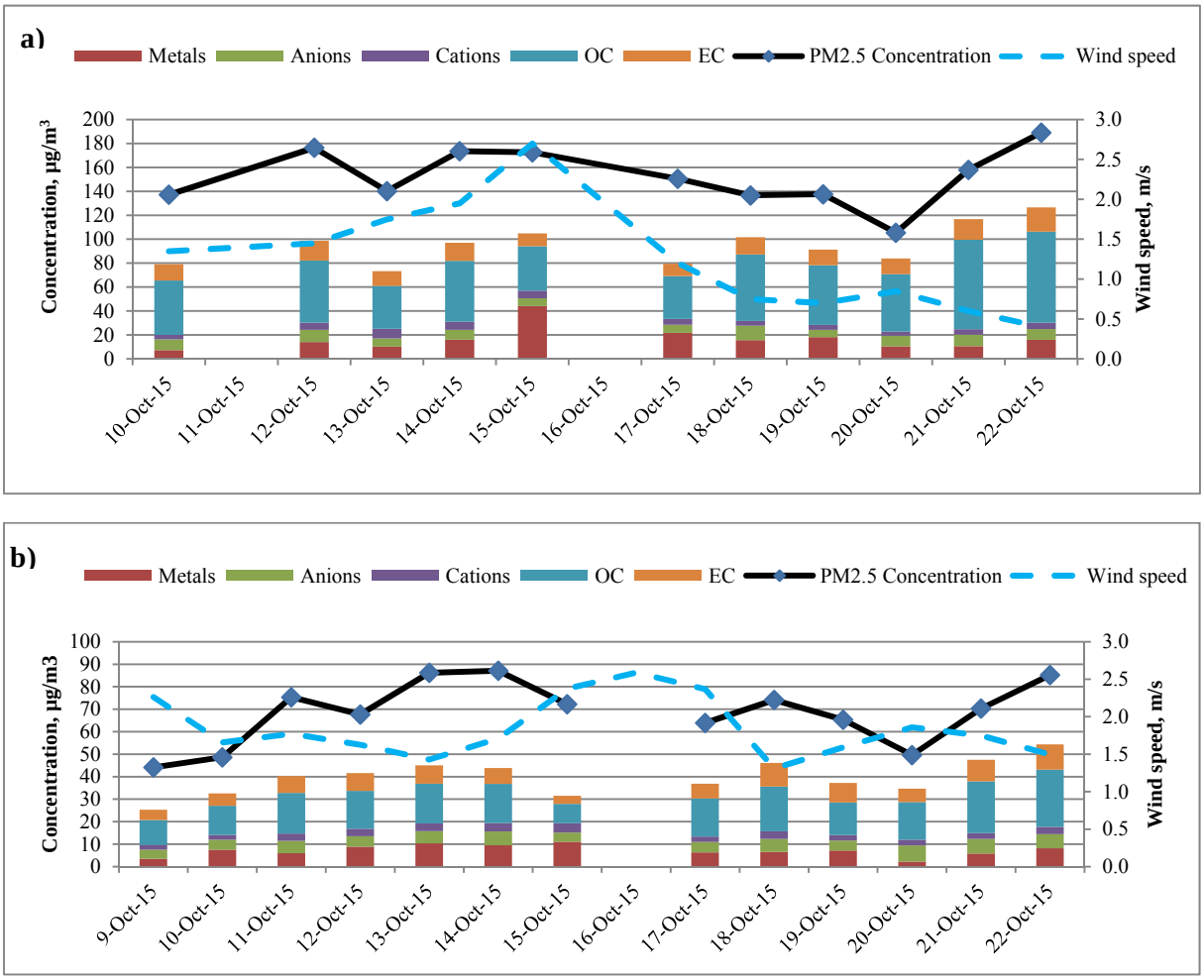

Figure 15. Relation between wind speed and $PM_{2.5}$ mass concentration its components a) UB03 site; b) NAMEM site.

Correlations between $PM_{2.5}$ components and $PM_{2.5}$ mass concentration, wind speed or relative humidity were investigated (Table 6). At the UB03 site, $PM_{2.5}$ mass concentration correlated with OC,

EC and other toxic elements of As, Se, Pb. This implies specific sources of organic aerosols, black carbon and toxic elements near the monitoring site. On the other hand, more ionic components, metals and metalloids were correlated with $PM_{2.5}$ mass concentration at NAMEM site. Because the NAMEM site is located in office area and there is no specific local sources around 1 km of the site, these correlations imply that $PM_{2.5}$ is composed of the mixture of various emission sources. Although correlations between meteorological parameters of wind speed or relative humidity and some $PM_{2.5}$ components were observed, the reasons of these correlations are not clear. It is necessary to collect additional observation data in various meteorological conditions.

Table 6. Site specific correlations between the $PM_{2.5}$ components and $PM_{2.5}$ mass concentration, wind speed or relative humidity.

UB03 site		NAMEM site	
$PM_{2.5}$ mass concentration positivity correlated to follow parameters ($R^2 > 0.6$)			
OC, EC, F^-, Cl^-, NO_2^-, Cu, As, Se, Pb		OC, EC, F^-, $HCOO^-$, NO_2^-, NO_3^-, Mg^{2+}, Ca^{2+}, Li, Be, Na, Al, K, V, Fe, Ga, As, Se, Rb, Sr, Ag, Cd, Cs, Ba, Hg, Tl, Pb, U	
Wind speed			
Positive ($R^2 > 0.6$)	Negative ($R^2 < -0.6$)	Positive ($R^2 > 0.6$)	Negative ($R^2 < -0.6$)
PO_4^-, Ca, Li, Be, Al, Sc, V, Cr, Fe, Mn, Co, Rb, Sr, Mo, Hg, Th	VOCs, OC, EC, SO_4^{2-}, NH_4^+, As, Se, Ti	None	VOCs, OC, EC, F^-, $HCOO^-$, NO_2^-, NO_3^-, As, Se, Cd, Ti
Relative humidity			
Positive ($R^2 > 0.6$)	Negative ($R^2 < -0.6$)	Positive ($R^2 > 0.6$)	Negative ($R^2 < -0.6$)
CH_3COO^-, Br^-, SO_4^{2-}, NH_4^+	Ca, Li, Be, Al, K, Sc, V, Fe, Mn, Co, Cu, Sr, Cs	Br^-, SO_4^{2-}, NH_4^+	Ca^{2+}, Li, Be, Na, Mg, Al, K, V, Fe, Mn, Ga, Rb, Sr, Ag, Ba, Ti

Enrichment factor (EF) is often used to compare compositions of chemical elements in aerosol particles with those in reference materials on order to qualitatively identify their potential natural and/or anthropogenic sources (Taylor and McLennan, 1995). The EFs of $PM_{2.5}$ particles can be calculated as follows:

$$EF = [M(PM_{2.5}) / Al(PM_{2.5})] / [M(Crust) / Al(Crust)]$$

where $M(PM_{2.5})$ is a concentration of element M in $PM_{2.5}$, and M(Crust) is a concentration of element M in earth crust (Wedepohl, 1995). When the enrichment factor of an element is close to 1, it is likely that this element originates from natural sources, and when the enrichment factor is larger than 1, it is likely that the element was enriched by artificial or human sources. Table 7 shows averages of the EFs of metallic and metalloid elements in $PM_{2.5}$ at the UB03 and NAMEM sites. These elements are classified into 3 groups, namely, low enriched ($1 < EF < 10$), moderate enriched ($10 \leq EF < 100$) and high enriched ($100 \leq EF$). Ca, Fe, Ba, Cr, Mn, Sr, Li, V, Co, Cs, Tl and U at the UB03 and NAMEM sites are classified to low enriched elements. There elements were similar to earth crustal composition and were not significantly originated from anthropogenic emission sources. On the other hand, Se, Cd, Sb at the both UB03 and NAMEM sites and Zn, As, Hg at the UB03 sites are classified to high enriched elements. These elements are highly contaminated from local emission sources.

Higher EFs of Hg, Se, Ag would be originated from coal combustion sources in coal power plant and domestic heating or cooking. Trace element analysis of Carboniferous–Permian coal in inner Mongolia demonstrated that concentrations of V, Cu, Zn, Ga, Se, Sr, Ag, Cd, In, Ba and U were higher

than those of world hard coal and that Se, Ag, Pb and Bi were enriched in the coal (Duan, *et al.*, 2015). Mercury emission from coal seam fire in Inner Mongolia of China will impact on the global mercury inventory and mercury concentrations in the urban areas of northern China (Liang *et al.*, 2014). Zn is prominently originated from tire wear dusts (Councell *et al.*, 2004), and Sb is from break wear dusts (Iijima *et al.*, 2008). Therefore, high EFs of Zn and Sb in Ub would be originated from automobiles. Heavy traffic in Ulaanbaatar is one of causes of air pollution and may affect mortality in Ulaanbaatar (Allen, 2013).

When the EFs are compared with those in Tokyo from 1995 to 2004 (Furuta *et al.*, 2005), the EFs of V and Ni, fingerprint elements of oil combustion sources, for fine particles (< 2 μm) in Tokyo were 473 and 64.6, respectively. On the other hand, the EFs of V and Ni in UB were so very low that those are classified in low enriched elements. There are a number of oil combustion sources such as chemical industry and diesel ships in Tokyo bay area, whereas the oil combustion sources would be lower contribution in UB considering from the EFs. These unique characteristics are important to further clarification of source contribution of $PM_{2.5}$ in UB.

Table 7. Averages and standard deviations of enrichment factors of metallic and metalloid elements in $PM_{2.5}$ at the UB03 and NAMEM sites

UB03 site				NAMEM site			
Element	EF	**Element**	EF	**Element**	EF	**Element**	EF
Na	2.15 ± 2.47	**Pb**	28.7 ± 14.5	**Na**	0.65 ± 0.04	**Pb**	28.5 ± 12.2
Mg	1.27 ± 0.71	**Li**	1.61 ± 0.51	**Mg**	0.94 ± 0.05	**Li**	1.77 ± 0.17
Ca	1.32 ± 0.36	**Sc**	1.73 ± 0.20	**Ca**	1.39 ± 0.49	**Sc**	1.20 ± 1.48
Fe	1.18 ± 0.17	**V**	1.49 ± 0.28	**Fe**	1.39 ± 0.19	**V**	1.64 ± 0.24
Zn	117.5 ± 79.5	**Co**	1.31 ± 0.15	**Zn**	72.4 ± 51.7	**Co**	2.02 ± 2.21
Ba	1.44 ± 3.31	**Se**	241.7 ± 167.2	**Ba**	1.12 ± 0.17	**Se**	187.3 ± 114.0
Ni	13.5 ± 19.6	**Ag**	29.3 ± 18.6	**Ni**	4.5 ± 12.7	**Ag**	81.1 ± 23.7
Cu	12.4 ± 4.6	**Cd**	182.6 ± 115.5	**Cu**	25.1 ± 16.3	**Cd**	219.7 ± 117.6
Cr	5.63 ± 6.84	**Sb**	216.2 ± 79.6	**Cr**	8.58 ± 9.26	**Sb**	550.1 ± 424.3
Mn	1.53 ± 0.28	**Cs**	1.11 ± 0.35	**Mn**	2.37 ± 0.81	**Cs**	1.13 ± 0.07
Ga	32.5 ± 27.1	**Hg**	205.8 ± 143.2	**Ga**	5.79 ± 0.95	**Hg**	72.5 ± 94.3
As	118.4 ± 76.8	**Tl**	5.98 ± 3.10	**As**	93.2 ± 55.1	**Tl**	4.30 ± 2.20
Sr	1.04 ± 0.27	**Bi**	64.1 ± 45.7	**Sr**	1.10 ± 0.17	**Bi**	72.5 ± 79.3
Mo	90.2 ± 132.0	**U**	3.14 ± 3.81	**Mo**	12.5 ± 6.2	**U**	1.62 ± 0.58

4. Conclusions

For the effective management of air quality, great importance is attached to the identification of the sources of ambient particulate matter, and to the quantification of the mass concentration of each source. The objective of this study was to provide useful data set of $PM_{2.5}$ mass concentration and their chemical components to investigate ambient fine particulate source apportionment study in Ulaanbaatar where the $PM_{2.5}$ source composition profiles were not available till now. In the future work we will analyze the monitoring data sampled in January 2016 and conduct Positive Matrix Factorization (PMF) analysis for source apportionment of $PM_{2.5}$ after complete our second batch of sample analysis which were collected in January, 2017. So far, we have found out some findings as follows in our data set;

- Carbonaceous components were most dominant contribution (approx. 40 %) to $PM_{2.5}$ in UB followed by metallic and metalloid components and ionic components (approx. 10 %). The contribution of OC in UB are remarkably higher than those at ambient and roadside stations in Japan. This implies higher emission originated from combustion process in coal power plant and domestic use in Gers.
- Local meteorology is an important to determine characteristics of $PM_{2.5}$ in UB. For example, high concentrations of geological elements concentration were observed at the UB03 site when daily average wind speed was higher than 2 m/s. The strong wind generated soil dust at the UB03 site, but at the NAMEM site, there is no influence of soil generated dust in downtown area and PM mass concentration was decreased. The organic carbon exhibited higher concentrations during calm conditions suggesting emissions from local sources
- At the UB03 site, $PM_{2.5}$ mass concentration correlated with OC, EC, As, Se, Pb. This implies specific emission sources the monitoring site. $PM_{2.5}$ concentration was correlated to most of analyzed components at the NAMEM site, which may be originated from mixture of various emission sources.
- The enrichment factors at the UB03 and NAMEM sites demonstrated that Se, Cd, Sb at the both UB03 and NAMEM sites and Zn, As, Hg at the UB03 sites were highly enriched. These elements would be highly contaminated by local emission sources such as coal combustion, tire wear and brake wear dusts.

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Influence of long-range transport on air quality in northern part of Southeast Asia during open burning season

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Abstract

Smoke haze episode occurs annually in northern part of Southeast Asia (NSEA) during the hot dry season. Concentration of air pollutants including particulate matter with diameter less than 10 μm (PM_{10}), CO, NO_x and O_3 are 2–3 times higher than those in the rest of the year. Open burning is a major source of air pollution in this area. This study aims to investigate an impact of long-range transport on air quality in the NSEA during open burning season and to estimate the origin of pollutants by comparing of PM aging at two different stations. Chiang Mai (CM) city was set as the receptor site. Based on the air transport paths to the origin from 3-day backward trajectory during Feb–Apr for 3 years (2012–2014), air masses were clustered into two groups for lower altitudes (500 m AGL) and three groups for higher altitudes (1,000 m and 1,500 m AGL). Moreover, three groups of air masses from different area were also observed from a total three attitude levels clustering. The result of air masses clustering indicated that the major air masses through CM were transported from the southwest direction of CM and the highest level of pollutants was originated from India region. The results revealed that the rising of air pollutant concentration in the area was impacted from long-range transport. However, the variation of pollutant concentration was observed in CM area. It was mainly impacted from changing of pollutant sources (number of open burning) and metrological conditions within a day. Ratios of ions (SO_4^{2-} and NO_3^- with K^+) found in PM were used to predict their ages. The $\frac{SO_4^{2-}}{K^+}$ ratio in Yangon (YG) was 2 times higher than that in CM. It suggested that the age of PM at CM was fresher than that in YG. Therefore, it revealed that CM was not a receptor site receiving pollutants from YG. Characteristic of PM in CM was a mix of aged

PM from long distance source and fresh PM emitted from local open burning, while only aged PM from long distance source (mostly across the sea) was found in YG.

Key words: Air pollution, Backward trajectory, Biomass burning, Long-range transport, Particulate matter aging

1. Introduction

Regarding annual smoke haze problem in Northern Thailand in the dry season, open burning including forest fire and agricultural waste burning are the major source. This air pollution problem causes serious degradation of economic, environment and health. Bad air quality is related to increasing of air pollutant concentration both in form of particulate matter and gaseous pollution such as carbon monoxide (CO), nitrogen oxide (NO_x) and volatile organic compounds (VOCs). Moreover, the secondary pollutants including ozone (O_3), which is toxic to human health and vegetation, is also increasing in this period. Concentrations of particulate matter at a size of less than 10 μm (PM_{10}) and O_3 are frequently exceeded Thailand National Ambient Air Quality Standard at 120 μg/m^3 (24 hrs average) and 200 μg/m^3 (hourly concentration) for PM_{10} and O_3, respectively. However, the pollutants contributed in Northern Thailand not only come from regional source but also from transportation of atmospheric pollutants over long distance (long-range transport) (Kim Oanh and Leelasakultum, 2011). Trajectories are well known to be good indicators of large-scale flow and can be useful for studying the potential sources of regional.

Measurement of CO along with O_3 can provide evidence for the impact of anthropogenic pollution (Lin *et al*., 2011 and Sikder *et al.,* 2011). CO is one of the O_3 precursors which is selected as an indicator of long-range transport because its lifetime (1–2 months) is longer than other O_3 precursors (Suthawaree *et al*., 2008). Moreover, NO_x which is defined as the sum of the species NO and NO_2, can also lead to O_3 production. The lifetime of NO_2 ranges from around 8 hrs in a typical planetary boundary layer to a few days in the upper troposphere (Tie *et al*., 2001; Beirle *et al*., 2011; Zien *et al*., 2014). Additionally, PM_{10} also has –3 days lifetime contribution in atmosphere (McMurry *et al*., 2004). Therefore, measurement of air pollutants such as PM_{10}, CO, O_3 and NO_x coupled with backward trajectory analysis should provide information concerning effect of long-range transport on air pollution.

Large amount of organic and inorganic particles were derived from biomass burning. Potassium (K) compounds, important inorganic particles, are commonly used as markers for biomass burning because it does not degrade during transportation (Freney *et al*., 2009; Tao *et al*., 2016). In general, physical and chemical properties of biomass burning particles have been changed based on the aging processes after emission. Normally, potassium chloride (KCl) were the most abundant inorganic particle type in the young smoke and then most of KCl particles have been converted to potassium sulfate (K_2SO_4) and potassium nitrate (KNO_3), through gas-particle reaction, with only minor KCl left (Li *et al*., 2003; Kong *et al*., 2015). Gao *et al*. (2003) found increasing of SO_4^{2-} and NO_3^- composition but decreasing of chloride ion (Cl^-) composition in the plum at downwind fire. Choung *et al*. (2015) indicated that the concentration of SO_4^{2-} and NO_3^- in PM at a size of less than 2.5 μm ($PM_{2.5}$) increased when it is transferred to far source region. Therefore, the ratio of ions such as $\frac{Cl^-}{K^+}$, $\frac{NO_3^-}{K^+}$ and $\frac{SO_4^{2-}}{K^+}$ would help to compare the aging of PM and indicate between near source and receptor area.

The aims of this study are to estimate the influence of long-range transport on air quality in northern part of Southeast Asia (NSEA) during open burning season, and to investigate origins of air pollution by comparing ratio of some ion species in PM based on backward trajectory and direction of air mass movement. Data obtained from this research will be useful for local and governmental organizations in term of environmental management, especially to improve air quality in the area.

2. Method

2.1 Monitoring stations

Base on backward trajectory, air masses arriving Chiang Mai Province during February–April was mainly originated from the southwest direction of the province (Kim Oanh and Leelasakultum, 2011; Chantara *et al*., 2012; Wiriya *et al*., 2013). Chiang Mai (CM), Thailand was therefore chosen as the receptor site and Yangon (YG), Myanmar, located upwind of CM, was selected as near source sampling site (Figure 1). Chiang Mai is the largest province in Northern part of Thailand. It is located 700 km north of Bangkok (capital city). The province is situated between latitude 17°15′–20°06′ N and longitude 98°05′–99°21′ E at elevation of 310 meters above sea level (ASL). This area consists of valleys and hills and forests. Mountain ranges are located in a north-south direction. North of the province is connected to Myanmar. The province covers an area of 20,107 km^2. The major land use patterns are forest (82.6 %) and agricultural land (14.6 %) (National Statistical Office. 2012). Population density is 81.6 people/km^2, in which the highest population density is observed at Chiang Mai city (1,563 people per km^2) (National Statistical Office. 2010). Chiang Mai presents a tropical wet and dry climate which it is characterized by the monsoon. The south-west monsoon usually arrive Thailand during the end of May–November. It brings stream of warm moist air from the Indian Ocean causing abundant rain (rainy season). Then the north-east monsoon starts from mid-November until mid-February and brings cool air from northern Vietnam/China which presents cool season. The period from mid-February until the end of May is the transition period between both monsoons and intensive thermal low therefore the hottest weather is observed in this period. The annual average temperature is 27.0 ºC and the highest temperature is observed in April (39.2 ºC), while the lowest is in January (11.0 ºC). The annual average humidity is 71 %. An annual number of rainy days are 122 days with 1,075 mm the total rainfall (Northern Meteorological Center, Thailand. 2014).

Yangon province is the largest economic center of Myanmar. This area is located in Lower Myanmar at the convergence of the Yangon and Bago Rivers, and is surround Andaman Sea in the south (Yangon City Development Committee. 2016). The province is situated between latitude 17°45' - 16°12' N and longitude 95°41'–96°49' E at elevation of 30 meters above sea level (ASL). The area within 40 km of Yangon International Airport (17 km north of YG city hall) is covered by croplands (85 %), oceans and seas (5 %), forests (3 %), built-up areas (3 %), and lakes and rivers (2 %). YG is the most densely populated region in Myanmar (723 people/km^2 in 2014). The climate is a tropical monsoonal character, with three distinct seasons: a rainy season from June to October, a cooler and drier winter from November to February, and a hot dry season from March to May. The annual mean temperature and precipitation are 27.4 ^oC and 2,681 mm respectively (Climatemps, 2016 and Weatherspark, 2016).

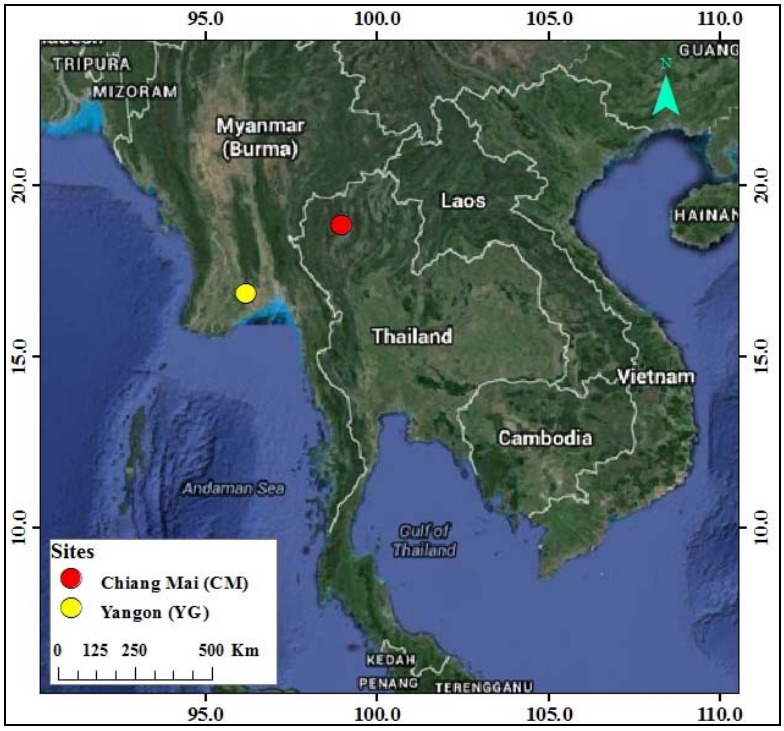


Figure 1. Map of study area.

