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Acid Deposition Monitoring Network in East Asia (EANET)

EANET Science Bulletin

Acid Deposition Monitoring Network in East Asia

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Acid Deposition Monitoring Network in East Asia (EANET)

Objectives

- Create a common understanding of the state of acid deposition in East Asia;

- Provide useful inputs for decision making at local, national and regional levels with the aim of preventing or reducing adverse impacts on the environment caused by acid deposition; and

- Contribute to cooperation on issues related to acid deposition among the participating countries.

Cover photo is provided by Dr. Mingqun Huo of the Network Center for the EANET

EANET Science Bulletin

Volume 5

The EANET Science Bulletin is published by the Network Center for the Acid Deposition Monitoring Network in East Asia (EANET) regularly. The following Editorial Board was established by the Network Center for the EANET to publish the EANET Science Bulletin (Vol.5):

Preface

The EANET Science Bulletin (Volume 5)

This is Volume 5 of the EANET Science Bulletin. The Bulletin is published to share the scientific findings from the research activities within EANET and to provide a platform for scientists from participating countries to publish their scientific and technical research results relevant to the EANET activities.

This volume is comprised of the Reports of the EANET Fellowship Program (2016-2018), Joint Projects of the EANET with Participating Countries, as well as Scientific and Technological Research Papers from Participating Countries.

This Science Bulletin follows the Medium Term Plan (MTP) for the EANET (2016-2020) that was adopted at the Seventeenth Session of the Intergovernmental Meeting (IG17) on the EANET in 2015. The Bulletin would contribute to a better understanding of the state of acid deposition with atmospheric pollutants and their impacts on the environment in East Asia. It can also contribute to promoting research activities on atmospheric transport modeling as well as to improve capacity building for tackling various air pollution issues such as Ozone/PM2.5 problems in participating countries.

Quality assured/quality controlled monitoring data and research results utilizing such data are very important and useful to assist policy makers in planning air pollution control and mitigation measures by improving the scientific and technical knowledge in not only each participating country but also in whole East Asia.

I hereby express my sincere appreciation to all authors of the research papers and the members of the Editorial Board for this Bulletin. I look forward to their continuous cooperation and concern for the EANET activities.

Dr. Shiro Hatakeyama
Director General
Asia Center for Air Pollution Research (ACAP)
Network Center for the EANET

June 2020

EANET Objectives

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Message from the Editor

As one of the activities of the EANET, promotion of research activity is specified in the Medium Term Plan for the Acid Deposition Monitoring Network in East Asia (2016-2020). In this regard, EANET encourages participating countries to conduct research on the state of acid deposition and publish it in the Science Bulletin.

The Science Bulletin (Vol.5) includes reports of the EANET research fellowship program (2016-2018), joint projects of the EANET with the participating countries, scientific and technological research papers from participating countries and technical notes. The articles only represent opinion of authors.

Reports of the EANET Research Fellowship Program (2016-2018)

The EANET Science Bulletin (Volume 5) is issued once every three years in line with the Medium Term Plan (2016-2020) approved at the Seventeenth Session of the Intergovernmental Meeting (IG17) of the EANET which was held in Bangkok, Thailand, 2015.

Therefore, the research reports of the EANET Fellowship Research Program for 2016-2018 are included in this Bulletin.

Comparison of observed and modeled NO_2 in China during the summer, 2013-2015: sensitivity of meteorological conditions and NO_X emission constrains

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Abstract

The causes of inter-annual variation of tropospheric NO_2 vertical column densities (VCDs) observed by satellite over China during summer time (June, July, and August) from 2013 to 2015 were investigated with the aid of chemical transport model in this study. For the purpose, the modeling system was developed and validated. We validated the modeled tropospheric NO_2 VCDs with OMI retrievals for the summer time of the year 2013 that model simulations reproduced the observed NO_2 VCDs well over China with slope 1.15 and correlation coefficient 0.79. Then, using the modeling system, influences of meteorological condition were examined by a sensitivity study during the period of 2013-2015 with anthropogenic emissions fixed at 2013 levels. Combined with the changes in the OMI-observed NO_2 VCDs, the influences of NO_X emissions were also examined. There was an increase of NO_2 VCDs by 4.26% for the year 2014 compared to 2013. The impacts of meteorological conditions and NO_X emissions on the increase of tropospheric NO_2 VCDs for 2014 were 2.95% and 1.31%, respectively. There was a decrease of NO_2 VCDs by 4.45% for the year 2015 compared to 2013. The meteorological fields contributed a 3.57% increase to the tropospheric NO_2 VCDs for 2015 while the NO_X emission reduction resulted in an 8.02% decrease. Therefore, this study inferred that the NO_X emission had an increase in 2014 and reduction in 2015 during the summer compared to the year 2013 levels. The large decreases of observed NO_2 VCDs in 2015 were mainly due to the declines of NO_X emissions while the influence of meteorological condition was relatively small. The satellite retrievals were further used to constrain the surface NO_X emissions using an inverse approach based on an assumption of the linear relationship between tropospheric NO_2 VCDs and surface NO_X emission represented in the modeling system. The summer time total of OMI-based top-down NO_X emissions over China for 2013, 2014, 2015 were 1.43, 1.48, 1.27 TgN, respectively. With the implementation of emission reduction control measures, a significant NO_X emission reduction in the 2015 summer was identified over China. Compared to the top-down NO_X emission of 2013 summer time, the provincial NO_X

emission variation had a spatial inconsistency with obvious increase or decrease in different regions (the largest decrease of 26.97% for Shanghai, the largest increase of 22.56% for Shan'xi) for the 2014 summer time, while there was an obvious decline in Eastern part of China for the year 2015.

Keywords: Tropospheric NO_2 VCDs; Meteorological conditions; NO_X emissions; Air quality model; OMI satellite

1. Introduction

Nitrogen oxides (NO_X=NO+NO_2) is an important trace gas in the atmosphere that is mainly emitted from anthropogenic sources (e.g., fuel combustion and human-induced biomass burning) and natural sources (e.g., soil and natural fires). NO_X plays a key role in atmospheric chemistry involved in the formation of ozone and secondary particulate matter. With the rapid economic growth and energy consumption over the past few decades, China's NO_X emissions have increased by 70% from 1995 to 2004 (Zhang et al., 2007) and 89% from 2000 to 2008 (Kurokawa et al., 2013), which leads to a more and more serious atmospheric pollution, such as acidic rain, haze and high ozone concentration that are harmful to human health and ecosystem.

The China's government aimed to reduce NO_X emissions by 10% from 2011 to 2015 for the Twelfth Five-year Plan and started to implement new emission control measures. Thus, a NO_X emissions decrease of 21% during this period has been evaluated in the bottom-up inventory (Liu et al, 2016). However, emission inventories are quantitative estimates of air pollutants and traditionally developed in a "bottom-up" approach which collects information from statistics and surveys that may have large uncertainty and often cannot update timely (Streets et al, 2003; Zhang et al., 2009a). The development of remote sensing technology promotes real-time observations of the trace gases and aerosols in the atmosphere. Satellite instruments, such as the Global Ozone Monitoring Experiment (GOME), SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographY (SCIAMACHY), Ozone Monitoring Instrument (OMI), and GOME-2, keep providing global remote sensing measurements of tropospheric NO_2 vertical column densities (VCDs) at different spatial resolution and time scales, which can be applied to assess the uncertainty of the bottom-up emission inventory (Han et al.,2009,2011) and constrain on the surface NO_X emissions (Martin et al., 2003, 2006; Lamsal et al., 2011; Mijling et al., 2012).

The main objective of this work is to investigate the causes of inter-annual variation of tropospheric NO_2 VCDs over China during summer time (defined as June, July, and August in this study) from 2013 to 2015 using both OMI satellite observations and Community Multi-scale Air Quality (CMAQ) modeling system. This paper provides a description of methodology in Sect. 2 and the results and discussion in Sect.3. First, the CMAQ modeling system was developed and validated for the year 2013 summer using OMI retrievals and EANET observations as presented in Section 3.1 and Section 3.2, respectively. Second, the influences of meteorological conditions and emission on inter-annual variation of NO_2 VCDs were examined by CMAQ simulations with fixed NO_X emissions in Sect. 3.3. Third, top-down NO_X emissions from 2013 to 2015 were

estimated using OMI-derived NO_2 VCDs and CMAQ simulations and year-to-year variation of NO_X emissions were analyzed at a national and provincial level in Sect. 3.4. The inverse estimation approach provides necessary information about the recent status of emissions over China. Section 4 presents a summary including future plans for this study.

2. Methodology

2.1 Model Description

The air quality modeling is helpful to understand how air pollutants react, transport, and deposit in the atmosphere and can be used to analyze the spatial-temporal variation of air pollutants which affect ambient air quality. The CMAQ modeling system has been extensively used in air quality simulation and analysis (Liu et al., 2010; Wang et al., 2010). Therefore, in this work, the three-dimensional CTM simulations were conducted by the Weather Research and Forecast (WRF) Model v3.5.1. and US EPA/Models-3 CMAQ v5.0.1. For the meteorological fields, the WRF run was performed with the National Centers for Environmental Prediction (NCEP) 1°×1° FNL (final) operational global analysis data. For chemical system, the CB05 gas-phase mechanism and the AERO6 aerosol mechanism were applied in this study. The lateral boundary conditions for CMAQ were taken from GEOS-Chem 2°×2.5° global simulations. For initial condition, the default data sets of CMAQ were used in this study. Therefore, a 30-day spin-up run were conducted before the three-month simulation to minimize the influence of the default data. Figure 1 shows the model domain of this study. Horizontal resolutions both for WRF and CMAQ are 36km×36km. For vertical resolutions, 23 sigma levels from surface to tropopause (about 100 mb) were defined for WRF and simulated meteorological fields were interpolated into 14 layers of the CMAQ.

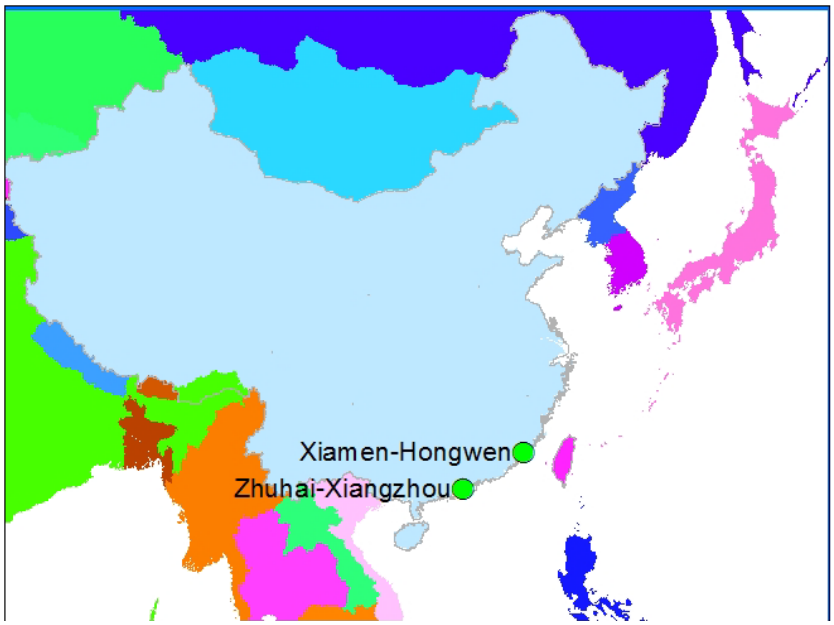

Figure 1. Simulation domain. The green circles represent the EANET sites in China which have available NO_2 and PM_{10} observation data from 2013 to 2015.

2.2 Bottom-up Emission Inventory

Anthropogenic emissions for the year 2013 in China were derived from the Multi-resolution Emission Inventory of China (MEIC model, http://www.meicmodel.org) developed by Tsinghua University that contained about 700 emitting sources with updated activity data and emission factors (Li et al., 2014, Zheng et al., 2014, Liu et al., 2015). For the other Asian countries, anthropogenic emissions for the year 2010 were taken from the MIX emission inventory (Li et al., 2015). Biogenic emissions from soil as an important natural NO_X source were obtained by the MEGAN v2.1 (Guenther et al., 2012) in an hourly variation with the meteorological conditions as input for the year 2013, 2014, and 2015, respectively.

2.3 OMI-satellite NO_2 VCDs retrievals

The OMI, a UV/Vis nadir spectrometer onboard NASA's EOS-Aura satellite (launched on 15 July 2004) provides columns of O_3, SO_2, NO_2, HCHO by measuring the solar radiation backscattered in UV-VIS radiances from 270 to 550nm with a spectral resolution of 0.5nm (Levelt et al.,2006) and has been utilized in many studies. It provides daily global coverage around 13:45 local time with a spatial resolution of 13km×24km at the nadir and coarser resolution at larger viewing angles.

Daily NO_2 VCDs from the Dutch OMI NO_2 (DOMINO) data version 2.0 product (Boersma et al,2011) developed by the Royal Netherlands Meteorological Institute (KNMI) were used in this work, available at TEMIS (www.temis.nl). The retrieval of tropospheric NO_2 VCDs based on three steps as followed: (1) retrieve a NO_2 slant column densities (SCDs) using the differential optical absorption spectroscopy (DOAS) technique, (2) separate the stratospheric and tropospheric NO_2 SCDs with the TM4 model assimilation, (3) convert the tropospheric NO_2 SCDs to VCDs using the tropospheric air mass factor (AMF). To reduce the retrieval errors, the observation data with cloud radiance fraction larger than 50%, surface albedo values larger than 0.3, solar zenith angle value larger than 70 degrees and those affected by row anomaly were filtered out. Errors in the retrieval of tropospheric NO_2 VCDs mainly caused by the AMF calculation were approximated to be ~10^{15} molec cm^{-2}, with a relative error of 25% (Boersma et al., 2011). To make comparison, the satellite observations were interpolated into the model grid by area weighted method based on the information of latitude and longitude.

2.4 EANET Observation

Since 1998, the Acid Deposition Monitoring Network in East Asia (EANET, http://www.eanet.asia/) provides continuous monitoring of deposition and air concentration including gaseous and particulate phase chemistry in 13 countries of the region, which has been widely used in comparison of chemical deposition and evaluation of ozone and relevant species in model simulation (Han et al., 2008; Wang et al., 2008). In this work, surface NO_2 and PM_{10} concentration simulated by the WRF-CMAQ model system were compared with available EANET NO_2 and PM_{10} observations in China (see Figure 1).

2.5 Numerical Experiment

2.5.1 Model simulation with fixed emissions of the year 2013

The inter-annual variation of NO_2 VCDs over China is mainly influenced by meteorological fields and the intensity and distribution of emission sources. In order to separate the influence between the two factors, sensitivity simulations were carried out with the aid of chemical transport model CMAQ.

First, to evaluate the influence of meteorological fields, the CMAQ simulation was conducted during the summer time from 2013 to 2015 with fixed NO_X emissions at the year 2013 levels and the WRF meteorological fields for the corresponding year, respectively. The difference of simulation from 2013 to 2015 represented the influence of meteorological conditions for the corresponding year, which was shown in the following equation.

$$\partial\Omega_i^{\mathrm{met}} = \frac{\Delta\Omega_i^{\mathrm{met}}}{\Omega_{2013}^{\mathrm{met}}} = \frac{\Omega_i^{\mathrm{met}} - \Omega_{2013}^{\mathrm{met}}}{\Omega_{2013}^{\mathrm{met}}} \quad (1)$$

Where i represented the year 2014 or 2015, $\Omega_{2013}^{\mathrm{met}}$ was the result of model simulation at the year 2013, $\Delta\Omega_i^{\mathrm{met}} = \Omega_i^{\mathrm{met}} - \Omega_{2013}^{\mathrm{met}}$ was the absolute changes of model simulation caused by only the variation of meteorological condition between the year i and 2013, and $\partial\Omega_i^{\mathrm{met}} = \frac{\Delta\Omega_i^{\mathrm{met}}}{\Omega_{2013}^{\mathrm{met}}}$ represents the relative changes.

Second, we used the OMI-satellite observation data to review the NO_2 VCDs during 2013–2015 summer time. We assume the satellite observation as the "real" tropospheric NO_2 VCDs and analysis the relative changes from 2013 to 2015, which is descried as $\frac{\Delta\Omega_i^{\mathrm{t}}}{\Omega_{2013}^{\mathrm{t}}}$ in the equation(2).

$$\partial\Omega_i^{\mathrm{t}} = \frac{\Delta\Omega_i^{\mathrm{t}}}{\Omega_{2013}^{t}} = \frac{\Omega_i^{\mathrm{t}} - \Omega_{2013}^{\mathrm{t}}}{\Omega_{2013}^{t}} \quad (2)$$

Similarly, $\Omega_{2013}^{\mathrm{t}}$ was the result of OMI-derived tropospheric NO_2 VCDs at the year 2013, $\Delta\Omega_i^{\mathrm{t}} = \Omega_i^{\mathrm{t}} - \Omega_{2013}^{\mathrm{t}}$ represents the absolute changes of tropospheric NO_2 VCDs for the year i compared to 2013, and the ratio $\partial\Omega_i^{\mathrm{t}} = \frac{\Delta\Omega_i^{\mathrm{t}}}{\Omega_{2013}^{\mathrm{t}}}$ represents the relative inter-annual variation.

Third, considering the changes in tropospheric NO_2 VCDs observed by satellite from 2013 to 2015, the influence of NO_X emissions on inter-annual variation of NO_2 VCDs over China $\partial\Omega_i^{\mathrm{e}}$ can be investigated through the following equation.

$$\partial\Omega_i^{\mathrm{e}} = \partial\Omega_i^{\mathrm{t}} - \partial\Omega_i^{\mathrm{met}} \quad (3)$$

What needs to be emphasized is that, we mainly focus on the major factors emissions and meteorological fields, the non-linear chemical mechanisms like deposition process, chemical reaction process, and others which also influences the NO_2 VCDs is not considered in this study.

Following this method, we can examine the major reason of inter-annual variation of NO_2 VCDs over China during summer time from 2013 and 2015 and identify the inter-annual variation trend of the NO_X emissions. There is one point that, the CMAQ modeling system was validated only for the year 2013 by comparing the model simulation results with bottom-up NO_X emission inventory as input and OMI-derived tropospheric NO_2 VCDs over China. The available EANET observation data were also used to evaluate the model results. The results are shown in Sec.3.1. In this study, based on the validation results for the year 2013, we assume that with proper emissions and meteorological fields, our CMAQ can reproduce the inter-annual variation of OMI-derived NO2 VCDs. In the future study, we will perform inter-annual simulations with emissions and meteorological fields of target year and compare the results with corresponding OMI observations.

2.5.2 Top-down constraints of NOX emissions

To analyze the year-to-year variation of NO_X emissions over China from 2013, a quantitative inverse estimation approach was applied in this study. Because of the short lifetime of NO_X, less than one day to a few days in the boundary layer, satellite NO_2 observations are closely correlated to surface NO_X emissions (Beirle et al., 2003; Martin et al., 2003; Zhang et al., 2009b). Following the method of Martin et al., (2003, 2006), this study assumed a linear relationship between tropospheric NO_2 VCDs and local NO_X emissions. The top-down surface NO_X emissions E_t for the summer time (June, July, and August) of the period from 2013 to 2015 were determined from the local retrieved NO_2 VCDs Ω_t, model simulations Ω_a with a prior emissions E_a through the following equation.

$$E_t = \alpha \Omega_t$$

$$\alpha = \frac{E_a}{\Omega_a}$$

where the coefficient α is a linear relationship between NO_X emissions and NO_2 VCDs that accounts for an effective NO_X lifetime, the NO_X chemistry reactions, NO_X transport as presented in that CMAQ model. The sensitivity simulation designed in the section 2.5.1 for the year 2013, 2014 and 2015 using the fixed anthropogenic emissions of 2013 over China to obtain the coefficient α for each year, respectively. This study used the linear relationship in the above equation to infer top-down NO_X emissions from retrieved tropospheric NO_2 VCDs Ω_t. Following this method, top-down NO_X emissions from 2013 to 2015 were estimated at a national and provincial level. The emission constrains from satellite could provide necessary information about the recent status of NO_X emission over China.

3. Results and discussion

3.1 Model Validation for the year 2013 Based on the Comparison with OMI Satellite Observation

The model reproducibility of tropospheric NO_2 VCDs over China during the summer time for the year 2013 was examined by sampling the valid data of OMI footprint. To make comparison, the CMAQ-simulated tropospheric NO_2 VCDs were obtained from surface to 10 km height averaged between 13:00 and 14:00 local time. The spatial distributions of CMAQ-simulated and OMI-retrieved tropospheric NO_2 VCDs over China and Japan were compared in Figure 2. The CMAQ simulated spatial distribution pattern was very similar with that of OMI observation which both showed a high concentration over polluted industrial regions such as Central East China. The average of simulated NO_2 VCDs with the bottom-up emission inventory was estimated as 2.34 x 10^{15} molec cm^{-2} over China, which over-predicted the OMI-derived NO_2 VCDs by 25.8% in the 2013 summer. Differences were also found over Japan, where the MIX emission inventory was applied for CMAQ. The differences between CMAQ and OMI were possibly due to the uncertainty of data sets used in the bottom-up emission inventory such as the activity data, emission factor, spatial allocation proxy, and other factors such as the meteorological field, deposition process, chemical reaction process and so on.

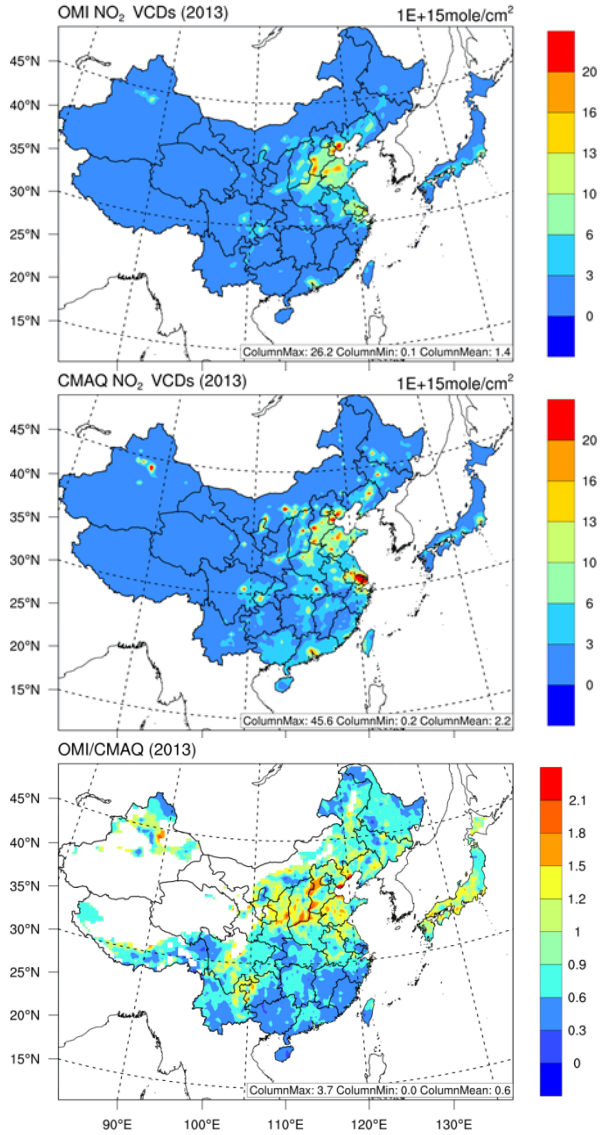

Figure 2. Spatial distributions of tropospheric NO_2 VCDs from OMI satellite

observations (top), CMAQ simulations (middle) averaged over the summer time for the year 2013. A ratio of OMI and CMAQ NO_2 VCDs was also presented (bottom) and grid cells where that modeled NO_2 VCDs below 1015 molec cm^{-2} were not plotted.

The relationships between CMAQ and OMI tropospheric NO_2 VCDs over China, the Greater Beijing region including Beijing, Tianjin and Hebei provinces (hereafter referred to as Jing Jin Ji), Yangtze River Delta and Pearl River Delta regions for the 2013 summer were further analyzed using the scatter plots. As shown in Figure 3, the correlation coefficients (R) ranged from 0.79 to 0.87, with slopes ranging from 0.96 to 1.61, which indicated that the model results were generally in good consistency with satellite retrievals. Over the whole China, the data points were above the standard 1:1 line, with slopes 1.15 and correlation coefficients from 0.79. The relationships for the key regions were also discussed here. For the Jing Jin Ji region, the CMAQ modeling showed a better agreement with the OMI-derived NO_2 VCDs with the slope 0.96 and correlation 0.81. The CMAQ modeling over the Yangtze River Delta region was 0.61 times larger than the OMI-derived NO_2 VCDs with a high correlation 0.82. The results for the Pearl River Delta region showed that the CMAQ modeling produced tropospheric NO_2 VCDs that were over-predicted by factors 0.27 compared to the OMI retrievals. In conclusion, the modeling results can be used for further studies.

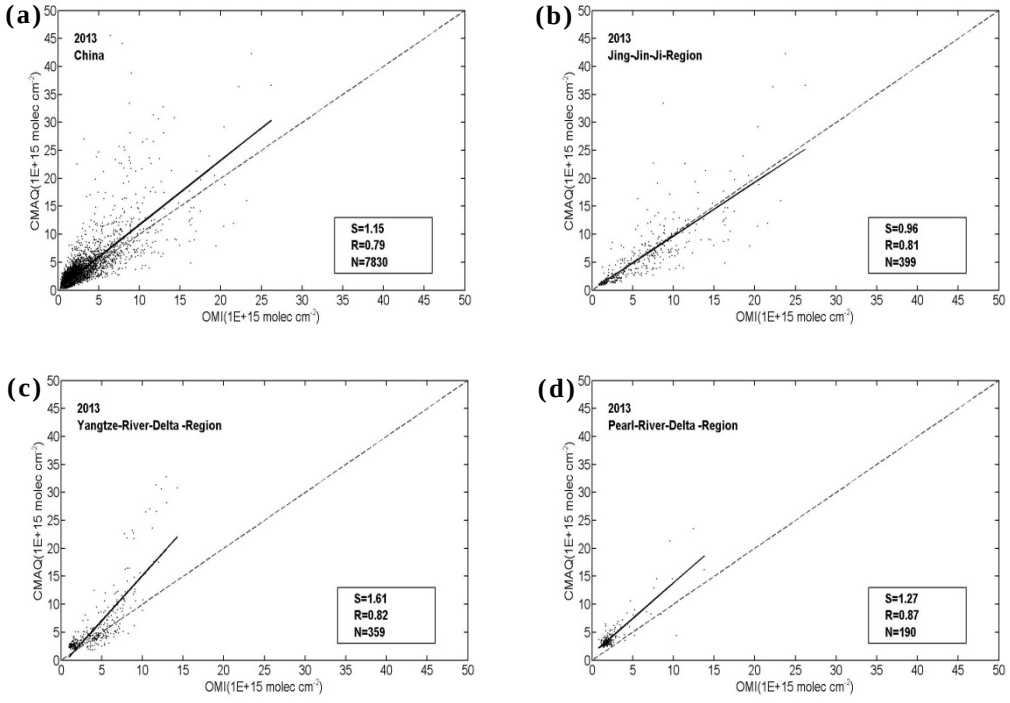

Figure 3. Scatter plots (solid lines) that reflected the correlation between the modeled and satellite tropospheric NO2 VCDs over (a) China (excluding the ocean area), (b) Jing Jin Ji region, (c) Yangtze River Delta region and (d) Pearl River Delta region in 2013 summer. Dashed lines was the standard plot y=x.