Figure 2 shows the location of CM observation stations comprise of Air Quality Monitoring (AQM) station and Acid Deposition Monitoring Network in East Asia (EANET) stations. The AQM station is located at CM city hall (18°50'N, 98°58'E), which is 5 km south of the city center. The major land use around this station consists of community area, paddy fields and forests. EANET station (18°46'N, 98°56'E) is located in Chiang Mai University, Mae Hia campus. It is about 5 km and 10 km in the southwest direction of city center and AQM station, respectively. This area is surrounded by agricultural area and forest, therefore it is classified as sub-urban area. At YG, the EANET station is installed at Department of Meteorology and Hydrology Mayangone (16°30'N, 96°07'E). It is located in the North of YG city hall with approximately 10 km in distance. It is surrounded by residential area, and therefore it was characterized as urban site based on EANET criteria.

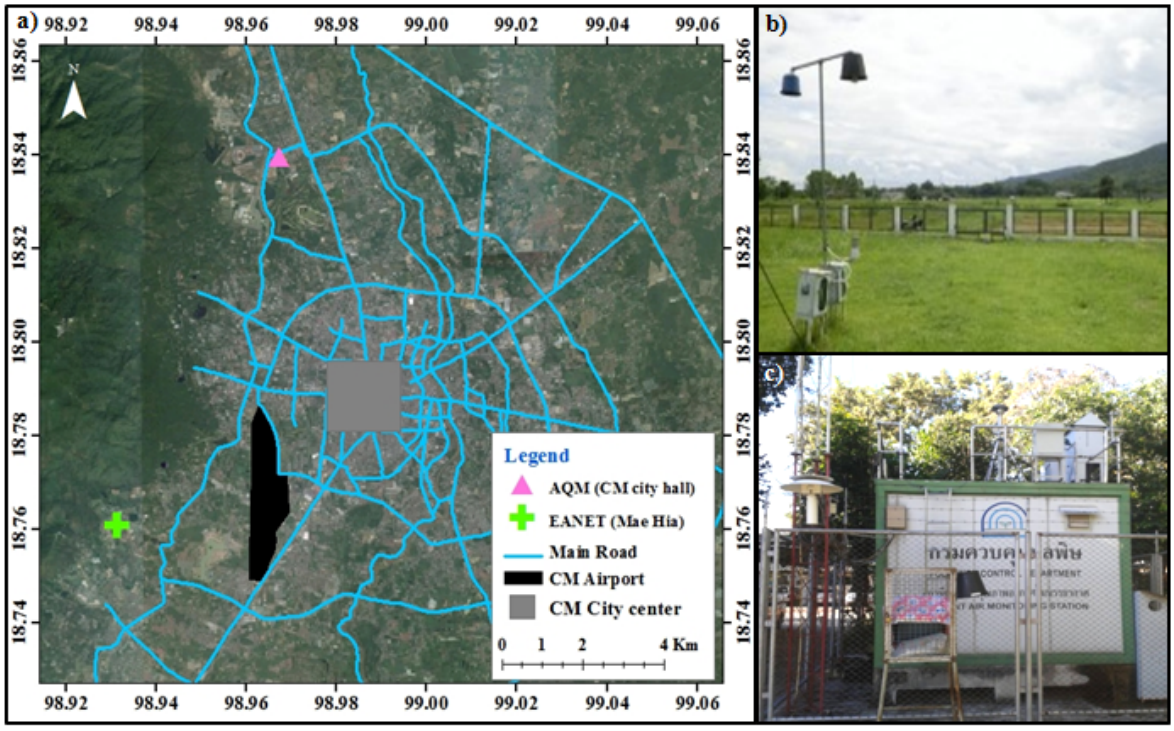


Figure 2. Location of CM station a) observation stations; b) EANET station; c) AQM station.

2.2 Monitoring data

2.2.1 Air pollutants

The hourly CO, O_3, NO_x and PM_{10} concentrations from 2012 to 2014 measured at the AQM station were obtained from Pollution Control Department (PCD), Thailand. PM_{10} concentration was measured continuously using a Taper Element Oscillating Microbalance (TEOM). Ultraviolet (UV) and chemiluminescence techniques were employed for O_3 and NO_x measurement, respectively. The non dispersive infrared (NDIR) Gas Filter Correlation photometry was used for CO measurement.

Missing data of all 3-year monitoring data used in this study was < 3 % for PM_{10} and O_3, and about 10 % for CO and NO_x.

2.2.2 Water soluble ions

Three year data (2012–2014) of water-soluble ions (Na^+, K^+, NH_4^+, Cl^-, NO_3^-, SO_4^{2-}) from dry deposition analysis (particulate phase) collected from CM and YG EANET stations was provided from Asia Center for Air Pollution Research (ACAP). The samples were collected by using the four-stage filter pack. Each sample was collected continuously for 10days and 15days for CM and YG station, respectively. The samples were extracted and analyzed for water-soluble ion content by using Ion Chromatograph (IC) (EANET. 2000) (Chantara *et al*., 2012). Outlier data of water-soluble ions were eliminated by using (statistic) Q-test. At 95 % confidence level 4 % (CM) and 6 % (YG) of the data were rejected.

In order to subtract effects of sea salt ion, only non-sea-salt (nss) ions were used for nearby source analysis. Equation (1), nss ion is calculated from subtraction of total measured ion with sea–salt (ss) ion. ss-ion and nss-ion were calculated using equation (2) and (3), respectively. (Radojevic and Bashkin, 2003) (EANET. 2015).

$$[nss-ion_x] = [ion_x] - [ss-ion_x] \quad (1)$$

$$[ss-ion_x] = \frac{X_{sea}}{Na^+_{sea}} * [Na^+] \quad (2)$$

$$[nss-ion_x] = [ion_x] - \frac{X_{sea}}{Na^+_{sea}} * [Na^+] \quad (3)$$

Where [nss-ion$_x$] is non-sea-salt part of interest ion; [ion$_x$] is total measurement of interest ion; [ss-ion$_x$] is sea-salt part of interest ion; [Na^+] is sodium concentration; $\frac{X_{sea}}{Na^+_{sea}}$ is the ratio of interest ion and sodium ion in sea water. The ratios are shown in Table 1.

Table 1. Sodium and component ratio in sea water (Radojevic and Bashkin, 2003).

$\frac{X_{sea}}{Na^+_{sea}}$	Ratio value by mole
$\frac{SO_4^{2-}}{Na^+}$	0.12
$\frac{Cl^-}{Na^+}$	1.16
$\frac{K^+}{Na^+}$	0.021

2.3 Air mass trajectories and air pollutants analysis

Backward trajectories of the data in the dry season (February to April) during the period of 2012–2014 were determined in order to identify the origin and transport paths of air masses and their arrival to CM and YG. 3-day backward trajectory was normally used to determine the regional of air pollutant distributed to Chiang Mai City from air mass movement (Somporn *et al.,* 2012) (Soppitaporn *et al.*, 2013) because it could distinguish the origin of air mass for both local and long-range area in which helpful to estimate the influence of the air mass source on pollutants contributed in the city. The origins of air mass calculated from one day backward trajectory were observed at around the border of Thailand (Wan *et al*., 2013), while 5-day backward trajectory presented very long distance of the origin (India, Bangladesh, Myanmar and Andaman seas) which could not illustrate the local effect (Kim Oanh *et al.,* 2011). Therefore, 3-day backward trajectories were calculated in this study by using *TrajStat-Trajectory Statistics* program version 1.2.2.6 which was developed by Yaqiang Wang in 2008 (Wang *et al*., 2009). This program is using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model as an external process to calculate trajectories, and Euclidean distance clustering method as well as a geographic information systems (GIS) technique for spatial data management, visualization and analyses. The program was available at http://www.arl.noaa.gov/ready/hysplit4.html. The meteorological input for the trajectory model was the GDAS (Global Data Assimilation System) and meteorological data (1° x 1°). In this study, the backward trajectories were calculated 4 times per day (end point) at 2:00, 8:00, 14:00 and 20:00 Local Sidereal Time (LST) (19:00, 1:00, 7:00 and 13:00 UTC, respectively) for total of 268 days during a period of 3 months (February–April) for 3 years (2012–2014). Additionally, 3 levels of an arrival altitude were set at 500, 1000 and 1500 m AGL in order to evaluate the variance existing among trajectory paths at different heights. Therefore, 3,216 backward trajectories per stations were calculated.

Concentrations of pollutants in each calculated time were individually matched with the clustered trajectories in order to analyze air pollutant with the categorized trajectory paths. In case of arrival attitudes, there were assumed that pollutant concentrations in each attitude were similar in this study.

2.4 Statistical analysis

The Spearman's correlation among air pollutants was calculated. One way ANOVA (analysis of variance) was used for comparison of air pollutants among the transport paths of air masses. Moreover, Q test was employed to subtract outlier concentrations.

3. Results and discussion

3.1 Backward trajectory analysis

The selected study period (February–April) is a period of open burning in the NSEA. Consequently smoke haze pollution is observed annually. The 3-day backward trajectories were employed using the data of the study period (February–April in 2012–2014) at CM AQM station in order to explain the variability and the influence of transport pathways on air pollutant contribution to air quality in CM province. Figure 3 illustrates the clustered analysis results of 3-day backward trajectories in CM at three arrival attitudes. Air mass patterns were classified based on their origins and directions of air masses before arriving at the destination (CM). For reliability of clustering, number of source regions was considered from clustering pattern which percentage of air mass contributed in all paths were greater than 10 %. Therefore, 3 clusters were classified in this study namely India (ID), Myanmar (MM) and Thailand (TH) paths. ID path was originated from northern of India and passed over Bengal bay and Myanmar through Chiang Mai from the southwest. MM path was initiated from India ocean or Myanmar continental area and directly moved northeast before arriving at the site. Both ID and MM paths were represented long-range transport of air massed in this study. Whereas TH path, was symbolized regional air mass trajectories which came from Thailand continental area or border of Thailand and Myanmar, then directly moved to the receptor. There were different air mass patterns with the arrival attitude. Two groups of air masses were clustered for 500 m AGL, in which 88 % and 12 % of the trajectories were from MM and TH, respectively (Figure 3a)). Meanwhile, three air movement paths were classified for 1000 m and 1500 m AGL, from MM, TH and ID (Figure 3b)–3c)). The results suggested that air mass path originated from very long distance (> 1000 km) such as ID is mainly contributed at high attitude level of air masses.

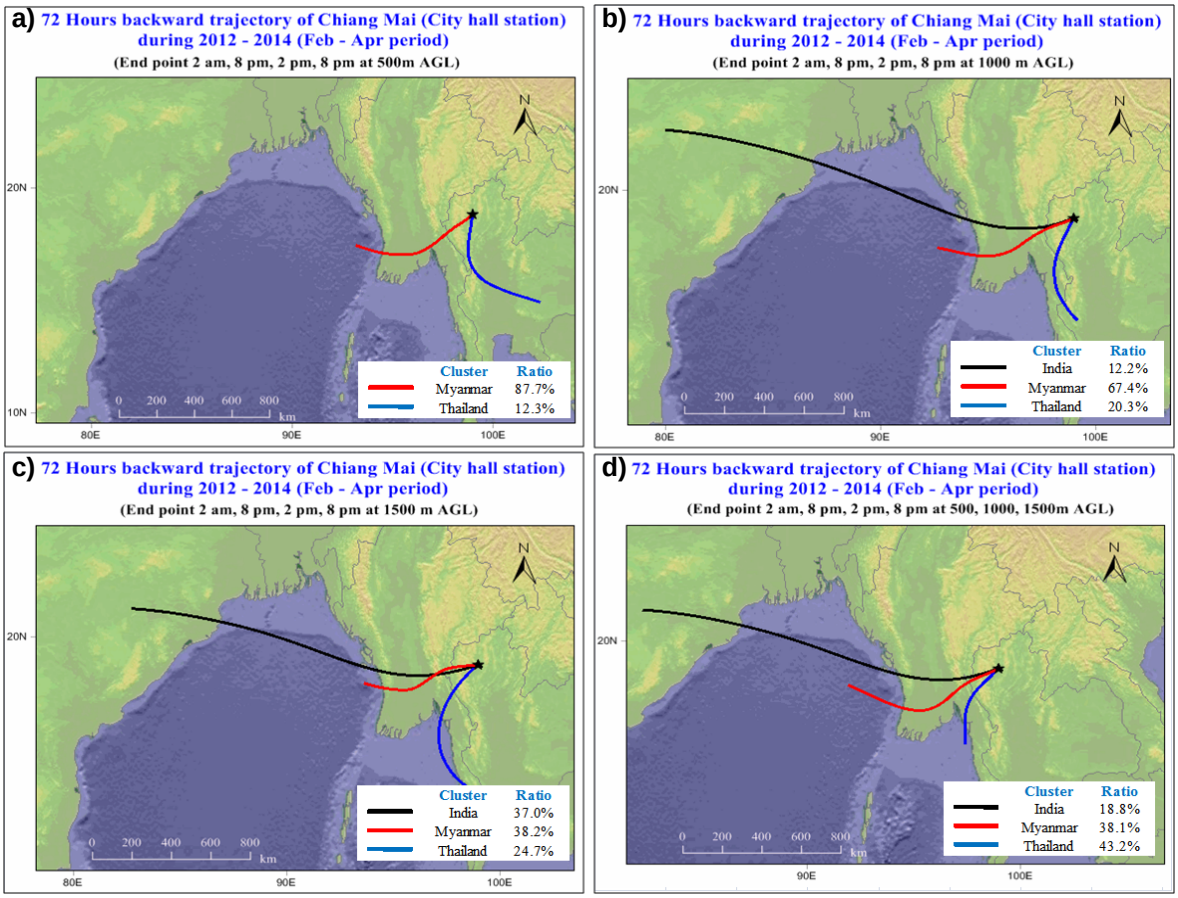


Figure 3. Cluster of 3-day backward trajectories during February to April (2012–2014) arrival at CM, a) 500m AGL; b) 1,000m AGL; c) 1,500m AGL; d) total 3 attitudes.

In case of the total four attitude level backward trajectories, three groups of air mass paths were clustered (Figure 3d)). Air masses paths to CM were mainly from Western border of TH (43 %) and MM continental area (38 %). However, all paths illustrated that air mass through CM came from southwest direction. The results of air mass patterns were well agreed with the previous study (Chantara *et al.*, 2012) conducted at the same study site. They reported that the main air mass of the 3-day backward trajectories in the dry season from 2005 to 2009 came from the southwest direction of Chiang Mai. Therefore, those results illustrated that the air pollutants in Chiang Mai were influenced by activities along the southwest direction through India continental area.

3.2 Air pollutants variation during 2012–2014

Concentrations of air pollutants, including PM_{10}, O_3, CO and NO_x, daily rainfall and wind speed in CM province during 2012–2014 were calculated.

3.2.1 Overview of air pollutant concentrations and some meteorological data of Chiang Mai city

Data of air pollutants, rainfall and wind speed from 2012 to 2014 were plotted (Figure 4). Rain amount and wind speed during the study period (red boxes) were relatively lower than another period of the year. The average wind speed in CM city was in a range of 15–30 km/hr. This value is classified as gentle to moderate breeze in Beaufort scales. Even though increasing of wind speed was presented in April but it was only a few days during mid to the end of month due to thunderstorm effect. Moreover, the geography of the CM which is located in the basin surrounded by high ranges of mountain is supporting

accumulation of air pollutants in the area. The concentrations (µg/m^3) of PM_{10}, O_3, CO and NO_x during February–April in 2012–2014 were 84.5 ± 43.8, 68.5 ± 16.3, 814.2 ± 453.8 and 36.6 ± 12.1, respectively. They were 2-3 times higher than the rest of the year (Table.2).

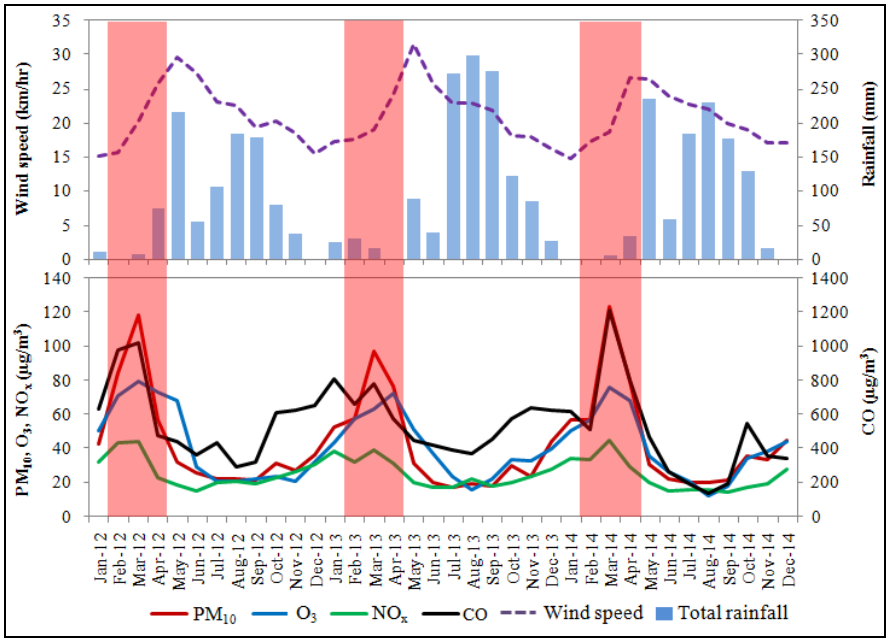


Figure 4. Monthly concentrations of air pollutants, wind speed and rainfall of CM city in 2012–2014. Hatched period shows open burning season in the NSEA.

Table 2. Air pollutant concentrations in CM AQM station during 2012–2014.

Pollutants		Concentration (µg/m^3)			
		PM_{10}	O_3	CO	NO_x
Feb–Apr	Mean ± SD	84.5 ± 43.8	68.5 ± 16.3	814.2 ± 453.8	36.6 ± 12.1
(study period)	Min	18.6	25.5	105.6	6.6
	Max	275	112	3,241	76.5
May–Jan	Mean ± SD	30.0 ± 13.9	32.4 ± 15.4	451.2 ± 294.9	21.8 ± 7.7
	Min	4.7	4.4	0.0	7.4
	Max	96.0	99.1	1,554	48.0

Hourly average concentrations of air pollutants from February to April 2012–2014 were plotted (Figure 5). PM_{10}, CO and NO_x concentrations presented the same diurnal variation pattern, in which two peaks were observed. The maximum concentrations was found around 8:00–9:00 LST, and decreasing in the afternoon and the secondary peak was found around 20:00 LST. Due to increasing of boundary layer height in the afternoon from increasing of temperature, the pollutants were dispersed and diluted (Zhao *et al.*, 2016). Whereas, the maximum O_3 concentration was observed in the afternoon period because O_3 is produced during presence of solar radiation (Gao *et al.*, 2016).

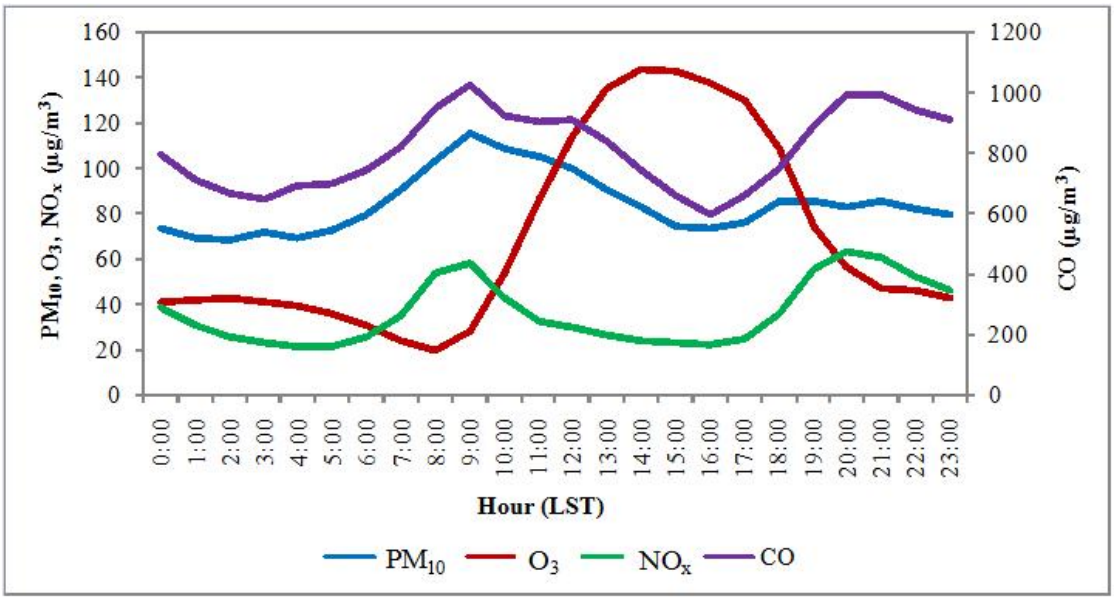


Figure 5. Diurnal variation of hourly average air pollutant concentrations of CM in a period of February–April (2012–2014).

Table 3 shows correlations among air pollutants during the study period. Significant correlations ($p < 0.01$) were observed for most of all air pollutants, which were illustrated that those pollutants should come from similar source. However, low correlations were observed between O_3 and other pollutants. It probably because O_3 can be generated from various sources such as biomass burning and traffic emission through series of physical processes and complex photochemical reactions therefore the formation mechanism is nonlinearly related to precursors (Gao *et al.*, 2016).

Table 3. Air pollutants correlation during February–April period 2012–2014 in CM.

Pollutants	Correlation		
	PM_{10}	O_3	CO
O_3	0.480**		
CO	0.529**	0.209**	
NO_x	0.750**	0.105	0.580**

*Note: ** significant correlation with $p < 0.01$*

3.2.2 Air pollutant concentrations associated with classified trajectories

According to the result from backward trajectory analysis for CM station, the variation of air pollutants classified with air mass paths was analyzed in this section. Pollutant concentrations and the clustered backward trajectories (3,216 trajectories) from same arrival times (end point) were matched in order to categorize the origin of pollutants with air mass movement. Figure 6 illustrates mean concentrations of PM_{10}, O_3, CO and NO_x associated with each air mass category. Significant differences ($p < 0.05$) of among clusters were found from PM_{10}. The highest concentration was observed from ID path (94.4 ± 55.4 µg/m^3), then followed by MM path (86.8 ± 48.4 µg/m^3) and TH path (78.3 ± 47.9 µg/m^3). Concentration of NO_x from ID cluster (45.1 ± 30.8 µg/m^3) was comparable to that from MM cluster (43.3 ± 29.5 µg/m^3). Nevertheless, the concentration from both clusters was higher than that from TH cluster (39.3 ± 26.5 µg/m^3). CO concentrations in descending order were ID (876 ± 603 µg/m^3) > TH (849 ± 597 µg/m^3) > MM (820 ± 548 µg/m^3). There were no significant difference among cluster observed for CO as

well as O_3. Mean concentrations of O_3 for all three sites were comparable in the range of 85.9–92.2 µg/m^3. The results of this study indicated that the highest air pollutant concentration came from ID cluster, while almost of air movement path involved in CM Province moved from TH and MM clusters.