3.2 Model Validation for the year 2013 Based on the Comparison with EANET Observation

Figures 4 and 5 compared the temporal variations of surface NO_2 and PM_{10} concentration from June 1st to August 31st in 2013 simulated by the CMAQ model with observation at the EANET Zhuhai-Xiangzhou and Xiamen-Hongwen sites. For the Zhuhai-Xiangzhou site, the modeled hourly NO_2 and PM_{10} concentration showed good agreement with the observations. The modeled NO_2 concentration at Zhuhai-Xiangzhou basically underestimated the observation by 22.5% during the period. The model overestimated the PM_{10} during the whole three months especially in polluted days, leading to a large overprediction of total mass concentration by 45.8%. For the Xiamen-Hongwen site, the modeled hourly NO_2 and PM_{10} concentration greatly overestimated by a factor of 2 and 2.3, respectively. There was also a large discrepancy in the temporal variation. There might be large uncertainty in the meteorological fields as well as emissions for these two sites.

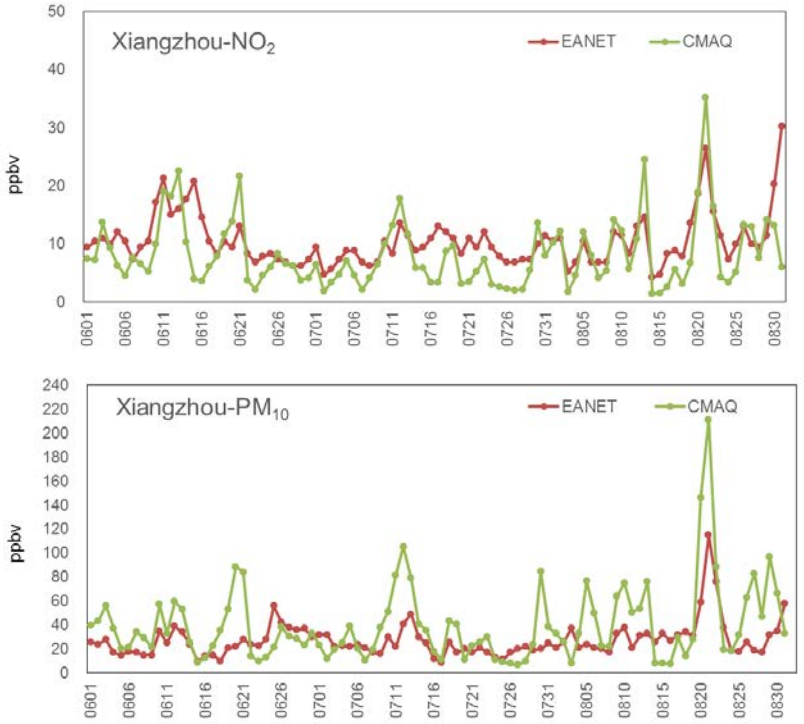

Figure 4. Temporal variations of NO2 and PM10 from June 1th to August 31th in 2013 at Zhuhai-Xiangzhou site

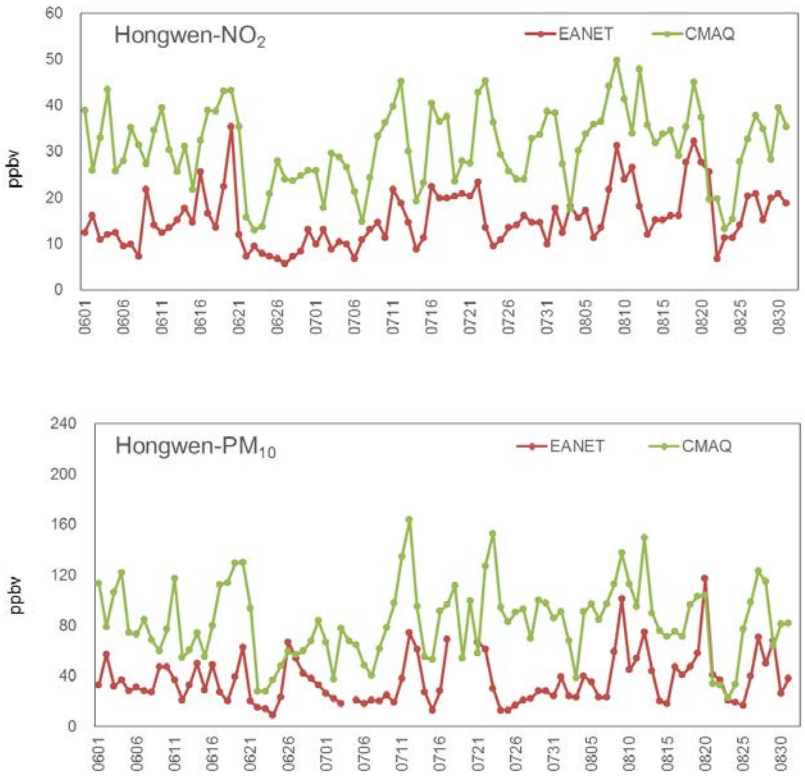

Figure 5. Temporal variations of NO2 and PM10 from June 1th to August 31th in 2013 at Xiamen-Hongwen site

3.3 Inter-annual Variation of OMI Tropospheric NO_2 Retrievals and Influence of Meteorological Conditions and emission

First, the tropospheric OMI NO_2 column retrievals were analyzed during the summer from 2013 to 2015 (see Figures 6a-c (right)). The seasonal averaged OMI NO_2 VCD retrievals for the summer in 2013, 2014 and 2015 over China were 1.86, 1.94, and 1.78 x 10^{15} molec cm^{-2}, respectively. It explained that compared to 2013 summer levels, there was an increase of NO_2 VCDs by 4.26% for 2014 and a reduction by 4.45% for 2015. An obvious decline of NO_2 VCDs over most region of China was observed by satellite instruments for the year 2015. To further investigate the impacts of meteorological conditions, three year simulations in summer were conducted by using the fixed emissions for the year 2013 with the WRF meteorological fields for 2013, 2014 and 2015, respectively, as described in section 2.5.1. The spatial distributions of CMAQ simulated NO_2 VCDs using the same emissions were presented in Figures 6a-c (left) which showed the similar spatial distribution pattern during 2013 and 2015. Relative difference of CMAQ simulated (left) and OMI observed (right) NO_2 VCDs between 2014 and 2013 and between 2015 and 2013 were also shown in Figures 6d and 6e, respectively. The seasonal averaged CMAQ NO_2 VCD retrievals for the summer in 2013, 2014 and 2015 over China were 2.34, 2.40, 2.42 x 10^{15} molec cm^{-2}, respectively. These results indicated that meteorological conditions in 2014 and 2015 tended to increase the NO_2 VCDs over China compared to those in 2013. Following the equation (2) in section 2.5.1, the results show that due to the contribution of meteorological conditions, the NO_2

VCDs increased by 2.95% and 3.57% for the year 2014 and 2015 compared to the 2013 levels, respectively. And following the equation (3) in section 2.5.1, it can infer that the tropospheric NO_2 VCDs increased by 1.31% in 2014 and decreased by 8.02% in 2015 resulting from the NO_X emission variation since the year 2013. It suggested that majority of contribution to increase NO_2 VCDs in 2014 was from meteorological conditions, but NOx emissions also affected the increase of NO_2 VCDs. On the other hand, the large decreases of observed NO_2 VCDs in 2015 were due to the decreases of NO_X emissions which canceled the influences by meteorological condition increasing NO_2 VCDs. The year-to-year variation of NO_X emissions will be investigated in the next section.

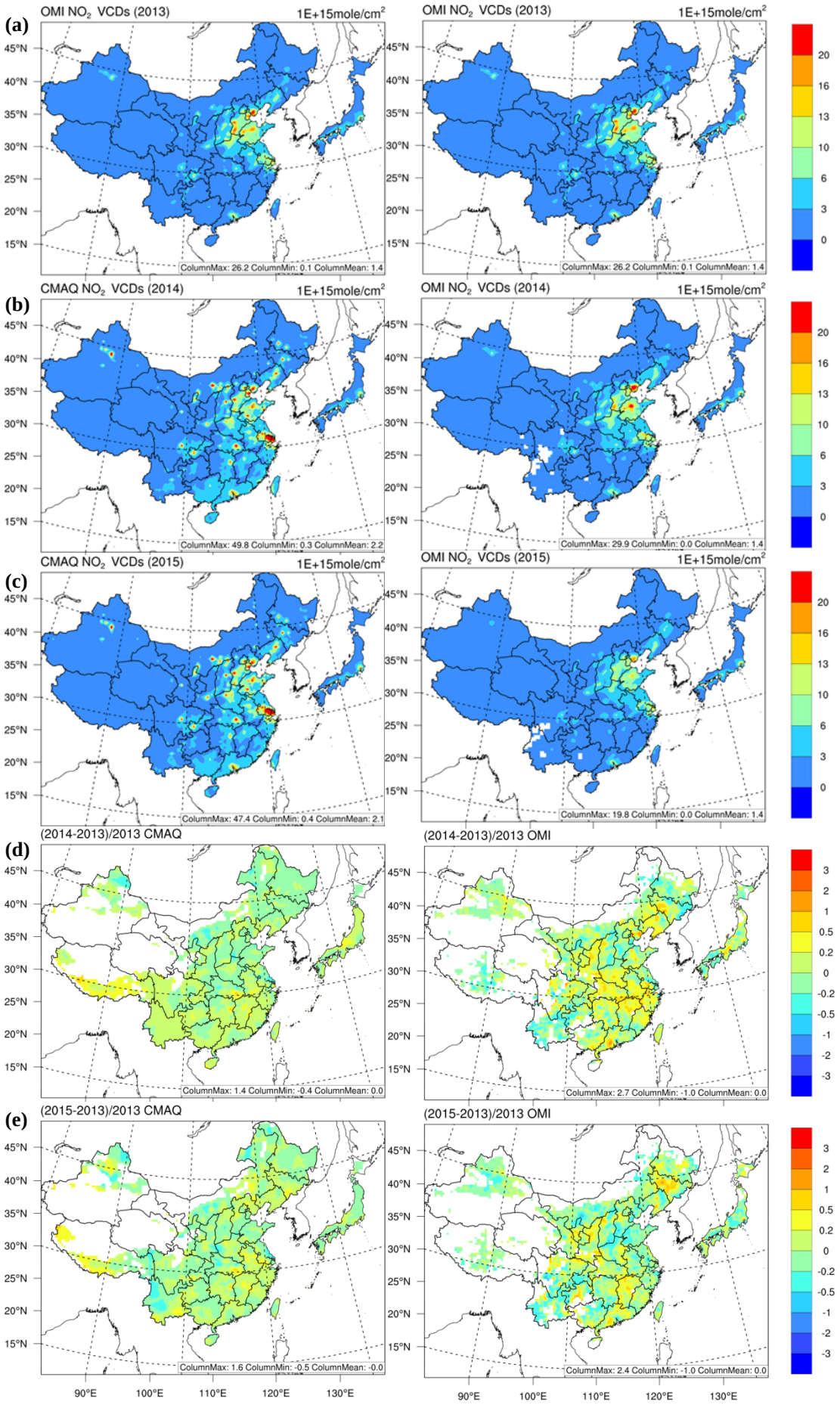
(a)
OMI NO2 VCDs (2013)
1E+15mole/cm2
ColumnMax: 26.2 ColumnMin: 0.1 ColumnMean: 1.4
OMI NO2 VCDs (2013)
1E+15mole/cm2
ColumnMax: 26.2 ColumnMin: 0.1 ColumnMean: 1.4
(b)
CMAQ NO2 VCDs (2014)
1E+15mole/cm2
ColumnMax: 49.8 ColumnMin: 0.3 ColumnMean: 2.2
OMI NO2 VCDs (2014)
1E+15mole/cm2
ColumnMax: 29.9 ColumnMin: 0.0 ColumnMean: 1.4
(c)
CMAQ NO2 VCDs (2015)
1E+15mole/cm2
ColumnMax: 47.4 ColumnMin: 0.4 ColumnMean: 2.1
OMI NO2 VCDs (2015)
1E+15mole/cm2
ColumnMax: 19.8 ColumnMin: 0.0 ColumnMean: 1.4
(d)
(2014-2013)/2013 CMAQ
ColumnMax: 1.4 ColumnMin: -0.4 ColumnMean: 0.0
(2014-2013)/2013 OMI
ColumnMax: 2.7 ColumnMin: -1.0 ColumnMean: 0.0
(e)
(2015-2013)/2013 CMAQ
ColumnMax: 1.6 ColumnMin: -0.5 ColumnMean: -0.0
(2015-2013)/2013 OMI
ColumnMax: 2.4 ColumnMin: -1.0 ColumnMean: 0.0
45°N
40°N
35°N
30°N
25°N
20°N
15°N
90°E
100°E
110°E
120°E
130°E
20
16
13
10
6
3
0
3
2
1
0.5
0.2
0
-0.2
-0.5
-1
-2
-3

Figure 6. Spatial distributions of tropospheric NO2 VCDs from CMAQ simulations using the fixed emissions in 2013 (left) and OMI satellite observations (right) averaged over the summer time for (a) 2013, (b) 2014, and (c) 2015. Relative differences of tropospheric NO2 VCDs of CMAQ (left) and OMI (right) for the summer in (d) 2014 and (e) 2015 compared to the 2013 levels were also presented. Grid cells where NO2 VCDs of the year 2013 were below 1015 molec cm-2 were not plotted.

3.4 Interannual Variation of OMI-based top-down NO_X Emission

The distributions of NO_X emissions constrained by OMI observed NO_2 VCDs and CMAQ were estimated during the summer for the year 2013, 2014, and 2015 following the approach described in section 2.5.2. The top-down emissions in 2013, 2014 and 2015 were shown in Figs. 7a-c. The relative differences between 2014 and 2013 and between 2015 and 2013 were shown in Figs. 7d-e that presented a slight change in NO_X emissions for 2014 and a strong decline for 2015 compared to the 2013 levels over China. The summer time total of satellite-constrained national NO_X emissions for 2013, 2014, and 2015 were 1.43, 1.48, 1.27 TgN, respectively. These results indicated that the NO_X emissions in China during the summer time slightly increased in 2014, while decreased greatly in 2015 compared to 2013.

The OMI-based top-down estimates for 2013, 2014 and 2015 summer time were compared in a provincial level as shown in Figure 8. In this study, the satellite-constrained NO_X emissions in 2014 had a spatial inconsistency with obvious increase or decrease in different regions (the largest decrease of 26.97% for Shanghai, the largest increase of 22.56% for Shan'xi, and an increase of 15.95% for Beijing) compared to the posterior 2013 emissions. For the year 2015, there was a strong decrease in the Eastern part of China with 25.48% for Tianjin, 22.46% for Zhejiang, 19.26% for Henan, 17.34% for Shanxi, 15.23% for Shandong, 14.66% for Hebei and only a little increase of 1.11% for Shanghai. The reduction in NO_X was presumably the result of the implementation of emission reduction control measures such as, the installation of denitration devices for power plants, etc.

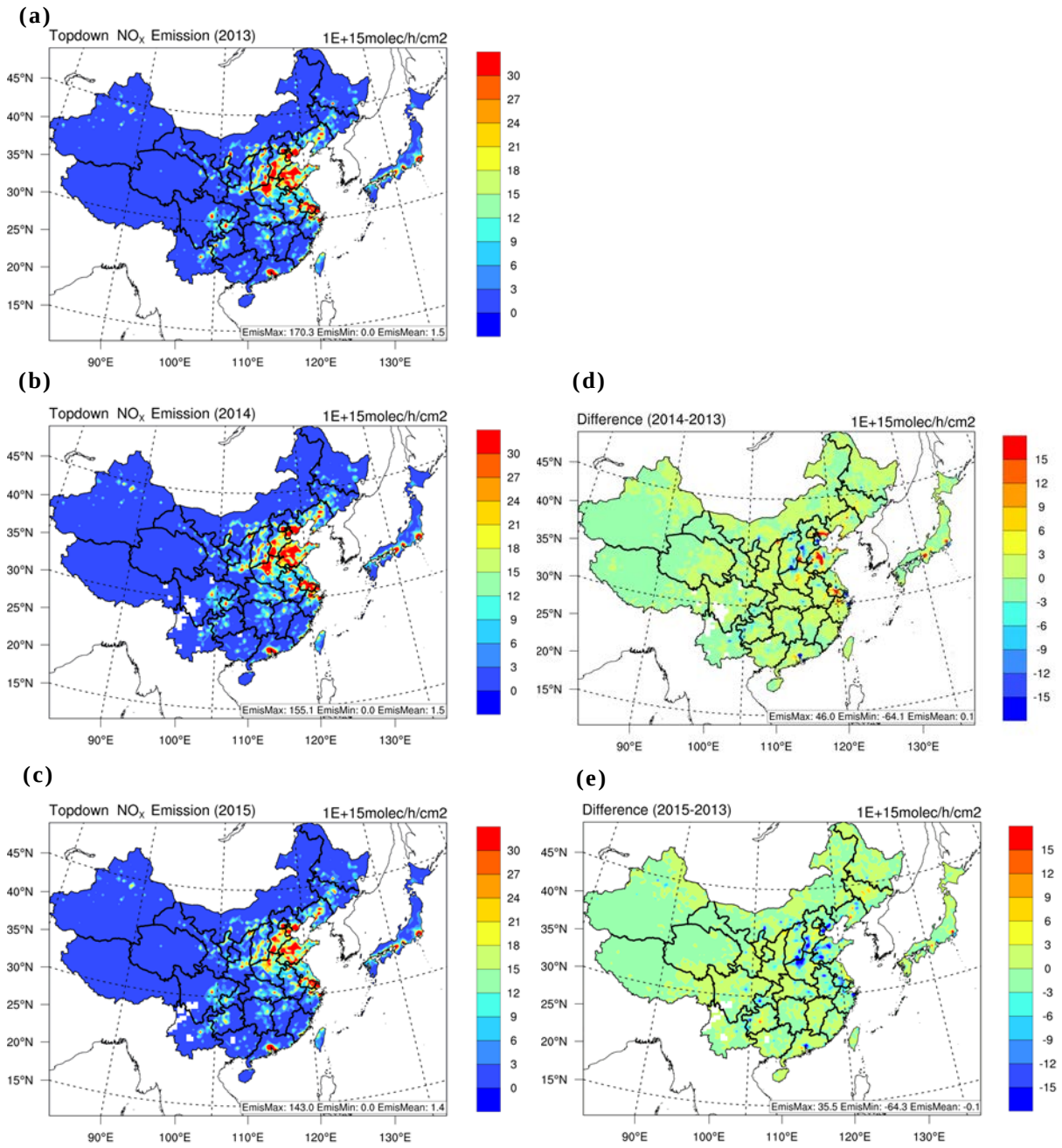

Figure 7. Spatial distributions of top-down NOX emissions for the year (a) 2013, (b) 2014 and (c) 2015 constrained by OMI satellite observations during the summer and the relative difference of (d) 2014 and (e) 2015 top-down emissions compared to the 2013 ones.

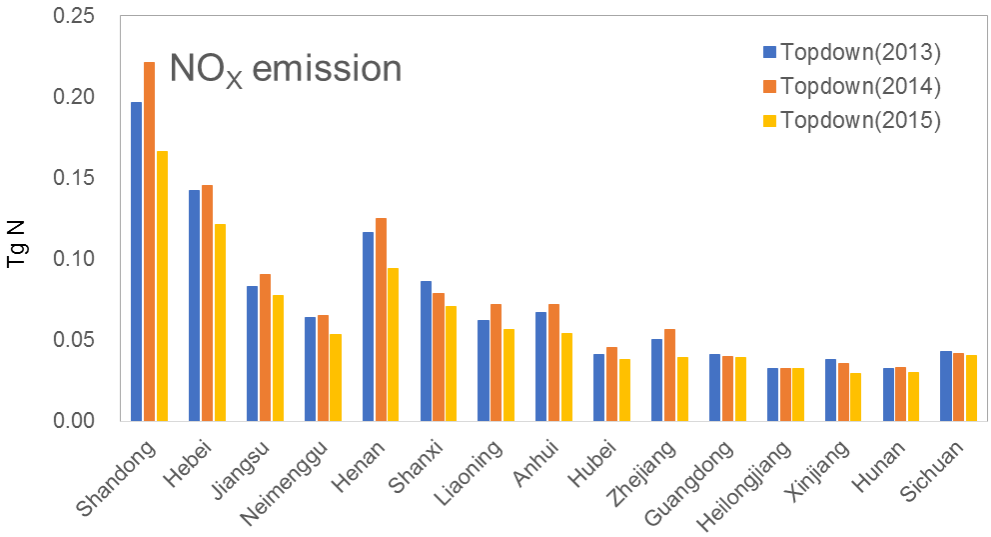

Figure 8. Provincial summer time totals (only top 15 in China is shown here) of OMI-based top-down emission for 2013, 2014 and 2015.

4. Conclusion

We examined the causes of inter-annual variation of OMI-observed NO_2 VCDs over China during summer time (June, July, and August) from 2013 to 2015 with the aid of a regional chemical transport model CMAQ. First, the CMAQ modeling system was developed and validated that the modeled tropospheric NO_2 VCDs in 2013 summer agreed well with OMI retrievals with the correlation coefficients (R) ranged from 0.79 to 0.87 and slopes ranging from 0.96 to 1.61 over China and the key regions, providing reliable validation of emission inventory. Available hourly observations of EANET sites were also used to evaluate the model results but more sufficient data were necessary for systematic comparison analysis.

Then, the influences of meteorological condition and emission on the inter-annual variation of NO_2 VCDs were examined by a sensitivity model simulation during the summer time of 2013-2015 with anthropogenic emissions fixed at 2013 levels. Compared to 2013 levels, an increase of NO_2 VCDs by 4.26% was observed by satellite for the year 2014, with the contribution of meteorological conditions and NO_X emissions variations 2.95% and 1.31%, respectively, which illustrated an increase of NO_X emissions in 2014. For the year 2015 compared to 2013, the meteorological conditions had a positive influence that brought an increase of NO_2 VCDs by 3.57%. On the other hand, for a significant NO_2 VCDs decline by 4.45% identified from satellite, it is indicated that the NO_X emission reduction contributed an 8.02% decrease to the NO_2 VCDs variation.

To further evaluate the influences of surface NO_X emissions, top-down NO_X emissions constrained by OMI observed NO_2 VCDs were estimated and analyzed. Top-down NO_X emissions were inferred from OMI observations in a simple inverse approach with a regional chemical transport model CMAQ based on an assumption of the linear relationship between tropospheric NO_2 VCDs and surface NO_X emission represented as

a ratio α. With the inverse approach applied, the summer time totals of top-down NO_X emission for 2013, 2014, 2015 were 1.43, 1.48, 1.27 TgN, respectively. For the year 2014, the satellite-constrained provincial NO_X emission had a spatial inconsistency of variations in different regions ((the largest decrease of 26.97% for Shanghai, the largest increase of 22.56% for Shan'xi) compared to the posterior 2013 emission. The top-down NO_X emission in 2015 had an obvious decrease in the Eastern part of China compared to the posterior 2013 emission. These results may have important policy implications.

Limitations in the current work applying a linear relationship between NO_X Emissions and NO_2 VCDs could introduce systematic error because of the non-linear chemistry reaction and non-ignorable NO_X transportation. The uncertainty of satellite retrievals and model simulation and the development of the non-linear relationship approach should be developed in the future study. This study focuses on summer time. Simulation at long time series should be conducted in the future study. Besides that, NO_2 VCDs are also influenced by aloft NO_X emissions from such as lightning and air planes. However, CMAQ simulation of this paper uses and the top-down method adjusts only surface emissions. This can be one of causes of uncertainty in our method and should be investigated in the future study.

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Investigation of atmospheric input and runoff discharge of sulfur and nitrogen compounds as the balance components of Komarovka river catchment by long-term observations at Russian EANET Primorskaya station (for 2005 – 2015)

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Abstract

The ultimate goal of ecological research for EANET is to evaluate an effect of atmospheric pollution on the properties of regional environment ecosystems and their changes under air pollutant impact. This study presents the first estimation of the annual input-output budget components of sulfur and nitrogen compounds at the natural catchment area of a small river in the Far East of Russia for 2005-2015. The assessment is based on the considered principal individual components of pollutant transfer: atmospheric wet and dry deposition fluxes and river runoff discharge.

The study was conducted for Komarovka river catchment where the Russian EANET ecological site Primorskaya was established in 2002. Most of the data for the calculations were collected as part of regular performing at the EANET program measurements. The procedures of atmospheric deposition flux estimation were done in line with EANET Technical documents.

Our results of budget components estimation demonstrate that the values of sulphur compounds (SOx) output from watershed were higher than their input for the same period for the most of years. In opposite, for nitrogen compounds (NHx and NOx) the atmospheric input exceeded the output at river catchment. Presented results can be applied for the further elaboration of pollutant migration in ecosystem and out as well as to be used for comparisons with similar studies at the other EANET ecological monitoring sites.

Keywords: Wet deposition, Dry Deposition flux, Runoff Discharge, Input/output balance

1. Introduction

To estimate a negative effect of acidification on ecosystems, it is necessary to know the exact amounts of acidifying compounds coming to and going from the system (e.g. chemical compound budgets). In this sense, a small river catchment, which is located far from direct emission sources and has no local anthropogenic influence like effluent or drainage due to plowing, can become a good model area for study.

The ecosystems in Russian Far East are suspected to be under the impact of changing transboundary air pollution and, hence, related effect of air pollutant loads. The present study is conducted for Komarovka river catchment at the Russian Far East where the EANET ecological site Primorskaya was established in 2002.

The river drains the forested area, which seems to be of a quasi-natural condition due to very limited human activities. There were no pollution sources of the river water found in the vicinity of observation site and upstream. Therefore, the levels of pollutant concentrations in stream water may depend on atmospheric inputs, including rainfalls over the watershed, snow melting waters, and dry deposition of gaseous and particulate matters.

The monitoring on wet deposition and dry deposition at the Primorskaya had been started in 2000-2002, however Dry Deposition Flux (DDF) of any pollutants have never been calculated for this site before and pollutant budget studies have never been performed for the catchment by now.

To do such studies for the first approach we used a simplified input-output budget calculation for pollutants. The budget estimation assumed: the input consists of total (dry and wet) atmospheric deposition while the output is presented solely by the discharge of compounds with the stream water. The budgets were calculated at annual basis for major sulfur and nitrogen compounds for 2005-2015.

The main objective of the research on Komarovka river catchment is to evaluate effects of atmospheric deposition on inland aquatic and soil properties changes. However, due to the short time of fellowship and large scale of the problem only several important tasks were addressed:

- To calculate total atmospheric deposition fluxes and surface water discharges of sulfur and nitrogen compounds for the Komarovka river catchment area.
- To evaluate inter-annual variations of concentrations and fluxes of sulfur and nitrogen compounds in atmospheric deposition (air and rainwater) and in river water.
- To evaluate input and output balance of sulfur and nitrogen compounds.

2. Data and methods

2.1 Wet and Dry Deposition Fluxes

The samples of atmospheric wet deposition used for estimation are taken on daily basis (24 hours from 9 a.m.) at open area of meteorological station at Primorskaya site. All the chemical analyses of samples were performed at Primorsky Environmental

Monitoring Center, Roshydromet (PCEM) in accordance to the existing techniques adopted in EANET. Ready-processed data on monthly and yearly wet deposition fluxes was acquired from yearly EANET Data Reports (EANET. 2006 - 2016)

Estimation of dry deposition flux was performed according to the Technical Manual on Dry Deposition Flux Estimation in East Asia (EANET. 2010). By using it, the Dry deposition flux of gaseous and particulate species is calculated by the inferential method as the product of air concentration and respective deposition velocity (Vd):

$$F_i = V_{d_i} \times C_i \qquad (1)$$

where F_i is flux of *i* species and C_i is concentration of *i* species. After hourly or daily deposition velocities are calculated, the deposition velocities are averaged in the time resolution of air concentration.

Air samples are taken at Primorskaya site twice a month (continuously using a four-stage filter pack). So concentrations of gaseous and particulate compounds are taken as average two-week values.

For calculation of deposition velocity, we used the Microsoft Excel macro program provided by EANET Network Center. According to the requirement for the input data, values of every parameter should be input into the excel table for every hour of studying period. Among meteorological data needed (Temperature [˚C], Relative Humidity [%], Wind Speed [m s^{-1}], Solar radiation [MJ m^{-2}], Precipitation [mm], Cloud coverage [0~10]), only solar radiation values were available on hourly basis and other parameters are all on daily basis. Therefore, after the changes of processing format for calculation, the averaged parameter values ("Daily") were applied for the program performance. Additionally we compared results that were acquired by using "Daily" parameters and those by "Hourly" parameters that were made from daily values by interpolation.

The type of vegetation growing on the investigated territory also plays a significant role for DDF estimation. The most of the territory of the Komarovka river catchment is covered with deciduous forest. Open areas are negligible. Therefore, we assumed the catchment area fully covered by forests and additional calculations for open (grass type) areas were not to be carried out.

Data on vegetation was acquired from EANET Data Report 2006 (EANET. 2006). Values of land-use categories were used for Vd calculations as follows in Table 1.

Table 1. Values of land-use categories

Ref Height	40 m
WS (wind speed) Height	40 m
Canopy height	30 m
Displacement Height	0.7Canopy height
Roughness height	1.05 m (EANET. 2010)

2.2 Chemical compounds runoff

Water sampling for analysis of concentrations and water discharge measurements are carried out 5 times a year with the correspondence to main stream hydrological regime periods (spring flood, summer pluvial high waters, and baseflow or mean water

periods in winter or summer) of the Komarovka River. Additionally, data on daily water discharge for 2005, 2010 and 2015 were obtained at the hydrological station "Tsentralny", situated right downstream the water sampling point.

For runoff flux estimation, an interval-representative (I-R) method was used. Daily runoff flux was calculated directly for the days of sampling as a product of concentration by water discharge. For the whole periods between sampling, the average of the two closest consequent values of compound concentrations and water discharge was taken.

Data for 2008 are missing from calculations because water discharge values were not estimated for that period. Water sampling in 2005 started only in September, so data from that year is also excluded from the balance assessments.

With available data on daily water discharge for 2005, 2010 and 2015, we tried to apply L-Q method. We used the equation of dependence of runoff (Load - L) on water discharge (Q). Based on the available values of compound concentrations in water and the respective water discharge (for all the years from 2005 to 2015), dependencies were estimated. The most indicative form for those dependencies is expressed by the power function (2).

$$L = aQ^n \qquad (2)$$

where L is observed runoff by river (Load), mmol s^{-1}; Q is water discharge, m3 s^{-1}; a, n – approximation parameters.

The (2) equation of the function was used for estimation of the daily water discharge values. Annual runoff from the catchment area for 2005, 2010 and 2015 was calculated based on the sum of daily runoff values.

2.3 Input and output balance

For the first approach, we used a simplified input-output budget calculation for pollutants. The budget calculation assumed: the input consists of total (dry and wet) deposition while the output is presented solely by the discharge with the stream water.

Using hydrological modeling on GIS software (GRASS ver. 7.0.5), catchment area of the Komarovka River upstream of the sampling point was estimated as approximately 155 km^2

3. Results and discussion

3.1 Wet Deposition Flux

The pH level of rainwater can be considered the main indicator of atmospheric acidification. At Primorskaya the average annual pH in the atmospheric precipitation ranged from 4.63 (2011) to 5.47 (2014). No statistically significant trend was found for pH, but one can see a tendency to a gradual increase in the pH value for the period from 2005 to 2015 (Figure 1). The similar increasing trend of low significance (α=0.1) can be seen for the similar characteristic – pAi. pAi is the hypothetical pH of atmospheric precipitation water if no neutralization takes place for both sulfuric and nitric acids. Numerically:

$$pA_i = -log\left(\left[nss\,SO_4{}^{2-}\right] + \left[NO_3{}^{-}\right]\right) \quad (3)$$

Seasonal variations can be traced by the monthly pH fluctuations with maximum values in January and February and minimum in September-October (Figure 2). It is also possible to distinguish an increasing tendency for the pH on monthly values since 2008.

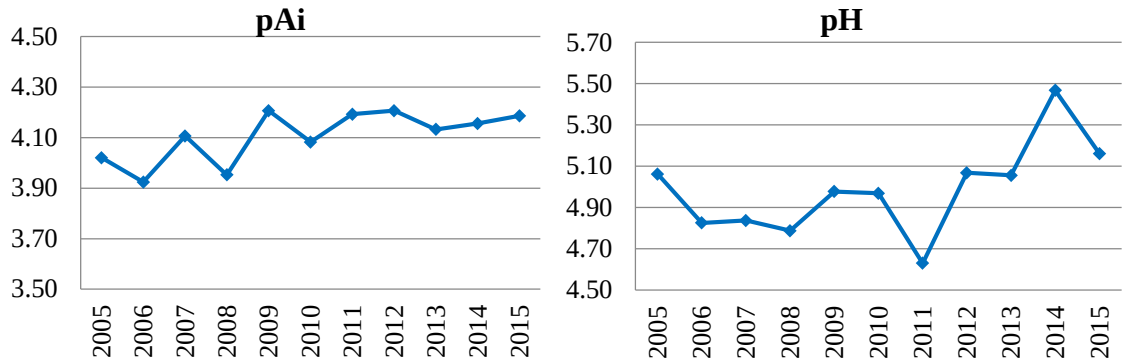


Figure 1. Changes in average annual pH and pAi of atmospheric precipitation

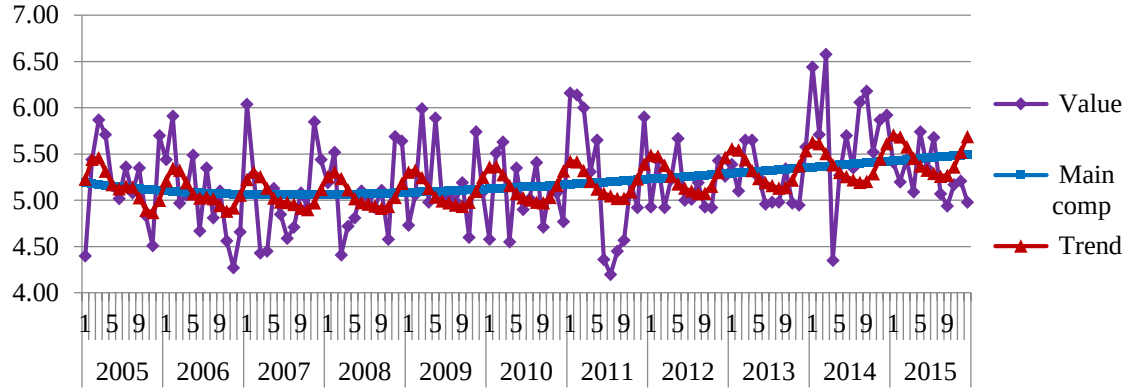


Figure 2. Changes in monthly mean pH values of atmospheric precipitation by MSC-E trend tool (Shatalov et al, 2015)

Significant trends in the weighted average annual concentrations (according to the Mann-Kendall test) for the compounds were observed neither for precipitation, nor for the substances concentrations in atmospheric precipitation, nor for their wet deposition. However, it is possible to notice weak decreasing tendencies in concentrations for sulfur and nitrogen compounds (Figure 3). For the values of wet deposition, such tendencies aren't observed (Figure 4).

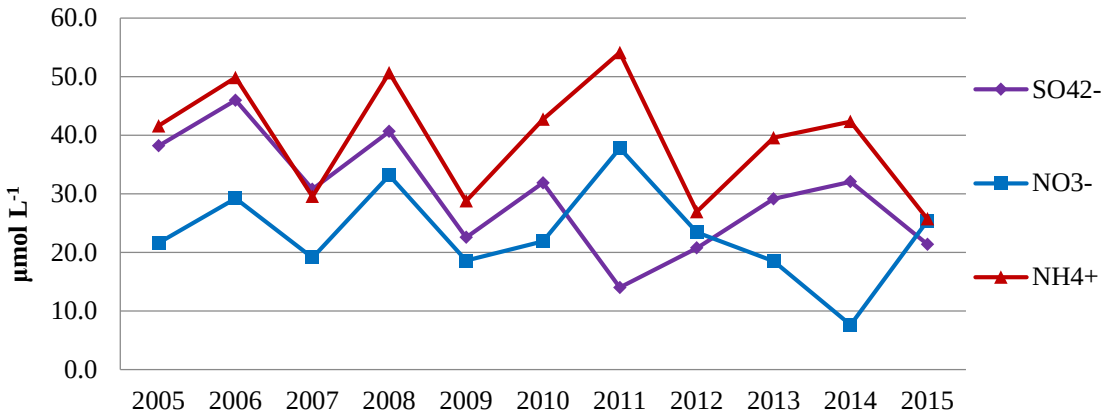


Figure 3. Changes in the average weighted annual concentrations of sulfates and nitrates in atmospheric precipitation

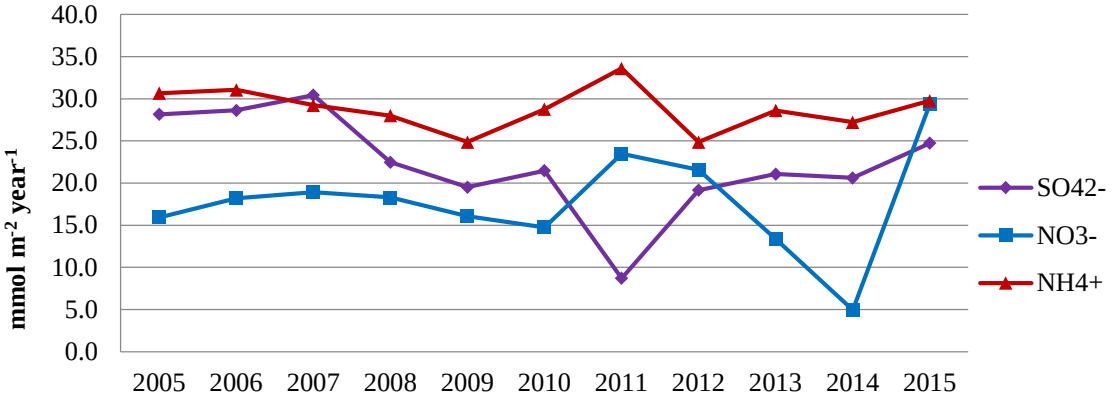


Figure 4. Changes in the amount of wet deposition flux of sulfates, nitrates and ammonium

3.2 Dry Deposition Flux

The most of the reported meteorological data we have were only daily average. So at the first we had to compare whether their use will bring a mistake or bias of calculations. The comparison was made between the two kinds of results for 2010-2015: the one was calculated with daily average values while approximated hourly data (obtained daily average values were interpolated for 24 hours with 1 hour step) were applied for another approach. As can be seen at Figure 5, the values for "daily" calculations exceeded "hourly" with the maintaining a high correlation. The overestimation of DDF by using “daily” values was about 22% on average. This could be caused both by excessive averaging of values without taking into account the intraday changes of Vd speed when using "daily" data, and by program inadequacy due to adopting the model Macros file for "daily" data application.

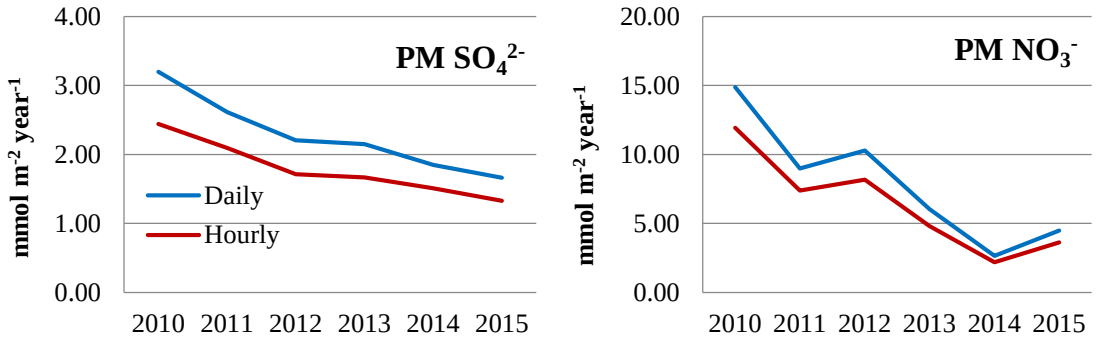
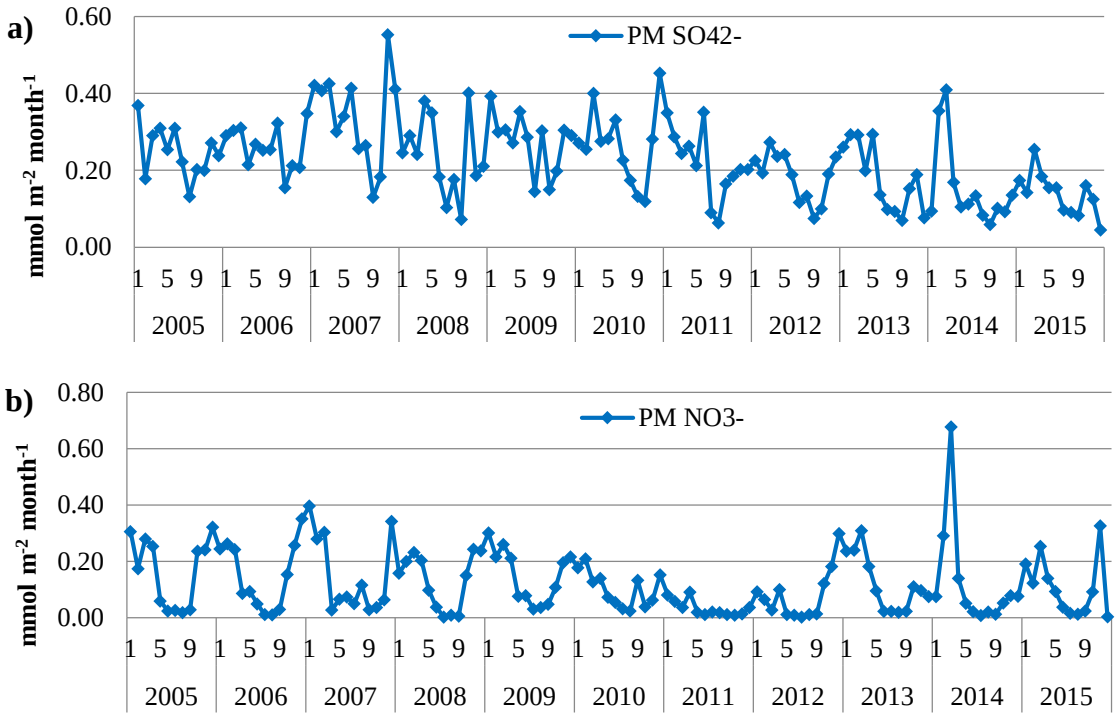


Figure 5. Comparison of the results of dry deposition flux calculations using daily and hourly data

Monthly values of the dry deposition fluxes of sulfur and nitrogen compounds carried by aerosol particles, calculated using the daily parameter values show a decreasing tendency. The maximum sulfate flux by particles is 0.55 mmol m^{-2} $month^{-1}$ (October 2007), the minimum - 0.04 mmol m^{-2} $month^{-1}$ (December 2015) (Figure 6, a). The maximum flux of nitrate was 0.68 mmol m^{-2} $month^{-1}$ (March 2014), the minimum - 0.04 mmol m^{-2} $month^{-1}$ (December 2015) (Figure 6, b). The maximum flux of ammonium was 0.96 mmol m^{-2} $month^{-1}$ (October 2007), the minimum - 0.02 mmol m^{-2} $month^{-1}$ (December 2015) (Figure 6, c).

For nitrogen compounds, a seasonal dependence is evident with the highest values during the cold season and the lowest during the warm period (Figure 6). The reason for it is yet to be determined and needs to be investigated further.



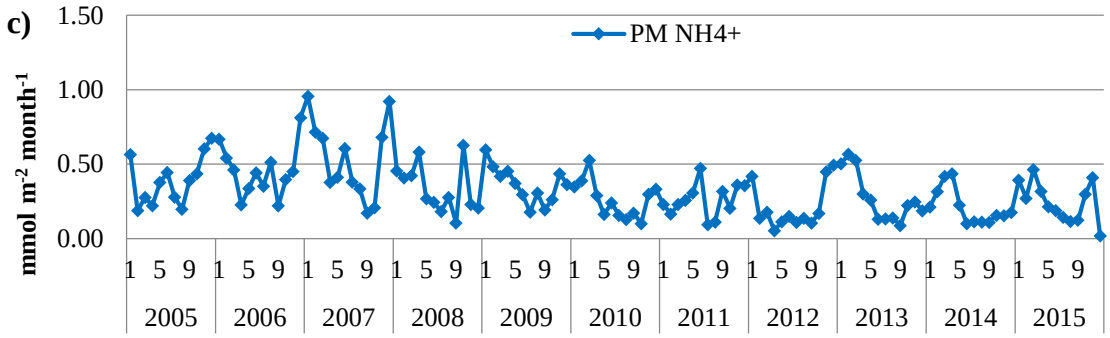


Figure 6. Fluctuations of average monthly values of dry deposition flux values for PM sulfate ions(a), nitrate ions(b) and ammonium ions(c)

Gaseous components make the major contribution into the DDF (Figure 9). Thus, for sulfur compounds, the total contribution of sulfur dioxide is about 75%. For nitrogen compounds it is even higher: the contribution of ammonia is about 77%, and the contribution of nitrogen dioxide is 80%.

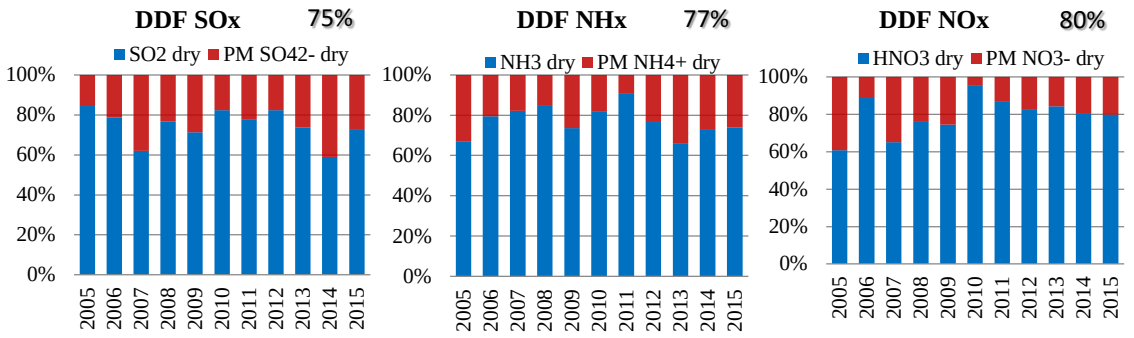


Figure 7. Gaseous and PM contribution to dry deposition fluxes for SOx, NHx and NOx

The Mann-Kendall test for annual values of the DDF showed declining trends with high level of significance for sulfates, ammonium PM ($\alpha = 0.01$), nitrates and gaseous sulfur dioxide ($\alpha = 0.05$) (Table 2), although trend in air concentrations is detected ($\alpha = 0.05$) only for sulfates.

Table 2. Results of Mann-Kendall Test and Sen's Slope Estimates for the annual DDF

Time series	n	Test Z	Significance level, α	Q
SO_2	11	-2.18	0.05	-0.819
HNO_3	11	0.62	-	0.202
NH_3	11	-1.40	-	-1.397
SO_4^{2-}	11	-2.80	0.01	-0.168
NO_3^-	11	-2.34	0.05	-0.060
NH_4^+	11	-2.65	0.01	-0.275
SOx summ	11	-2.18	0.05	-0.956
NOx summ	11	0.47		0.146

NHx summ	11	-1.71	0.1	-1.672

* Mann-Kendall Test and Sen's Slope Estimates for the Trend of Annual Data performed with MSExcel template Makesens 1.0

3.3 Wet and Dry Deposition Fluxes

Comparison of deposition fluxes shows that for the most of the years wet deposition exceeds the dry ones, sometimes by several times (Figure 8). However, for some years dry deposition can be equal or exceed wet deposition regardless of the amount of annual precipitation. For example, it is observed in 2011 for sulfur compounds and in 2014 for nitrogen oxides (due to a sharp decrease of the wet deposition); or in 2010 for nitrogen oxides, and 2007, 2011 for ammonia compounds (due to the increase of the dry deposition flux because of either high concentration of airborne compounds as in 2007 or favorable meteorological conditions like in 2010).

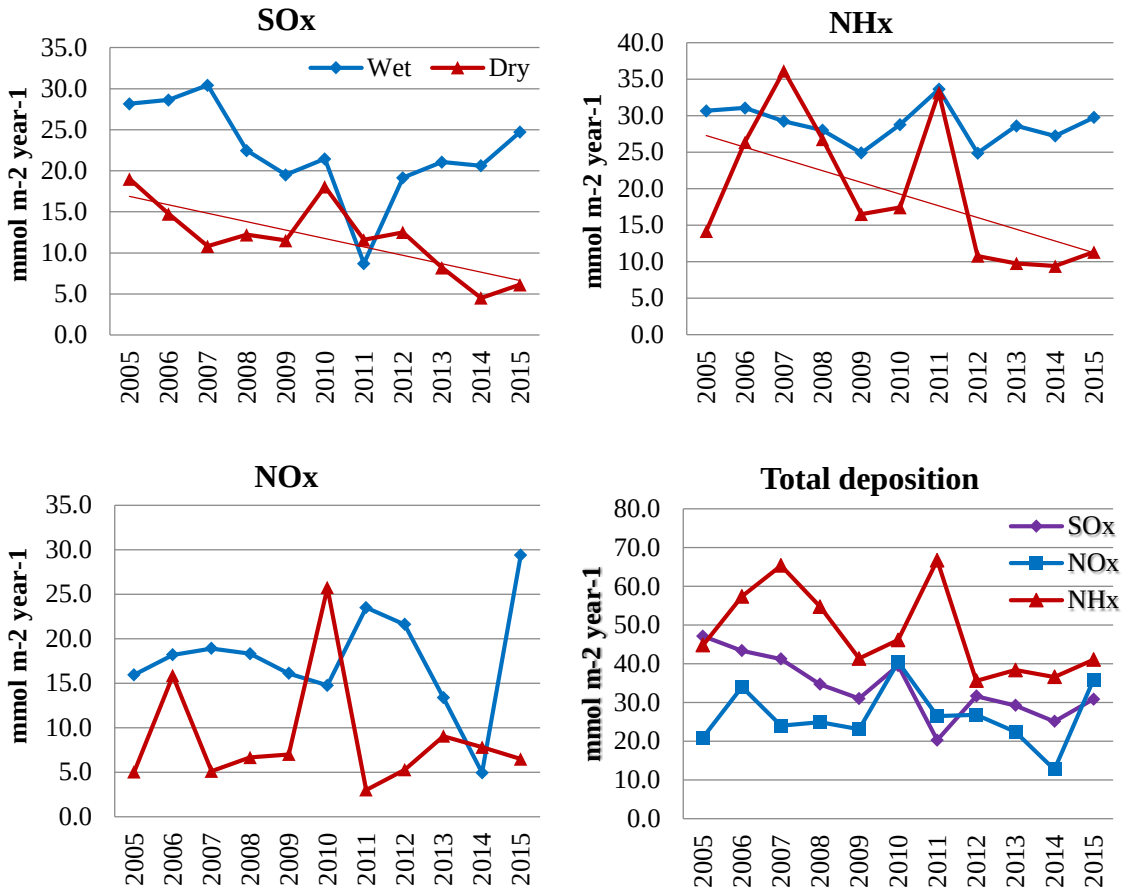


Figure 8. Comparison of the dry and wet deposition fluxes and total deposition

3.4 Chemical compounds runoff

A sharp increase in the chemical compounds runoff from the catchment area can be noted for 2012 (Figure 9, b) due to extreme water discharge (Figure 9, a). This may be due to both a real increase in runoff and the considerable high errors of applied I-R method.