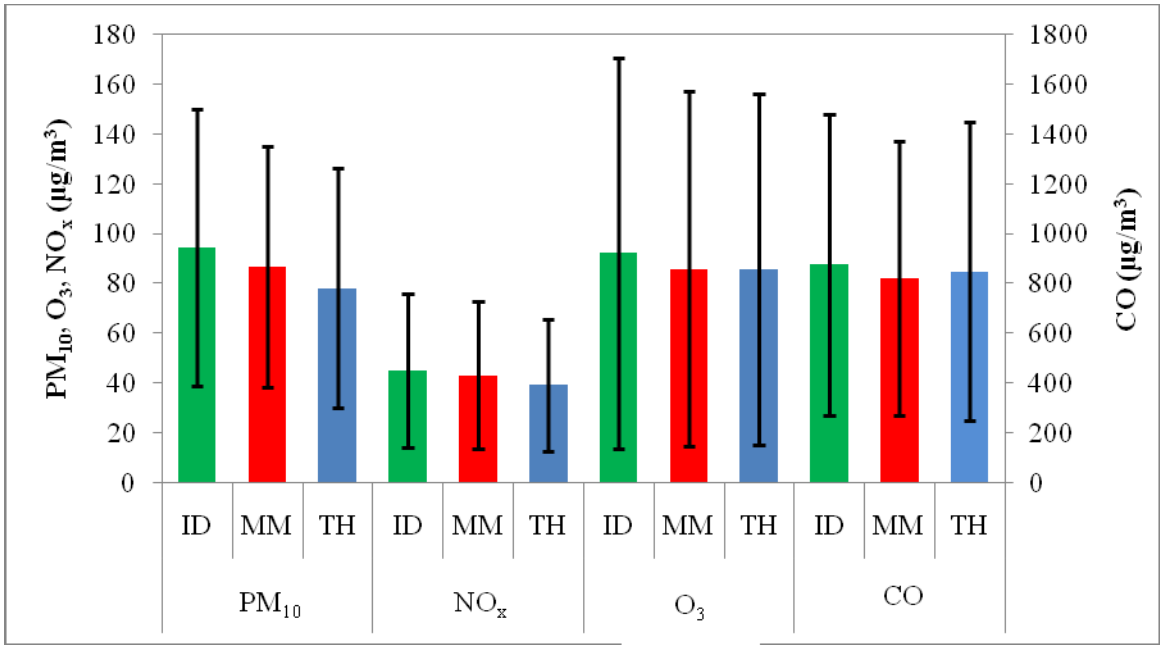


Figure 6. Mean concentrations of air pollutants categorized by air mass pattern.

According to diurnal patterns of air pollutants within a day. Consequently arrival times of air masses were applied to classify the pollutant concentrations in order to estimate influence of air mass paths on the pollutants contributed in the site at different arrival times. Table 4 presents mean concentrations of air pollutants in each arrival time and categorized air masses. Concentrations of PM_{10} and NO_x from MM and ID paths at 2:00 were comparable with those from regional path (TH path). Significant differences among the paths were observed at 8:00, 14:00 and 20:00 hours. It might be because low concentrations of long-range transport pollutants in the early morning due to low number of open burning in night time (Reid *et al.*, 2012). The pollutant concentrations influenced from regional source were observed in the early morning while the daylong impacts of long-range transport were observed until evening. Impacts from both regional and long-range sources were observed on CO concentration in CM because difference of CO concentration was varied all the time, but no significant difference among trajectories paths was observed. In case of O_3 concentration, a significant difference among clusters was distinguished in the afternoon. It probably because amount of O_3 in this period was generated from photochemical reaction of long-range transport ozone precursors such as NO_x and CO, while local O_3 or O_3 generated from local precursors were existed in another period of day.

Table 4. Mean concentrations of air pollutants in each arrival time with air mass clusters.

Pollutants	Arrival time	Concentrations at various arrival altitude (μg/m^3)							
		500 m		1000 m			1500 m		
		MM	TH	ID	MM	TH	ID	MM	TH
PM_{10}	2:00	69.2^a	62.4^a	87.4^a	64.5^{ab}	63.7^b	73.4^a	63.4^a	66.9^a
	8:00	108^a	70.5^b	147^a	100^b	91.9^b	118^a	92.7^b	99.8^b
	14:00	85.9^a	63.3^b	113^a	81.1^b	73.3^b	93.9^a	74.2^b	80.2^{ab}
	20:00	85.4^a	67.5^b	104^a	83.6^b	72.4^b	92.3^a	78.2^a	78.2^a
NO_x	2:00	26.4^a	23.0^a	29.1^a	25.5^{ab}	24.2^b	27.0^a	24.0^a	26.5^a
	8:00	55.9^a	42.1^b	68.1^a	53.8^b	47.0^b	62.0^a	49.1^b	50.2^b
	14:00	25.4^a	20.9^b	29.1^a	24.9^b	22.4^b	26.2^a	23.8^a	23.9^a
	20:00	66.4^a	42.6^b	85.3^a	65.5^b	45.0^c	72.5^a	61.1^{ab}	53.9^b
O_3	2:00	56.0^a	64.4^a	55.1^a	55.3^a	63.8^a	56.7^a	59.2^a	54.2^a
	8:00	26.6^a	23.8^a	21.3^a	27.0^a	26.3^a	24.6^a	28.7^a	24.8^a
	14:00	195^a	166^b	211^a	192^{ab}	180^b	205^a	182^b	185^b
	20:00	72.9^a	89.0^b	70.2^a	71.4^a	89.4^a	71.7^a	72.9^a	81.0^a
CO	2:00	646^a	863^b	688^a	652^a	717^a	701^a	606^a	712^a
	8:00	982^a	757^a	1158^a	946^{ab}	855^b	1047^a	893^a	910^a
	14:00	763^a	657^a	801^a	754^a	710^a	795^a	665^a	801^a
	20:00	1015^a	881^a	1307^a	991^b	871^b	1085^a	934^a	960^a

Note: ^{a,b,c} significant difference from statistic test ($p < 0.05$)between cluster in the same level

3.3 Aging comparison analysis

According to the result of 3-day backward trajectory, the major air masses to CM Province during February to April came from southwest direction (Myanmar). Therefore, aging of PM at CM and YG stations were compared by using the ratio of water-soluble ion concentrations in PM.

3.3.1 Water-soluble ions variation

By using Table 1, concentrations of water-soluble ions (K^+, Cl^- and SO_4^{2-}) from both stations were listed in non-sea-salt (nss) and sea-salt (ss) fractions and presented in Table 5. The result indicated that values of nss-Cl^- were negative for both stations, which mean that it was influenced by sea salt. Thus the concentration of nss-Cl^- was excluded in this study. In case of NH_4^+ and NO_3^-, sea salt subtraction was not necessary because of less influence from sea salt fraction.

Monthly ion concentrations (nss-K^+, NH_4^+, NO_3^- and nss-SO_4^{2-}) from both stations during the period of 3 years (2012–2014) are shown in Figure 7. Those ions concentrations were significantly correlated ($p < 0.5$) with of PM10 concentrations at CM-AQM station, except for NO_3^- from YG station.

Table 5. Monthly water-soluble concentrations in 2012–2014 at CM and YG stations.

Stations	Month	Ion concentrations (nmol/m^3)								
		[t-SO_4^{2-}]	[ss-SO_4^{2-}]	[nss-SO_4^{2-}]	[K^+]	[ss-K^+]	[nss-K^+]	[t-Cl^-]	[ss-Cl^-]	[nss-Cl^-]
CM	Jan	8.66	0.25	8.41	9.19	0.09	9.10	3.13	4.80	-1.67
	Feb	13.93	0.55	13.39	13.55	0.20	13.35	3.58	10.63	-7.05
	Mar	26.71	0.44	26.27	18.96	0.16	18.80	2.89	8.51	-5.62
	Apr	9.95	0.56	9.39	11.69	0.20	11.49	2.70	10.75	-8.05
	May	6.23	0.37	5.85	2.02	0.14	1.89	3.40	7.23	-3.83
	Jun	5.30	0.29	5.01	1.74	0.10	1.64	4.72	5.53	-0.82
	Jul	3.58	0.35	3.23	3.77	0.13	3.65	4.34	6.77	-2.43
	Aug	2.00	0.25	1.75	3.30	0.09	3.21	3.10	4.81	-1.71
	Sep	1.35	0.38	0.99	2.02	0.14	1.88	1.57	7.44	-5.87
	Oct	19.18	0.41	18.77	6.42	0.15	6.27	1.50	7.94	-6.44
	Nov	5.08	0.32	4.75	4.37	0.12	4.25	3.73	6.27	-2.54
	Dec	6.61	0.28	6.33	2.85	0.10	2.74	3.71	5.49	-1.78
YG	Jan	40.72	1.27	39.45	32.76	0.46	32.30	14.16	24.57	-10.41
	Feb	31.70	1.06	30.63	19.26	0.39	18.87	6.67	20.59	-13.92
	Mar	63.65	1.64	62.01	17.54	0.59	16.94	8.19	31.76	-23.58
	Apr	20.90	0.94	19.97	10.97	0.34	10.63	5.87	18.12	-12.25
	May	26.58	1.14	25.44	6.58	0.41	6.16	9.08	22.07	-13.00
	Jun	3.45	0.32	3.13	3.44	0.11	3.33	9.59	6.11	3.48
	Jul	18.28	2.31	15.97	3.68	0.84	2.84	17.61	44.67	-27.06
	Aug	5.48	1.19	4.82	4.34	0.43	3.95	13.76	23.11	-9.35
	Sep	12.82	1.05	11.81	6.65	0.38	6.27	6.16	20.31	-14.15
	Oct	14.35	0.65	13.74	5.18	0.23	4.95	4.82	12.49	-7.67
	Nov	21.58	0.88	20.71	18.60	0.32	18.28	7.74	16.93	-9.19
	Dec	7.07	0.35	6.89	36.00	0.13	35.88	12.31	6.83	5.48

Note: total (t-); sea-salt fraction (ss-); non-sea-salt (nss-)

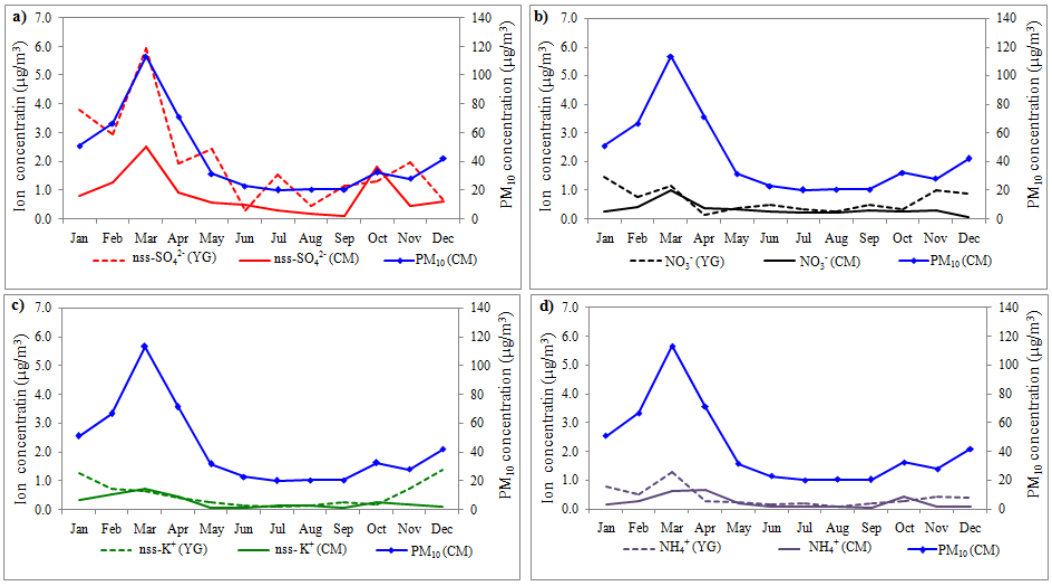


Figure 7. Monthly ion and PM_{10} concentrations at CM and YG stations a) nss-SO_4^{2-}; b) NO_3^- ; c) nss-K^+; d) NH_4^+.

Table 6. Ion concentrations in 2012–2014 at Chiang Mai and Yangon stations.

Stations		Concentrations (µg/m³)							
		Feb–Apr				May–Jan			
		nss-SO_4^{2-}	NO_3^-	nss-K^+	NH_4^+	nss-SO_4^{2-}	NO_3^-	nss-K^+	NH_4^+
Chiang Mai	Mean	1.68^{ax}	0.62^{ax}	0.58^{ax}	0.50^{ax}	0.57^{bx}	0.26^{bx}	0.15^{bx}	0.14^{bx}
	SD	1.83	0.61	0.58	0.55	0.82	0.21	0.17	0.19
	Max	6.63	2.54	2.07	2.16	4.05	0.86	0.73	1.03
	Min	0.04	0.05	0.04	0.04	0.00	0.00	0.00	0.00
	n	25	25	25	25	65	65	65	65
Yangon	Mean	4.03^{ay}	0.76^{ax}	0.66^{ax}	0.77^{ax}	1.49^{by}	0.57^{ay}	0.46^{ay}	0.29^{ay}
	SD	4.20	0.92	0.56	0.81	1.97	0.65	0.87	0.31
	Max	12.13	3.47	2.06	3.15	6.62	2.79	5.21	1.35
	Min	0.10	0.00	0.02	0.00	0.00	0.00	0.00	0.00
	n	16	16	16	16	49	49	49	49

Note: ^{a,b} and ^{x,y} significant difference ($p < 0.05$) of the same ion between study periods and stations, respectively.

Table 6 presents ion concentrations of study period (February–April) and normal period (May–January) of 2012–2014. Concentrations of nss-SO_4^{2-}, NO_3^-, NH_4^+ and nss-K^+ in µg/m³ at CM station in study period were 1.68, 0.62, 0.50 and 0.58, respectively. Meanwhile, the concentration (µg/m³) of those ions in YG were 4.03 (nss-SO_4^{2-}), 0.76 (NO_3^-), 0.77 (NH_4^+) and 0.66 (nss-K^+). The concentrations in study period were approximately 3 times higher than those in normal period for both stations.

According to result of ion concentration comparing and correlation result between PM_{10} and water-soluble ions, there were indicated that increasing of the ion concentrations during February–April should be influenced from open burning (forest fire and agricultural burning) and relate to the rising of air pollutant concentrations in CM.

3.3.2 Ion ratio comparison analysis

Flaming and fresh smoke includes large numbers of particles containing crystals of KCl (Li *et al*., 2003). Then they were converted to K_2SO_4 and KNO_3 through gas-particle reaction during transportation in the atmosphere (Freney *et al*., 2009) (Kong *et al*., 2015). Therefore, the concentration ratio of $\frac{nss-SO_4^{2-}}{nss-K^+}$ and $\frac{NO_3^-}{nss-K^+}$ between both EANET stations (CM and YG) were used in order to compare the aging of PM and indicate between near source and receptor area. Figure 8 shows the ratio of ions between CM and YG stations. The ratio of $\frac{nss-SO_4^{2-}}{nss-K^+}$ at YG station was around 2 times higher than that at CM station. Whereas, no significant difference of $\frac{NO_3^-}{nss-K^+}$ ratios between both stations was found. According to instability of NO_3^- form in PM, it is easily interchanged with HNO_3 form in aerosol due to temperature variation (Chuang *et al*., 2015). Therefore, NO_3^- concentration of both stations should not be related with the aging of PM. Based on the ratio values of SO_4^{2-} and K^+, aging of PM at YG station was older than that at CM station.

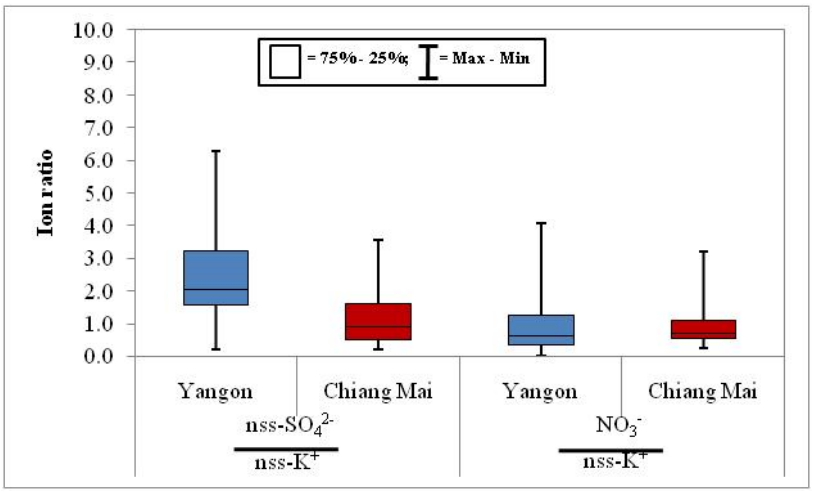
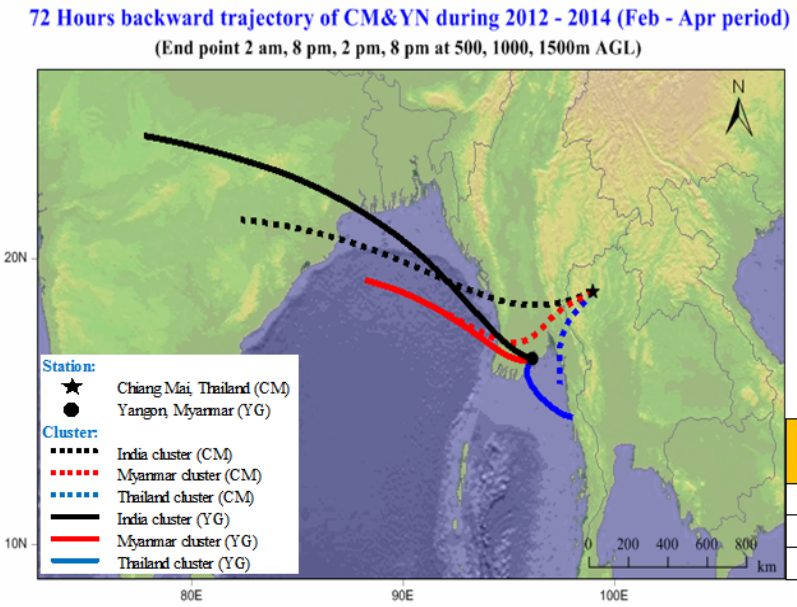


Figure 8. The ratios of $\frac{nss-SO_4^{2-}}{nss-K^+}$ and $\frac{NO_3^-}{nss-K^+}$ at Chiang Mai and Yangon stations during February–April in 2012–2014.

3.3.3 Air mass movement analysis

The 3-day backward trajectories were employed in order to indicate the origin of PM in YG due to air mass movement. Total of 3,216 trajectories were classified, in which three main paths of air masses, namely ID, MM and TH, were presented (Figure 9). Approximately 50 % of the air movement paths to YG were started in Bengal bay and passed over only a short distance of MM continental area (MM cluster) with a few open burning sources. Moreover, about 50% of air masses to YG, including Western of TH (39 %) and ID continent area (11 %), were originated from neighbor countries and those air masses transported across the sea before arrival YG site. Additionally, there was no air mass moving from middle MM continent in the north of YG site which was one large open burning source in Southeast Asia. Those 3 paths of air masses suggested that PM in YG were mainly transported across the Bengal and Andaman Sea and there were no additional of fresh PM originated along the air mass movement. Meanwhile, all of air movement patterns to CM were found passing over MM continental area, the PM should be mixing of

aged PM from long distance source and fresh PM originated from open burning in the continent along the air pathway to CM. Therefore, the aging of PM in YG was older than that in CM.



Cluster	Trajectories (%)	
	Chiang Mai	Yangon
Thailand	43.2%	38.7%
India	18.8%	11.1%
Myanmar	38.1%	50.1%

Figure 9. Cluster analysis of backward trajectories of Chiang Mai and Yangon during February to April (2012–2014) at 3 arrival attitudes and 4 synoptic times per day.

4. Conclusion

Open burning season (February to April) in the northern part of Southeast Asia was found to associate with high of pollutant level and bad air quality. This period of the year 2012–2014 was selected as the study period. Analysis of 3-day backward trajectories during study period indicated that biomass burning in the southwest direction of Chiang Mai as well as the north of India were potential sources of pollutants contributed in Chiang Mai Province. Moreover, long-range air movements were related to high level of pollutants in this area. The diurnal variation of air pollutants occur mainly because of number of biomass burning and meteorological factors changing within a day. Trajectory analysis suggested that a part of aerosols found at Chiang Mai station come from Myanmar continent, however the degree of particulate aging in Chiang Mai was fresher than that in Myanmar base on ions ratio comparison between both sites. This finding was related to the geography of the area and air movement pattern to monitoring stations. In Chiang Mai Province located in the middle of Southeast Asia continent, the PM had been mixed from aging and fresh PM along transportation to receptor site. Whereas, Yangon located at the Andaman coast, air masses with PM directly arrived from long distance source without additional of fresh PM from middle of Myanmar continent area was found. Therefore, this study revealed that Chiang Mai was not a receptor site receiving pollutants from Yangon.

Acknowledgements

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Joint Projects of the EANET with Participating Countries

Scientists from the EANET participating countries and the Network Center have been promoting joint research projects for various topics. Progress reports of the ongoing projects are introduced in this chapter.

Joint Research Project with Institute of Atmospheric Physics, Chinese Academy of Sciences: The model intercomparison study for Asia Phase III (MICS-Asia Phase III)

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Abstract

With the remarkable development of atmospheric model simulations, spatial and temporal variations of pollutant concentrations and depositions in East Asia have been depicted by atmospheric models. These outcomes are very effective for the regions where numbers of monitoring sites are limited in East Asia. However, there are inevitable uncertainties in the results of atmospheric models associated with emission inventories and modelling parameters. In order to have a common understanding of model performance and uncertainties in Asia, MICS-Asia Phase I project launched in 1998. Then, the subsequent activities of the Phase II and III has been organized by Asia Center for Air Pollution Research (ACAP) jointly with International Institute for Applied Systems Analysis (IIASA) and Institute of Atmospheric Physics, Chinese Academy of Sciences (CAS/IAP), respectively. The Phase III is composed of 3 main research topics, namely, (1) Multi scale model intercomparison for wet and dry depositions, temporal and spatial variations of ozone, $PM_{2.5}$ compositions and secondary organic aerosol, source contributions, (2) Development of the emission inventories commonly used in model intercomparison and intercomparison of emission inventories and (3) Model intercomparison for air quality/climate change interaction. The new scientific findings obtained by the MICS-Asia Phase III will be drafted in the special issues of an international journal.