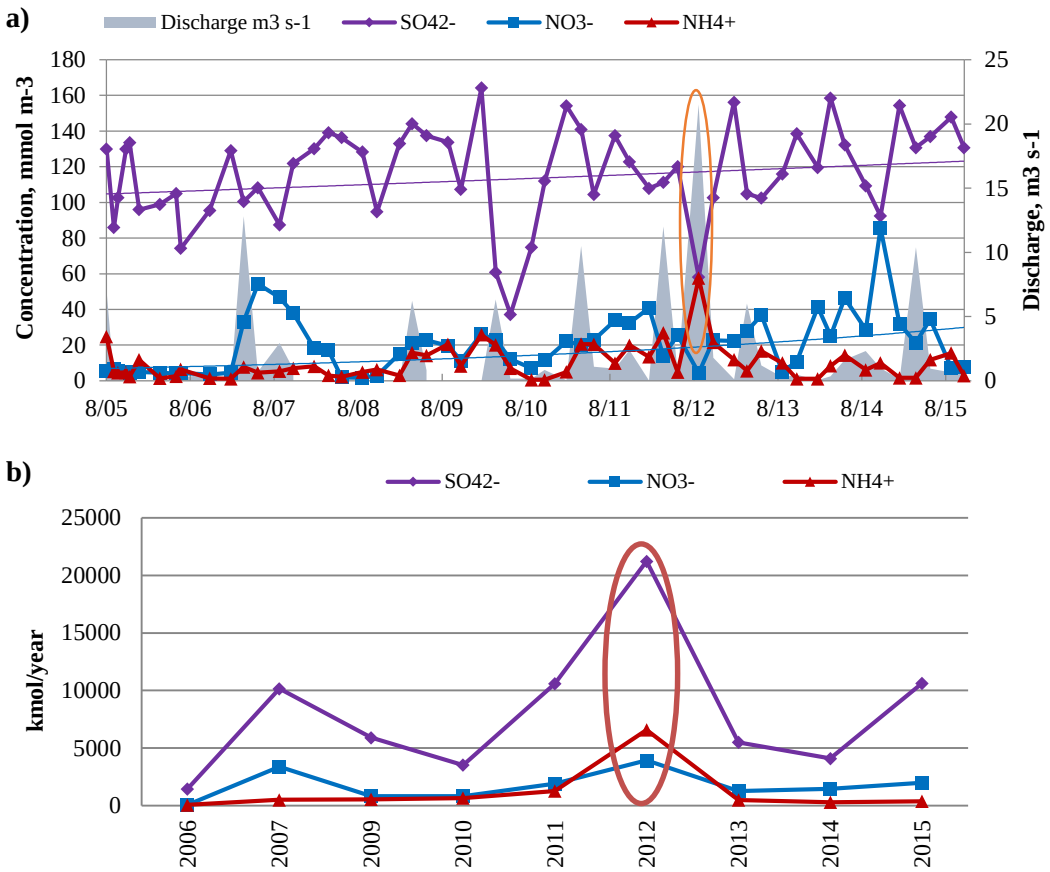
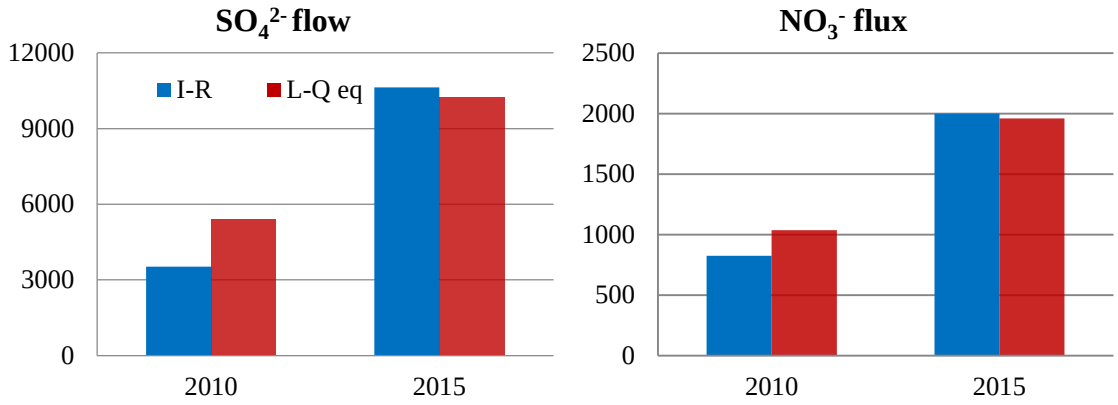


Figure 9. Time series of chemical compounds concentrations (a) and Estimated annual chemical compounds runoff (b)

To check the latest hypothesis, another estimation method was used (L-Q method) for 2010 and 2015. The two of applied calculation methods can provide both very close results (2015 for nitrate ions) and differing by several times (2015, for ammonium ions) (Figure 12). L-Q method for flow calculation of compound runoff seems able to provide more reliable results than interval-representative flux (I-R) method (Ide et al, 2003). However, it would be useful to continue comparing methods using the increased frequency of sampling and water discharge measurements.



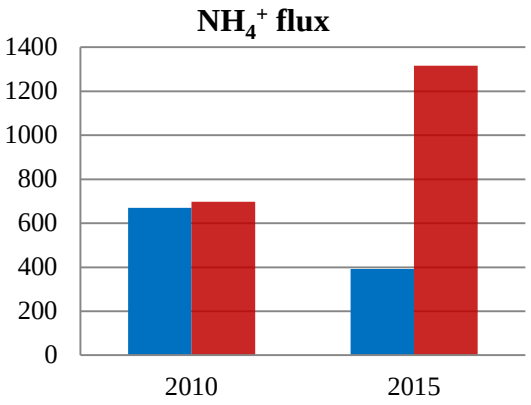


Figure 10. Comparison of results by I-R (blue) and L-Q (red) methods of annual runoff calculations for Komarovka river basin (kmol year^{-1})

3.3 Input and Output balance

Water balance of the catchment area showed that the amount of precipitation is approximately twice higher than the calculated water discharge (Figure 11). Thus, we can assume both a high level of evaporation, groundwater discharge and other ways of runoff.

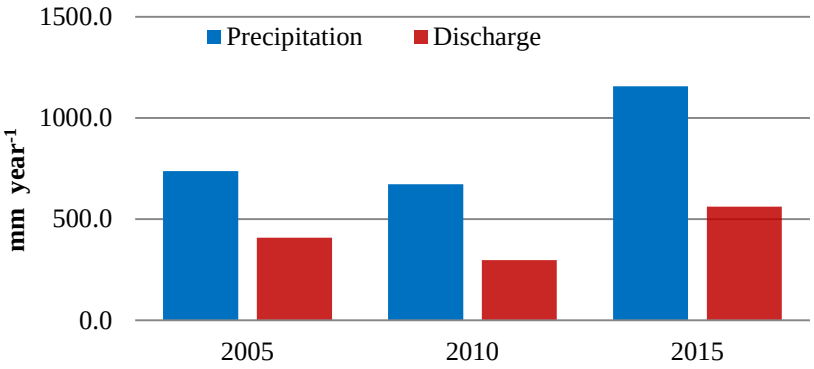


Figure 11. Water input/output budget of the Komarovka river basin for 2005, 2010 and 2015

There is a periodic change in the input-output interrelation of sulfur compounds with a predominance of either deposition flux or runoff (Figure 12). On average, during the study period, a sulfur compounds output (51.3 kmol km^{-2} year^{-1}) prevails over the input (34.1 kmol km^{-2} year^{-1}). For nitrogen oxides, input exceeds the output: average falls during the study period (26.8 kmol km^{-2} year^{-1}) is more than twice higher than the runoff (10.9 kmol km-2 year-1). Input of the ammonia for all years is higher than the runoff. With the exception of 2012, when, due to a sharp increase in output, it was more than deposition: the average input (47.4 kmol km^{-2} year^{-1}) is significantly higher than the runoff (7.5 kmol km^{-2} year^{-1}) during the study period.

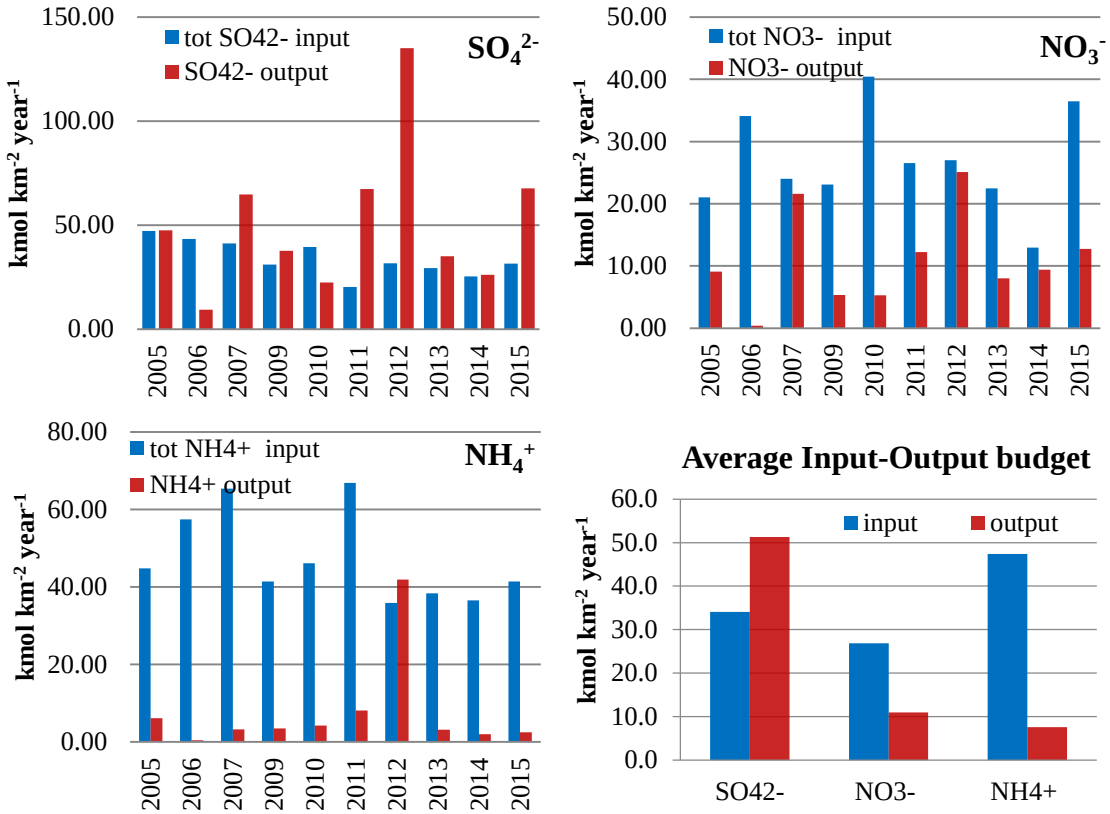


Figure 12. Comparison of input and output of chemical compounds and their multiyear average annual input-output budget

A similar case with higher sulfur compounds runoff compared to input were highlighted at in Europe (Vuorenmaa et al, 2017) and North America (Mitchell et al, 2013). In those cases, it was assumed that sulfates were mobilized from the soils of the catchment areas, where they had been accumulated for the previous period of high emissions and deposition of pollutants. There were no studies on the input-output balance for Komarovka watershed before, so it's yet problematic to determine the source of sulfur compounds in the runoff yet.

4. Conclusion

No significant or evident trends of pollutants concentrations were found in wet deposition. Declining tendencies of annual dry deposition can be retrieved for sulfur and nitrogen compounds with the use of Mann-Kendall test, although there are almost no trends in air concentration time series. Decreasing of total SOx DDF can be supposed.

The relative contribution of gaseous SO_2 to the Dry Deposition amount of oxidized sulfur is about 75%.

Annual wet deposition is usually higher than dry one but in some years dry deposition was estimated to contribute more into total input of pollutants.

Due to the small number of surface water samples the application of interval-representative method can give overestimated or underestimated results.

Budget component estimation demonstrates that the output of sulphur compounds (SOx) were higher than the input for the most of years at river catchment. In opposite,

for nitrogen compounds (NHx and NOx) the input exceeded the output.

Future perspectives

In the future, it is necessary to continue the research on the effect of atmospheric deposition at the Komarovka River catchment. In particular, the study on the chemical compound budgets could include the parts of chemical and biological cycling of pollutants.

It is worthwhile to refine the data processing and introduce new experimental data, in particular: to calculate the dry deposition by hourly data, to increase the number of samples of surface water, to use the daily data of river water discharge, to collect data on land use of different categories. It is advisable to introduce additional fluxes into the account of the balance of substances in the form of throughfall depositions, soil solutions, etc.

Acknowledgements

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The impacts of exposure to fine particulate matter on premature mortality in Bangkok

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Abstract

Fine particulate matters ($PM_{2.5}$) cause a potential adverse impact to human health, including premature mortality under long-term exposure. Based on the available of $PM_{2.5}$ monitoring concentrations, this study estimated the premature mortality attributable to $PM_{2.5}$ in Bangkok, Thailand from 2010 to 2017. The study identified population in Bangkok is exposed to $PM_{2.5}$ annual average concentrations of 14-55 $\mu g/m^3$. Then, the annual concentration of $PM_{2.5}$ and premature death attributable to $PM_{2.5}$ were estimated at district level. Results showed that in 2017, if the $PM_{2.5}$ annual average concentration in Bangkok had reduced to World Health Organization Guideline level of 10 $\mu g/m^3$, it would have lower the risk of premature mortality attributable to $PM_{2.5}$ for 71, 74, 46 and 73 percent of the four $PM_{2.5}$ related disease for adults chosen for this study: Chronic obstructive pulmonary disease (COPD), Lung Cancer (LC), Ischemic heart disease (IHD) and Stroke, respectively. The findings of this study suggest that strict controls of $PM_{2.5}$ concentration emission in Bangkok are required to reduce the numbers of mortality attributable to $PM_{2.5}$.

Keywords: $PM_{2.5}$ concentration, Long-term exposure, Premature mortality, Bangkok

1. Introduction

Long-term exposure to ambient fine particulate matter which aerodynamic diameter less than or equal to 2.5 μm so called $PM_{2.5}$ is a one of the major risk factors to harmful human health and death. World health Organization (WHO) reported 4.2 million premature deaths due to cardiovascular and respiratory disease, and cancers worldwide per year in 2016 contributed by exposure to ambient $PM_{2.5}$ in both cities and rural areas which cause (WHO. 2018). Chronic exposure to $PM_{2.5}$ contributes to the risk of developing cardiovascular and respiratory diseases, as well as of lung cancer. The numbers of premature deaths attributable to $PM_{2.5}$ are investigated due to the increase of ambient $PM_{2.5}$ and the growth and ageing of population. Many studies estimated the global premature mortality due to $PM_{2.5}$ by analyzing satellite-retrieved $PM_{2.5}$ concentrations. Recent studies estimated long-term $PM_{2.5}$ induced premature

deaths in South and Southeast Asia with high resolution during 1999-2014 (Shi, et al., 2018a) using an integrated exposure-response (IER) model (Apte, et al., 2015) based on the satellite-retrieved $PM_{2.5}$ concentrations, population density, and baseline mortality data. However, the application of fixed uniform baseline mortality for each disease across continental regions might overlook differences among countries overtime. The quantitative analysis of the contributions of the drivers to the trend in mortality at city level has not been performed. The changing trends in premature deaths in relation with $PM_{2.5}$ exposure levels, population, or baseline mortality at city level have not been investigated. Therefore, there is no information on the factors influencing the changes in premature deaths and the impact of $PM_{2.5}$ control measures under the county standards on $PM_{2.5}$ induced premature death.

Bangkok, the capital city of Thailand recently experience $PM_{2.5}$ episodes characterized by high concentration of $PM_{2.5}$ due to combustion emissions from multiple sources and metrological condition from January to April every year. Transportation-related sources are the major contributions to the $PM_{2.5}$ induced poor air quality in Bangkok. In Bangkok the population is exposed to $PM_{2.5}$ annual average concentrations of 14-55 $\mu g/m^3$. The country $PM_{2.5}$ monitoring data has become available for public since 2011. World Health Organization Air Quality Guidelines estimate that reducing annual average $PM_{2.5}$ concentrations from levels of 35 $\mu g/m^3$, common in many developing cities, to the WHO guideline level of 10 $\mu g/m^3$, could reduce air pollution-related deaths by around 15% (WHO. 2006). Thailand National Ambient Air Quality Standard for $PM_{2.5}$ annual average concentration is 25 $\mu g/m^3$. But still, awareness of public health is limited. Therefore, it is needed to assess the impact of $PM_{2.5}$ exposure to public health.

This study estimated multi-year (2010-2017) premature deaths associated with $PM_{2.5}$ exposure in Bangkok, the capital city of Thailand and explored the changes in $PM_{2.5}$ concentrations, baseline mortality and the trends of the premature deaths during 2010-2017. The long-term trends of $PM_{2.5}$ concentration and the changes in the contributions of each health outcome at each district were also analyzed. The results show the health benefits of controlling the $PM_{2.5}$ concentrations based on the Thailand National $PM_{2.5}$ ambient air quality standard and WHO guidelines.

2. Method and data source

2.1 Ambient air quality monitoring data

The ambient $PM_{2.5}$ monitoring data in Bangkok used in this study operated by Pollution Control Department (PCD) from 2016-2018 on hourly basis. The ambient $PM_{2.5}$ data covered 12 monitoring stations located in 9 districts as presented in Figure 1. The ambient air monitoring locations are classified in 2 types; general area road site. As mandated in the National Environmental Quality Act BE 2535 of Thailand in Part 7, the PCD has authority to conduct monitoring, inspection and control. PCD under the Ministry of Natural Resources and Environment is the instrumental institution for the air quality monitoring implementation in Thailand. Monitoring of ambient air quality in Thailand has been conducted since 1983 by the Thailand PCD. As of 2018, the PCD ambient air quality monitoring network in Bangkok consists of 12 continuous ambient air quality monitoring stations. The monitoring objectives are: i) to determine whether

the general levels of pollutants in the atmosphere are likely to be harmful, ii) to evaluate long term air pollution trends from which to formulate policy with regard to abatement action, iii) to assess effectiveness of air pollution control program and mitigation measures implemented to prevent adverse effects of air pollutants, iv) to provide information on a problem of particular concern for which actions are needed, v) to serve as source data on which to base planning decisions and for use on specifically project, vi) to isolate individual rogue emissions to justify control action, and vii) to provide information for correlation with medical studies. The monitoring data is required to meet the QA/QC procedure with a goal to achieve valid and reliable air quality monitoring data. QC procedures are implemented such as calibrations, checks, replicate samples, routine self-assessment, and external audits. Generally, the road site location is set for approximately 1.5 meters away from main road. The road site sampling location consists of Thon Buri, Bang Khun Thian, Wong Thonglang, Din Deang and Pathum Wan. The general sampling site is set for approximately 15 meters away from main road and avoids obstruction of the building for at least 30-degree angle. The general sampling sites consist of Phayathi, Thon Buri, Bangna, Wang Thonglang, Bang Kapi, Din Deang and Yan Nawa.

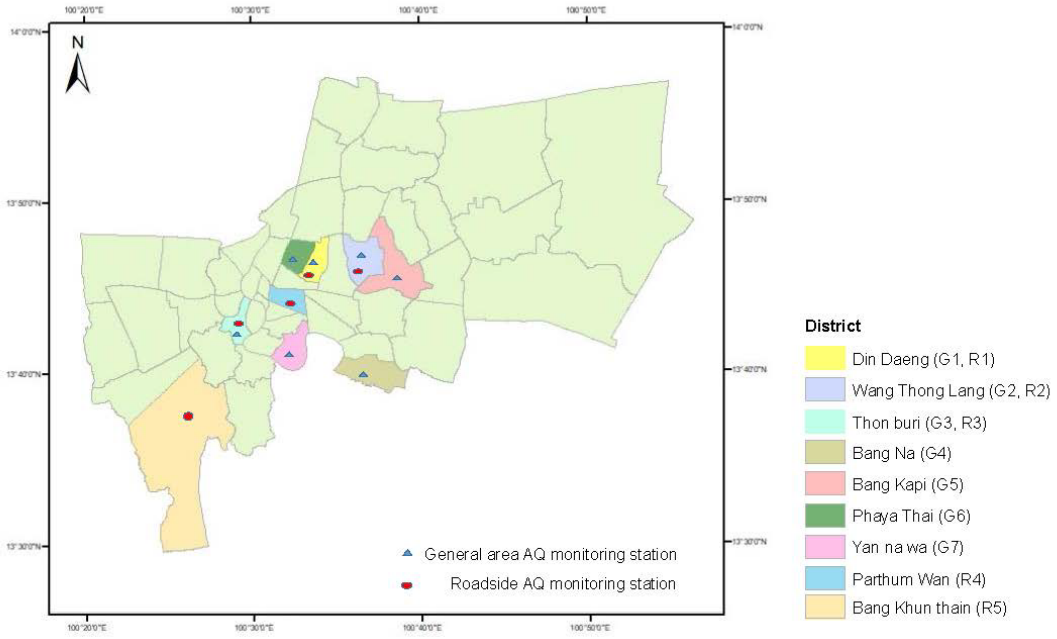


Figure 1. PCD ambient air monitoring network in Bangkok

2.1 Mortality and population data

The Ministry of Public Health (MoPH), Thailand is the responsible national agency that oversees issues related to health. However, healthcare is administered at the regional, provincial (Provincial Health Office), district (District Health Office/District Hospitals) and sub-district levels (Sub-district Health Promotion Hospitals). Health data for individuals is collected and checked at the sub-district levels before being sent to the district level where the data is cleansed and verified. The data is then sent to the provincial level to be processed and analyzed, before being aggregated at the regional

level and the aggregated data is quality-checked and analyzed at the national level. The health data that is collected can be classified into 3 types: health resources (human resources, health facilities, supplies and equipment, and financial resources), health services (health promotion, immunization, treatment), and health status (vital statistics, mortality and morbidity). Cause of death is categorized according to the International Statistical Classification of Diseases and Related Health Problems, 10th revision (ICD10). In this study, number of deaths by each district was obtained with supplementary information, including residential location, place of death, age, sex and primary cause of death. Number of populations by each district is derived from Nation Statistical Report.

2.2 Ambient $PM_{2.5}$ data estimation

The estimation of $PM_{2.5}$ concentrationdue to laccking of data is nessesary to determine the premature mortality of a particular disease attributable to $PM_{2.5}$ exposure. In this study, the $PM_{2.5}$ data before 2011 was estimated by equation (1). The $PM_{2.5}$ average concentration was estimated for each district. It was estimated using $PM_{2.5}$ to PM_{10} relationship for each district as follows:

$$PM_{2.5i} = \frac{\overline{PM_{2.5m}}}{\overline{PM_{10m}}} \times PM_{10i} \qquad (1)$$

where $PM_{2.5i}$ is the estimated $PM_{2.5}$ concentration ($\mu g/m^3$) on hourly basis, i is the hourly unit, $\overline{PM_{2.5m}}$ and $\overline{PM_{10m}}$ represent the mean concentration of $PM_{2.5}$ and PM_{10}, respectively in a month for a district, and PM_{10i} is the hourly monitoring PM_{10} concentration ($\mu g/m^3$) in a district.

2.3 Premature mortality assessment

The health effects data provided by hospitals under the jurisdiction of the MoPH, categorized according to ICD10 as previously described. Within the classification of diseases the data sub-set for descriptive statistical analysis was used. Five $PM_{2.5}$ related disease were chosen for this study: chronic obstructive pulmonary disease (COPD) ICD10 classification J40-J44, J47, lung cancer (LC) for adults age older than 30 years old ICD10 classification C33-C34, ischaemic heart disease (IHD) ICD10 classification I20-I25, stroke ICD 10 Classification I60-I63, I65-I67, I69.0-I69.3 for adults age older than 30 years old, and acute lower respiratory disease (ALRI) ICD10 classification J10-J20 for children age younger than 4 years old, all of which have direct adverse health effects links to ambient $PM_{2.5}$ exposure.

However, with developing economic situation in the country, affordable healthcare has helped prevent premature mortality from ALRI, resulting in a decline in the baseline mortality rate of infants. The increasing number of premature deaths attributable to ALRI was relatively small compared with the other disease, especially at the district level, despite the flowing population and increasing $PM_{2.5}$ concentrations. Therefore, further analysis of premature mortality from ALRI at district level could be neglected in this study.

Premature mortality due to a particular disease attributable to $PM_{2.5}$ exposure was estimated using the traditional epidemiological exposure-response relationship (Apte, et al., 2015) (Shi, et al., 2018b).

$$M = Y \times \frac{RR-1}{RR} \times P \qquad (2)$$

where M is the premature mortality due to a particular disease for a district attributable to $PM_{2.5}$ exposure, Y is the baseline mortality rate of a disease in each district of Bangkok or the reported annual mortality rate for a disease in a district. RR represents the relative risk of a disease for a specific $PM_{2.5}$ concentration, P represents the annual exposed population (i.e., above 30 years of age in a district based on a proportion to the total population in the district.

The baseline mortality for each disease varied temporally and among districts provided by Ministry of Public Health in Thailand. The extrapolation of RR to the monitored $PM_{2.5}$ concentrations for each disease was estimated based on existing epidemiological studies of the integrated exposure-response (IER) functions for ambient $PM_{2.5}$ exposure:

$$RR = 1 + \alpha[1 - \exp(-\gamma(x - x_0)^{\delta})], \ if\ x > x_0 \qquad (3)$$

where x is the annual mean of $PM_{2.5}$ concentration and x_0 is the threshold concentration below which no additional risk is assumed. For each disease category, x_0 represents the theoretical minimum risk concentration (range 2.4-5.9 μg/m^3). The parameters α, γ and δ are specific to each disease and were determined using a stochasting fitting progress (Apte, et al., 2015). The mean values of RR for each disease provided as a look-up table from SI Table S2 to estimate the number of premature mortalities (Apte, et al., 2015). With regarding the RR used in this study, it should be noted that the generated estimates of premature mortality attributable to $PM_{2.5}$ carry some uncertainty. The health effect used in this study come mainly from studies conducted in Western Europe and North America. The applicable of the results generated by this study for assessments in Thailand can be associated with additional uncertainties and should be considered with caution. In addition, the calculations do not account for multiple exposure causes or multipollutant scenarios.

2.4 Rate of change

The annual rate of change (ARC) was determined to compare the variations in $PM_{2.5}$ induced premature mortality from each disease between 2010 and 2017 at district level. ARC is defined as follows (Shi, et al., 2018b);

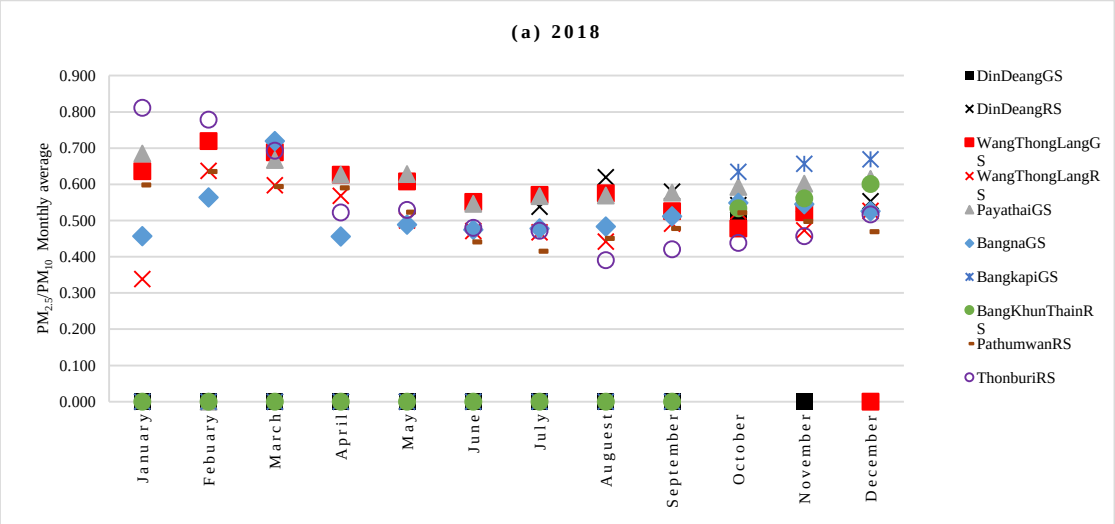
$$ARC = \frac{B-A}{A} \times \frac{1}{t} \times 100\% \qquad (4)$$

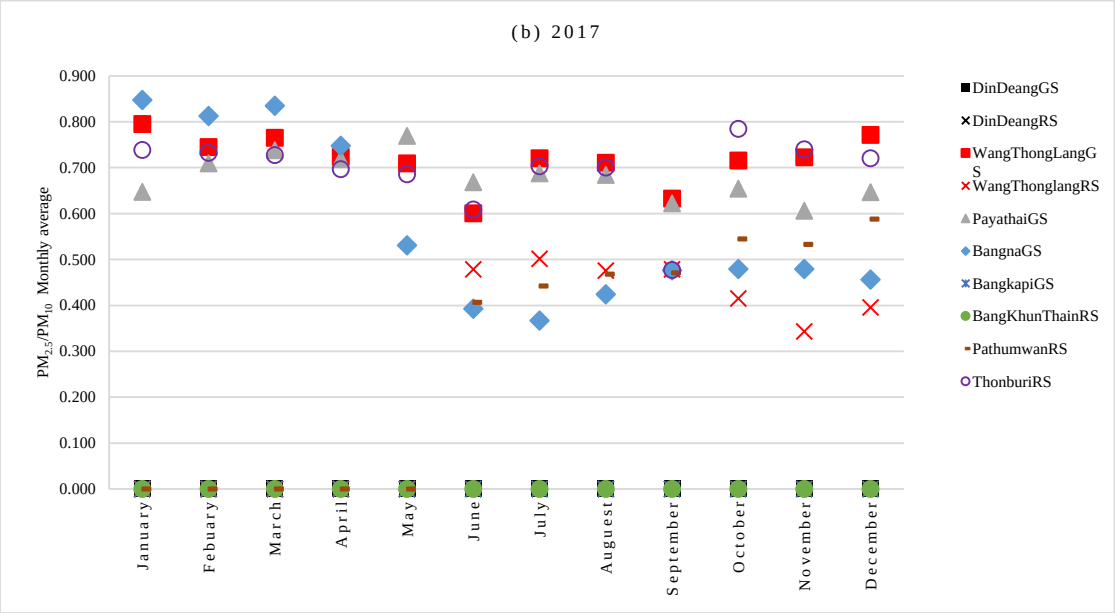
where A and B are premature mortality in 2010 and 2017 respectively, and t is the length of this study period.

3. Results and discussion

3.1 $PM_{2.5}$ data investigation

The monthly $PM_{2.5}$ to PM_{10} ratio for each district was estimated for 2016-2018 as presented in Figure 2. Most of the ratio slowly decrease from February to June and then gradually increase from July to December. The ratio for general site is slightly higher than those observed in the road site monitoring location at Wang Thong Lang District in 2018 and 2017.





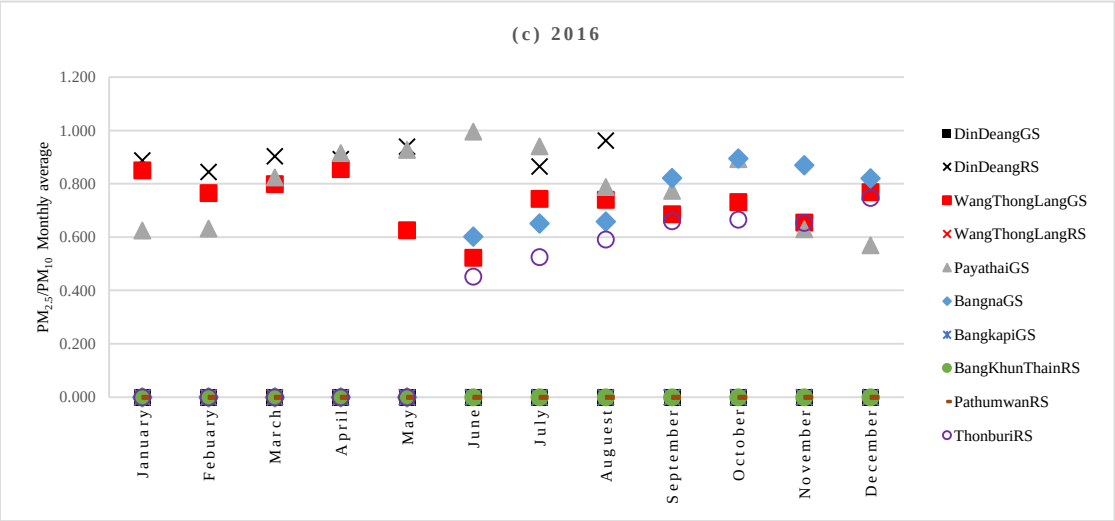


Figure 2. $PM_{2.5}/PM_{10}$ monthly ratio variation

Changes in annual mean of $PM_{2.5}/PM_{10}$ ratio for each district mean the annual ratio fluctuated annually. Therefore, the changes in annual ratio over the entire districts from 2010 to 2018 were quantified. Overall, the ratio showed the decreasing trend from 2016 to 2018 and presented in Figure 3. However, the ratio is not available for every district before 2016. The ratio from all available districts in 2016 were obtained for the estimation of unavailable $PM_{2.5}$ concentration in other districts from 2010 to 2015. Because of the variations in the trend of the ratio from general and road monitoring site, the ratio from the same monitoring location type is applied to estimate the $PM_{2.5}$ concentration for the same monitoring location type. For example, the ratios from DinDeang and Thonburi road sites in 2016 are applied to estimate the $PM_{2.5}$ hourly concentration in WangThongLang, Bangkhuntian and Pathumwan road sites at a specific month from 2010 to 2016. On the other hand, the ratios from WangThongLand general site in 2016 is applied to estimate the $PM_{2.5}$ hourly concentration in Bangkapi, Bangna, DinDeang, Thonburi and Yannawa at a specific month from 2010 to 2016.

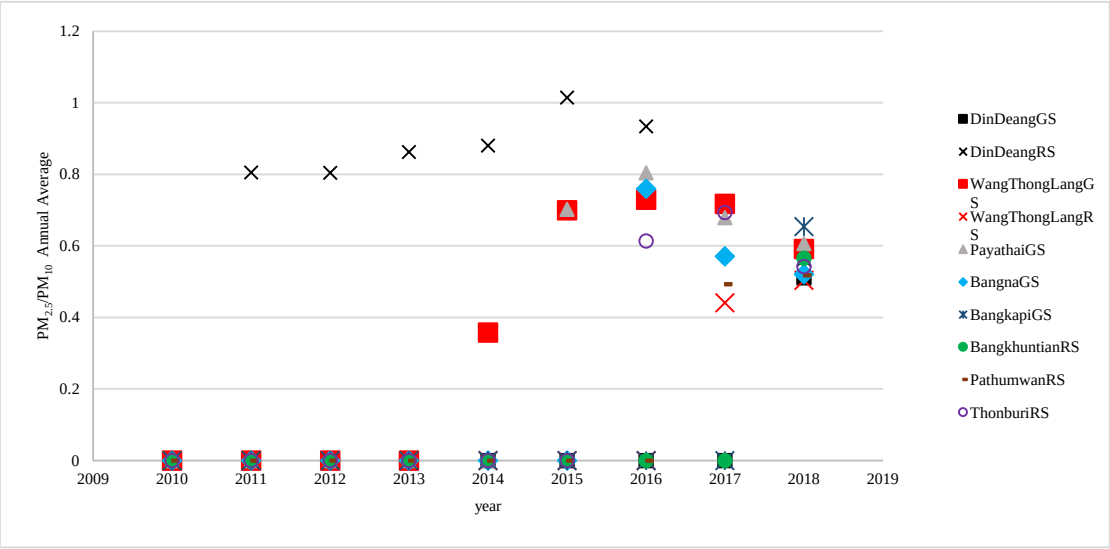


Figure 3. $PM_{2.5}/PM_{10}$ annual average

The annual average of $PM_{2.5}$ for each district was estimated as presented in Figure 4. The study identified population in Bangkok is exposed to $PM_{2.5}$ annual average annual average concentrations of 14-55 μg/m^3 from 2010-2017.

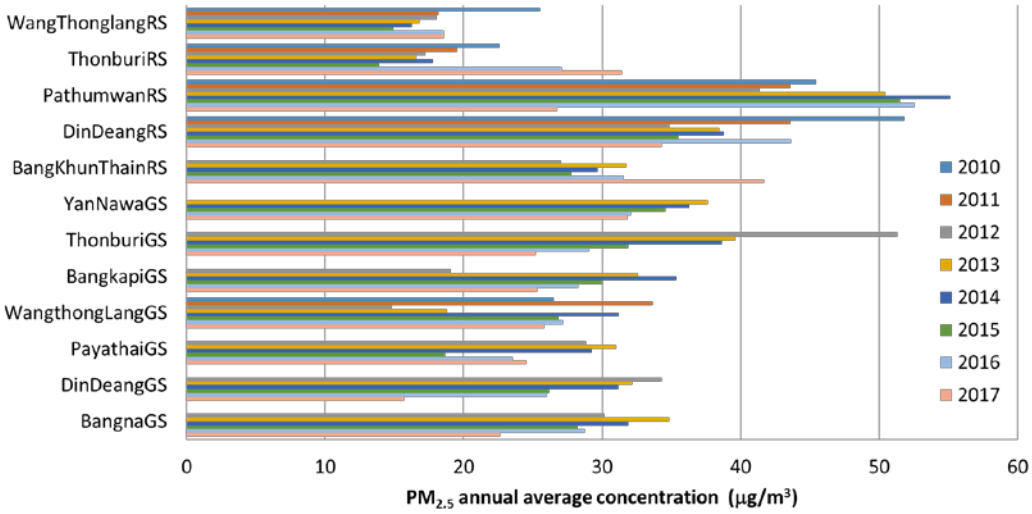
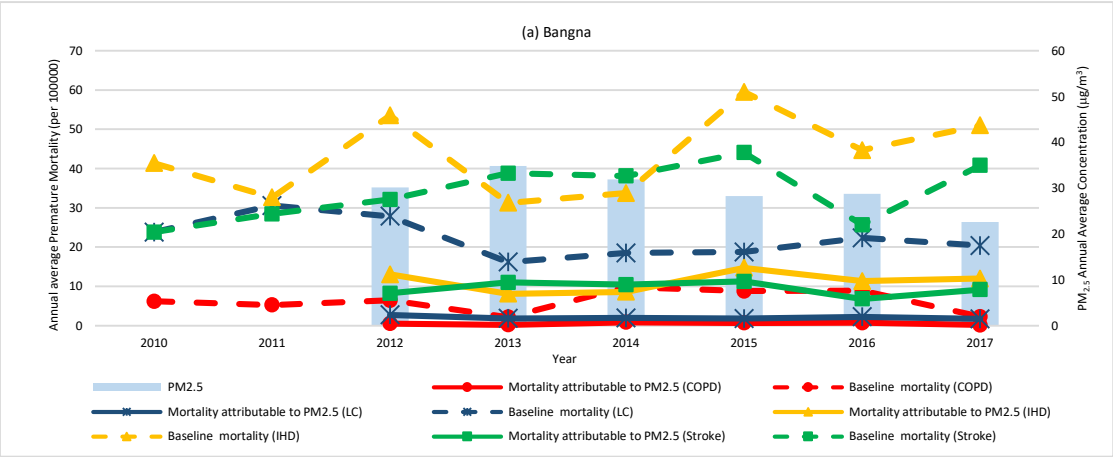
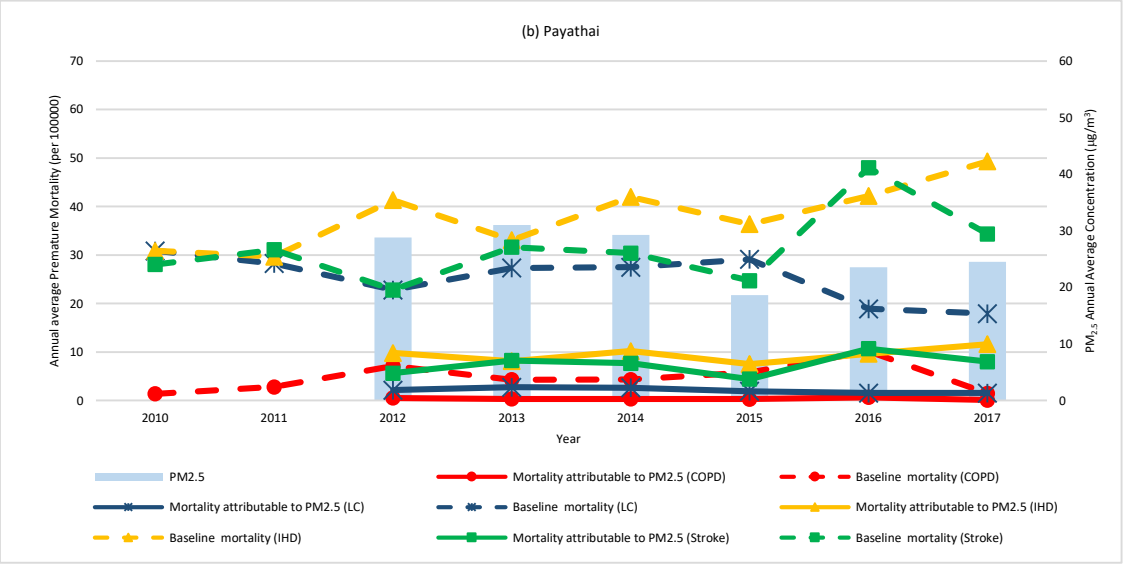


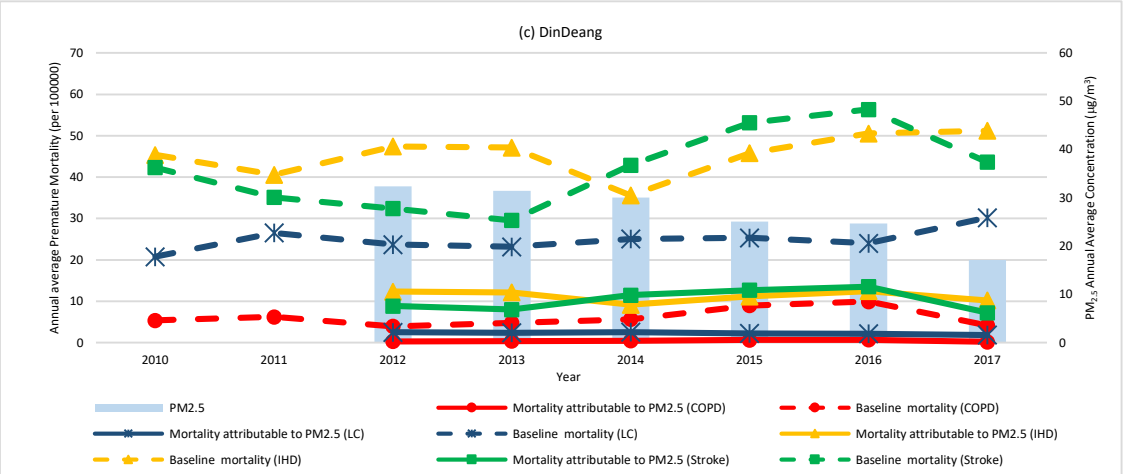
Figure 4. $PM_{2.5}$ annual average concentrations

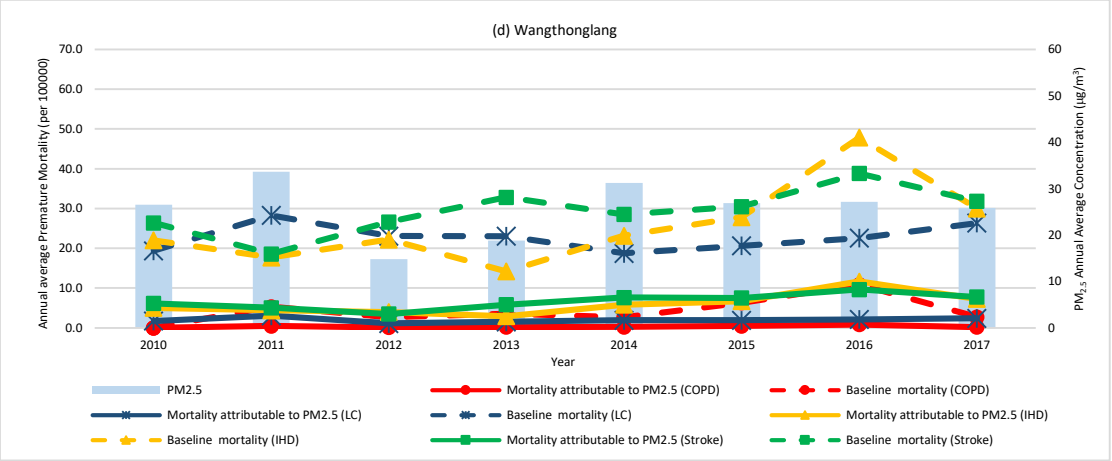
3.2 Long-term analysis of premature mortality

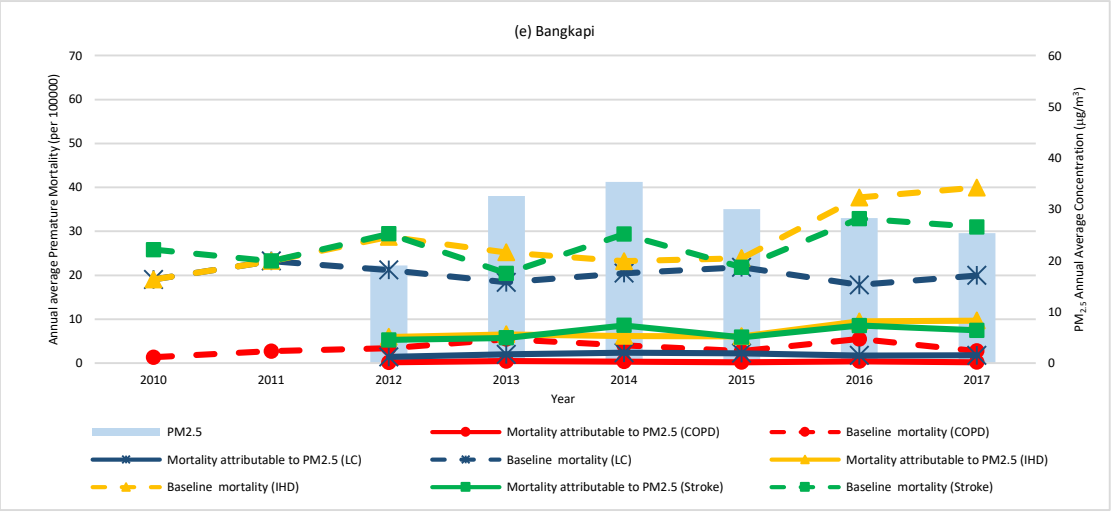
The premature mortality attributable to $PM_{2.5}$ for COPD, LC, IHD and Stroke were estimated in the districts, where $PM_{2.5}$ available. For example, at Thonburi district general area, the average of premature mortality for COPD attributable to $PM_{2.5}$ is estimated 1.3, 1.1, 0.7, 1.0, 0.9 and 0.7 (per 100000) in 2012, 2013, 2014, 2015, 2016 and 2017, respectively as presented in Figure 5f. The baseline premature mortality for COPD, LC, IHD and Stroke in the districts were estimated from 2010-2017. For example, at Bang Khun Thain district road site area, the average of baseline premature mortality for COPD is estimated in average of 4 (per 100000) from 2010-2017 as presented in Figure 5h.Change in $PM_{2.5}$ concentration, population, and the baseline mortality for each disease in the different districts of the Bangkok area means the premature mortality fluctuated annually. Therefore, the long-term tendency change in annual premature death and their percent contributions were qualified over the 2 locations from 2010 to 2017. Overall, $PM_{2.5}$ -induced premature mortality in Bangkok showed an increasing trend from 2010 to 2017 as presented in Figure 5.

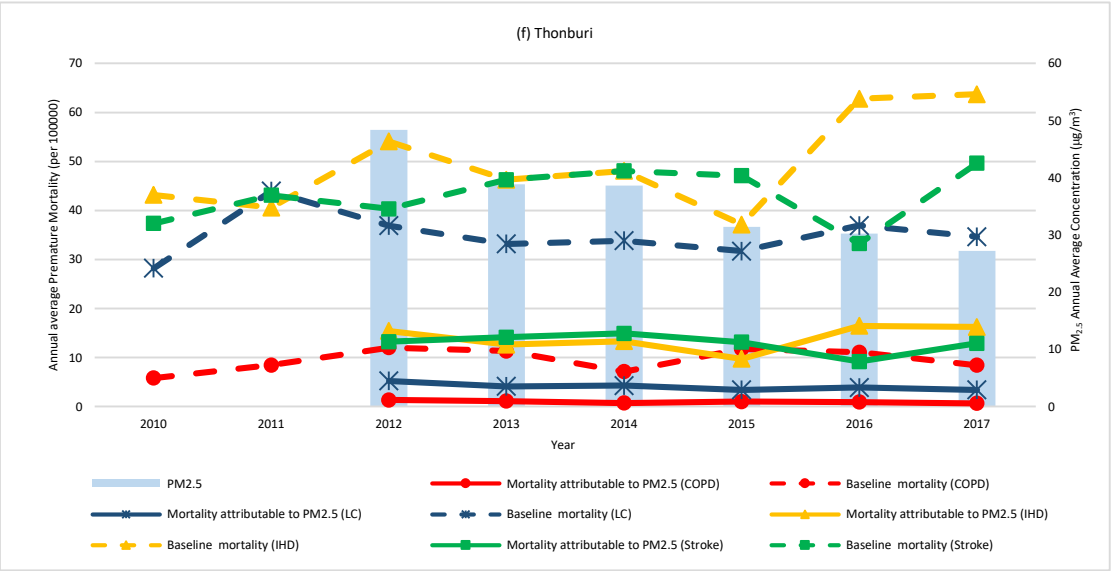

(a) Bangna
Annual average Premature Mortality (per 100000)
PM2.5 Annual Average Concentration (µg/m³)
Year
PM2.5
Mortality attributable to PM2.5 (COPD)
Baseline mortality (COPD)
Mortality attributable to PM2.5 (LC)
Baseline mortality (LC)
Mortality attributable to PM2.5 (IHD)
Baseline mortality (IHD)
Mortality attributable to PM2.5 (Stroke)
Baseline mortality (Stroke)

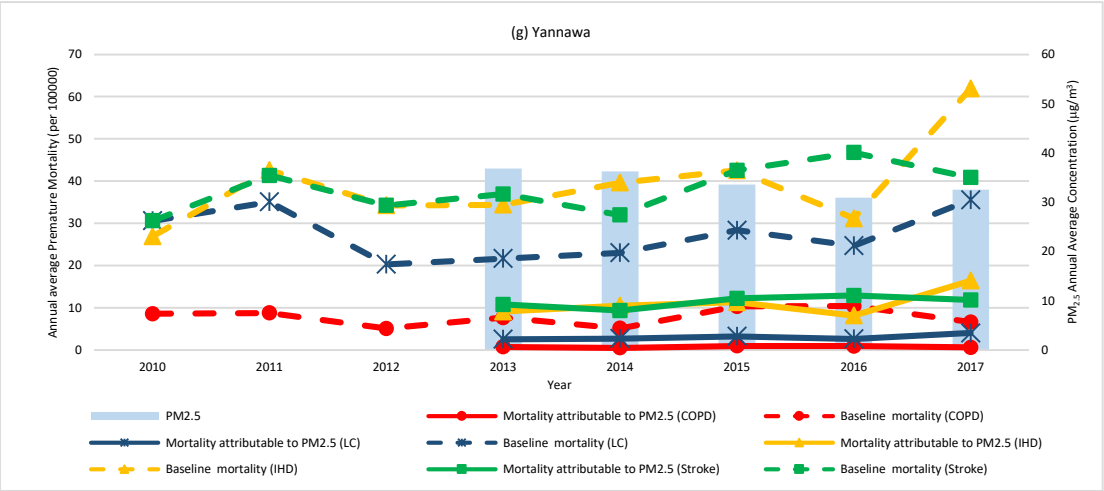

(b) Payathai
Annual average Premature Mortality (per 100000)
PM2.5 Annual Average Concentration (µg/m³)
Year
PM2.5
Mortality attributable to PM2.5 (COPD)
Baseline mortality (COPD)
Mortality attributable to PM2.5 (LC)
Baseline mortality (LC)
Mortality attributable to PM2.5 (IHD)
Baseline mortality (IHD)
Mortality attributable to PM2.5 (Stroke)
Baseline mortality (Stroke)

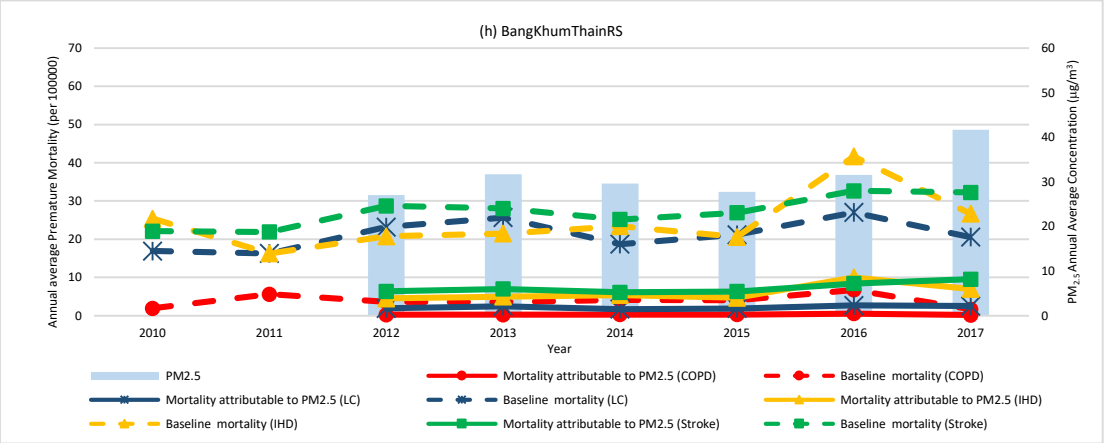

(c) DinDeang
Annual average Premature Mortality (per 100000)
PM2.5 Annual Average Concentration (µg/m³)
Year
PM2.5
Mortality attributable to PM2.5 (COPD)
Baseline mortality (COPD)
Mortality attributable to PM2.5 (LC)
Baseline mortality (LC)
Mortality attributable to PM2.5 (IHD)
Baseline mortality (IHD)
Mortality attributable to PM2.5 (Stroke)
Baseline mortality (Stroke)

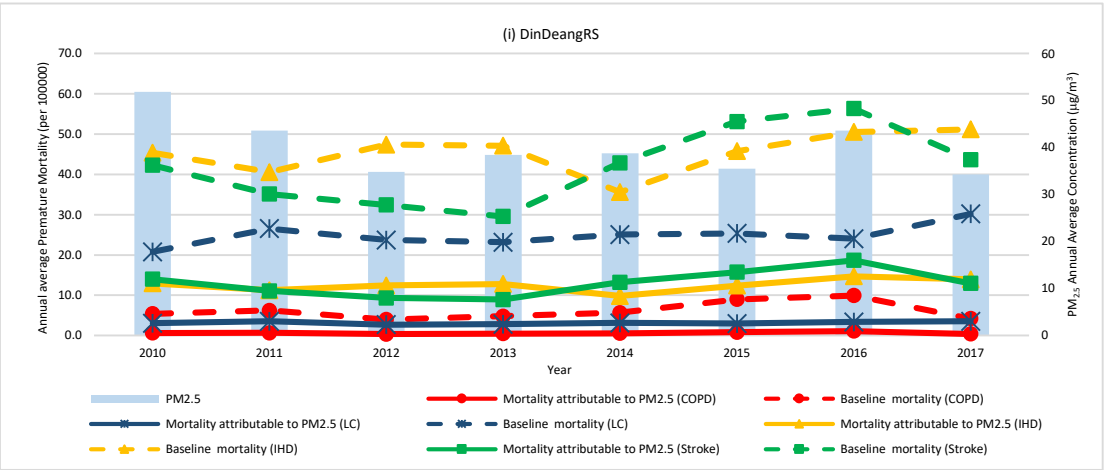

(d) Wangthonglang
Annual average Premature Mortality (per 100000)
PM2.5 Annual Average Concentration (µg/m³)
Year
PM2.5
Mortality attributable to PM2.5 (COPD)
Baseline mortality (COPD)
Mortality attributable to PM2.5 (LC)
Baseline mortality (LC)
Mortality attributable to PM2.5 (IHD)
Baseline mortality (IHD)
Mortality attributable to PM2.5 (Stroke)
Baseline mortality (Stroke)