Key words: $PM_{2.5}$, Ozone, Atmospheric deposition, Atmospheric modeling, Climate change

1. Introduction

Due to the remarkable improvement of computational technologies and development of community models for atmospheric chemistry, atmospheric numerical models are becoming common tools for the study of air quality. Recently, atmospheric model simulations are often used for the analysis of observed wet and dry deposition in the East Asia at the EANET (Acid Deposition Monitoring Network in East Asia) monitoring sites (Kuribayashi *et al.*, 2012) (Lu *et al.*, 2010) (Morino *et al.*, 2011). In addition, numerical models can provide both spatial and temporal variations of pollutant concentration and deposition in the model domain, which is very effective for the regions where numbers of monitoring sites are limited like wide area of East Asia (Task Force on Research Coordination of EANET. 2015).

However, there are inevitable uncertainties in the results of atmospheric models associated with emission inventories, boundary conditions, meteorological data, physical and chemical processes in the model, etc. Furthermore, even though the same community model (such as Community Multiscale Air Quality Model (CMAQ) and Weather Research and Forecasting Model coupled with Chemistry (WRF-Chem)) and input data is used, large variability often appears in the simulated results if settings of parameters and selections of modules in the model are different.

To take action for faced issues of atmospheric modeling, model inter-comparison studies are very effective approaches. We can obtain the information to improve the model reproducibility of observations and understand the cause of uncertainties. In addition, through the model inter-comparison studies, we can build the community of researchers of modeling and emission inventories, which is essential for the study of air quality in East Asia.

In order to have a common understanding of model performance and uncertainties in Asia, a model intercomparison study on long-range transport and deposition of sulfur, so called MICS-Asia Phase I, was carried out during the period from 1998 to 2000. Eight models participated in the Phase I study. The outcome of the model intercomparison exercise was discussed at the 3rd Workshop on the Transport of Air Pollutants in Asia, held at International Institute for Applied Systems Analysis (IIASA) in September 2000. There are several publications conducted by the Phase I study (Carmichael *et al.*, 2001) (Carmichael *et al.*, 2002) (Ichikawa *et al.*, 2001).

As it was concluded at the 4th Workshop on the Transport of Air Pollutants in Asia held at IIASA in October 2001, further model intercomparison study (MICS-Asia Phase II) would be useful to improve the understanding of the long-range transport of air pollutants in Asia. While the Phase I focused exclusively on sulfur compounds, it was recognized that a wider perspective could yield important insights including nitrogen compounds, ozone and aerosols to be critical for effective control of various environmental problems. According to the outline of the Phase II study, participants prepared necessary common data of boundary condition, emissions and meteorological field, and then carried out preliminary model simulations after the 5th Workshop. At the 6th Workshop held at IIASA in February 2004, the specifics of Phase II was discussed, taking into account the preliminary model intercomparison results reported by the participants. From the 7th (February 2005) to 9th (February 2007) Workshop, preparation of scientific papers of an overview of the MICS-Asia Phase II and some model intercomparision results regarding ozone, aerosols, deposition and relationship with global model had been discussed. The findings in the MICS-Asia

Phase II activity were published in the special issues of Atmospheric Environment in May 2008 (Carmichael & Ueda, 2008) (Carmichael et al., 2008).

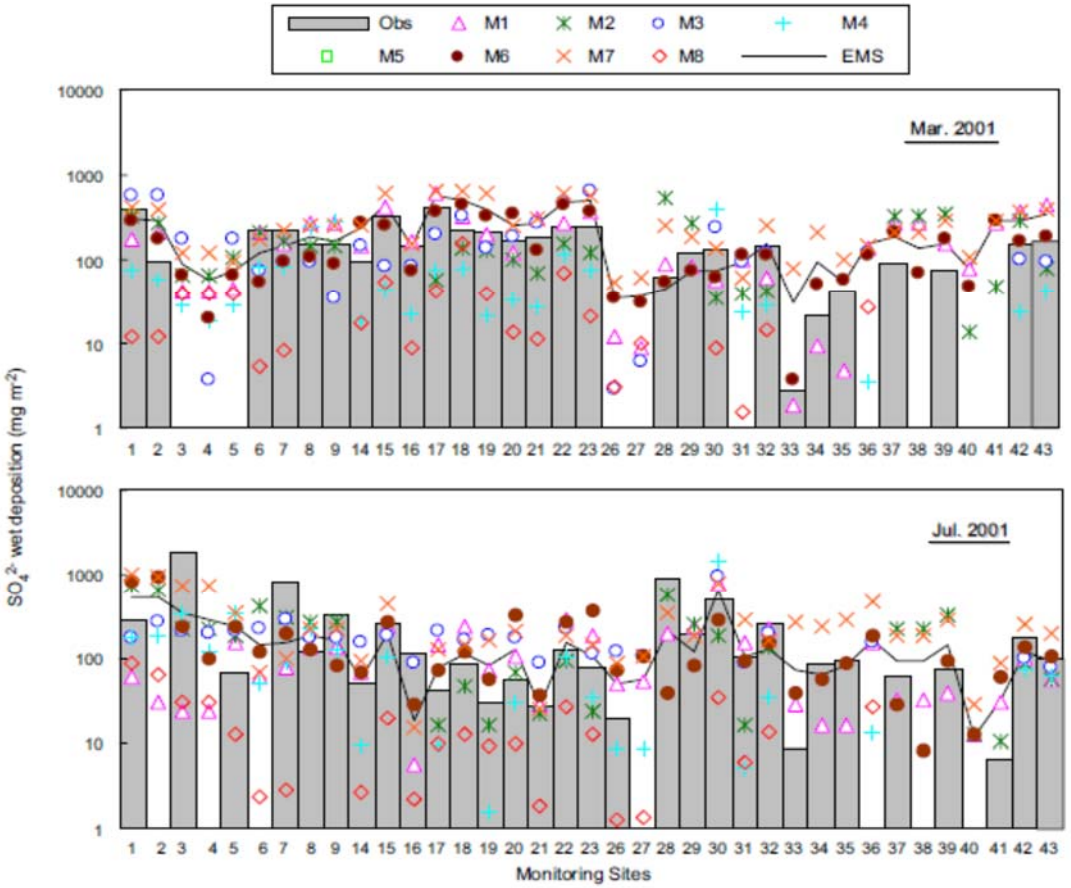

Figure 1. The predicted and observed monthly mean sulfate wet deposition amounts (as sulfate) at various EANET sites for March and July 2001. The solid line shows the ensemble mean predictions (EMS). The numbers of x-axis correspond to the EANET observation locations. Symbols mean simulated wet deposition amount by each models. (Carmichael et al., 2008).

Figure 1 shows the predicted and observed monthly mean sulfate wet deposition amounts at various EANET sites for March and July 2001. At many sites, the sulfate wet deposition amounts in March were higher than those in July because of seasonal wind and distribution of atmospheric pressure. The ensemble means reproduced these seasonality and regional distribution. The correlation coefficient between the ensemble means and measured values for the sulfate wet deposition amounts in March 2001 was 0.73 and that in July 2001 was 0.47.

In line with the conclusion in the 9th MICS-Asia Workshop, each participant had been proceeding to the complementary study for Phase II activities. From the 9th (February 2007) to 11th (February 2009) Workshops, the concrete contents to be conducted in the Phase III in accordance with the interest and progress of each participant was discussed. All participants gave presentations concerning their modeling works and interesting topics to be conducted in MICS-Asia as the Phase III project. It was highlighted that the Phase III should include multi-scale simulations, with sensitivity analyses for urban and regional scale emissions changes, source-receptor relationships for the regional scale and some activities that provide meaningful insights for the hemispheric

transport of pollution.

2. New joint research activities on modeling studies in Asia for EANET

MICS-Asia Phase III is co-organized by Asia Center for Air Pollution Research (ACAP), Japan and Institute of Atmospheric Physics, Chinese Academy of Sciences (CAS/IAP), China. It aimed for inviting more atmospheric modelling researcher and related atmospheric chemistry researches in East Asian countries and developing activity of atmospheric modelling. MICS-Asia Phase III project is one of EANET additional activities and a part of activity of Joint International Center on Air Quality Modeling Studies (JICAM), which was agreed to establish between ACAP and CAS/IAP in 2011.

At the beginning of the Phase III, the 1st International Workshop on Atmospheric Modeling Research in East Asia was held in Dalian, China in March 2010. Overview of recent modelling and monitoring studies in East Asia and the activity of the Phase III were considered in the 1st Workshop. At the 2nd Workshop on Atmospheric Modeling Research in East Asia in Sanya, China in December 2010, three research topics of the Phase III were adopted and the leader of each topic was identified as follows.

Topic 1: Multi scale model intercomparison (Leader: Zifa Wang, CAS/IAP, China)
Topic 2: Development of the emission inventories commonly used in model intercomparison and intercomparison of emission inventories (Leader: Jung-Hun Woo, Konkuk University, Republic of Korea)
Topic 3: Model intercomparison for Air Quality/Climate Change interaction (Leader: Gregory Carmichael, University of Iowa, USA)

At the 3rd Workshop on Atmospheric Modeling Research in East Asia in Chengdu in September 2011, all the participants had group discussion to materialize more concrete work plan for each of the three topics. Based on the discussion, the leaders prepared the draft Work Plan for MICS-Asia Phase III, and it was further discussed at the International Workshop on MICS-Asia III in Beijing in July 2012 to finalize the Work Plan.

3. Work Plan of MICS-Asia Phase III

After the 3rd Workshop, the Work Plan of MICS-Asia Phase III has been modified according to progress of each topics. This section introduces the current Work Plan for each research topic.

(Topic 1: Multi scale model intercomparison)

Topic 1 aims to evaluate strengths and weaknesses of current air quality models for air quality prediction, and provide techniques to reduce uncertainty and improve performance in Asia. The outcomes will provide answers the following key questions, including:

- ❖ Assessing the ability of models to reproduce pollutants concentrations under highly polluted conditions (e.g. Regional Haze);
- ❖ Quantifying uncertainties of each process (emissions, chemical mechanisms, transportations and depositions) and model resolutions (horizontal and vertical) on air

quality modeling. In particular, uncertainties in key boundary layer parameters, which have both direct and indirect impacts on modeling results through transporting meteorological fields and providing parameters in physical-chemical modules, respectively, need to be addressed;

- ❖ Investigating the air quality responses to specific emissions perturbations in a common case.

Topic 1 determined the following 3 domains and simulation periods. The 1st domain (D1:45 km mesh) covers Northeast, Southeast, and South Asia. All participating models should conduct full year simulation in 2010 for initial choice and 2008 and 2009 simulation for optional. These simulation data are analyzed for seasonal variation and inter-annual variability of wet deposition (S/N ratio, nitrogen species deposition, etc.) and atmospheric concentrations (O_3, $PM_{2.5}$, Black Carbon, etc.). It also focuses on severe pollution case in January 2013 and all models should consider influences of emission variation (e.g. heating period) and meteorological condition. The 2nd domain (D2: 15 km mesh) covers Northeast Asia covering Northeast and Southeast China, Korea, and Japan or Southeast Asia. The simulation period of D2 should be consistent with that for the 3rd domain. Third domain (D3: 5 km mesh) covers Beijing and surroundings or other megacities such as Tokyo, Seoul, Pearl River Delta depending on each participant's interest. The simulation periods of D3 are four episodes in 2010, and each episode should simulate for 10 days. It should also simulate severe pollution case in January 2013.

The participating models of topic 1 are global models of Chemical Atmospheric general circulation model for Study of atmospheric Environment and Radiative forcing (CHASER), Goddard Earth Observing System (GEOS)-Chem, etc. and regional models of CMAQ, WRF-Chem, Nested Air Quality Prediction Modeling System (NAQPMS), etc.

For the input data, all models should use the following common data:

- ❖ Gridded emission data include Asian anthropogenic emissions datasets developed by the Topic 2 group (Mosaic Asian Anthropogenic Emission Inventory for the MICS-Asia (MIX inventory)), global anthropogenic emissions from newest TF HTAP (Task Force on Hemispheric Transport of Air Pollutants) datasets which include MIX inventory, and natural emissions datasets of biogenic, biomass burning, dust, seasalt, volcano emissions, etc.
- ❖ As for meteorological fields, participants should use the same meteorological model (WRF) which will be prepared by CAS/IAP to drive their air quality models. The participants who have to use other meteorological fields are requested to compare them with the reference meteorological fields.
- ❖ As for boundary conditions, information on global model output for boundary conditions of D1 during 2008 and 2010 will be provided by CHASER, GEOS-Chem, and other global models by using the newest TF HTAP emission inventory.

The observational data used for validation of modelling results are provided by ACAP for EANET data from 2008 to 2010 and 2013 and by CAS/IAP for Chinese station data in 2008, 2010 and 2013. The vertical profile data is obtained by disclosed data of Light Detection and Ranging, Laser Imaging Detection and Ranging (LIDAR), ozone sounding and air craft measurements.

(Topic 2: Development of the common emission inventories and intercomparison of emission

inventories)

Topic 2 aims to evaluate strengths and weaknesses of current emission inventory and provide techniques to reduce uncertainty and improve performance in Asia. It also aims to develop the common emission inventories used in model intercomparison, which called MIX inventory. The detailed specification of MIX inventory are shown below. Figure 2 illustrates the framework of anthropogenic inventory for MICS-Asia Phase III.

- Anthropogenic emission inventories was prepared by integration of national inventories of China (Multi-resolution Emission Inventory for China (MEIC)), India, Japan (Japan Auto-Oil Program Emission Inventory-Data Base (JEI-DB)), Republic of Korea (Clean Air Policy Support System (CAPSS)), and other countries (Regional Emission inventory in ASia (REAS) version 2). Aircraft and international shipping emissions were also included to the anthropogenic inventories. Most elaborated and precise national activities data are used for this development.
- GFED (Global Fire Emissions Database) data are used for biomass burning emission inventory. The considered emission sectors are deforestation, savanna, forest, agricultural, and peat fires.
- MEGAN (Model of Emissions of Gases and Aerosols from Nature) data was used for biogenic emission inventory, and dust, seasalt, volcano, and naturally produced NO_x emission inventories by research data base were also used.
- For future emissions, growth factors from Greenhouse Gas - Air Pollution Interactions and Synergies (GAINS) for 2020 and 2030, which include projections from Chinese (Tsinghua and Energy Research Institute (ERI)) and Indian (The Energy and Resources Institute (TERI)) groups are used. Baseline and control scenario should be provided.

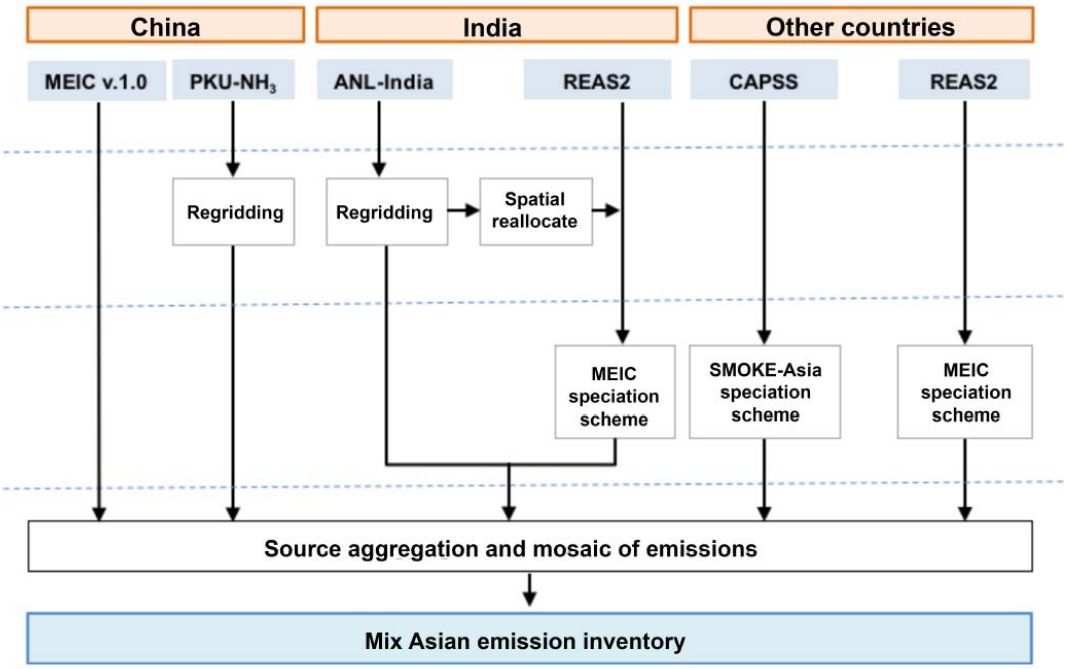

Figure 2. Schematic methodology for MIX emission inventory development.

MEIC: Multi-resolution Emission Inventory for China, PKU NH_3: NH_3 emission inventory by Peking University, ANL: Argonne National Laboratory, REAS: (Regional Emission inventory in Asia, SMOKE: Sparse Matrix Operator Kernel Emissions.

(Topic 3: Model intercomparison for Air Quality/Climate Change interaction)

Topic 3 aims to share best practices in Air Quality/Climate Change modeling in Asia and to evaluate strengths and weaknesses of current Air Quality/Climate Change models (leads to improvements in Air Quality models and predictions). Topic 3 will analyze Air Quality/Climate Change quantities such as air concentrations, depositions, Aerosol Optical Depth, etc. of Short-lived Climate Pollutants (SLCP) such as ozone and black carbon and other pollutants which also affect solar radiation such as sulfate and organic carbon aerosol.

The participating models of topic 3 are online-coupled models of WRF-Chem, WRF-CMAQ, etc. and offline models of CHASER, GEOS-Chem, CMAQ, Comprehensive Air Quality Model with Extensions (CAMx), Regional Air Quality Model 2 (RAQM2), etc. Because model domain was the same as Topic 1, the participating model of Topic 1 can also join Topic 3. The detailed activities of Topic 3 are shown as follows.

- Multi-model estimates for aerosol components such as black carbon, sulfate, organic carbon, dust, and nitrate and ozone by sector. Online-coupled models can provide on-line calculation of aerosol optical properties and radiative forcing, as well as coupled feedbacks to the climate/weather system. Offline models can provide simulation of aerosol and ozone fields and then be used to calculate aerosol optical properties and radiative forcing by common radiative transfer model. The final output will provide multi-model estimates of SLCP distributions & deposition for use in health, ecosystem & climate studies for base case in 2008 and 2010 and selected emission scenarios:
- Multi-model estimates of Direct Radiation Forcing at surface, atmosphere and top of atmosphere due to anthropogenic SLCPs by sector. Emission reduction scenario regarding the climate change perspective is also conducted. The final output will provide multi-model estimates of radiative forcing (direct + other elements) and regional responses for base case and selected emission scenarios.
- Sensitivity analysis of the impacts of model resolution, emissions, optical properties and chemical or physical mechanisms such as deposition, convection, cumulus scheme, aerosol microphysics will be conducted.

4. Current status and future works

After the work plan for each research topic was finalized, activity of Topic 2 was preceded by other topics because common emission inventory is essential for conducting model intercomparison. Development of MIX inventory had been conducted by researchers of Topic 2 and the results were published in an international journal (Li et al, 2017).

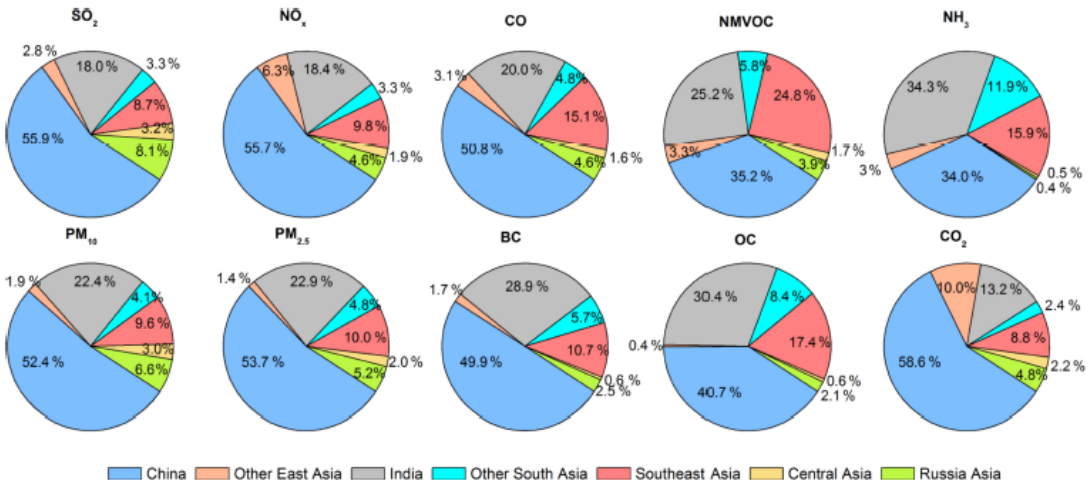

Figure 3. Emissions distributions by Asian regions in 2010 (Li et al, 2017).

Figure 3 shows emission distributions of anthropogenic pollutants by Asian regions in 2010. China is the largest contributor for most of anthropogenic pollutants, and its emission exceeded 50 % contributions of total Asian emission for SO_2, NO_x, CO, PM_{10}, $PM_{2.5}$, and CO_2. India is the largest contributor for NH_3 emissions (34 % of total Asian emission) and the 2nd largest contributor for all other species (13–30 %) followed by China. Southeast Asia and Other South Asia contribute more than 20 % to total NMVOC (Non-methane volatile organic compounds), NH_3, OC (Organic carbon), and CO emissions and around 10 % for other species, which is attributed to a high contribution from biofuel emissions. Contributions from other Asian regions are less than 10 % for all species. The gridded emission data will be disclosed after relevant researches of the Phase III are published.

After distribution of MIX inventory to participants of the Phase III, simulated results using common data of boundary condition, emissions and meteorological field, were submitted by 11 groups for model inter-comparison by the end of 2015. At the 7th Workshop on Atmospheric Modeling Research in East Asia in Chengdu in February 2016, preliminary results of model inter-comparison for temporal and spatial variations of ozone, aerosols and their precursors, wet and dry depositions, and relationship between air pollution and climates were reported. Persons in charge for leading analysis were also assigned for various topics such as wet and dry deposition, ozone, source-receptor relationship, $PM_{2.5}$ and secondary organic aerosols, emission inventories, and inter-actions between air pollution and climate change. Then, they had been implemented further analysis of the submitted results.

In November 2016, discussion workshop for promoting preparation of papers for the special issues of MICS-Asia III was held in Beijing, China. It was decided that the special issues for MICS-Asia III will be drafted in an academic journal of Atmospheric Chemistry and Physics. At this discussion workshop, progress of the analysis of model intercomparison results for each topic such as wet and dry depositions, temporal and spatial variations of ozone, $PM_{2.5}$ compositions and secondary organic aerosol (SOA), source receptor analysis and air quality and climate change were reported and discussed. Then, the future plan for preparation of the papers for special issue of MICS-Asia III were discussed. For the next step, this work shop also discussed new simulations in 2013 with fine spatial resolutions for northeast China.

It has passed 20 years since MICS-Asia Phase I launched. The 8th International Workshop

on Atmospheric Modeling Research in East Asia is decided to hold at IIASA, Austria in March 2017. At the workshop, the results of further analysis after the discussion workshop will be reported and detailed contents of scientific papers will be discussed. It will also hold an event to celebrate the 20th year anniversary of MICS-Asia project.

Acknowledgements

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Joint Research Project with Asian Institute of Technology, Thai Pollution Control Department and JICA Research Institute: Characteristics of ionic and carbonaceous components in precipitation and particulate matter in Japan and Thailand

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Abstract

Bangkok, one of the mega cities in South East Asia, has been suffered serious air pollution issues due to rapid economic growth, urbanization, and motorization. In order to design appropriate air pollution countermeasures, information on major contributing sources of air pollutants such as PM and O_3 are required. The Thai Pollution Control Department (PCD) continuously monitors PM and gases and ionic components of total suspended particle and precipitation. However, there are lack of other component data of PM and precipitation, which are necessary to analyze source apportionment of PM. This study has implemented monitoring of chemical components of $PM_{2.5}$, coarse particles (> 2.5 μm) and precipitation at the selected sites in Bangkok since September 2015. By using these monitoring data, temporal variations of chemical components of $PM_{2.5}$, coarse particles and precipitation will be clarified. These observation data will be used for validation of atmospheric model simulation in Bangkok and for socioeconomic analysis of $PM_{2.5}$ and O_3 by using model results and input on air pollution mitigation measures in Bangkok Metropolitan Area.

Key words: $PM_{2.5}$, Ozone, Wet deposition, Source apportionment, Atmospheric modeling

1. Introduction

Air pollution has become a serious issue in many urban cities of East Asian countries due to rapid economic growth, urbanization, and motorization. Bangkok, one of the mega cities in South East Asia, also has been suffered serious air pollution issues. PM_{10} and $PM_{2.5}$ are the most important air pollutant in both urban and rural areas, the concentration levels are as high as or higher than those in North America and European countries (Guo *et al.*, 2014). The maximum concentrations of daily average of PM_{10} and $PM_{2.5}$ in Bangkok are 188 and 101 μg/m^3 in 2015. The concentration of PM_{10} exceeded the standard in 27 out of 29 provinces and the concentration of $PM_{2.5}$ exceeded the standard in 9 out of 10 provinces of Thailand in 2015 (Pollution Control Department. 2015). The major sources of particulate matter in Bangkok are transportation sectors and industrial sectors.

Bangkok is also suffered high ozone (O_3) pollution because of large emission of air pollution and year-round active photooxidation. The level of ozone in all 25 provinces exceeded the standard in 2015. The maximum 1 hour average of O_3 was 125 ppb, which was a 4 % decrease from the level detected in 2014. The maximum 8 hour average was 97 ppb, which decrease 5 % from the previous year. In addition, the city also has been facing problem with acid deposition where acidity of rain water was reported to increase (EANET. 2015).

Considering aforementioned air pollution situation, mitigation measures to reduce PM and O_3 pollution in the city are urgently required. However, in order to design appropriate policies, information on major contributing sources of PM and O_3 are required. The Thai Pollution Control Department (PCD) continuously monitors PM_{10}, $PM_{2.5}$, O_3, SO_2, NOx, CO, VOC, and ionic components of total suspended particle and precipitation. However, there are lack of data of ionic components of $PM_{2.5}$ and carbonaceous components in PM and precipitation, which are necessary to analyze source apportionment of $PM_{2.5}$ and removal processes of PM. Moreover, the wet and dry deposition fluxes of acidic and carbonaceous substances can be used to assess potential impacts on the ecosystem and climate change.

Considering the current situation in Bangkok, we determined the main objectives of the project as shown below:

(1) Monitor chemical components of $PM_{2.5}$, coarse particles (> 2.5 μm) and precipitation at the selected sites in Bangkok over a year.
(2) Clarification of temporal variations of chemical components of $PM_{2.5}$, coarse particles and precipitation at selected sites in Bangkok.
(3) Model development and simulations (a receptor model and/or a chemical transport model) to identify the sources of $PM_{2.5}$ and O_3 in Bangkok.
(4) Socioeconomic analysis of $PM_{2.5}$ and O_3 by using model results and input on air pollution mitigation measures in Bangkok Metropolitan Area.

Since 2014, the collaborative research project of “A Study on Urban Air Pollution Improvement in Asia” has been conducted by Asia Center for Air Pollution Research (ACAP), the Asian Institute of Technology (AIT) and Japan International Cooperation Agency Research Institute (JICA-RI). The project has been also supported by Thai national research counterparts including the PCD, Environmental Research and Training Center (ERTC), King Mongkut’s University of Technology Thonburi (KMUTT) and Ladkrabang (KMITL). In this article, the overview of atmospheric monitoring and primitive results are presented.

2. Monitoring methods

2.1 Sampling site

Long term monitoring was conducted at the two sites which represent urban and suburb area of Bangkok. One is located at the rooftop of the PCD in urban area of Bangkok, and the other is at the rooftop of the ambient laboratory of AIT in Pathumtani, suburb area of Bangkok. The location map of both sites is shown in Figure 1. The detailed explanation of sampling sites is as follows.

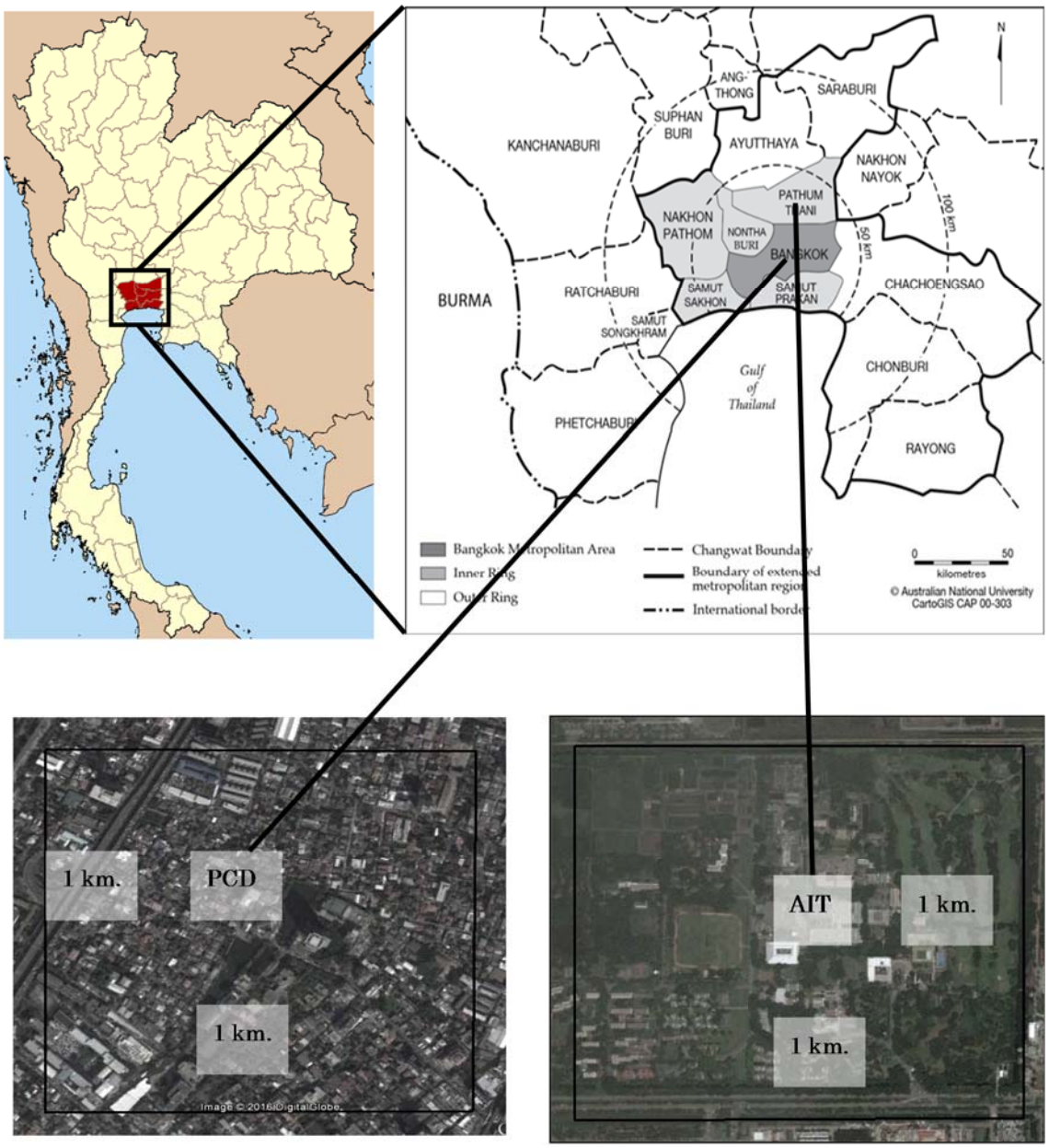

Figure 1. Location of monitoring sites at PCD and AIT.

The rooftop of PCD building is located at 13.8º N and Longitude 100.5º E and 64 m high above the ground. The building is mainly surrounded by houses, commercial places, and institutions within a radius of 5 km. It is approximately located of 0.75 km away from the main road (Paholyothin, Rd) which has heavy traffic congestion during rush hours. Sky train line is located above this road.

The ambient laboratory at AIT is located at 14.1º N and 100.6º E and 6 m high above the ground. This site is surrounded by many canals, rice paddies and other crops fields, as well as some small and medium industries. A mixed industrial estate is located about 8 km to the North (Navanakhon Industrial Estate) and the other was about 6 km to the South (Thai industry). AIT is located approximately 500 m away from the main road (Paholyothin, Rd) and is about 40 km from the Bangkok city center. It is situated at the upwind of the Bangkok city during the dry season.

2.2 Measurement of gases and particulate matter components

The five-stage and two-stage filter pack air samplers were used to collect weekly ambient air samples, i.e., coarse particles (> 2.5 μm), fine particles ($PM_{2.5}$) and gaseous substances. Air samples were collected on various types of filter: quartz filter (FC) for the coarse PM, Teflon filter (F0) for the $PM_{2.5}$, Polyamide filter (F1) for HNO_3, SO_2, HCl and NH_3, Acid impregnated filter (F2) for SO_2 and HCl, and Alkaline impregnated filter (F3) for NH_3 in the five-stage filter pack. This sampling method is basically followed by EANET protocol (EANET. 2013) but a $PM_{2.5}$ cascade impacter and a mass flow controller (flow rate: 2 L/min) were installed in the system. The schematic picture of the five-stage filter pack is shown in Figure 2 and the pictures of PCD and AIT site equipment are shown in Figure 3.

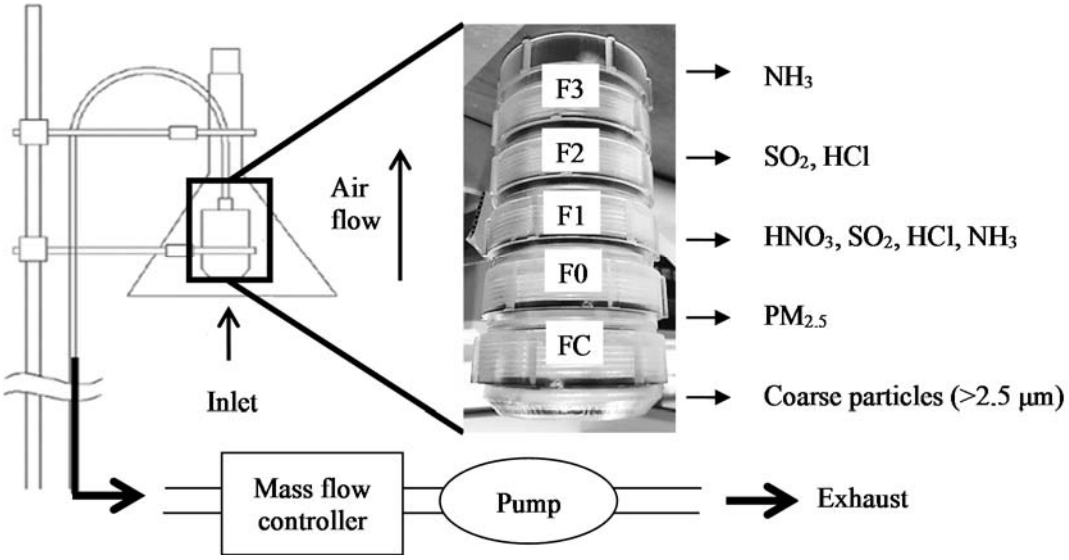

Figure 2. Schematic diagram of the five-stage filter pack.

Figure 3. The pictures of AIT (left) and PCD (right) site equipment.

Filters from the two-stage filter pack are used for EC/OC analysis. Weekly sampling is conducted from September 2015 to February 2016 in original plan. Before sampling, filters for mass were placed into petri dish and then keep at 22 ± 2 ˚C and 40 ± 5 % in desiccator for 24 hours in microbalance room before and after weighing. The filter holder was sealed into a polyethylene bag and furthers an aluminum-coated bag for avoiding the contamination and sunlight. The sealed mounted holder was kept in a transportation box at approximately 10 ˚C by using ice gel during shipping to a monitoring site to avoid evaporation of the substances during transportation. Shipping interval of the filter packs from the laboratory to the site was weekly and the samples were kept in refrigerator until analysis. When some samples are shipped Japan periodically, the sample were kept low temperature by coolant.

The filters were used to analyze for mass concentrations, ion components (SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, Na^+, K^+, Ca^{2+}, Mg^{2+}) and carbonaceous components (Organic carbon (OC), Elementary carbon (EC) and Black carbon (BC)). Ionic components were determined by Ion Chromatography (Dionex ICS-1500). OC and EC were determined by the Interagency Monitoring of Protected Visual Environments (IMPROVE) thermal/optical reflectance (TOR) protocol using a Desert Research Institute (DRI) model 2001 carbon analyzer, and BC is determined by Dual-Wavelength Optical Transmissometer Data Acquisition System (Magee Scientific, Model OT21). Weekly concentration of each composition in coarse particle components and $PM_{2.5}$ fractions were determined, respectively.

2.3 Measurement of atmospheric deposition

Wet deposition samples were collected by a wet-only sampler (Ogasawara Keiki Co. Ltd., US330) which is customized for sampling carbonaceous components. To avoid the adhesion of particles on the wall of the collecting funnel as much as possible, a special glass collecting funnel and filter unit was designed and installed in the wet-only sampler (Figure. 4). In the wet-only sampler, a quartz-fiber filter unit was installed at the bottom of the collecting funnel using a clamp. Collecting tank in the refrigerator was connected to the filter unit using a tube. The funnel was covered with a mesh sieve to prevent contamination from vegetation and insect debris. The wet-only sampler was equipped with a rain sensor that released the opening mechanism at the first sign of precipitation and automatically closed the lid after the rain stopped. The detailed description of OC and EC measurement is shown in our previous study (Huo *et al.*, 2016).

For the measurement of dry deposition, the five-stage filter pack method was used as

explained in the previous section. After sampling, the filters of FC, F0, F1, F2 and F3 were extracted by solvent and were shaken for 1 hour using automatic shaker followed by the Technical Manual on Air Concentration Monitoring in East Asia. The chemical analysis was carried out after the extraction for each 2 weeks.

The concentrations of components in rainwater were calculated using the equation provided in the Technical Manual on wet deposition monitoring (EANET. 2010a). The wet deposition fluxes were determined using the following equation:

$$\text{Wet deposition flux } (\mu eq/m^2/week) = (C_{Ai} \times V) / A \quad \text{(Eq. 1)}$$

[C_{Ai}: ion concentration (μeq/L), V: Rain volume (L), A: Area of glass funnels (m^2)]

The dry deposition fluxes were determined by the Inferential method. The methodology is described in the Technical Manual on Dry Deposition Flux Estimation in East Asia in detail (EANET. 2010b). A deposition flux was calculated from the air concentration and deposition velocity shown in the following equation:

$$F_i = V_{di} \times C_i \quad \text{(Eq. 2)}$$

[F_i: flux of i species (nmol/m^2/week), C_i: concentration of i species (nmol/m^3), V_{di}: deposition velocity of i species (m/s)]

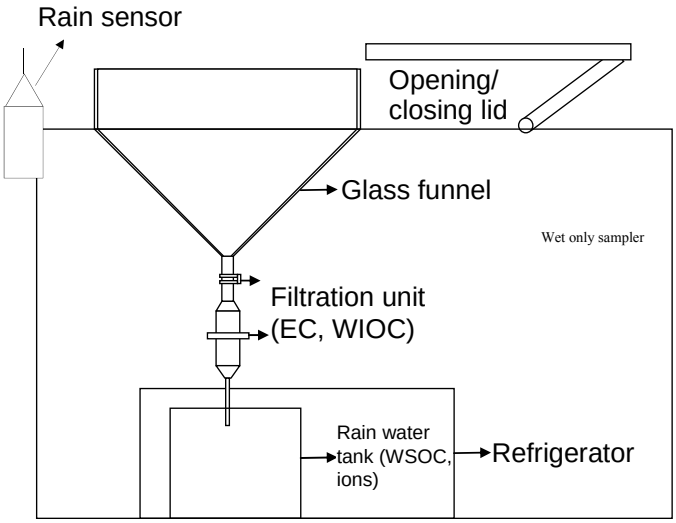

Figure 4. Schematic diagram of the wet only sampler.

For meteorological data to calculate dry deposition velocity, wind speed (m/s), temperature (ºC), precipitation (mm), relative humidity (%), cloud coverage and solar radiation (W/m^2) data were collected from Don Mueang airport standard meteorological station (located within a distance of 15 km from PCD) and Pathumthani agrometeorological station (located within a distance of 4 km from AIT). Solar radiation data was taken from energy laboratory at AIT. The meteorological data was 1-hour data for September to October 2015 and 3-hour data for other months. Therefore, the hourly data was prepared using interpolation of measured values. The land coverage in 1 km around the sampling site using google map to identify the fraction of different land use types such as tree cover, grass, water, and building surfaces.

3. Primitive results

3.1 PM mass concentrations

Figures 5 and 6 show the time variations of weekly average mass concentrations at the PCD and AIT sites for the wet period from 1 September to 8 November 2015, and dry period from 9 November 2015 to 29 February 2016, respectively. At PCD site, the average concentrations of $PM_{2.5}$ and coarse particles in the wet period were 24 ± 4 μg/m^3 and 59 ± 21 μg/m^3, while these in dry period were 31 ± 11 μg/m^3 and 53 ± 14 μg/m^3. The highest concentration of coarse particles was 82 μg/m^3 in the week of 9 to 16 November 2015, and that of $PM_{2.5}$ was 50 μg/m^3 in the week of 22 to 29 February 2016.

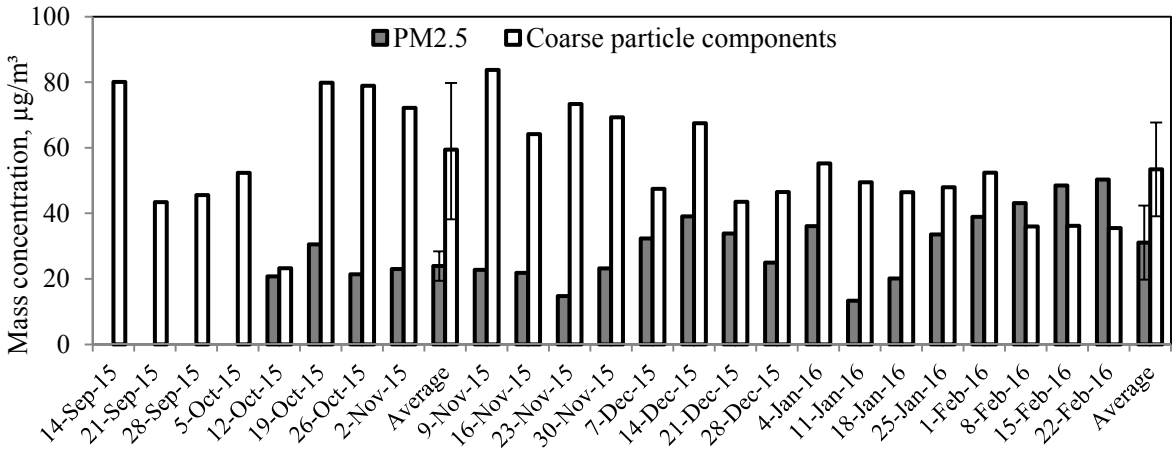

Figure 5. Time variations of weekly PM mass concentration at PCD.

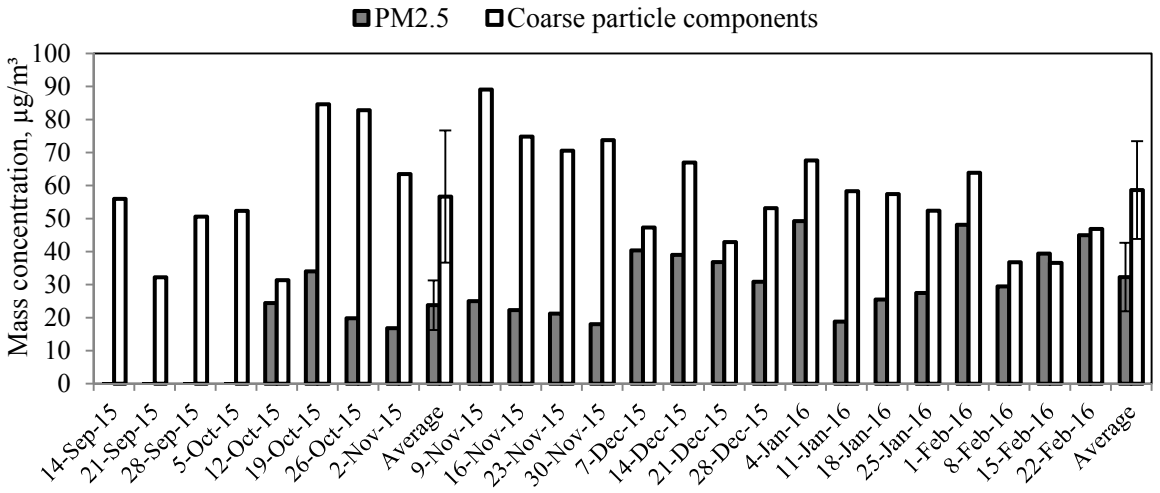

Figure 6. Time variations of weekly PM mass concentration at AIT.

At the AIT site, the overall average mass concentrations of $PM_{2.5}$ and coarse particles in the wet period were 23 ± 7 μg/m^3 and 56 ± 20 μg/m^3, respectively. The averages in the dry period were 32 ± 10 μg/m^3 and 56 ± 15 μg/m^3. The highest weekly levels of $PM_{2.5}$ was observed in the week of 4 to 11 January 2016 of 49 μg/m^3, while the highest values of coarse particles was 88 μg/m^3 in the week of 9 to 16 November 2015. The minimum level of $PM_{2.5}$ and coarse particles were 17

$\mu g/m^3$ (2 to 9 November 2015) and was 31 $\mu g/m^3$ (12 to 19 October 2015). Many activities took place around AIT campus including festivals, agriculture rice straw field burning, trash burning that cause the fluctuation of the PM mass concentrations. Comparing the PM levels at both sites, coarse particles at AIT was lower than PCD in the wet season, but was slightly higher during the dry period, while the $PM_{2.5}$ at AIT and PCD were in similar range which may suggest the large influence of the regional transport and formation processes of fine PM.