(e) Bangkapi
Annual average Premature Mortality (per 100000)
PM2.5 Annual Average Concentration (µg/m³)
Year
PM2.5
Mortality attributable to PM2.5 (COPD)
Baseline mortality (COPD)
Mortality attributable to PM2.5 (LC)
Baseline mortality (LC)
Mortality attributable to PM2.5 (IHD)
Baseline mortality (IHD)
Mortality attributable to PM2.5 (Stroke)
Baseline mortality (Stroke)


(f) Thonburi
Annual average Premature Mortality (per 100000)
PM2.5 Annual Average Concentration (µg/m³)
Year
PM2.5
Mortality attributable to PM2.5 (COPD)
Baseline mortality (COPD)
Mortality attributable to PM2.5 (LC)
Baseline mortality (LC)
Mortality attributable to PM2.5 (IHD)
Baseline mortality (IHD)
Mortality attributable to PM2.5 (Stroke)
Baseline mortality (Stroke)


(g) Yannawa
Annual average Premature Mortality (per 100000)
PM2.5 Annual Average Concentration (µg/m³)
Year
PM2.5
Mortality attributable to PM2.5 (COPD)
Baseline mortality (COPD)
Mortality attributable to PM2.5 (LC)
Baseline mortality (LC)
Mortality attributable to PM2.5 (IHD)
Baseline mortality (IHD)
Mortality attributable to PM2.5 (Stroke)
Baseline mortality (Stroke)


(h) BangKhumThainRS
Annual average Premature Mortality (per 100000)
PM2.5 Annual Average Concentration (µg/m³)
Year
PM2.5
Mortality attributable to PM2.5 (COPD)
Baseline mortality (COPD)
Mortality attributable to PM2.5 (LC)
Baseline mortality (LC)
Mortality attributable to PM2.5 (IHD)
Baseline mortality (IHD)
Mortality attributable to PM2.5 (Stroke)
Baseline mortality (Stroke)


(i) DinDeangRS
Annual average Premature Mortality (per 100000)
PM2.5 Annual Average Concentration (µg/m³)
Year
PM2.5
Mortality attributable to PM2.5 (COPD)
Baseline mortality (COPD)
Mortality attributable to PM2.5 (LC)
Baseline mortality (LC)
Mortality attributable to PM2.5 (IHD)
Baseline mortality (IHD)
Mortality attributable to PM2.5 (Stroke)
Baseline mortality (Stroke)

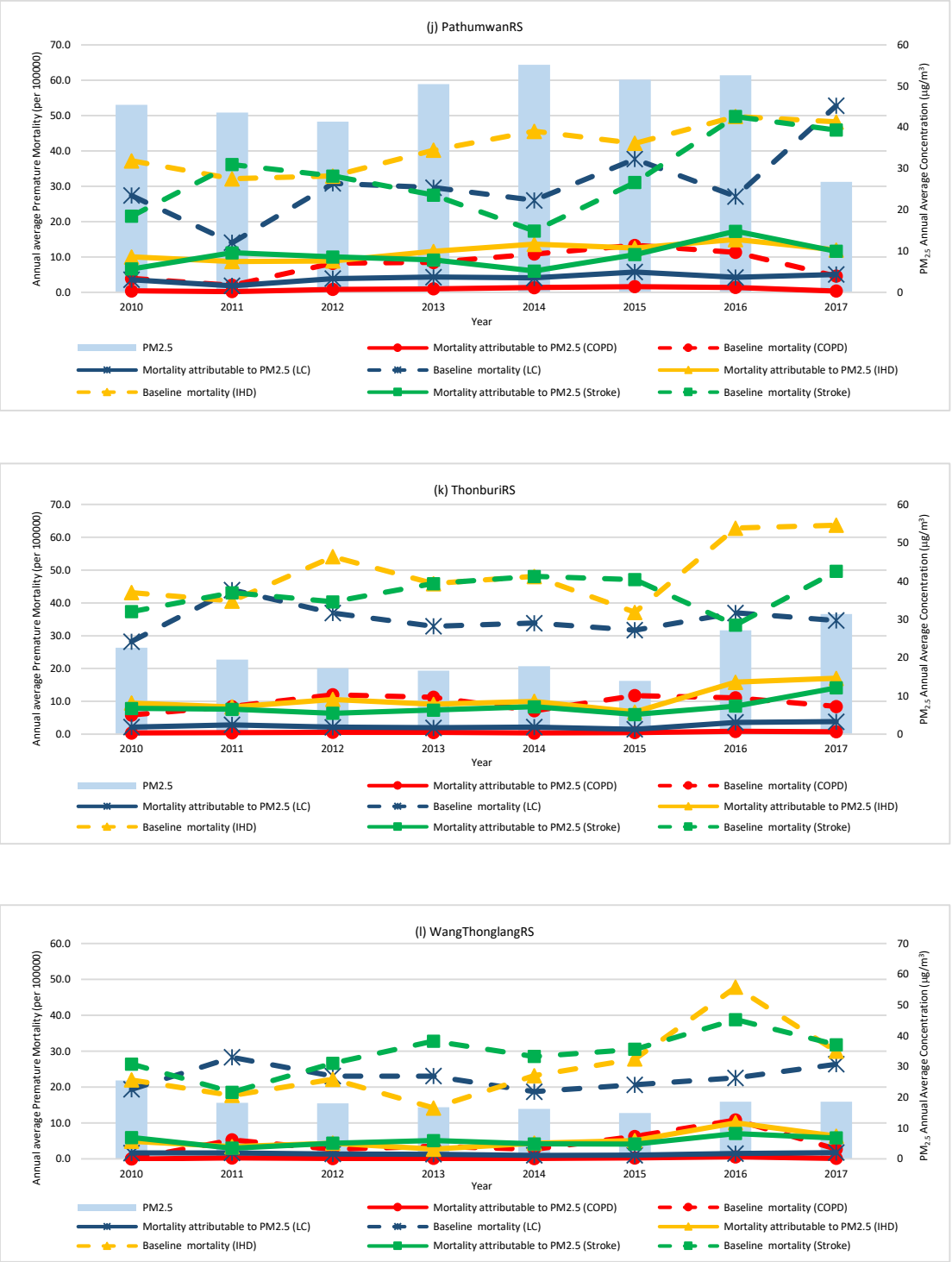


Figure 5. Long-term analysis of premature mortality

Premature mortality from all four studies diseases increased gradually from 2010-2017, however, the largest increments occurred in stroke and IHD, followed by LC and COPD. Because of the variation in the trend of the four disease and their baseline mortality amounts, their relative contributions to total premature mortality also varied during the study period. The estimated premature mortality in the proportion of the contributions of the diseases to baseline premature deaths showed that COPD and LC varied little annually. Both IHD and Stroke increased their contributions from 2010 to

2017 with minor increase in their proportions.

3.3 Annual rate change from 2010-2017

$PM_{2.5}$ annual average concentration presents decreasing tendencies except in BangKapi, Wangthonglang, Thonburi and BangKhunThain. Population exposure to $PM_{2.5}$ is decreasing in every district except in BangKhunThain. The premature mortality induced by $PM_{2.5}$ presents incremental tendencies for COPD, LC, IHD and Stroke in Thonburi and Bangkapi district. Premature deaths presents incremental tendencies for IHD except that in Bangna and Dindeang district. Summary of annual rate change from 2010-2017 is present in Table 1.

Table 1. Annual rate of change from 2010-2017

District	$PM_{2.5}$ (%)	Population (%)	Baseline mortality (%)				Premature mortality induced by $PM_{2.5}$ (%)			
			COPD	LC	IHD	Stroke	COPD	LC	IHD	Stroke
Bangna	-4.1	-1.1	-7.9	-1.8	2.9	8.9	-11.6	-6.4	-1.4	1.8
DinDeang	**-7.8**	-1.0	-2.8	5.7	1.6	0.4	-6.1	-4.4	**-2.1**	**-3.0**
Payathai	-2.5	-0.8	0.8	**-5.2**	7.5	2.8	**-13.4**	-4.6	3.1	7.0
WangThongLang	-0.3	-0.4	**-6.9**	4.5	4.5	2.6	-8.0	5.2	5.6	3.3
Bangkapi	5.5	-0.2	**12.8**	0.6	13.7	2.5	1.3	4.7	10.5	6.9
Thonburi	-7.3	-1.4	5.6	2.9	5.9	4.1	-8.5	**-5.9**	0.9	-0.4
YanNawa	-2.3	-0.9	-2.9	2.0	**16.2**	4.2	-3.2	**11.8**	**16.7**	2.0
BangKhunThainRS	**9.0**	**2.1**	-1.8	2.7	0.7	5.7	-5.6	5.1	9.1	8.4
DinDeangRS	-4.2	-1.0	-2.8	5.7	1.6	0.4	-4.7	1.9	0.9	-0.9
PathumwanRS	-5.1	-1.8	2.2	**11.6**	3.7	**14.1**	-1.6	5.1	2.4	9.4
ThonburiRS	4.9	-1.4	5.6	2.9	5.9	4.1	**12.8**	9.2	10.0	10.0
WangThongLangRS	0.3	-0.4	-6.9	4.5	4.5	2.6	-6.0	0.7	12.2	**13.3**

Among the 4 disease, IHD and Stroke are the two primary contributors to total premature deaths in each district. In 2017, average premature mortality attributable to $PM_{2.5}$ were accounted for 0.3, 2.8, 11.6, and 9.8 (per 100000) for COPD, LC, IHD and Stroke respectively. Because of the variations in the trends of the four disease and their baseline amounts, their relative contributions to total premature mortality also varies. The study results show that, in 2017, if the $PM_{2.5}$ annual average concentration had reduced to WHO Guideline (10 $\mu g/m^3$), it would have lower the risk of premature mortality attributable to $PM_{2.5}$ for 71, 74, 46 and 73 percent for COPD, LC, IHD and Stroke respectively. Summary of annual average premeasure mortality (per 100000) attributable to each of the disease in each district, and their responding change (%) to WHO Guideline (10 $\mu g/m^3$) of $PM_{2.5}$ concentration annual mean is presented in Table 2.

Table 2. Annual average premeasure mortality (per 100000) attributable to each of the disease in each district, and their responding change (%) to 10 $\mu g/m^3$ of $PM_{2.5}$ concentration annual mean

District	COPD		LC		IHD		Stroke	
	2017	Reduce (%)	2017	Reduce (%)	2017	Reduce (%)	2017	Reduce (%)
Din Daeng	0.3	70	2.8	73	12.7	45	10.8	72
Wang Thong Lang	0.2	66	2.1	69	6.8	42	6.8	68
Thon buri	0.7	73	3.6	76	16.7	47	13.6	74
Bang Na	0.2	67	1.7	69	11.9	42	9.0	69
Bang Kapi	0.2	70	1.8	72	9.7	45	7.5	72
Paya Thai	0.1	69	1.5	72	11.5	44	7.9	71
Yan na wa	0.6	75	4.0	78	16.5	49	11.7	76
Pathum Wan	0.3	71	5.0	74	11.9	46	11.5	73
Bang Khun Thain	0.2	**79**	2.5	**81**	7.0	**53**	9.5	**79**
Average	**0.3**	71	**2.8**	74	**11.6**	46	**9.8**	73

4. Conclusion

This study quantifies the premature mortality from COPD, IHD, stroke and LC attributable to $PM_{2.5}$ exposure in Bangkok, Thailand during 2010-2017. In addition, the long-term trends and monthly patterns of $PM_{2.5}$ concentration were analyzed. With regardless of the air quality monitoring location type, $PM_{2.5}$ annual average concentration varies year by year due to local sources and meteorological condition of the area. Among the 4 disease studies, IHD and Stroke are the two primary contributors to total premature deaths in each district. In 2017, premature mortality due to COPD for districts attributable to $PM_{2.5}$ exposure is found in the average of 0.3 per 100000 population. Premature mortality due to IHD for districts attributable to $PM_{2.5}$ exposure is found in the average of 11.6 per 100000 population. Premature mortality due to LC for districts attributable to $PM_{2.5}$ exposure is found in the average of 2.8per 100000 population. Premature mortality due to Stroke for districts attributable to $PM_{2.5}$ exposure is found in the average of 9.8 per 100000 population. Premature mortality due to stroke presents the most significant number. Change in $PM_{2.5}$ concentration, population, and the baseline mortality for each disease in the difference districts mean the annual premature mortality fluctuated annually. The study results estimated that reducing annual average $PM_{2.5}$ concentration from the existing level 2017 to the WHO guideline level of 10 $\mu g/m^3$, could reduce $PM_{2.5}$ related deaths by around 46-73 percent.

Acknowledgements

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Study on the impacts of air pollution transport and its effects to human health

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Abstract

The impact of air pollution on human health is a complicated problem. In this report, we describe the main health effects of exposure to ozone, particulate matter, sulfur dioxide and nitrogen dioxide. Evidence shows that through these actions, air quality has improved. However, some pollutants such as ozone and particulate matter ($PM_{2.5}$), still remains high and exceeds the air quality standards. In the early days of abundant resources and minimal development pressures, little attention was paid to growing environmental concerns in Cambodia. As a consequence, the government established Air Quality Guidelines, the Air Pollution Index to improve air quality. The air pollution comes mainly from transportation, industrial emissions, and open burning sources. Among them, transportation contributes the most to air pollution. This paper reviews the results of the ambient air quality and studies related to air pollution and health impacts by estimating the premature mortality caused by exposure of $PM_{2.5}$ and ozone in Cambodia.

Keywords: $PM_{2.5}$, Ozone, Risk Assessment, Premature Mortality, Cambodia

1. Introduction

The air pollution is a major environmental health problem affecting everyone in developed and developing countries alike. There are two main types of air pollution – ambient air pollution (outdoor pollution) and household (or indoor) air pollution which refers to pollution generated by household combustion of fuels (caused by burning fuel such as coal, wood or kerosene) using open fires or basic stoves in poorly ventilated spaces. Both indoor and outdoor air pollution can contribute to each other, as air moves from inside buildings to the outside, and vice versa. Household air pollution kills 4 million people a year and tends to affect countries including countries in Africa and Asia (WHO, Factsheets), where polluting fuels and technologies are used every day particularly at home for cooking, heating and lighting. Women and children, who tend to spend more time indoors, are affected the most. The main pollution are (1) particulate

matter, a mix of solid and liquid droplets arising mainly from fuel combustion and road traffic; (2) nitrogen dioxide from road traffic or indoor gas cookers; (3) sulphur dioxide from burning fossil fuels; and (4) ozone at ground level, caused by the reaction of sunlight with pollutants from vehicle emissions.

The pollutant that affects people the most is particulate matter (PM). While particles with a diameter of 10 microns or less, (PM_{10}) can penetrate and lodge deep inside the lungs, the even more health-damaging particles are those with a diameter of 2.5 microns or less ($PM_{2.5}$). These particles are so small comparing a human hair of which diameter is less than 100μm or so. $PM_{2.5}$ can penetrate the lung barrier and enter the blood system. They can increase the risk of heart and respiratory diseases, as well as lung cancer. Ozone is a major factor in causing asthma (or making it worse), and nitrogen dioxide and sulfur dioxide can also cause asthma, bronchial symptoms, lung inflammation and reduced lung function (WHO, Factsheets).

So how polluted air can start to affect our health? For $PM_{2.5}$, WHO guidelines says that the annual average concentration of 10 $\mu g/m^3$ or less is acceptable level of risk. To encourage cities to reduce air pollution, even if they are unable to meet the ideal safe levels, WHO has set three interim targets for cities. These are: 15 $\mu g/m^3$ (interim target 3); 25 $\mu g/m^3$ (interim target 2); 35 $\mu g/m^3$ (interim target 1). Many cities are now exceeding the very upper level of interim target 1.

As well as affecting our health, pollutants in the air are also causing long-term environmental damage by driving climate change, itself a major threat to health and well-being.

Most sources of outdoor air pollution are well beyond the control of individuals and demand action by cities, as well as national and international policymakers in sector like transport, energy waste management, buildings and agriculture. The Guidelines apply worldwide and are based on expert evaluation of current scientific evidence for: particulate matter (PM) ozone (O_3) nitrogen dioxide (NO_2) and sulfur dioxide (SO_2), in all WHO regions.

1.1 Particulate matter

PM affects more people than any other pollutant. The major components of PM are sulfate, nitrates, ammonia, sodium chloride, black carbon, mineral dust and water. It consists of a complex mixture of solid and liquid particles of organic and inorganic substances suspended in the air. The more health damaging particles are those with a diameter of 2.5 microns or less ($PM_{2.5}$), which can penetrate and lodge deeply inside the lungs. Chronic exposure to particles contributes to the risk of developing cardiovascular and respiratory diseases, as well as of lung cancer. PM_{10} is measured and daily or annual mean of the concentration is calculated usually as the index of air quality in the past. Routine air quality measurements typically describe such PM concentrations in terms of micrograms per cubic meter ($\mu g/m^3$). When sufficiently sensitive measurement tools are available, concentrations of fine particles ($PM_{2.5}$ or smaller), are also reported.

There is a close, quantitative relationship between exposure to high concentrations of small particulates (PM_{10} and $PM_{2.5}$) and increased mortality or morbidity, both daily and over time. Conversely, when concentrations of course and fine particulates are

reduced, it is expected that related mortality would also go down – presuming other factors remain the same.

This allows policymakers to project the population health improvements that could be expected if particulate air pollution is reduced. Small particulate pollution have health impacts even at very low concentrations indeed no threshold has been identified below which no damage to health is observed. Therefore, the WHO 2005 guideline limits aimed to achieve the lowest concentrations of PM possible.

Guideline values: $PM_{2.5}$: 10 µg/m annual mean, 25 µg/m 24-hour mean
PM_{10}: 20 µg/m annual mean, 50 µg/m 24-hour mean

In addition to guideline values, the Air Quality Guidelines provide interim targets for concentrations of PM_{10} and $PM_{2.5}$ aimed at promoting a gradual shift from high to lower concentrations. If these interim targets were to be achieved, significant reductions in risks for acute and chronic health effects from air pollution can be expected. Progress towards the guideline values, however, should be the ultimate objective.

The effects of PM on health occur at levels of exposure currently being experienced by many people both in urban and rural areas and in developed and developing countries although exposures in many fast-developing cities today are often far higher than in developed cities of comparable size. "WHO Air Quality Guidelines" estimate that reducing annual average particulate matter (PM_{10}) concentrations from levels of 70 µg/m^3, common in many developing cities, to the WHO guideline level of 20 µg/m^3, could reduce air pollution related deaths by around 15%. However, even in the European Union, where PM concentrations in many cities do comply with Guideline levels, it is estimated that average life expectancy is 8.6 months lower than it would otherwise be, due to PM exposures from human sources.

In developing countries, indoor exposure to pollutants from the household combustion of solid fuels on open fires or traditional stoves increases the risk of acute lower respiratory infections and associated mortality among young children; indoor air pollution from solid fuel use is also a major risk factor for cardiovascular disease, chronic obstructive pulmonary disease and lung cancer among adults. There are serious health risks not only from exposure to PM, but also from exposure to ozone (O_3), nitrogen dioxide (NO_2) and sulfur dioxide (SO_2).

As with PM, concentrations are often highest largely in the urban areas of low- and middle-income countries. Ozone is a major factor in asthma morbidity and mortality, while NO_2 and SO_2 also can play a role in asthma, bronchial symptoms, lung inflammation and reduced lung function.

1.2 Ozone (O_3)

Guideline values: O_3 100 µg/m^3 8-hour mean

The recommended limit in the 2005 Air Quality Guidelines was reduced from the level of 120 µg/m^3 in previous editions of the "WHO Air Quality Guidelines" (2000) based on recent conclusive associations between daily mortality and lower ozone concentrations. Ozone at ground level – not to be confused with the ozone layer in the upper atmosphere – is one of the major constituents of photochemical smog. It is formed

by the reaction with sunlight (photochemical reaction) of pollutants such as nitrogen oxides (NOx) from vehicle and industry emissions and volatile organic compounds (VOCs) emitted by vehicles, solvents and industry.

As a result, the highest levels of ozone pollution occur during periods of sunny weather. Excessive ozone in the air can have a marked effect on human health. It can cause breathing problems, trigger asthma, reduce lung function and cause lung diseases. (Inhalation of ozone could lead to breathing problems, cough, inflammation of the respiratory airways, aggravation of lung diseases such as asthma, chronic bronchitis and progression to lung damage (United State Environmental protection Agency, 2014).

In Europe it is currently one of the air pollutants of most concern. Several European studies have reported that the daily mortality rises by 0.3% and that for heart diseases by 0.4%, per 10 $\mu g/m^3$ increase in ozone exposure.

1.3 Nitrogen dioxide (NO_2)

Guideline values: NO_2: 40 $\mu g/m^3$, annual mean, 200 $\mu g/m^3$, 1-hour mean

The current WHO guideline value of 40 $\mu g/m^3$ (annual mean) was set to protect the public from the health effects of gaseous pollutants. As an air pollutant, NO_2 has several correlated activities. At short-term concentrations exceeding 200 $\mu g/m^3$, it is a toxic gas which causes significant inflammation of the airways. NO_2 is the main source of nitrate aerosols, which form an important fraction of $PM_{2.5}$ and, in the presence of ultraviolet light, of ozone. The major sources of anthropogenic emissions of NO_2 are combustion processes (heating, power generation, and engines in vehicles and ships). NO_2 at concentrations currently measured (or observed) in cities of Europe and North America.

1.4 Sulfur dioxide (SO_2)

Guideline values: SO_2: 20 $\mu g/m^3$, 24-hour mean, 500 $\mu g/m^3$, 10-minute mean

A SO2 concentration of 500 $\mu g/m^3$ should not be exceeded over average periods of 10 minutes duration. Studies indicate that a proportion of people with asthma experience changes in pulmonary function and respiratory symptoms after periods of exposure to SO_2 as short as 10 minutes. The 2005 revision of the 24-hour guideline for SO_2 concentrations from 125 to 20 $\mu g/m^3$ was based on the following considerations.

Health effects are now known to be associated with much lower levels of SO_2 than previously believed. A greater degree of protection is needed. Although the causality of the effects of low concentrations of SO_2 is still uncertain, reducing SO_2 concentrations is likely to decrease the risk of adverse effect on human health by exposure to it (WHO, 2005). SO_2 is a colorless gas with a sharp odor. It is produced from the burning of fossil fuels (coal and oil) and the smelting of mineral ores that contain sulfur. The main anthropogenic source of SO_2 is the burning of sulfur containing fossil fuels for domestic heating, power generation and motor vehicles.

SO_2 can affect the respiratory system and the functions of the lungs, and causes irritation of the eyes. Inflammation of the respiratory tract causes coughing, mucus secretion, aggravation of asthma and chronic bronchitis and makes people more prone to infections of the respiratory tract (WHO, 2005). Hospital admissions for cardiac disease and mortality increase on days with higher SO_2 levels. When SO_2 combines with water, it forms sulfuric acid; this is the main component of acid rain which is a cause

of deforestation.

1.5 Health consequences of environmental air pollution

The adverse effects of air pollution are well established and documented. Aside from the respiratory symptoms and allergic reactions that may be associated with the air pollution, it may seem difficult to link directly to some unexpected diseases to the effect of air pollution as the causative agent. Some of the diseases which can be attributed to the outdoor air pollution are as follows:

1.5.1 Respiratory diseases

Respiratory diseases are common manifestation of air pollution. The milder symptoms include runny nose, sore throat, cough and allergy. Serious condition includes asthma, pulmonary emphysema, pneumonia, chronic bronchitis. Chronic obstructive pulmonary disease (COPD) (Faustini A., 2013), pulmonary emphysema, pulmonary fibrosis. At early stage of life, air pollution could predispose children to development of childhood asthma. (Loftus A., 2014), (Ding G., 2014). Since the industrial revolution, the rate of pollution of our environment has increased tremendously with the negative impact threatening our existence in terms of the human health, and that is also related to the climate changes. Aside from active and passive smoking, predisposing factors to lung cancer include: exposure to NOx, coal fumes (Seow W.J., 2014).

1.5.2 Cardiovascular diseases

Several researches conducted from the last two decades have proved beyond reasonable doubt that air pollution can trigger irregular heart rhythms, strokes, heart attack especially in the people at risk of these conditions. (Gold R.D. and Samet M.J., 2013). Researchers have also attributed both short and long time increase in level of the following Pollutants to increase in hospitalization for cardiac diseases 'PM_{10}', '$PM_{2.5}$' (Wang Y. E. M., 2014), Ozone (Almeida S.M., 2014), SO_2, NO_2 (Zhao A., 2014). Recent studies by (Brucker N., 2014) evaluated possible effects of occupational exposure to air pollutants among taxi drivers and they found that exposure to polycyclic aromatic hydrocarbon (PAH) is an important factor that leads to atherosclerosis among the studied groups.

1.5.3 Diabetics

Diabetes is classified into two types according to the underlying mechanisms: type 1 which is insulin dependent diabetics and it usually presents in younger age. Type 2 diabetes in contrast often presents in the middle-aged and the elder adults. Longitudinal studies in human (Hathout E.H., 2002) show that increase in ozone exposure may be a contributory factor to increase in incidence of diabetes type 1 and PM_{10} may be a specific contributory factor to development of type 1 diabetics before the age of 5. Follow up studies of his research in 2006 conformed to his earlier result but ruled out the possibilities of SO_2, NO_2 and PM_{10} as a contributory factor to childhood asthma. In these studies researchers measured the exposure of air pollutant from birth until diagnosis. Moreover, researchers have also shown in series of experiment the relationship between road traffic pollutants and development of type diabetics.

Predisposing factors to diabetic type 2 are: $PM_{2.5}$ (Chen H., 2013), PM_{10}, NO_2 (Eze I.C., 2014).

1.5.4 Cancer

Recently, World Health Organization announced the classification of air pollution as class I human carcinogen due to cumulative evidence by the researchers across the world that air pollution is a predisposing factor to nasopharyngeal, lung, head and neck cancer (Wong I.C., 2014).

1.6 Risk assessment on air pollution

The 2013 assessment by WHO's International Agency for Research on Cancer (IARC) concluded that outdoor air pollution is carcinogenic to humans, with the particulate matter component of air pollution most closely associated with increased cancer incidence, especially lung cancer. An association also has been observed between outdoor air pollution and an increase in cancer of the urinary tract/bladder.

Ambient outdoor air pollution in both cities and rural areas was estimated to cause 3.7 million premature deaths worldwide per year in 2012; this mortality is due to exposure to small particulate matter of 10 microns or less in diameter (PM), which cause cardiovascular and respiratory disease, and cancers.

People living in low- and middle-income countries disproportionately experience the burden of outdoor air pollution with 88% (of the 3.7 million premature deaths) occurring in those countries, and they are the greatest burden in the WHO Western Pacific and Southeast Asian regions. The latest burden estimate reflects the very significant role of the air pollution which plays in cardiovascular illness and premature deaths – much more than previously understandings by scientists.

WHO Expert Consultation: Risk communication and intervention to reduce exposure and to minimize the health effects of air pollution which is the two-day consultation on 12–14 February 2019 at WHO headquarters in Geneva suggested: 1) 4.2 million deaths every year as a result of exposure to ambient (outdoor) air pollution, and 2) 3.8 million deaths every year as a result of household exposure to smoke from dirty cook stoves and fuels (1.4 million deaths due to stroke, 1.8 million deaths due to lung disease and cancer, 2.4 million deaths due to heart disease.)