3.2 Carbonaceous component concentrations in PM

The average EC and OC concentrations in PM at the AIT and PCD sites are shown in Figure 7. The average EC and OC concentrations of AIT in $PM_{2.5}$ were 4.56 ± 1.60 $\mu g/m^3$ and 6.70 ± 3.88 $\mu g/m^3$, respectively, while those in concentration of coarse particles were 1.49 ± 0.33 $\mu g/m^3$ and 2.45 ± 0.45 $\mu g/m^3$, respectively. The EC and OC concentrations in $PM_{2.5}$ at the PCD site were 3.49 ± 1.24 $\mu g/m^3$ and 5.17 ± 3.04 $\mu g/m^3$, respectively, and those in coarse particles at PCD were 1.31 ± 0.34 $\mu g/m^3$ and 2.23 ± 0.59 $\mu g/m^3$, respectively.

The ratio of EC to total carbon (TC, EC + OC) can be used to indicate the both sources of combustion and also the potential of wet removal. The ratio of EC to TC in $PM_{2.5}$ at AIT site in dry period was approximately 0.4, while in wet period was 0.5. The higher EC/TC ratio likely indicate the diesel vehicle exhaust while lower EC/TC may indicate open burning of biomass burning or solid waste burning that frequently occurred in the dry season around the AIT site. However, in the wet season part of water-soluble organic carbon (WSOC) may be washed out that changes the EC/TC ratio for PM. Therefore, in future studies the data of WSOC in wet deposition should also be considered.

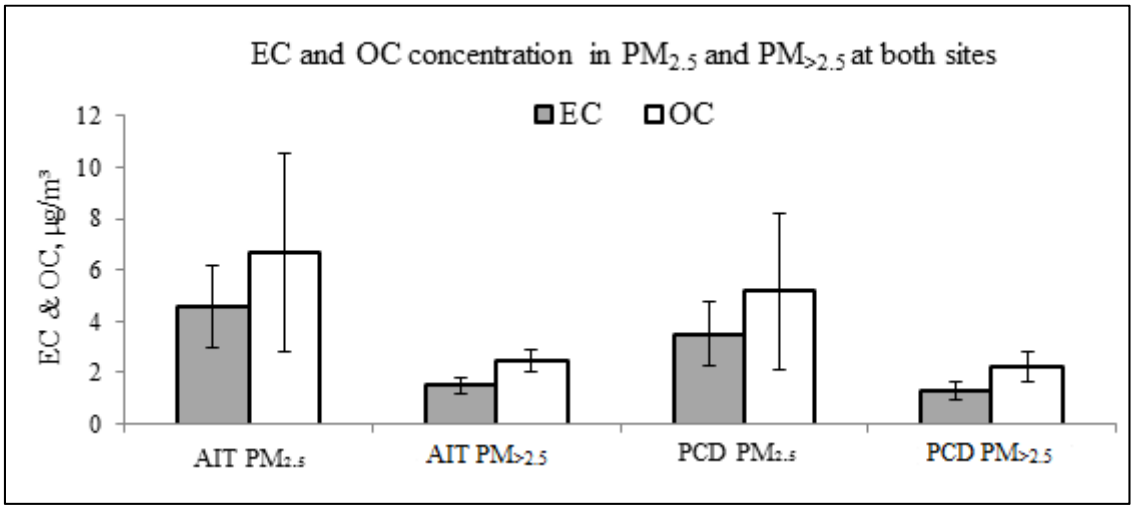

Figure 7. Average EC and OC concentrations in PM at the AIT and PCD sites (September 2015 to February 2016).

3.3 Ionic component concentrations in PM

Figures 7 and 8 show the average concentrations of ionic component in PM in wet and dry period the AIT and PCD sites, respectively. At AIT site, the average ionic concentration for the wet and dry period of $PM_{2.5}$ was observed the largest for SO_4^{2-}, 2.48 $\mu g/m^3$ in wet period and 3.08 $\mu g/m^3$ in dry period, while PO_4^{3-} was the least of 0.01 $\mu g/m^3$ in both seasons. NH_4^+ was the major cation that contributed the highest in wet season of 1.04 $\mu g/m^3$ and in dry period of 1.38 $\mu g/m^3$. The

average ion concentration in coarse particle was the largest for NO_3^- of 1.35 µg/m^3 in wet period and 2.99 µg/m^3 in dry period. Ca^{2+} was the major cation in coarse particle, and the average concentrations were 1.24 µg/m^3 in wet period and 2.20 µg/m^3 in dry period.

At the PCD site, the average ion concentration in $PM_{2.5}$ was the largest for SO_4^{2-} which was 3.08 µg/m^3 in wet period and 3.93 µg/m^3 in dry period. PO_4^{3-} contributed the least of 0.01 µg/m^3 in wet period and 0.03 µg/m^3 for $PM_{2.5}$ in dry period. NH_4^+ largely contributed in both periods, and the average were 1.08 µg/m^3 in wet and 1.42 µg/m^3 in dry period. The average ionic concentrations in coarse particle and was the largest for NO_3^- of 1.58 µg/m^3 in wet period and 3.27 µg/m^3 in dry period. Ca^{2+} was the highest cation in coarse particle with the level of 1.09 µg/m^3 in wet period and 1.57 µg/m^3 in dry period.

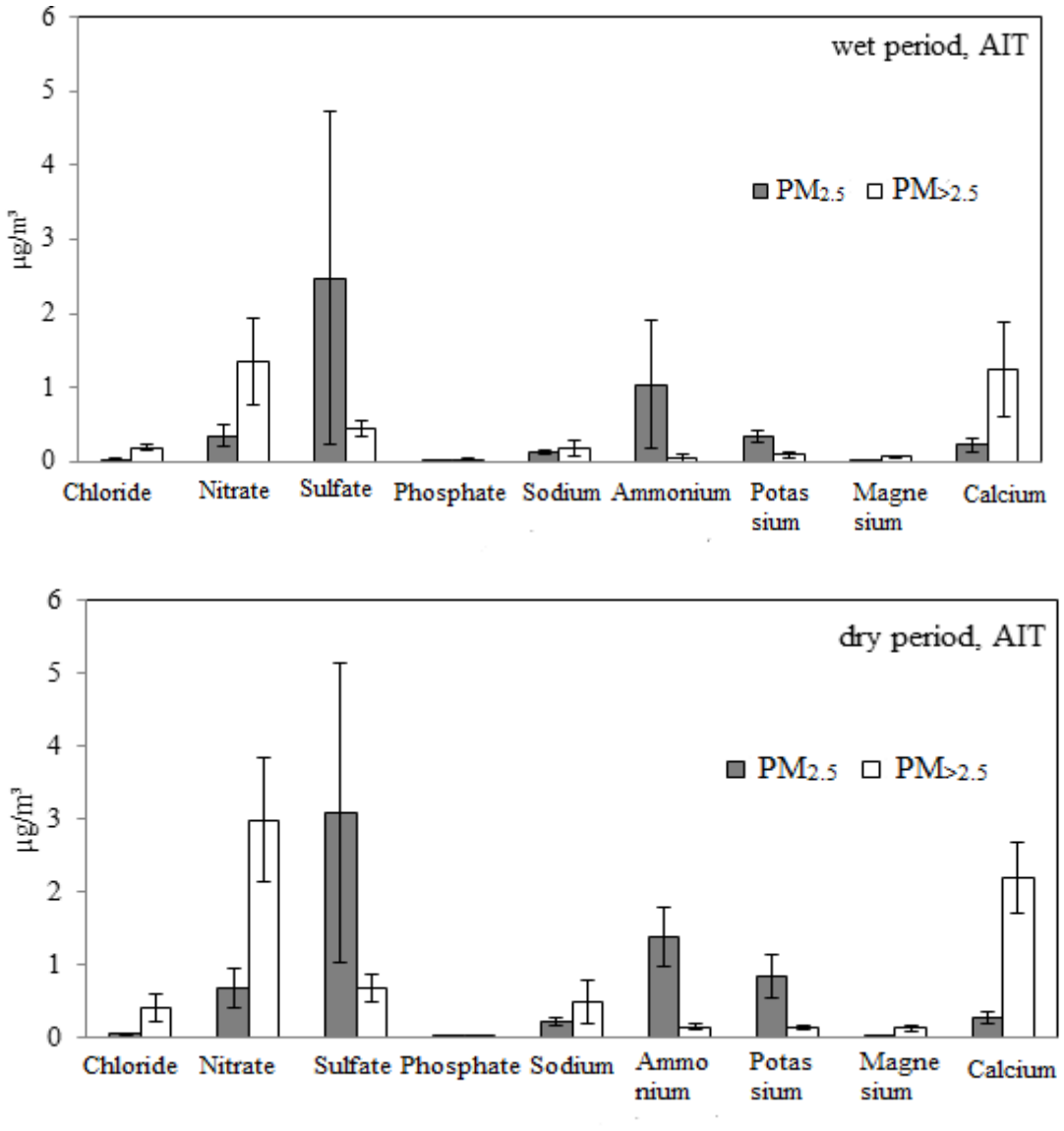

Figure 8. Average concentrations of ionic component in PM in wet (September 2015 to November 2015) and dry period (November 2015 to February 2016) at the AIT site.

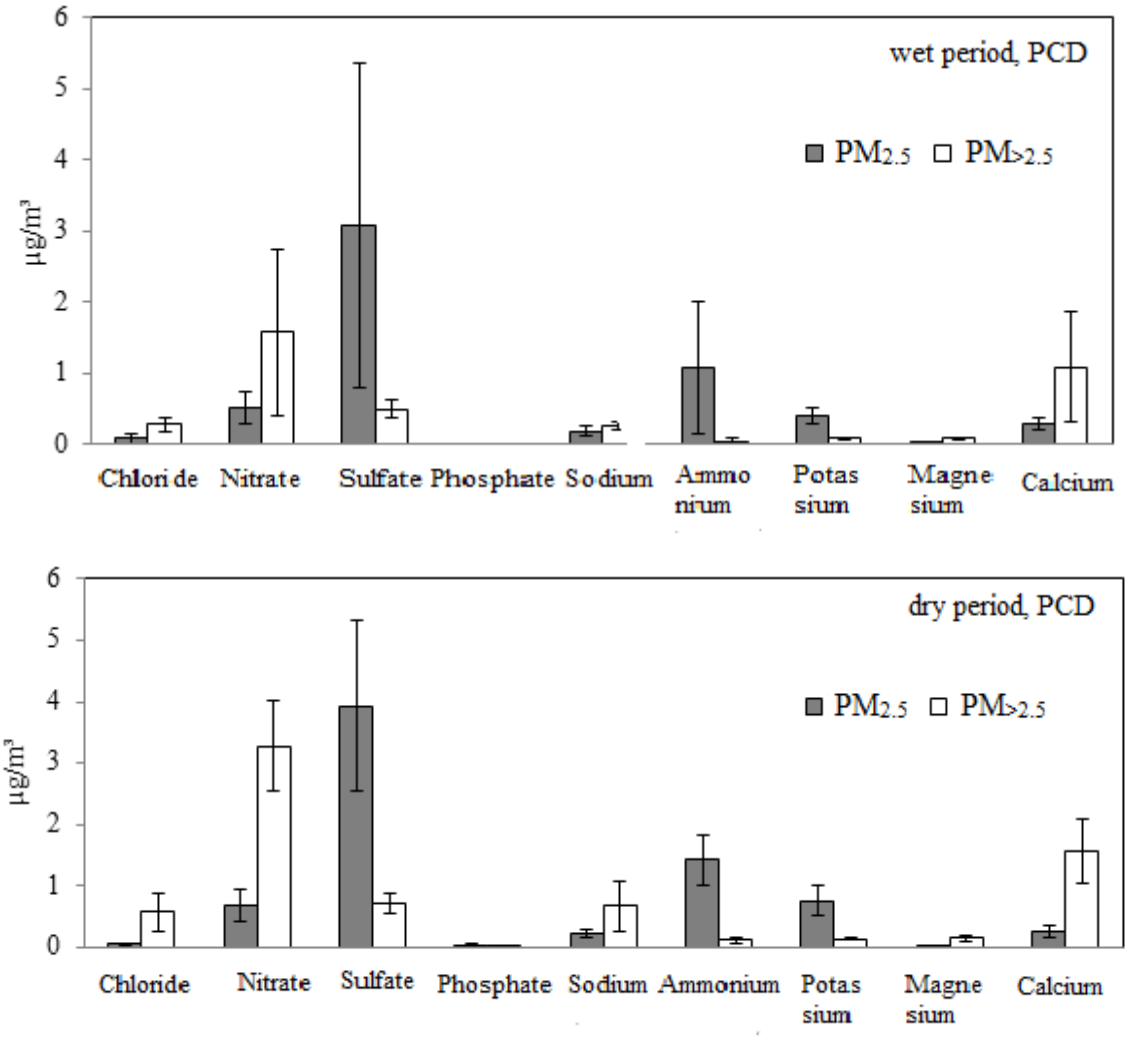

Figure 9. Average concentrations of ionic component in PM in wet (September 2015 to November 2015) and dry period (November 2015 to February 2016) at the PCD site.

4. Future works

The monitoring of $PM_{2.5}$, coarse particles and precipitation in Bangkok is to be implemented in February 2017 because it is necessary to characterize atmospheric composition in wet season and dry season. After the scheduled monitoring, receptor models of Chemical Mass Balance (CMB) and Positive Matrix Factorization (PMF) will be applied by using all PM monitoring data of the project period (September 2015 to February 2017). From these outcomes, the significant emission sources of PM in Bangkok Metropolitan Area may be identified, which provides important information for development of sector classified emission inventory. Temporal variations of wet and dry depositions for ionic and carbonaceous components are also evaluated by using previously described methodology. This results is important for clarification of atmospheric removal process in atmospheric modelling study.

In the next step, emission inventory of gaseous and particulate substances in Bangkok Metropolitan Area in 2015 will be developed. The existing emission inventory will be updated by

incorporating additional emission sectors of mobile source, painting, ship, oil tank, farm, livestock and fertilizer, and so on. By using updated emission inventory, the modeling simulation in Bangkok Metropolitan Area from 2015 to 2016 will be conducted by using chemical transport model (CTM) of WRF-CAMx. The observation data will be used for validation of modelling results. As consequence of model analysis, the temporal spatial distribution of PM and other air pollutants will be obtained. This output will be quite important for air pollution mitigation in Bangkok and other mega cities in South East Asia.

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Progress report on Joint Research Project with Japan, Thailand and Malaysia on catchment analysis

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Abstract

Effect of sulfur (S) deposition on terrestrial ecosystems is still one of the important issues to be investigated in East Asia. Based on a scientific community of Acid Deposition Monitoring Network in East Asia (EANET), dynamics of S derived from atmospheric deposition are investigated in forest catchments in Niigata, Japan, Nakhon Ratchasima, Thailand, and Sabah and Sarawak, Malaysia. To clarify S dynamics in the forest ecosystems, analysis of S isotopic ratio is applied for rainwater, soil water and stream water in addition to measurement of the fluxes. Moreover, the accumulated data in two historical sites in Japan and Thailand showed different reactions of the ecosystems to reduction of the atmospheric S input. The knowledge obtained by the project will be informative for discussion on a possible impact of S deposition in the region.

Key words: Forest catchment, Sulphur isotope, Japanese cedar, Dry evergreen forest, Tropical rainforest, Rehabilitated forest

1. Introduction

Atmospheric deposition of sulfur (S) compounds may gradually decrease according to the recent emission inventories in East Asia (e.g. Lu *et al.,* 2010). However, the S deposition level is still high and cumulative load of S is quite large in the EANET region. Since S deposited on ecosystems may be retained in soil and/or cycled in the soil-plant system, manifestation of its effect may be delayed (e.g. Mitchell and Likens 2011; Kobayashi *et al.*, 2012). Moreover, several rivers/lakes for monitoring on inland aquatic environment in the East Asian countries showed

pH-declining trend with SO_4^{2-}-increasing trend (EANET. 2011). Effect of S deposition on terrestrial ecosystems is one of the important issues to be investigated in East Asia.

Scientists from the Network Center (NC) and the EANET countries have been promoting the catchment-scale analysis in different types of forests, namely in Kajikawa site, Niigata, Japan, in Sakaerat site, Nakhon Ratchasima, Thailand and in Danum Valley site, Sabah, Malaysia. Taking account of the background above, dynamics of S derive atmospheric deposition was studied in these catchment sites. The research team successfully obtained the grant for the study from the international research fund, Asia Pacific Network on Global Change Research (APN) from 2012 to 2015 (ARCP2013-13CMY-Sase). In this report, we will introduce recent progress on the catchment projects in Japan, Thailand and Malaysia, based on the final project report for the APN Project (Sase *et al.,* 2015).

2. Method

2.1 Site description

The study sites were established in four forest catchments in Japan, Thailand and Malaysia, as shown in Table 1. Fluxes of ions including SO_4^{2-} had been studied by previous projects since 2002, 2005 and 2008 in Kajikawa, Sakaerat and Danum Valley sites, respectively. However, the surveys in these sites were mostly finished in 2010/2011. In 2012, the study sites were reactivated for the APN project and the rehabilitated forest in Bintulu was added as a new site for the APN project.

Table 1. Study forest catchments in Japan, Thailand and Malaysia.

Site	Kajikawa	Sakaerat	Danum Valley	Bintulu
Country	Niigata, Japan	Nakhon Ratchasima, Thailand	Sabah, Malaysia	Sarawak, Malaysia
Forest type	Japanese cedar	Dry evergreen forest (DEF)	Tropical rainforest	Rehabilitated Forest
Start year	2002	2005	2008	2012

2.2 Project outline

The surveys were conducted in the existing three catchments above and also in the Bintulu Rehabilitated Forest in Sarawak, Malaysia. In order to determine S dynamics in the forest ecosystems, analysis of S isotopic ratio is applied for rainwater, soil water and stream water in addition to measurement of the fluxes. The data obtained by the project will explain the possible impacts of S deposition on the forests. Since nitrogen (N) deposition is also quite high in the region, its relation to acidification/eutrophication should also be discussed. The fluxes of S and N by rainfall outside forest canopy (RF), throughfall (TF) (+ stemflow, SF), soil solution (SS), and stream water (SW) have been measured in Kajikawa, Sakaerat and Danum Valley sites. In the case of Bintulu site, the fluxes from SW may not be able to be estimated, since the stream is flowed not continuously but just seasonally. The S isotopic ratio was measured for SO_4^{2-} in these water samples. In Sakaerat, Danum Valley, and Bintulu, the ion-exchange resin samplers were also applied for these purposes. Detailed methods for measurement of the fluxes and S isotopic ratio were described in the EANET Science Bulletin Volume 3 (Sase *et al*., 2013).

3. Progress of the project

3.1 Major outcomes from the APN project

Based on the field surveys and data analysis, characteristics of S dynamics have been clarified in the four study forests (Sase *et al*., 2015).

- S loads from atmosphere: The S deposition amount was larger in the following order, Kajikawa (29 kg S ha^{-1} $year^{-1}$) > Bintulu (19) >> Sakaerat (5.8) > Danum Valley (3.6).
- SO_4^{2-} concentration in stream water (SW): The SW concentration was higher in the following order, Kajikawa (101 μmol_c L^{-1}) > Bintulu (73) > Danum Valley (39) >> Sakaerat (6.3).
- The SW concentration in Sakaerat site was significant low compared to that of the deposition (41.9 μmol_c L^{-1}, as the weighted-mean of TF+SF), suggesting retention of S in the forest ecosystems.
- S isotopic analysis of SW suggested various possible sources of S in these forest ecosystems. Not only atmospheric deposition but also geology and biological fractionation should carefully be considered as possible S sources in SW.

The results above are introduced also in the Third Periodic Report of the State of Acid Deposition in East Asia (PRSAD3) with some more data. For the detailed discussion, the final project report for the APN project (Sase *et al*., 2015) should be referred. As a scientific deliverable, discussion on stream water chemical properties and material budgets in Danum Valley site was published in Journal of Tropical Ecology (Yamashita *et al*., 2014).

3.2 Different reactions of the catchments to reduction of atmospheric inputs

Among four study sites, Kajikawa site in Japan and Sakaerat site in Thailand are historical sites, where the regular surveys started in 2002 and 2005, respectively. In this section, the results from these historical sites are highlighted.

Climate plots in Nakajo meteorological station near Kajikawa site and Sakaerat site were shown in Figure 1. Although monthly precipitation amounts fluctuated slightly in Nakajo station, the amounts were always larger than 100 mm and the lines of temperature and precipitation amount did not cross each other. Therefore, the climate in Kajikawa site can be considered as a perhumid condition through a year. On the other hand, the monthly precipitation amounts in Sakaerat site are dynamically changed and the lines of temperature and precipitation amount crossed each other in February/March and in November. The period when the line of precipitation amount is below the line of temperature can be considered as the dry season in the climate plot. In the case of Sakaerat site, the period from November to February seems to be the dry season. The rest of a year can be considered as the wet season and particularly the period from April to October can be considered as a perhumid period. Thus, the climate in Sakaerat site has a distinct dry season depending on the precipitation pattern (Sase *et al*., 2012), and this may affect deposition processes of air pollutants and their biogeochemical processes in forest ecosystems.

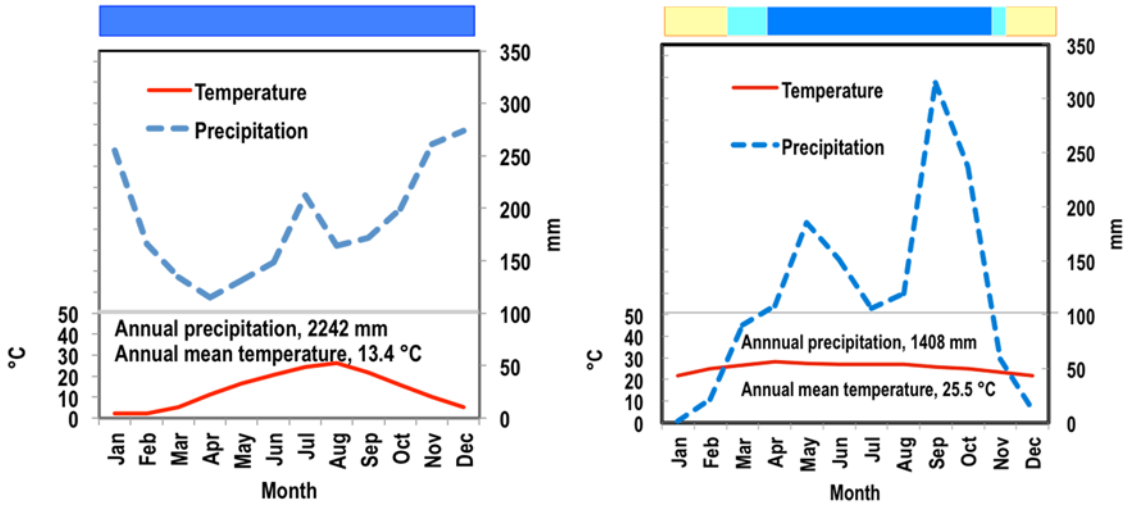

Figure 1. Climate plots in Nakajo meteorological station near Kajikawa site in Niigata Prefecture, Japan (left) and in Sakaerat site in Nakhon Ratchasima Province, Thailand (right) (after Sase *et al*., 2012). Blue, light blue, and yellow colors indicate a perhumid period in wet season, wet season and dry season, respectively.

The S deposition amounts by RF and TF+SF in Kajikawa site and Sakaerat site were shown in Figure 2. In the case of S, since canopy interactions, such as uptake or leaching on leaf surface, may be little (Lindberg and Lovett 1992), the deposition amounts by TF+SF can be considered as total deposition. The S deposition amounts by TF+SF have been declining significantly in both sites ($p = 0.0020$ and 0.0023 in Kajikawa and Sakaerat, respectively, by seasonal Mann Kendall test). The deposition amounts of non-sea salt (nss) S also showed declining trends (detailed data is not shown here), although the peak was observed in a different year in Kajikawa site. The deposition amounts of nss-S peaked in 2006 and declined thereafter in Kajikawa site ($p = 0.050$), which was similar to the SO_2 emission trend in China (Lu *et al*., 2011). The national emission trends of SO_2 (Kurokawa *et al*. 2013) and the EANET wet deposition data in Bangkok and Puthumtani (EANET. 2013) showed declining trends over the last decade. Therefore, the trends of S deposition amounts observed in two forest sites appear to reflect the regional atmospheric conditions, respectively.

The pH and SO_4^{2-} concentrations in SW in Kajikawa site and Sakaerat site were shown in Figure 3. The increasing trend of the SW pH in Kajikawa site became significant since 2007 ($p = 0.0015$ by Mann Kendall test). The SO_4^{2-} concentrations in SW showed a significant declining trend during the observation period ($p < 0.0001$). Alkalinity and NO_3^- concentrations showed significant increasing trends (detailed data are not shown here, $p < 0.0001$ and $p = 0.0023$, respectively). Since a clear drop of SO_4^{2-} concentrations in SW can be seen after 2006 (see the lower left case of Figure 3.), the SW chemistry appeared to reflect the reduction of the deposition sensitively. The decline of SO_4^{2-} concentrations in SW may contribute to the increasing trends of pH and alkalinity. In this aspect, SW seems to be recovered from acidification with reduction of the atmospheric S input. However, NO_3^- concentrations showed an increasing trend, suggesting that unused N in ecosystems was leached into SW gradually. Although the deposition amounts of dissolved inorganic N (DIN, as sum of NO_3^- and NH_4^+) by TF+SF showed the significant declining trend ($p = 0.025$), canopy interactions, especially uptake/consumption of DIN on leaf canopy should be taken into account for precise discussion on the trend (Sase *et al*., 2008). On the other hand, trees in Kajikawa site have been matured gradually for 40 years after plantation, and N uptake rate by trees appeared to be declining gradually. Possibility of so-called N saturation should carefully be

assessed taking account of tree growth in Kajikawa site.

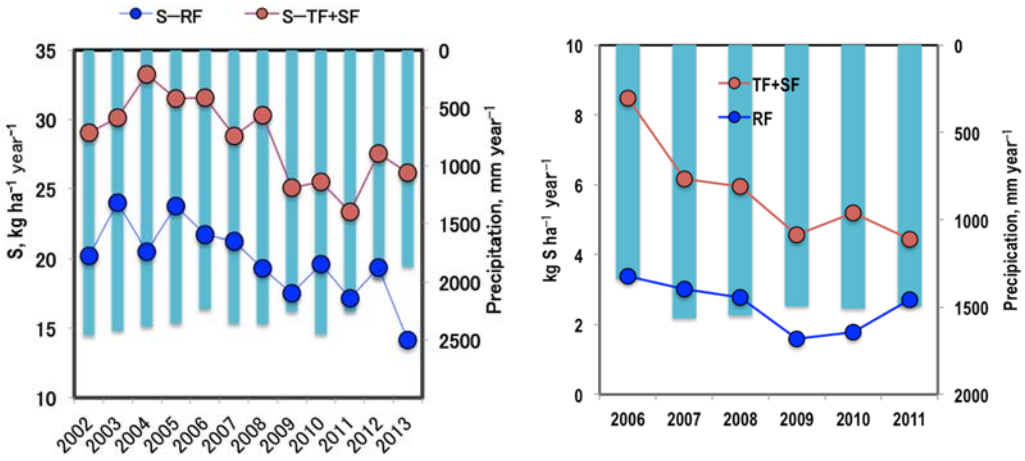

Figure 2. Sulfur deposition by rainfall (RF) and throughfall + stemflow (TF+SF) in Kajikawa site in Niigata Prefecture, Japan (left) and Sakaerat site in Nakhon Ratchasima Province, Thailand (right). Light blue column charts show the annual precipitation amounts.

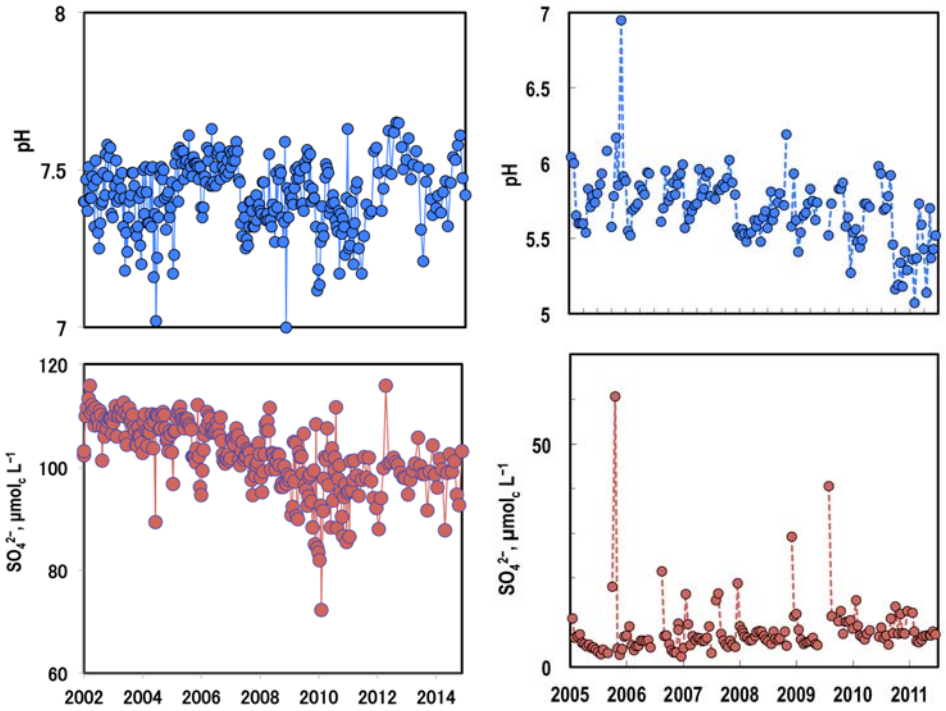

Figure 3. pH (upper) and SO_4^{2-} concentrations (lower) in stream water in Kajikawa site, Niigata Prefecture, Japan (left) and in Sakaerat site, Nakhon Ratchasima Province, Thailand (left).

On the other hand, the SW pH in Sakaerat site declined significantly ($p < 0.0001$) with increase of the SO_4^{2-} concentrations ($p < 0.0001$). Most of the other ion constituents, such as NO_3^-, Cl^-, Na^+, Ca^{2+}, and Mg^{2+}, showed significant increasing trends of their concentrations, while NH_4^+ showed the declining trends. The SW seems to be acidified, even though the atmospheric S input. As mentioned above, SO_4^{2-} concentrations in SW in Sakaerat site (6.3 $\mu mol_c\ L^{-1}$, the weighted mean) were significant low compared to those of the deposition (41.9 $\mu mol_c\ L^{-1}$, as the weighted-mean of TF+SF), suggesting retention of S in the

forest ecosystems. It seems that S accumulated in the ecosystems has been leached with other ions as the atmospheric input declined. As shown in Figure 1, two high precipitation periods can be seen during the wet season in Sakaerat. Although the detailed data is not shown here, contribution of the precipitation in the late wet season to the annual precipitation became larger in the forest plot in Sakaerat site. More water can be flowed into SW effectively in the late wet season. The wetter condition in the late wet season may also induce dissolution of accumulated S in the ecosystems, as suggested by Mitchell and Likens (2011).

The trends of atmospheric deposition and SW chemistry in Kajikawa site and Sakaerat site were summarized with possible interpretations in Table 2. Even though the atmospheric S input declined in both sites, the SW chemistry showed different reactions probably depending on the climate. It was suggested that difference in climate and consequent diversity of ecosystems should be taken into account for assessment of effect of atmospheric deposition.

Table 2. Summary of the trends of atmospheric deposition and stream water chemistry in Kajikawa site and Sakaerat site.

	Kajikawa	Sakaerat
Atmospheric deposition	Decline of S deposition	Decline of S deposition
Stream water	Increase of alkalinity Decline of SO_4^{2-} concentration Increase of NO_3^- concentration	Decline of pH Increase of SO_4^{2-} concentration
Possible interpretations	Recovery from acidification Progress of nitrogen saturation	Progress of acidification

3.3 Project workshop in Bangkok, Thailand

The APN Project Workshop 2014 on Sulphur Dynamics in East Asian Forests was held on 18-19 December 2014 in Bangkok, Thailand, to share the outcomes from the project and discuss current environmental issues and future research topics in the region. The workshop was organized by the Asia Center for Air Pollution Research (ACAP) in cooperation with the Royal Forest Department (RFD) of Thailand. The workshop was attended by approx. 45 scientists/experts from institutes, universities or agencies in Thailand, Malaysia and Japan. Ten speakers made presentations on their research outcomes and the panel discussion was held for the current atmospheric environmental issues and the future research topics and collaboration. As the current issues, possible effect of PM and tropospheric ozone was highlighted in the discussion. The workshop was very fruitful and informative for all the participants.

3.4 Remarks

The grant supporting the project has been finished in August 2015. Therefore, some of the field surveys have tentatively stopped in Thailand and Malaysia. However the data analysis is still ongoing. Several scientific papers are now under preparation/submission. New research topics have been discussed in a process of the data analysis. We may get a new grant for the new research topics and reactivate the surveys in these study sites in the near future.

Acknowledgements

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Scientific and Technological Research Papers from Participating Countries

For our cooperation on research activities, we have some research papers including technical note from our participating countries that have been utilized the EANET data. Therefore, they are included in this part of the EANET Science Bulletin.

Evaluation of tropospheric ozone concentrations over EANET station areas based on remote sensing GOME-2 high-resolution ozone profile datasets

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Abstract

The numbers of global atmospheric composition parameters been retrieved from satellite data are available to use for regional air quality assessment. The scientific data institutes like O3M SAF of EUMETSAT provides several products on retrieved contents of trace gases and aerosols in the atmosphere including high resolution ozone concentration profiles. In order to understand the limitations of using the ozone profile product for assessment of ozone concentration over EANET region, a comparison of near surface satellite data with automatic monitor measurements at EANET station Mondy was performed. As demonstrated by the results of comparison at different scale of data averaging, there is no precise agreement between satellite data and instant values of automatic monitor, however, their mean values are suitable to be used for general air quality assessment at regional scale. The evaluation of mean levels of ozone concentrations was produced for the areas of remote EANET stations Terelj (Mongolia) and Jiwosi (Northern China) being presented as application example of proposed approach.

Key words: Ozone, Remote sensing, Regional pollution, Monitoring network

1. Introduction

There are a number of networks performing regular monitoring of near-surface ozone monitoring all over the world. Results of the monitoring of several networks are available for scientific community including an access through Internet portals of data sharing and integrating. For

example, Jülich Open Web Interface (JOIN) for accessing, analyzing and visualizing atmospheric composition database (https://join.fz-juelich.de/) provides the data collected for Tropospheric Ozone Assessment Report (http://www.igacproject.org/TOAR) of which the significant part of information over East Asia region is thedata from EANET stations. Besides numerous surface measurements there are several satellite-board instruments and platforms for obtaining global ozone content in the atmosphere like OMI (Ozone Monitoring Instrument) aboard AURA satellite. Total ozone column values retrieved from OMI data are available at Goddard Center (http://disc.sci.gsfc.nasa.gov/Aura).

It is important to be sure that in spite of various instrument and monitoring principles there is an agreement between different types of measurement data. Comparisons between ground-based and satellite ozone results done for different *in-situ* and satellite instruments were presented in recent papers (Chiou *et al.*, 2014).

The aim of this article is to explore a possibility to evaluate near surface tropospheric ozone levels at EANET sites with the use of the comparison results of data from automatic measurement monitor and ones retrieved from satellite based instrument. Three year of space remote sensing observations were compared for the different periods of data averaging. The seasonal variations were also approached.

2. Data description

Satellite Atmospheric Facility (SAF) for atmospheric composition and UV Radiation, a part of European Organisation for the Exploitation of Meteorological satellites (EUMETSAT) Network of SAFs, provides a number of near real time and offline products about atmospheric physical properties or chemical composition such as trace gas columns, ozone profiles, aerosols and surface UV data retrieved from Global Ozone Monitoring Experiment-2 (GOME-2) measurements. The GOME-2 is one of the new-generation European instruments carried on Metop satellites. The satellites have sun-synchronous orbits that means the satellite passes over any given point of the planet's surface at the same local solar time. In this paper we used high-resolution ozone profile product from the GOME-2 nadir scanning mode.

Data records for the period of 13/5/2013 to 31/12/2015 were used for comparison with surface measurements at EANET station Mondy and for assessment of mean levels of ozone concentrations at two EANET stations in China and Mongolia.

The high-resolution ozone profile product provides ozone concentrations as the vertical ozone profiles (*state vector*) that are given as partial ozone columns in Dobson Units (DU) in 40 layers from the surface up to 0.1 hPa. Height of the layer nearest to the surface may vary up to 2 km for the EANET stations considered in this paper. Ground pixel size is 40 x 80 km^2 or 40 x 40 km^2 (along track x cross track). It means ozone concentration is assumed as constant in the box of approximately 1km height and 40 x 40 km^2 in area. Date, time and geolocation for each ground pixel/retrieval are included in files of the product.

Also the product includes vector of retrieved errors for the *state vector* of ozone concentrations. Relative errors for data considered are in range of 10 % to 50 % approximately. More information can be found in (http://o3msaf.fmi.fi/products/ohp.html), including quality flags of the data.

Among the EANET areas the remote EANET station Mondy was chosen to apply the first approach comparison of EANET data with retrieved GOME-2 ozone profiles. Mondy is remote station

located in south-eastern Siberia to the south-west from Baikal Lake at the Russian-Mongolian border. Regular continuous atmospheric monitoring at the station was started in May 1999 with the measurements of precipitation chemistry and air concentrations by filter pack and ozone automatic monitor (AM) Dylec-1007-AHJ. The measurements of O_3 are performed based on UV absorption ozone analysis and the detection limit is set to 1 ppb according to EANET Manual (EANET. 2013). Temporal resolution of monitor data is 30 minutes of which raw measurements are available from data storage. Such higher time resolution of data allows us to compare short-term measurements of satellite and AM with time difference less than 15 minutes. However, for the extend of remote sensing Metop-A data period the in-situ measurements at Mondy station are not available for the weeks of 2014 and the end of 2015.

3. Co-location criteria of EANET and satellite data

Spatial resolution of GOME-2 ground pixel is 80 x 40 km^2 at the beginning of data records and 40 x 40 km^2 for majority of data. Pixels that cover Mondy station were selected for comparison. That is, there was approximately one *state vector* of ozone concentration at the area 40 x 40 km^2 that covered Mondy station for one day. All satellite data for the pixels of interest are approximately between UTC 2:30 and UTC 4:30, i.e. it is approximately from 10:30 to 12:30 local time. The satellite and EANET observations were selected as paired if the difference between the observation times was less than 15 minutes.

4. Results of comparison

Time series of retrieved GOME-2 data of ozone concentrations at the near-surface level at area of Mondy station are presented by black dots and correspondent errors of each retrieved *state vector* are shown in grey at Figure 1a. Figure 1b shows automatic monitor's measurements of 30 minutes resolution for whole day time series and GOME-2 data time series (without errors). Both time series demonstrate similar annual cycle with clear seasonal maximum in spring and minimum in autumn.

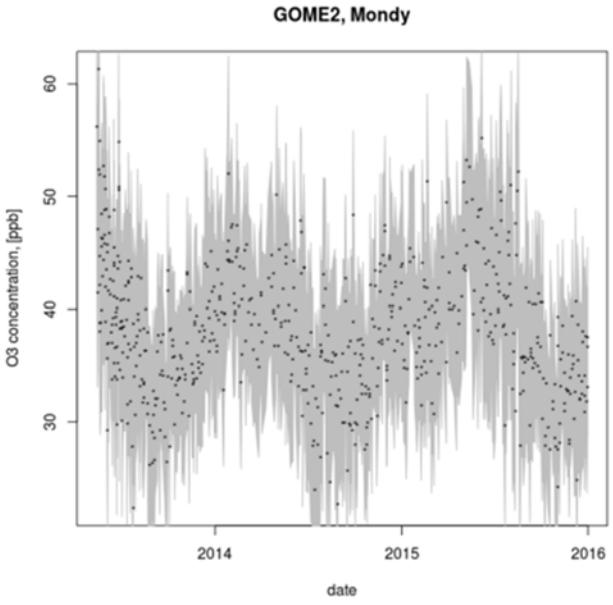

Figure 1a. Time series of GOME-2 data surface level of ozone concentrations at area of Mondy station with correspondent errors.

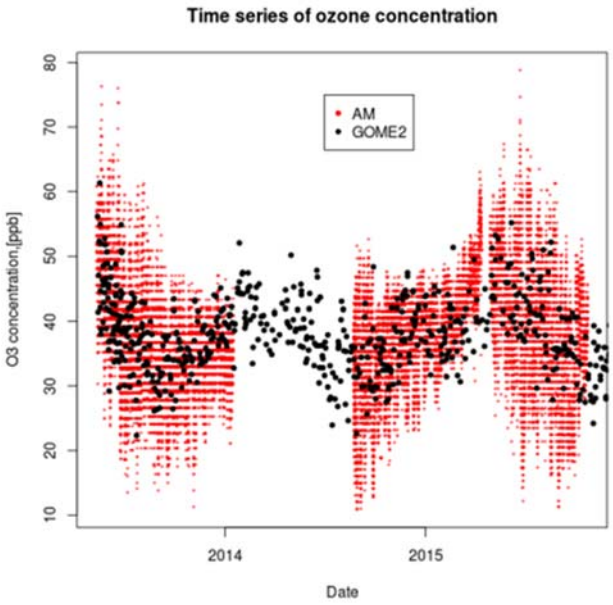

Figure 1b. Time series of AM and GOME-2 data surface level of ozone concentrations at Mondy station.

Box-and-whisker plot of inter-annual variations of raw data is presented at Figure 2. Both data sets have maximum in spring time and minimum concentrations in autumn. The most similarity in distribution is for months May, July, August and October, where mean values are close and almost coincide with medians. However, March, September, November, December and especially April do not fit well, which was also tested by t-test.

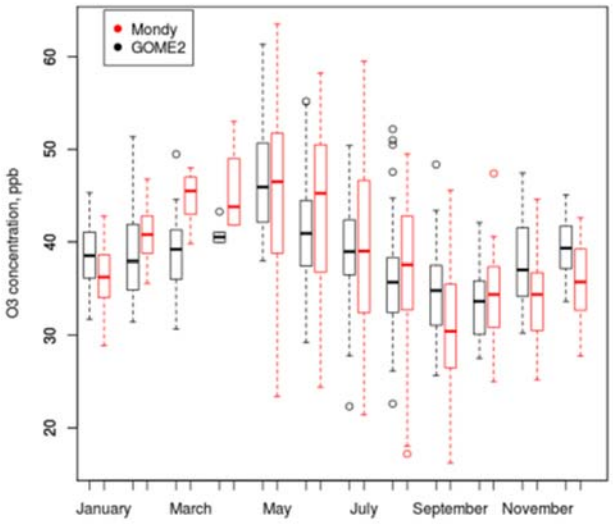

Figure 2. Annual variation of AM and GOME-2 monthly data of ozone concentrations (by box-and-whisker plot).

To quantify and compare annual variations, the first harmonics in a Fourier expansion of AM and GOME-2 time series were fitted with oscillation period 365.25 days according to model presented as Formula (1):

$$y = m + c\cos(\omega t - \varphi) \qquad (1)$$

where m is mean value of time series, c is an amplitude, φ is phase in radians. Table 1 presents parameters obtained as a result of harmonic regression. Mean values of both time series are very close and their difference is even less than automatic measurement detection limit 1 ppb. Phase values are also very close. For both time series the maximum of annual variation for ozone concentrations are detected in April and the minimum is at the end of October. These results are in agreement with box-and whisker annual plot (Figure 2) and other paper (Shamansky and Potemkin, 2011). Results derived from satellite data by harmonic regression are in good agreement with AM dataset and they may be used to obtain general characteristics of annual variations of ozone concentrations. We apply this regression approach to describe the variation characteristics at two other EANET sites.

Table 1. Results of harmonic regression for AM and GOME-2 time series.

	AM	GOME-2
Mean (m)	37.5 ppb	38.1 ppb
Amplitude (c)	6.4 ppb	4.4 ppb
Phase (φ)	5.8 radian	5.7 radian

5. Ozone concentrations assessment for Terelj and Jiwozi remote EANET stations

To evaluate mean ozone concentrations at other EANET sites, the proposed methodology described above was applied to satellite datasets for areas of two remote EANET stations, Terelj and Jiwosi, located in Mongolia and Northern China, respectively. The results of harmonic regression calculated with Formula (1) are presented in Table 2 for both stations. Mean values at stations seem to be different but phase is the same and very close to one of Mondy station. Maximum concentrations calculated according to harmonic regression are achieved at the middle of April and minimum concentrations - at the end of October.

Table 2. Parameters of harmonic regression for GOME-2 data for areas of Terelj and Jiwozi stations.

	Terelj, Mongolia	Jiwozi, China
Mean (m)	36.7 ppb	31.8 ppb
Amplitude (c)	4.6 ppb	5.7 ppb
Phase (φ)	5.9 radian	5.9 radian

Box-and-whisker plots of monthly values of ozone concentrations retrieved from GOME-2 data are shown at Figure 3 (a, b). There are clear seasonal changes to be depicted with a maximum at the end of spring and minimum in the middle of autumn.

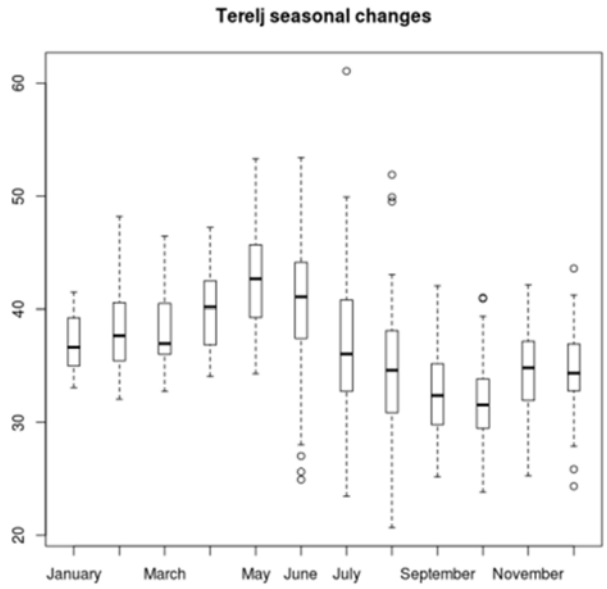

Figure 3a. Box-and-whisker plot of annual variations of ozone concentrations retrieved from GOME-2 data at Terelj area, Mongolia.