Air pollution has a disastrous effect on children. World widely, up to 14% of children aged of 5 – 18 years old have asthma relating to factors including air pollution. Every year, 543,000 children younger than 5 years old die by respiratory disease linked to air pollution. Air pollution is also linked to childhood cancers. Pregnant women are exposed to air pollution that can affect fetal brain growth. Air pollution is also linked to cognitive impairment in both children and adults.

WHO estimates that approximately 80% of outdoor air pollution related to premature deaths were ischemic heart disease and strokes, while 14% of deaths were chronic obstructive pulmonary disease or acute lower respiratory infections; and 6% of deaths were lung cancer. Some deaths may be attributed to more than one risk factor at the same time. For example, both smoking and ambient air pollution affect lung cancer. Some death of lung cancer could have been averted by improving ambient air quality, or by reducing tobacco smoking.

Figure 1. Effects on human health by air pollution (Air Pollution infographics, WHO)

Table 1. Air Quality Guidelines (WHO, 2006)

$PM_{2.5}$	1 year mean	10 ($\mu g/m^3$)
	24 h mean	25 ($\mu g/m^3$)
PM_{10}	1 year mean	20 ($\mu g/m^3$)
	24 h mean	50 ($\mu g/m^3$)
Ozone, O_3	8 h, daily maximum	100($\mu g/m^3$)
Nitrogen dioxide, NO_2	1 year mean	40 ($\mu g/m^3$)
	1 h mean	200($\mu g/m^3$)
Sulfur dioxide, SO_2	24 h mean	20 ($\mu g/m^3$)
	10 min mean	500 ($\mu g/m^3$)

We conducted the study to calculate the premature mortality caused by the exposure of $PM_{2.5}$ in Cambodia using the C-R function derived from the epidemiological studies so that the adverse effect of $PM_{2.5}$ on human health can be estimated and an scientific information link to effective environmental policy in Cambodia.

2. Method

2.1 CMAQ/REAS Modeling System

The Community Multi-scale Air Quality (CMAQ) model is used to simulate the spatial distribution and temporal variations of $PM_{2.5}$ components and ozone for the simulation domain. This model is driven by the meteorological field simulated by the Weather Research and Forecasting (WRF) model (Skamarock W. C., et.al., 2008). For the year 2020 scenarios using the three-dimensional atmospheric chemistry and transport modeling system that simulates ozone, acid deposition, visibility, and fine PM throughout the troposphere. Designed as a one-atmosphere model, CMAQ can address

the complex couplings among several air quality issues simultaneously across spatial scales ranging from local to hemispheric. The CMAQ source code is highly transparent and modular to facilitate extensibility through community development.

CMAQ is a third-generation air quality model that is designed for applications ranging from regulatory and policy analysis to understanding the complex interactions of atmospheric chemistry and physics (Nolte G.C., et.al., 2018). First-generation air quality models simulated air quality using simple chemistry at local scales, and Gaussian plume formulation was the basis for prediction. Second-generation models covered a broader range of scales (local, urban, regional) and pollutants, addressing each scale with a separate model that often focused on a single pollutant (e.g., ozone). Third-generation models, like CMAQ, treat multiple pollutants simultaneously up to continental or larger scales, often incorporating feedback between chemical and meteorological components. The Model-3/CMAQ system was first released to public in July 1998 and had a recent update release in October 2006. The model structure and input data treatment processes of CMAQ were presented at Figure 2 below.

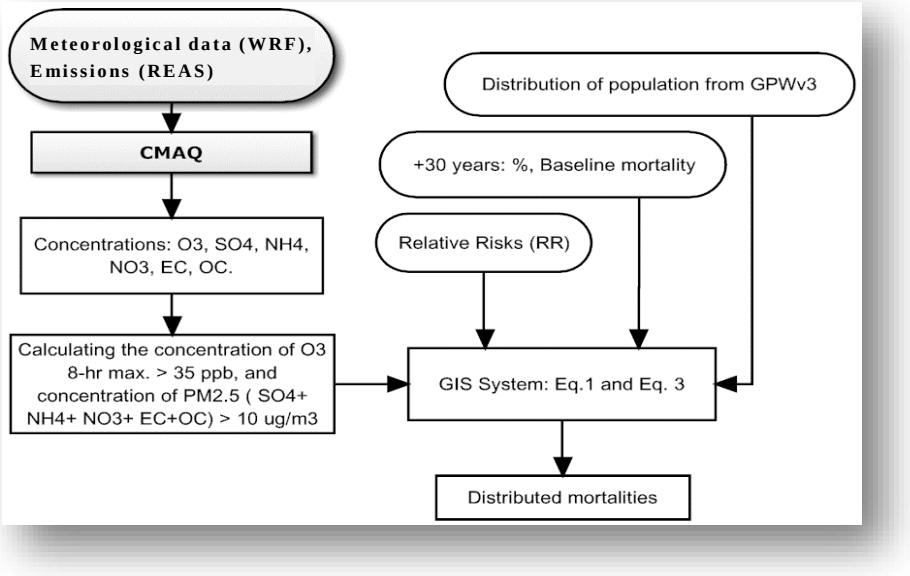

Figure 2. Research flowchart

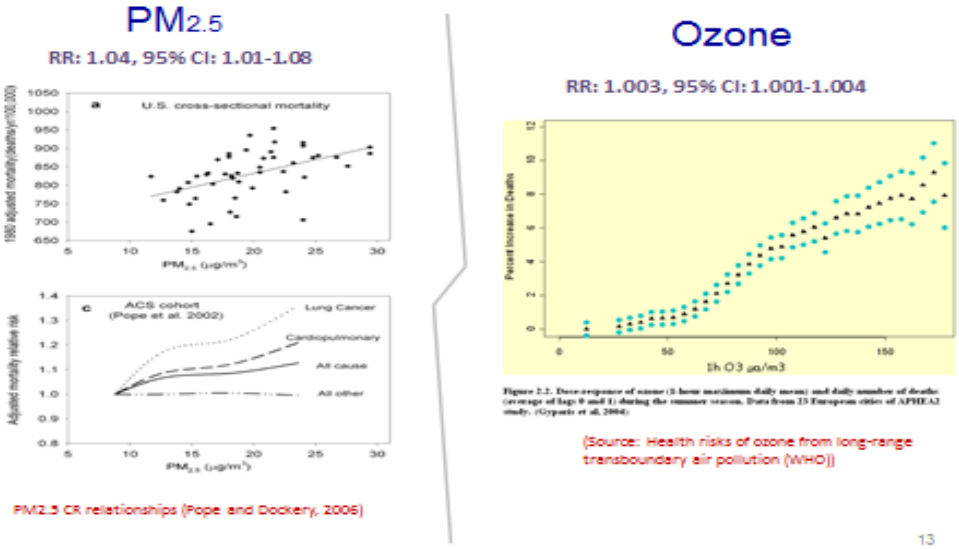

Figure 3. C-R function on epidemiological studies

2.2 $PM_{2.5}$: Exposure and Premature Mortality analysis

The distributed annual premature mortality rate in each grid cell is calculated as follows using Eq. (1) for $PM_{2.5}$ mean annual concentrations above 10µg/m^3 for the age group of 30 years and above:

$$mortality_{PM\ 2.5}(i,j,t)=pop(i,j,t)\ M_b(i,j,t)\ \beta_{PM2.5}\ \Delta PM_{2.5}(i.j,t) \qquad (1)$$

where mortality indicates premature mortality, i, j specify the location of grid cell within the simulation domain, t is the year of simulation, pop is the exposed population, Mb is the annual baseline mortality, β is the $PM_{2.5}$ CR coefficient, which can be calculated using Eq, (2). We use $PM_{2.5}$ mean annual concentration above 10 µg/m^3 since 10 µg/m^3 is the lowest level at which total, cardiopulmonary and lung cancer mortality have been shown to increase in WHO Air Quality Guidelines (2006). The value of β is 0.004.

$$\beta=\ln(RR)/\Delta C_{PM2.5} \qquad (2)$$

$\Delta C\ PM_{2.5}$ is the change in concentration.

According to Pope et al. (2002), an increase of annual average of $PM_{2.5}$, within a range from around 7.5 to 30 µg/m^3, caused a 4% (95% confidence interval: 1.01-1.08) increase in mortality rate for the age group of 30 years and above. This give the β a value 0.004 and $\Delta\ PM_{2.5}$ (i,j,t) is the change in the annual mean concentration above 10 µg/m^3 .

2.3 Ozone: Exposure and Premature Mortality analysis

The distributed annual premature mortality rate base on a RR value of 1.003 (95%CI: 1.001-1.004)(Anderson H.R., et al., 2004) 0.3% increase in daily premature mortality caused by a 10 μg/m^3 (5 pbb) change in 8 h maximum mean concentration above 70 μg/m^3 (35 pbb) (WHO, 2008) at each grid cell are calculated by summing the daily premature mortality, which can be calculated using the following function (USEPA 2006):

$$mortality_{O3}(i,j,n) = Y_0(i,j,n)[1 - \exp[-\beta_{O3}(\Delta O3(i.j,n)] \quad (3)$$

where n is the calculation day and Y0 is the daily mortality at certain ozone level where there is no statistically significant increase in mortality risk estimates was observed. We estimated it in our study by multiplying the population of the age group of 30 years and above by daily baseline mortality for age group. β is calculated from RR value of 1.003 from the previous study (cite the reference), which gives β a value 0.0003. ΔO3 is the change in ozone concentration calculated based on the daily maximum 8-h mean concentration above 35 ppb (or the value of the SOMO35 index of the day n) as follow:

$$SOMO35(i,j,n) = [\text{max 8 h mean} - 35]_n \quad (4)$$

The daily maximum 8-h mean concentration is the highest moving 8 h average to occur from hour 0:00-hour 23:00 in day.

2.4 Population Distribution

The information on the age and sex distribution of the population obtained from Cambodia Inter Censal Population Survey (CIPS) 2013 has been used to project the future age- and sex-stratified population. The results include population projections of national level and its provincial aggregated by sex five-year age groups and summary of some demographic indicators.

Table 2. The population by age and sex. Source: CIPS 201

Age Group	Sex		Total
	Male	Female	
0-4	674,274	629,314	1,303,588
5-9	748,334	726,316	1,474,650
0-14	797,569	745,592	1,543,161
15-19	788,186	769,818	1,558,004
20-24	811,231	802,710	1,613,941
25-29	666,403	676,517	1,342,920
30-34	580,996	629,941	1,210,937
35-39	361,176	373,794	734,970
40-44	388,882	455,941	844,823
45-49	366,219	406,380	772,599

50-54	292,251	369,099	661,350
55-59	198,455	300,049	498,504
60-64	156,355	234,264	390,619
65-69	111,775	160,882	272,657
70-74	80,798	121,192	201,990
75-79	51,190	79,618	130,808
80+	47,415	73,657	121,072
Total	**7,121,509**	**7,555,084**	**14,676,593**

3. Result

The premature mortality of $PM_{2.5}$ was calculated utilizing the C-R function of epidemiological studies and result of the Chemical Transportation Model (CMAQ). We used GIS software (Arc GIS 10) and data base of population distribution with the size of grid cell of 0.04167° (GPWv3; CIESIN 2005).

The monitoring of $PM_{2.5}$ in Cambodia started in 2017. The monitoring data of $PM_{2.5}$ in Phnom Penh in 2017 and 2018 is shown as Figure 4. The concentration of $PM_{2.5}$ is high in the dry season (November – April) and low in the rain season (May – October).

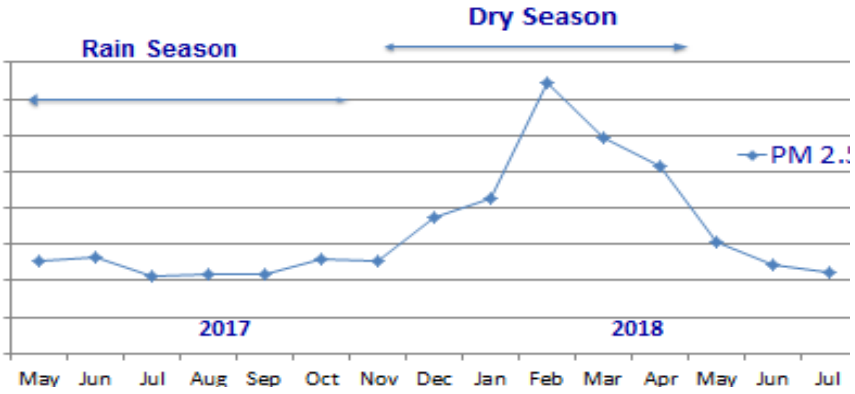

Figure 4. Graphic show Monthly Data of $PM_{2.5}$ monitoring 2017, 2018 in Phnom Penh

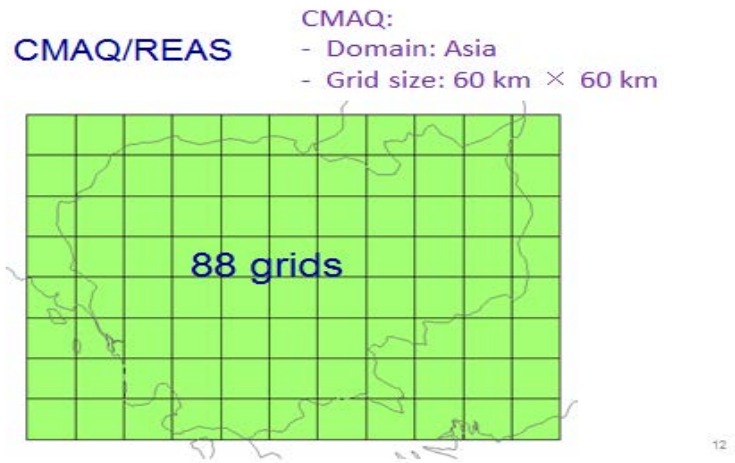

Figure 5. Grids size 60 km * 60 km

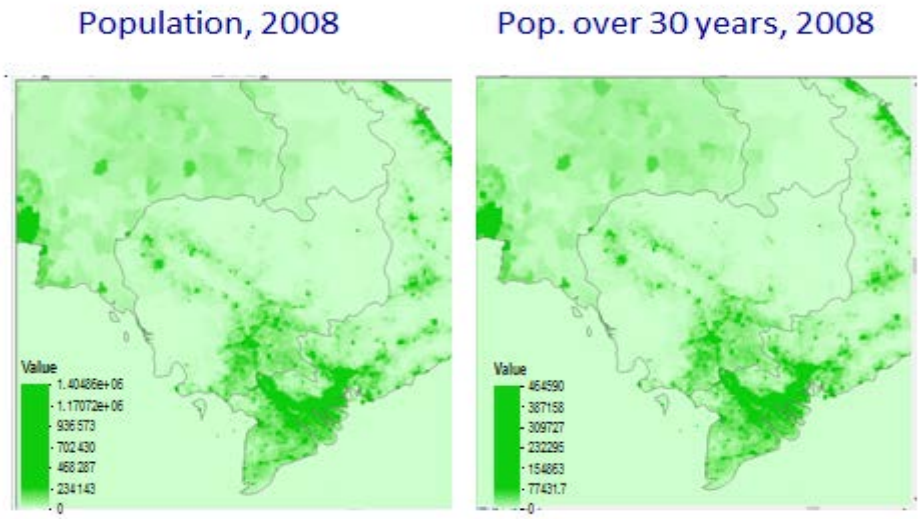

Figure 6. Pictures of ArcMap show the population 2008

We used the grid cell of 60 km×60 km for the calculation (Figure 4). The annual mean of concentration of $PM_{2.5}$ in each cell calculated by CMAQ is shown in Table 3. The population distribution of total age (left figure) and over 30 years (right figure) are shown in Figure 6.

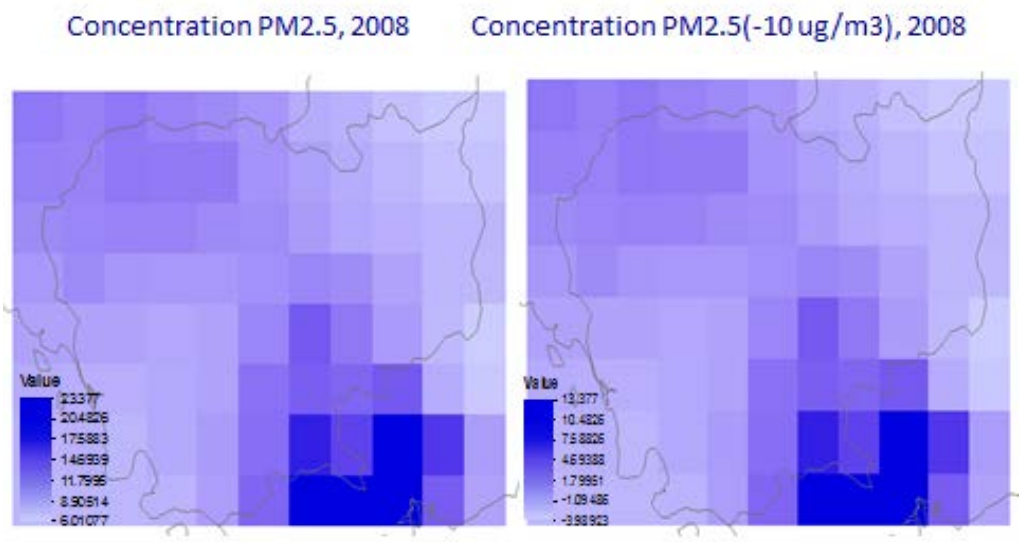

Figure7. Pictures of ArcMap show the Concentration $PM_{2.5}$ 2008 and $PM_{2.5}$(-10ug/m^3), 2008

Table 3: Annual mean of $PM_{2.5}$ in 2008 by CMAQ

No	ID	Long titude	Lati tude	PM2.5 ug/m3	No	ID	Long titude	Lati tude	PM2.5 ug/m3	No	ID	Long titude	Lati tude	PM2.5 ug/m3
1	4053	101.99	14.25	10.84	31	3861	106.60	13.39	7.76	61	3558	104.98	11.68	11.31
2	4054	102.56	14.30	10.47	32	3862	107.17	13.41	7.32	62	3559	105.54	11.71	12.56
3	4055	103.13	14.33	10.93	33	3863	107.73	13.42	7.31	63	3560	106.10	11.73	12.13
4	4056	103.70	14.37	10.17	34	3753	102.12	12.61	8.91	64	3561	106.65	11.75	12.87
5	4057	104.27	14.40	9.65	35	3754	102.68	12.65	9.83	65	3562	107.21	11.77	7.83
6	4058	104.85	14.43	9.11	36	3755	103.24	12.68	9.11	66	3563	107.77	11.78	6.35
7	4059	105.42	14.46	7.87	37	3756	103.81	12.72	9.00	67	3453	102.25	10.97	7.41
8	4060	105.99	14.48	7.48	38	3757	104.37	12.75	8.94	68	3454	102.80	11.01	7.47
9	4061	106.56	14.50	6.80	39	3758	104.93	12.78	9.36	69	3455	103.35	11.05	7.30
10	4062	107.13	14.52	6.17	40	3759	105.48	12.80	9.99	70	3456	103.90	11.08	8.07
11	4063	107.71	14.53	6.17	41	3760	106.05	12.82	9.51	71	3457	104.46	11.11	8.74
12	3953	102.04	13.70	10.38	42	3761	106.62	12.84	8.26	72	3458	105.01	11.14	11.53
13	3954	102.60	13.74	10.16	43	3762	107.18	12.86	7.25	73	3459	105.56	11.16	17.10
14	3955	103.17	13.78	10.95	44	3763	107.74	12.87	7.08	74	3460	106.12	11.19	14.50
15	3956	103.74	13.82	10.76	45	3653	102.16	12.06	8.34	75	3461	106.67	11.20	23.38
16	3957	104.31	13.85	10.74	46	3654	102.72	12.10	8.26	76	3462	107.23	11.22	15.36
17	3958	104.87	13.88	9.35	47	3655	103.28	12.14	8.31	77	3463	107.78	11.23	8.63
18	3959	105.44	13.90	8.51	48	3656	103.84	12.17	8.11	78	3353	102.29	10.43	7.15
19	3960	106.01	13.93	7.86	49	3657	104.40	12.20	8.32	79	3354	102.84	10.47	7.20
20	3961	106.58	13.95	7.23	50	3658	104.96	12.23	10.13	80	3355	103.39	10.51	7.23
21	3962	107.15	13.96	6.68	51	3659	105.52	12.25	12.96	81	3356	103.94	10.54	7.38
22	3963	107.72	13.98	6.60	52	3660	106.08	12.28	10.87	82	3357	104.49	10.57	8.68
23	3853	102.08	13.15	9.73	53	3661	106.64	12.30	8.80	83	3358	105.04	10.60	12.14
24	3854	102.64	13.19	10.03	54	3662	107.20	12.31	7.02	84	3359	105.59	10.62	20.21
25	3855	103.21	13.23	10.28	55	3663	107.76	12.32	6.01	85	3360	106.14	10.64	23.06
26	3856	103.77	13.27	10.14	56	3553	102.21	11.51	7.78	86	3361	106.69	10.66	22.73
27	3857	104.34	13.30	9.84	57	3554	102.76	11.55	7.71	87	3362	107.24	10.68	12.74
28	3858	104.90	13.33	9.57	58	3555	103.32	11.59	7.68	88	3363	107.79	10.69	8.30
29	3859	105.47	13.35	8.81	59	3556	103.87	11.62	8.00					
30	3860	106.03	13.37	8.09	60	3557	104.43	11.66	8.30					

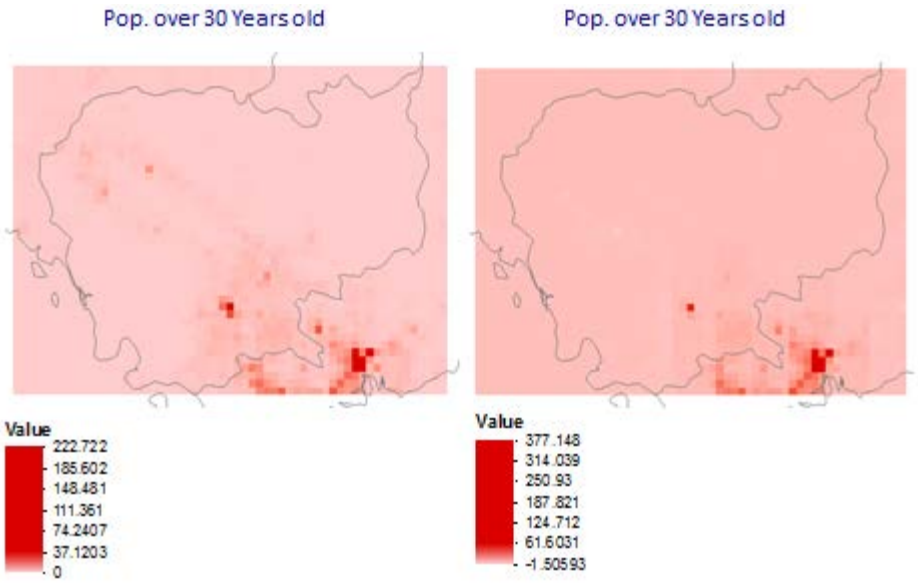

Figure 8. Pictures of ArcMap show Mortality of $PM_{2.5}$(left) and $PM_{2.5}$ (-10 ug/m^3)(right), 2008

The annual mean of the concentration of $PM_{2.5}$ in 2008 calculated by CMAQ is shown

in Figure 7. The concentration is relatively high in the southeast area of Cambodia facing Vietnam. The premature mortality caused by $PM_{2.5}$ was calculated for people over 30 years old (Figure 8). The calculation was done for the annual mean of $PM_{2.5}$ and the annual mean minus 10ug/m^3. The right figure (annual mean minus 10ug/m^3) shows a smaller number of premature mortality than left figure (annual mean) because of the linearity of the C-R function of $PM_{2.5}$.

Table 4. Premature Mortality of $PM_{2.5}$, 2008, Pop. over 30 Years old

TableMorPMPop08

Rowid	LONG_NAME	ZONE-CODE	COUNT	AREA	MIN	MAX	RANGE	MEAN	STD	SUM
1	Kingdom of Cambodia	68	1512	15.12	0	51.473156	51.473156	0.260102	1.401292	393
2	Lao People's Democratic Republic	69	81	0.81	0.002844	0.121409	0.118565	0.048121	0.043661	4
3	Kingdom of Thailand	71	256	2.56	0.021111	1.160119	1.139007	0.22701	0.142465	58
4	Socialist Republic of Vietnam	72	447	4.47	0.001728	222.722473	222.720745	1.588973	11.013727	710

Table 5. Premature Mortality of $PM_{2.5}$ (-10 ug/m^3), 2008, Pop. over 30 Years old

TableMorPM8M10

Rowid	LONG_NAME	ZONE-CODE	COUNT	AREA	MIN	MAX	RANGE	MEAN	STD	SUM
1	Kingdom of Cambodia	68	1512	15.12	-0.959218	18.005146	18.964364	0.071784	0.558992	109
2	Lao People's Democratic Republic	69	81	0.81	-0.117448	-0.004463	0.112985	-0.042092	0.037245	-3
3	Kingdom of Thailand	71	256	2.56	-0.700566	0.19956	0.900127	-0.010681	0.073628	-3
4	Socialist Republic of Vietnam	72	447	4.47	-1.505929	377.147644	378.653573	2.276111	18.666771	1017

Table 4 and 5 show the total number of the estimated premature mortality in Cambodia calculated by GIS. The number of the premature mortality of Lao, Thailand and Vietnam were also calculated partly included in the domain (see Figure 5). The premature mortality of $PM_{2.5}$ is 109 (Table 5), however, it is 393 (Table 4) if the concentration under 10ug/m^3 is included.

Table 6 indicates the aggregated relative risk (RR) of ozone for dairy maximum of 8-hour mean in each grid cell in the domain in 2008. Figure 9 shows the distribution of the RR in the domain (left figure) and calculated premature mortality using GIS. The distribution of the premature mortality due to ozone is relatively high in the northwest part of Cambodia facing Thailand, which is different from the distribution of the premature mortality of $PM_{2.5}$.

The total number of premature mortality of ozone in 2008 in Cambodia is 177 (Table 7).