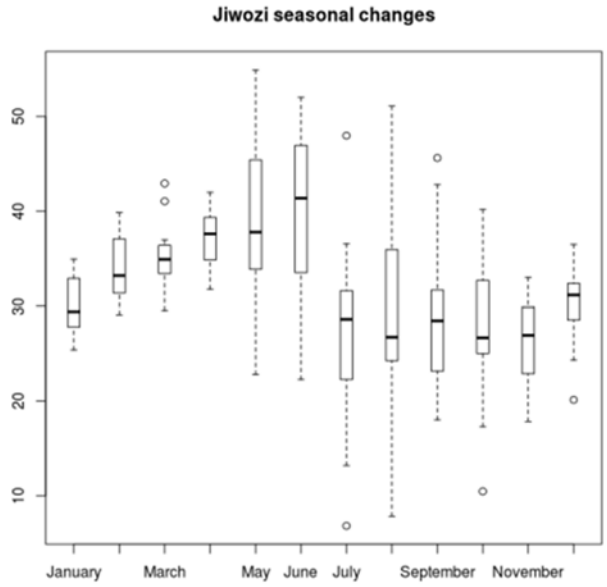

Figure 3b. Box-and-whisker plot of annual variations of ozone concentrations retrieved from GOME-2 data at Jiwozi area, China.

Seasonal median and mean values for areas of Terelj and Jiwozi are presented in Table 3. Ozone concentrations at Jiwozi are always lower for each season than at Terelj, however, the variance at Jiwozi is higher.

Table 3. Evaluated mean levels of ozone concentrations at Terelj and Jiwozi EANET stationsfor whole year and four seasons.

Season	Year		DJF		MAM		JJA		SON	
Station	Terelj	Jiwozi	Terelj	Jiwozi	Terelj	Jiwozi	Terelj	Jiwozi	Terelj	Jiwozi
Median, [ppb]	36.3	31.3	35.8	31.4	40.8	36.4	37.5	30.9	32.6	27.7
Mean, [ppb]	36.7	31.8	36.2	31.2	41.1	37.6	37.5	32.4	33.1	27.9
St.d., [ppb]	5.7	8.2	3.8	3.8	4.4	5.7	6.5	10.7	4.1	6.2

The results obtained from satellite data according to methodology above may be further used for regional pollution assessments.

6. Discussion

Values retrieved from satellite data and AM measurements being possible to compare are always related to approximately same period of the day, i.e. between 10:30 am and 12:30 pm of local time in cases of stations above because of sun-synchronous orbit of the satellite. As presented at Figure 4, the distributions of whole EANET data time series (presented in Figure 4 in red color) and

EANET data of measurement time nearest to satellite data (orange color) are similar (except for April values). The use of Kolmogorov-Smirnov test proves these two distributions are same at 0.05 confidence level. It is reasonable to assume that ozone concentration assessment based on midday measurement subsets and those based on whole time series will provide the same results, though near surface ozone concentrations are closer to its minimum values of their diurnal cycle at the hours when satellite data available (Shamanskyz and Potemkin, 2011). Therefore, the assessment values based on satellite data being available for midday period at this region are also valid as evaluation of mean levels of ozone concentrations. However, for other regions it may be necessary to take into account hours of day at which satellite data are available together with particular features of diurnal variation of ozone concentrations for the region.

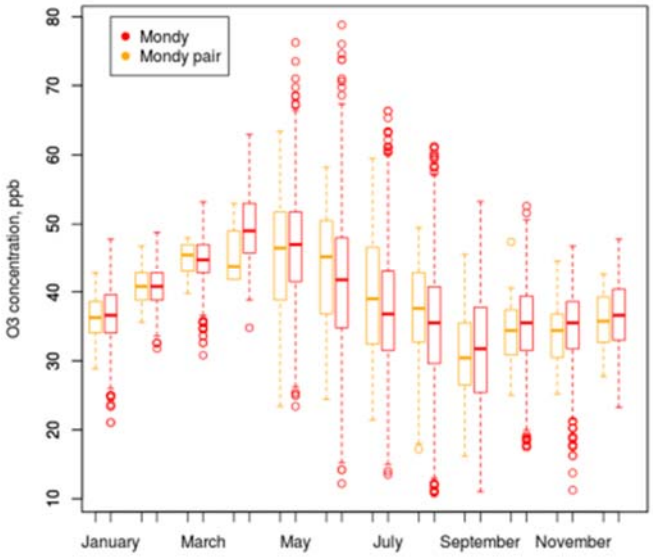

Figure 4. Box-and-whisker plots of AM monthly data at Mondy station. Whole time series of 30 minutes time resolution boxplots are in red color, subsets of time series paired with satellite data are in orange color.

Though general characteristics of satellite retrieved data and AM concentrations are similar, comparison of paired dataset demonstrates not so good agreement. The scatterplot of paired data selected according to co-location criteria is shown at Figure 5. The set of points forms a cloud and related correspondent correlation coefficient is very low (0.16).

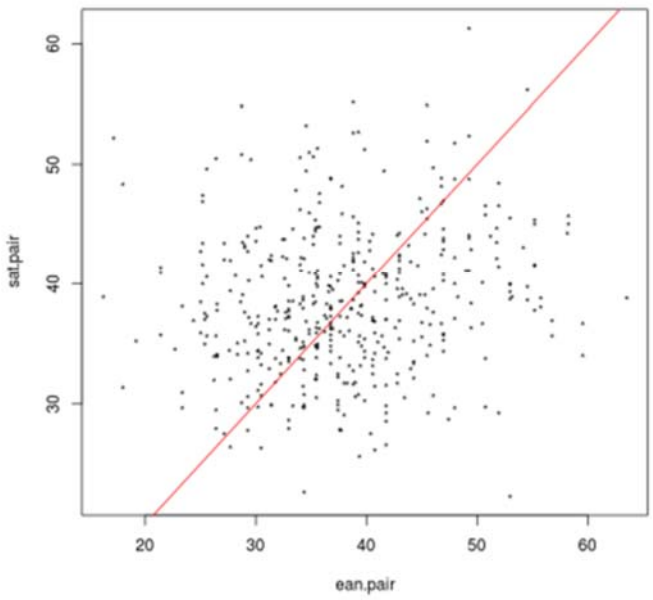

Figure 5. Scatterplot of co-located AM and GOME-2 data. Red line has slope 1 and intercept 0.

There are errors of retrieval included in ozone profile product (shown in grey at Figure 1a), but the errors may be twice lower compared to difference between paired AM and GOME-2 data. Histogram of differences between automatic monitor data at EANET station and retrieved data is presented at Figure 6. Errors are correspondent approximately to normal distribution with a maximum frequency around zero value.

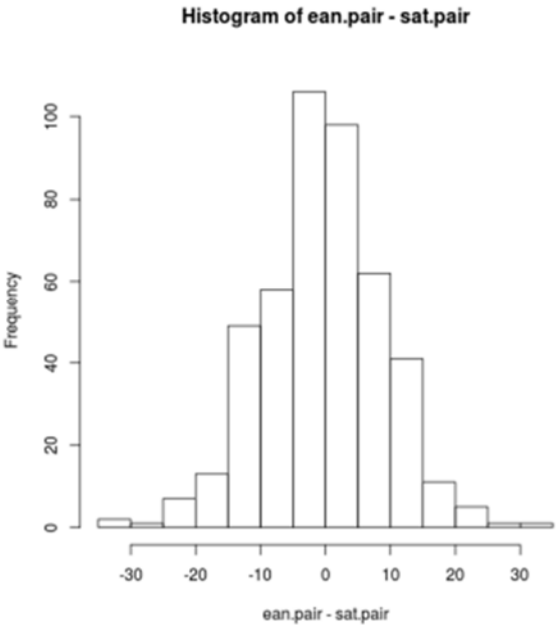

Figure 6. Histogram of differences between AM and GOME-2 paired data.

In case of Mondy station, an assessment of instant ozone concentration by satellite data may not coincide with near-surface measurements by automatic monitor even provided errors of retrieved data would be taken into account. Therefore it is not reasonable to use raw satellite data to obtain instantaneous values of ozone concentrations near surface for the stations described above. By applying moving average to data series, a correlation coefficient between AM and satellite datasets improves. The correlation coefficient increases rapidly to 0.5 for width of moving average window to be equals approximately two weeks. This period of averaging seems to be enough to remove a noise of satellite retrieval, hence, an average concentrations of ozone may be assessed by retrieved data.

7. Conclusion

High resolution ozone profile data, retrieved from GOME-2 provides ozone concentrations with global coverage for 40 layers of atmosphere up to 0.1 hPa including the one nearest to the earth surface. This information may be used to evaluate monthly average values and annual variations of ozone concentrations near surface, however, precise instant values retrieved from satellite data may not well agree with values measured by automatic monitor.

Annual changes of ozone for three remote EANET stations located in North-Western part of EANET region (Mondy in Russia, Terelj in Mongolia and Jiwozi in China) obtained from satellite data have the similar phases and approximately the same period of maximum concentrations (at the middle of April) and minimum concentrations (at the end of October) of the years, however, mean ozone concentrations at Jiwozi are lower than at Mondy and Terelj.

Next step of research is to compare all available EANET ozone data sets with concentrations from GOME-2 data to extend a comparison between data from EANET stations and GOME-2 data according to methodology discussed above to evaluate ozone characteristics over the whole EANET region and to obtain spatial clusters with similar ozone concentration levels within EANET region.

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Technical Note

Utilization of the EANET Data and the Spread of the EANET Research

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The EANET was established as a regional cooperative initiative to promote efforts for environmental sustainability. Monitoring activities at acid deposition monitoring site and ecological survey sites had started from March 1998 as the preparatory phase and from January 2001 as the regular monitoring with 10 participating countries including China, Indonesia, Japan, Malaysia, Mongolia, Philippines, Republic of Korea, Russia, Thailand, and Vietnam. Cambodia, Lao PDR and Myanmar joined the EANET in 2001, 2002 and 2005, respectively. The data on wet deposition monitoring were submitted from a total of 55 monitoring sites, including 20 remote, 14 rural and 21 urban sites in 2014. The filter pack method is used at 42 sites for measurements of air concentrations of pollutants. Automatic or manual gas monitors is also used for the ozone concentration. PM_{10} is measured by automatic or manual instruments at 21 sites.

The EANET has promoted public awareness activities on acid deposition and other priority chemical species, including their effects, control and mitigation measures to various levels of the community which includes policy makers, scientists, general public, youth, school children and others to share common understanding on atmospheric environmental issues among the scientific community and policy makers by exchanging information through a network of experts, particularly for school children and teachers through a number of activities.

Three reports for policy makers were published titled “Goals, Achievements and Way Forward”, “Clean Air for Sustainable Future” and “EANET and Clean Air for Sustainable Development”. The EANET has undertaken joint public awareness projects with participating countries to develop brochures and videotapes on acid deposition in the national language, and several times has held “Workshop on Public Awareness on Acid Deposition Problems” in the participating countries. An e-learning program on acid deposition problems was issued on the EANET website (http://www.eanet.asia/product/e_learning/index.htm) as one of the public awareness materials for environmental education and other public awareness activities. The capacity building workshops for policy makers were held in the EANET to raise awareness of policy makers of participating countries to the adverse impacts on the environment caused by acid deposition. The Factsheets titled “Country efforts and achievements in combating acid deposition” were developed by all participating countries of the EANET through collaboration and coordination with the Network Center and the Secretariat. The published public awareness materials were also issued on the EANET website (http://www.eanet.asia/product/index.html). It has been increasing in number of researchers who is promoting environmental study in Asia by utilizing annual published “Data Report”, “Periodic Report” which is published every 5 years, or “EANET Science Bulletin”. The EANET data is Asian

environmental archive that has been collected in accordance with international standards, however, it is not fully utilized. It is necessary to investigate the utilization of the EANET data in order to increase the research activities in East Asia.

Prior to the EANET, there is an ambient monitoring network named EMEP in Europe. The European Monitoring and Evaluation Programme (EMEP) is a scientifically based and policy driven programme under the Convention on Long-range Transboundary Air Pollution (CLRTAP) for international co-operation to solve transboundary air pollution problems. EMEP has a long history for about 20 years than the EANET has. In analyzing the data use of the EANET, comparison with the EMEP is effective. In this technical note, it was investigated literature number or author which quoted the network data or showed some relation with network, by using various search engines on Internet.

Figure 1 (a) shows the yearly variation of number of publication which quoted each network name of EANET and EMEP, by using Springer search engine. Main document investigated by the Springer search engine is Journal of Atmospheric Chemistry or Water, Air & Soil Pollution. The number is increased from 2001 when the regular monitoring has started to 2014, but number of publication the EANET is about several. On the other hand, EMEP showed nearly 10 times more publication in comparison with the EANET. Figure 1 (b) shows the same variation by using a research engine of the Science Direct. The Science Direct research engine mostly treated Journal of Atmospheric Environment. The number of publication of each network were different from Figure 1 (a), however, the trend and the magnitude of number differences are the same. Figure 1 (c) and 1 (d) are the trend of publication number using the Wiley On Line search engine and Google Scholar search engine, each. The Wiley On Line search engine mostly treats Journal of Geophysical Research. The Google Scholar search engine showed the results of most wide range including variety of journals, books, presentation materials, and memos. However, the search result of the Google Scholar finds out that the number of search hits includes much noise before 2000 when the EANET isn't active as shown in Figure 1 (d).

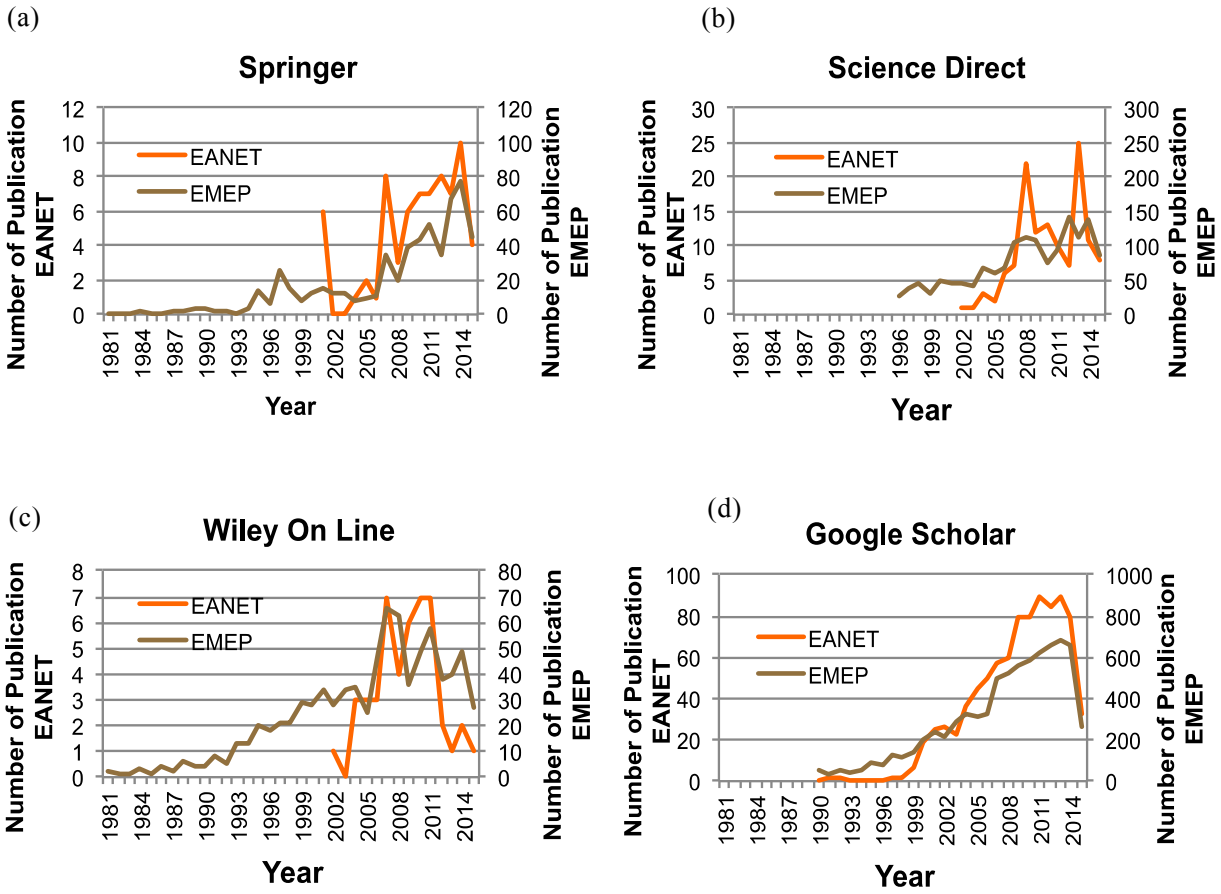

Figure 1. Yearly trend of publication number which quoted each network name of EANET and EMEP.

(a) results using Springer search engine
(b) results using Science Direct search engine
(c) results using Wiley On Line search engine
(d) results using Google Scholar search engine

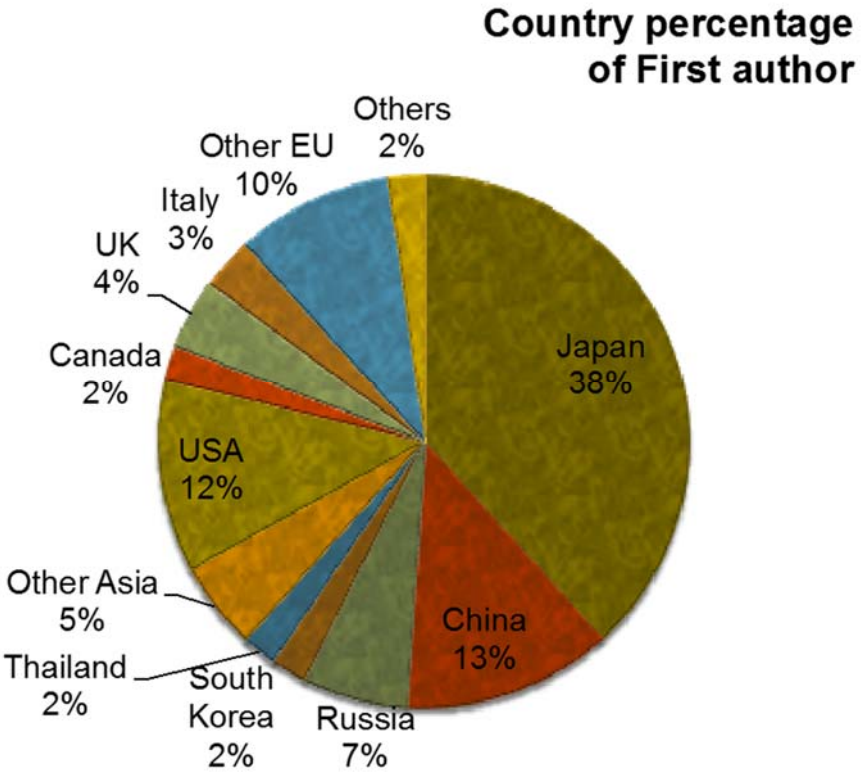

Figure 2. Country percentage of first author which quoted EANET. Research result of 94 articles using Springer search engine was shown. Other Asia involves Indonesia, Nepal, and India.

Figure 2 shows the country percentage of first author which quoted EANET. The first author's country of 94 articles from searched results using the Springer search engine was shown. From this graph, it became clear that the Japanese quoted EANET most. Chinese author was the second highest. There were a lot of American authors besides Asia. The number of Asian author except for Japan and China was very little, and this shows that the researcher who have an interest in the EANET is little in Asia.

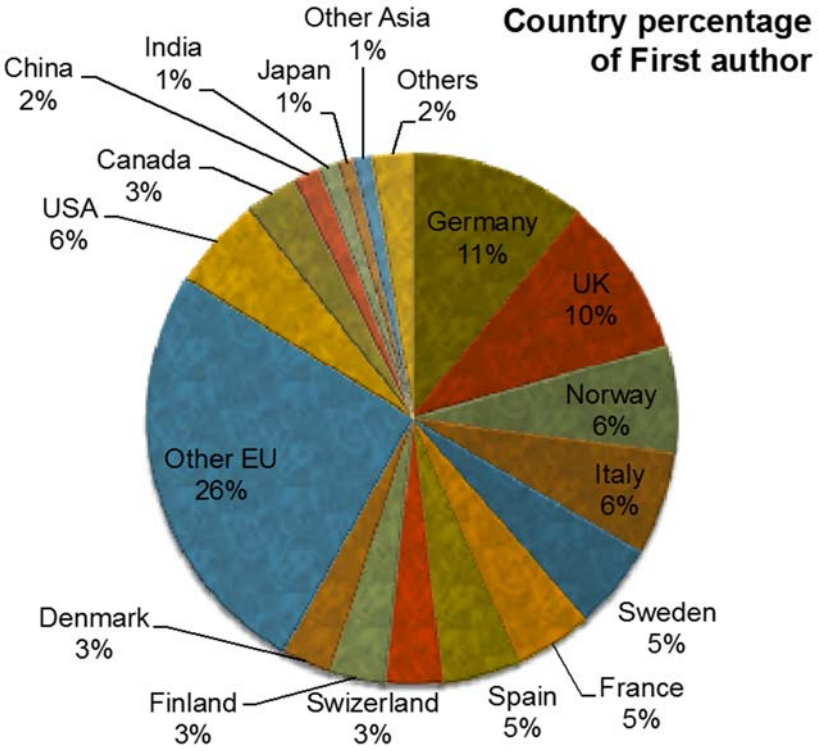

Figure 3. Country percentage of first author which quoted EMEP. Research result of 960 articles using Springer search engine was shown. Other EU involves Greece, Poland, Russia, Netherlands, Austria, Czech Republic, Belgium, Bulgaria, Hungary, Portugal, Croatia, Lithuania, Serbia, Turkey, Estonia, Ireland, Slovakia, Belarus, Latvia, Romania, Slovenia, Ukraine, Cyprus, Luxembourg, Macedonia, and Montenegro.

Figure 3 shows the country percentage of first author which quoted EMEP. The first author's country of 960 articles from searched results using the Springer search engine was shown. From this graph, it became clear that the Germany quoted EMEP most. The second highest is United Kingdom, but after the 3rd place, Norway, Italy, Sweden, France, Spain has been antagonistic. The sum of the percentage of European author becomes 84%, and it is clear that many European researchers have an interest in own nations data. As the reason that the number of Asian researcher who quote EANET was low, it might be due to low opportunity to grown up as a scientist. From this point of view, capacity building in East Asia is very important issue.

As references, articles in which "EANET" was quoted were listed below for 3 years starting from 2013. The Google Scholar was used as a search engine. The level of the quotation is various from the articles for which the EANET data was utilized to the articles which just introduces the EANET. Articles written in East Asian languages, including Chinese or Korean, has been searched several cases. In the case that the literature name was unknown, the articles were excluded from the list.

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