Table 6. Total daily RR of daily 8-hour mean, daily maximum of O3 in 2008 calculate by CMAQ

No	ID	Long titude	Lati tude	O3 conc	RR DAY	No	ID	Long titude	Lati tude	O3 conc	RR DAY	No	ID	Long titude	Lati tude	O3 conc	RR DAY
1	3260	106.16	10.10	1.06	0.32	53	3559	105.54	11.71	1.81	0.54	85	3858	104.90	13.33	2.24	0.67
2	3261	106.71	10.12	1.05	0.31	54	3560	106.10	11.75	1.62	0.48	86	3859	105.47	13.35	2.00	0.60
3	3262	107.26	10.14	1.66	0.50	55	3561	106.65	11.75	1.36	0.41	87	3860	106.05	13.37	2.06	0.62
4	3263	107.80	10.15	1.70	0.51	56	3562	107.21	11.77	1.18	0.35	88	3861	106.60	13.39	2.01	0.60
5	3353	102.29	10.45	2.20	0.66	57	3565	107.77	11.78	1.26	0.38	89	3862	107.17	13.41	1.76	0.53
6	3354	102.84	10.47	2.12	0.63	58	3653	102.18	12.06	2.80	0.84	70	3865	107.75	13.42	1.69	0.51
7	3355	103.39	10.51	1.99	0.59	59	3654	102.72	12.10	2.05	0.61	71	3955	102.04	13.70	3.12	0.93
8	3356	103.94	10.54	1.85	0.55	60	3655	103.28	12.14	1.87	0.56	72	3956	102.80	13.74	3.19	0.95
9	3357	104.49	10.57	1.32	0.40	61	3656	103.84	12.17	1.89	0.57	73	3955	103.17	13.75	2.86	0.86
10	3358	105.04	10.60	1.27	0.38	62	3657	104.40	12.20	1.87	0.56	74	3958	103.74	13.82	3.10	0.93
11	3359	105.59	10.62	1.36	0.41	63	3658	104.96	12.23	1.75	0.52	75	3957	104.31	13.85	2.81	0.84
12	3360	106.14	10.64	1.37	0.41	64	3659	105.52	12.25	1.84	0.55	76	3955	104.87	13.88	2.51	0.75
13	3361	106.69	10.66	1.40	0.42	65	3660	106.05	12.28	2.03	0.61	77	3959	105.44	13.90	2.24	0.67
14	3362	107.24	10.68	1.32	0.39	66	3661	106.64	12.30	1.89	0.56	78	3960	106.01	13.93	2.01	0.60
15	3363	107.79	10.69	1.11	0.33	67	3662	107.20	12.31	1.45	0.43	79	3961	106.58	13.95	1.58	0.47
16	3453	102.25	10.97	2.36	0.71	68	3663	107.76	12.32	1.46	0.44	80	3962	107.15	13.96	1.43	0.43
17	3454	102.80	11.01	2.22	0.66	49	3753	102.12	12.61	2.51	0.75	81	3965	107.72	13.98	1.83	0.55
18	3455	103.35	11.05	1.35	0.40	50	3754	102.68	12.65	2.26	0.68	82	4053	101.99	14.25	3.62	1.08
19	3456	103.90	11.08	1.24	0.37	51	3755	103.24	12.68	2.01	0.60	83	4054	102.56	14.30	3.37	1.01
20	3457	104.46	11.11	1.40	0.42	52	3756	103.81	12.72	2.06	0.62	84	4055	103.13	14.33	2.81	0.84
21	3458	105.01	11.14	1.39	0.42	53	3757	104.37	12.75	2.74	0.82	85	4056	103.70	14.37	2.87	0.86
22	3459	105.56	11.16	1.67	0.50	54	3758	104.93	12.78	1.94	0.58	86	4057	104.27	14.40	2.50	0.75
23	3460	106.12	11.19	1.58	0.47	55	3759	105.49	12.80	2.07	0.62	87	4058	104.85	14.43	2.19	0.65
24	3461	106.67	11.20	1.66	0.50	56	3760	106.05	12.82	2.06	0.62	88	4059	105.42	14.46	2.12	0.64
25	3462	107.23	11.22	1.40	0.42	57	3761	106.62	12.84	2.15	0.64	89	4060	105.99	14.48	1.76	0.53
26	3463	107.78	11.23	1.20	0.36	58	3762	107.18	12.86	1.82	0.54	90	4061	106.56	14.50	1.70	0.51
27	3553	102.21	11.51	2.61	0.78	59	3765	107.74	12.87	1.64	0.49	91	4062	107.13	14.52	1.79	0.54
28	3554	102.76	11.55	2.40	0.72	60	3855	102.08	13.15	2.93	0.88	92	4065	107.71	14.55	1.95	0.58
29	3555	103.32	11.59	1.63	0.49	61	3854	102.64	13.19	2.95	0.88	93	4154	102.52	14.85	3.58	1.07
30	3556	103.87	11.62	1.54	0.46	62	3855	103.21	13.23	2.45	0.73	94	4155	103.09	14.89	3.37	1.01
31	3557	104.43	11.66	1.62	0.48	63	3856	103.77	13.27	2.36	0.71	95	4156	103.67	14.92	3.11	0.93
32	3558	104.98	11.68	1.56	0.47	64	3857	104.34	13.30	2.52	0.75	96	4157	104.24	14.96	2.84	0.85

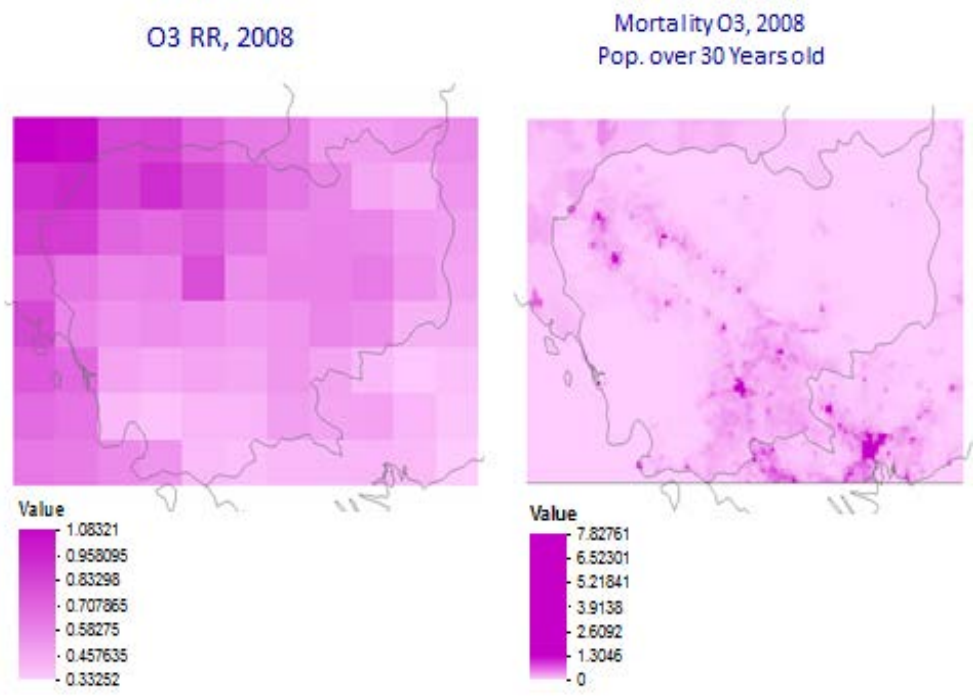

Figure 9. Pictures of ArcMap show RR (left) and Mortality of O_3 (right) 2008

Table 7. ArcMap show RR and Mortality of O_3 2008

ZonalCaMortality

Rowid	LONG_NAME	ZONE-CODE	COUNT	AREA	MIN	MAX	RANGE	MEAN	STD	SUM
1	Kingdom of Cambodia	1	8718	15.135417	0	3.657695	3.657695	0.020295	0.066866	177
2	Lao People's Democratic Republic	2	512	0.888889	0.000052	0.019815	0.019763	0.00629	0.005852	3
3	Kingdom of Thailand	3	1524	2.645833	0.000081	0.200232	0.200151	0.034707	0.023228	53
4	Socialist Republic of Vietnam	4	2766	4.802083	0.000038	7.827607	7.827569	0.050397	0.250928	139

4. Discussion

Our finding is that ozone and $PM_{2.5}$ contributed to increase annual premature mortality rate in Cambodia.

We estimated the premature mortality attributed to the following diseases:

- cardiopulmonary causes
- cardiovascular causes
- ischemic heart disease
- respiratory causes
- lung cancer

4.1 Premature Mortality

Premature Mortality caused by exposure to both ozone and $PM_{2.5}$ in Cambodia year 2008 are estimated to be about 393 premature deaths for $PM_{2.5}$ and 176 for Ozone. The estimated distributed premature mortality is caused by exposure to $PM_{2.5}$ annual mean concentration under and above 10 $\mu g/m^3$ and the daily maximum 8-h mean concentrations of ozone above 35 ppb for the age group of over 30 years. If we estimate the premature mortality of the concentration $PM_{2.5}$ above 10 ug/m^3, the result is 108 premature deaths (Figure 10).

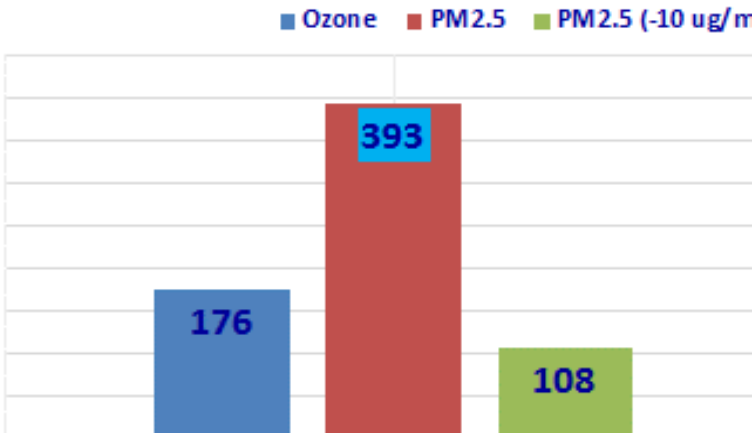

Figure 10. Premature mortality in Cambodia by air pollution (2008)

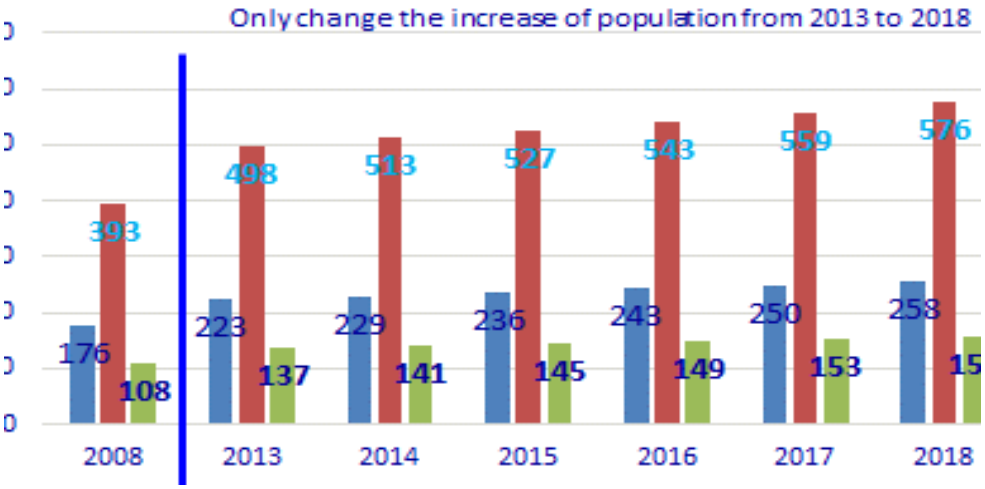

Figure 11. Premature mortality in Cambodia by air pollution 2008, 2013-2018

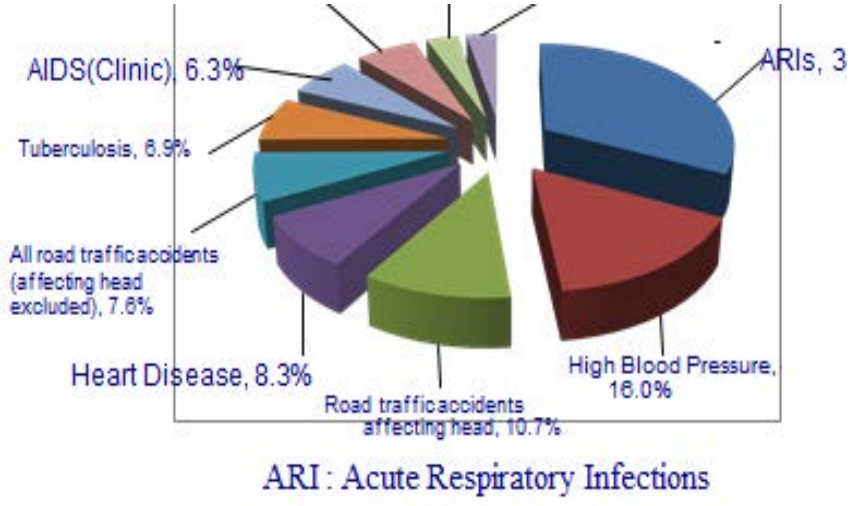

Figure 12. National Health Statistic Report 2011

We estimated the premature mortality of $PM_{2.5}$ and ozone in the future (2013 – 2018) assuming the population continue to increase constantly (Figure 11). It shows the premature mortality in 2018 will be 258 (ozone) and 576 ($PM_{2.5}$)/158($PM_{2.5}$ above 10 $\mu g/m^3$).

The positive associations of $PM_{2.5}$ with cardiorespiratory and lung cancer mortality indicated by the previous epidemiological studies are consistent with biological pathways through which $PM_{2.5}$ may influence health (Pun C.V., et al., 2016). $PM_{2.5}$ has been shown to deposit in the alveolar region of the lung and to move into interstitial spaces between cells and towards other organs. The mechanisms through which chronic $PM_{2.5}$ exposure may increase risk of respiratory disease mortality are poorly defined but may involve systemic inflammation and oxidative stress in the lung epithelial cells.

5. Conclusion

In conclusion, we estimated the number of premature deaths related to $PM_{2.5}$ and Ozone.

The result which are based on the CMAQ modeling system for year 2008 shows that elevated ozone and $PM_{2.5}$ concentration in Cambodia probably cause some risks on human health as well as other environmental risks. Our result shows that the number of deaths due to $PM_{2.5}$ on human health is greater than the effect of ozone for the age group over 30 years. The case of premature deaths in Cambodia caused by $PM_{2.5}$ is estimated as 393 and 108 (above 10 $\mu g/m^3$), and the case caused by ozone is estimated 176 for age group over 30 years.

Assessing premature mortality risks by exposure to elevated concentrations of $PM_{2.5}$ and Ozone in Cambodia, where rapid development and exploding urbanization have been increasing annually, involves unique challenges in the region that lacks adequate ozone and $PM_{2.5}$ epidemiological studies.

In this study, we only try to estimate the adverse effect on human health

quantitatively but not to detect the effect on each disease such as the risk of death from cardiovascular by ozone. We did, however, indicate clearly a significant increase of the risk of premature death including the respiratory causes in association with an increase in ozone concentration.

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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.

Cooperation for air quality improvement in four model cities in China

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Abstract

In order to improve the environment in China, air quality improvement cooperation projects are being promoted in cooperation between local cities in Japan and China. At the request of the central governments of Japan and China, joint research with four model cities (Chongqing, Xi'an, Xiamen, and Zhuhai) was conducted for four years from 2015. Asia Center for Air Pollution Research (ACAP) became the counter organization to solve their problems. The environmental issues of the four model cities were various, such as ozone and VOC. For each environmental issue, Japanese environmental technology and measurement analysis technology were introduced to achieve certain results that could be reflected in the policy, and at the same time, new issues also emerged. In this article, contents of joint research conducted in the four model cities are introduced.

Keywords: Ozone, VOC, Regional pollution, Remote sensing, real world driving emission

1. Introduction

In early 2013, very severe air pollution occurred over a wide area of mainland China. The main pollutant was PM2.5, which was a serious air quality situation, with some daily average values exceeding 0.50 mg m^{-3}. The Beijing government and other local governments have taken emergency measures such as temporarily shutting down large-scale factories, but air quality have not improved with the unfavorable weather conditions. Chinese central government, which took the situation seriously, started to develop an action plan to prevent air pollution while accelerating the establishment of a monitoring system. Since air pollution was not a temporary phenomenon but continued intermittently thereafter, the Ministry of the Environment Japan decided to continue cooperation in order to improve the Chinese air environment. With emphasis on the accumulation of experience and know-how in Japan's air pollution control measures in local governments, the government decided to implement the Japan-China Inter-City Cooperation Project for The Improvement of The Chinese Air Environment, focusing on cooperation between Japanese local governments and Chinese cities.

Based on the basic concept described above, the total number of 11 local governments (Toyama, Saitama, Tokyo, Nagano, Hyogo, Fukuoka, Kawasaki, Yokkaichi, Kobe, Kitakyushu, and Oita) in Japan that participated in the “City Framework for Intercity Cooperation” in response to calls from the Ministry of the Environment Japan. In addition, 13 local governments (cities) from China (Liaoning, Shanxi, Hebei, Jiangsu, Guangdong, Beijing, Shanghai, Tianjin, Shenyang, Dalian, Tangshan, Sakai, and Wuhan) participated in this project.

On the other hand, in the Sino-Japan Friendship Center for Environmental Protection (SJFC), which was designated as platform by the Ministry of Environmental Protection, China (current, Ministry of Ecology and Environment), had been proposed to cooperate with candidate 4 model cities (Chongqing, Xi'an, Xiamen and Zhuhai) apart from the above-mentioned cooperation. Since there was no counterpart on the Japanese side that could appropriately respond to requests from these 4 model cities, a new technical support organization (technical platform) was established in Asia Center for Air Pollution Research (ACAP) under the Japan Environmental Sanitation Center to meet the demands from the four cities. This activity was continued for 4 years from 2015.

2. Outline of cooperation for 4 model cities

Table 1 shows the outline of cooperation for the four model cities. Initially, the Chongqing City Environmental Protection Bureau and the Environmental Science Research Institute requested the impact of NOx from automobile exhaust, but they asked for cooperation on the bad odor derived from industrial factories and the VOC countermeasures that the central government is focusing on. In Xi'an, high-concentration ozone in summer and PM2.5 in winter are issues. The Institute for Environmental Science is working on the investigation of the cause of high-concentration ozone, and cooperation for the mechanism study of high-concentration ozone was requested. Xiamen City is one of the good air quality cities in China. However, the impact of NOx from automobile exhaust is large, and Xiamen environment monitoring center have been asking for cooperation on emission measures. Zhuhai City, located in the Pearl River Delta, had requested as a cooperating item to grasp the actual state of VOC emissions as a measure against ozone.

Table 1. Outline of cooperative items for the four model cities

Chongqing	Examination of VOC emission control measures derived from industrial activities
Xi'an	Study on ozone pollution control measures by analysis of high concentration ozone generation mechanism
Xiamen	Examination of exhaust gas regulation by utilizing big data from vehicle exhaust gas monitoring by remote sensing and actual running emission data using on board vehicle NOx/PM meter
Zhuhai	Support for the construction of a comprehensive system for measures against volatile organic compounds for ozone control

Technical cooperation was implemented in three ways: 1. status assessment and technical exchange, 2. seminars for local staff, and 3. training inviting local staff to

Japan. As shown in Table 2, the technical exchange reached 39 times in 4 years. A large-scale experiment was conducted in Xiamen in 2018, so the number of visits increased compared to others.

Table 2. Number of technical exchange (meeting and experiments)

	Chongqing	Xi'an	Xiamen	Zhuhai	Total
FY2015	1	1	1	1	4
FY2016	3	3	2	2	10
FY2017	3	2	3	3	11
FY2018	1	4	7	2	14
Total	8	10	13	8	39

Table 3 shows the number and location of technical seminars held locally. By inviting Japanese experts on the research issues requested in each city, technical seminars were held with participants from the four model cities gathered at the venue.

Table 3. Number and location of technical seminars

	Number	Location
FY2015	2	Chongqing, Xi'an
FY2016	0	
FY2017	2	Chongqing, Xiamen
FY2018	1	Zhuhai
Total	5	

Invitation training programs for visiting Japan were conducted as shown in Table 4 by inviting officials and environmental researchers from the four model cities, as well as staff from SJFC. In 2018, Xi'an and Zhuhai could not participate due to the circumstances of their problem.

Table 4. Invitation training programs for visiting Japan

	Number	Number of guests invited	Invited organization
FY2015	1	20	Chongqing, Xi'an, Xiamen, Zhuhai, SJFC
FY2016	4	20	Chongqing, Xi'an, Xiamen, Zhuhai, SJFC
FY2017	2	18	Chongqing, Xi'an, Xiamen, Zhuhai, SJFC
FY2018	1	14	Chongqing, Xiamen, SJFC
Total	8	72	

3. Details of the cooperative studies

Regarding the issues in the four model cities shown in Table 1, technical exchange meeting was held as shown in Table 2. Data analysis including observations was conducted in three cities except Zhuhai. This chapter focuses on the field observations and analysis results.

3.1 Chongqing

The purpose of the research is to examine the appropriate implementation method of VOC emission control measures derived from industrial activities. In order to effectively reduce VOC, the obtained results will be reflected in VOC emission

reduction guidance. As a model case, the environmental impact of VOCs emitted from automobile painting plants was taken up. This is to confirm the reduction of environmental impact by introducing VOC removal equipment. Therefore, the VOC concentration measurement in the air before and after the introduction of the VOC removal equipment was conducted. Figure 1 shows the spatial distribution of ethylbenzene concentration around the automobile paint factory building. The measurement was conducted on October 28 and 29, 2016, before the introduction of the VOC removal equipment.

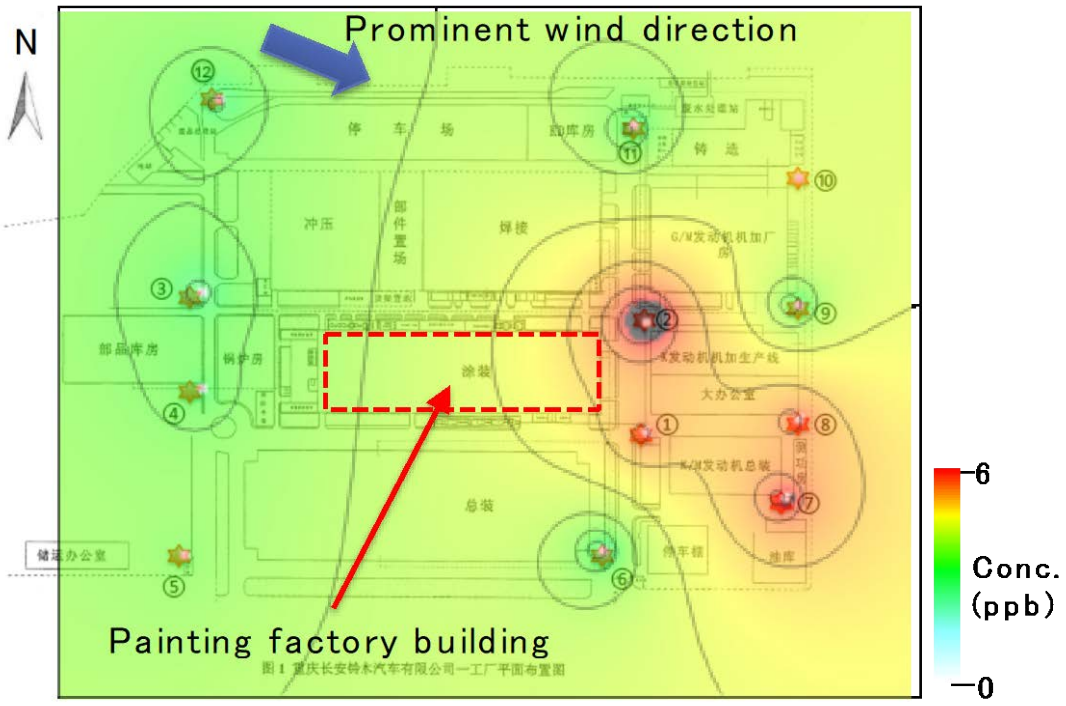

Figure 1. Spatial distribution of ethylbenzene concentration around automobile paint factory building measured on October 28 and 29, 2016

In the measurement, ambient air was collected from 12 locations around the paint building with a syringe, and the VOC concentration was measured with two types of sensor gas chromatograph (SGVA-P2 and SGEA-P2) from FIS Inc., Japan. The ambient air was collected every 3 hours at the same location, and the average concentration of 6 times is shown in Figure 1. A northwest prevailing wind was observed. A high concentration of ethylbenzene used as a solvent for paint was observed around the lee of the paint building.

Similar observations were conducted on February 9 and 10, 2017 after the introduction of the VOC removal system. Compared to the previous observation, the atmospheric stability was higher and the general pollution concentration such as PM2.5 concentration was higher, however, a clear decrease in the ethylbenzene concentration was confirmed. On the other hand, high concentrations of VOCs such as toluene, benzene, etc. were found in the places different from the production site in the factory yard. Although it was possible to control the emission amount of VOCs from the chimney (so called end-of-pipe emission), it becomes clear that the impacts of non-

organized emission, such as intentional disposal of solvents on the road and leakage from the exhaust pipe, were large.

To evaluate VOC management in factories more generally, Chongqing City created an evaluation table and devised a method for numerical evaluation. In order to verify that this evaluation table correctly evaluates VOC management, four Japanese VOC management experts were invited to Chongqing City to evaluate the same factories from the Japanese perspective. On May 7 and 8, 2018, the Japanese VOC management experts visited four factories to evaluate the management status of VOCs. Table 5 shows the comparison results. The results from the four factories judged by Japanese experts and the results independently evaluated by Chongqing City were almost the same.

Table 5. Evaluation of VOC emission management

	Evaluation results based on Chinese indicators	Japanese Expert Evaluation Results
Evaluation method	Assigning values based on normalization methods and grades	Assigning values by experts
Number of companies surveyed	15	4
Evaluation factor	· 8 Technical Indicators: processing effectiveness, maturity of technology, adaptability, Initial cost, running cost, Resource consumption, Construction period, secondary contamination · 5 Supervisory control indicators: Monitoring, Production control, Process supervision, Capacity building, Risk management	VOC emission facility data, Facilities and production management including improvement, Exhaust gas treatment technology (equipment), Social indicators, etc.
Evaluation results	· Each Company's VOC emissions are reduced and emissions reduction reached 89.3%, however, the difference between the technology and the company is large. · Effective technology: concentrated rotor + TNV > concentrated rotor + RTO > Photocatalyst > Activated carbon adsorption > Jets Scrubber · Regulatory compliance: car manufacturing companies are better than parts manufacturing industries.	· Chongqing Ford is the Best. Qin River is comparatively bad. · Each company has advanced and effective processing technology. But, environmental awareness is low. The recovery rate of the gas is not high yet. The need to reduce costs and implement energy. · As for the adsorption of activated carbon, recycling and reuse for VOC reduction were not carried out.
Comparison of evaluation results	As a result of both evaluations, the results of the company rankings are similar. The Japanese side evaluation confirms that the completion of the index system of the Chinese side is high, the value of each factor is appropriate, and the effect of the reduction is scientifically reflected.	

3.2 Xi'an

The purpose of the research is to study counter measures to control the ozone generation in summer through understanding of high concentration ozone generation mechanisms in Xi'an city and surrounding areas.

The ozone concentration varies in various ways under the influence of surrounding pollutants and weather (Minoura, 1999). Figure 2a shows the spatial distribution of the averaged potential ozone concentration (PO; sum of ozone and NO_2 concentrations) from April to September 2016 based on hourly data of 13 atmospheric measurement stations under the jurisdiction of Xi'an City. Since ozone reacts with NO and changes to NO_2 immediately, impacts of the NOx concentration is important in understanding the ozone concentration. PO is a conservative quantity not affected by concentration fluctuations due to ozone oxidation and photodissociation, and indicating the oxidizing ability in the environment and is an index of the degree of contamination. The PO

concentration is high during the day and low at night due to ozone generation. The spatial distribution of PO concentration using hourly values tends to be high during the day and low at night in the entire city of Xi'an, however the long-term average for half a year can be used to understand a regional characteristics of air pollution from distribution (Figure 2a). It was revealed that the PO concentration was significantly different between the 6th station (Xiaozhai) and the 12th station (Ganting Town), which were only 4 km away. In order to understand this cause, the situation around each station was investigated. As a result, it was found that the No. 6 station was in an office district lined with high-rise buildings, while the No. 12 station was installed in an open area near a large pond, and the situation of the diffusion of the contaminant including ozone was different.

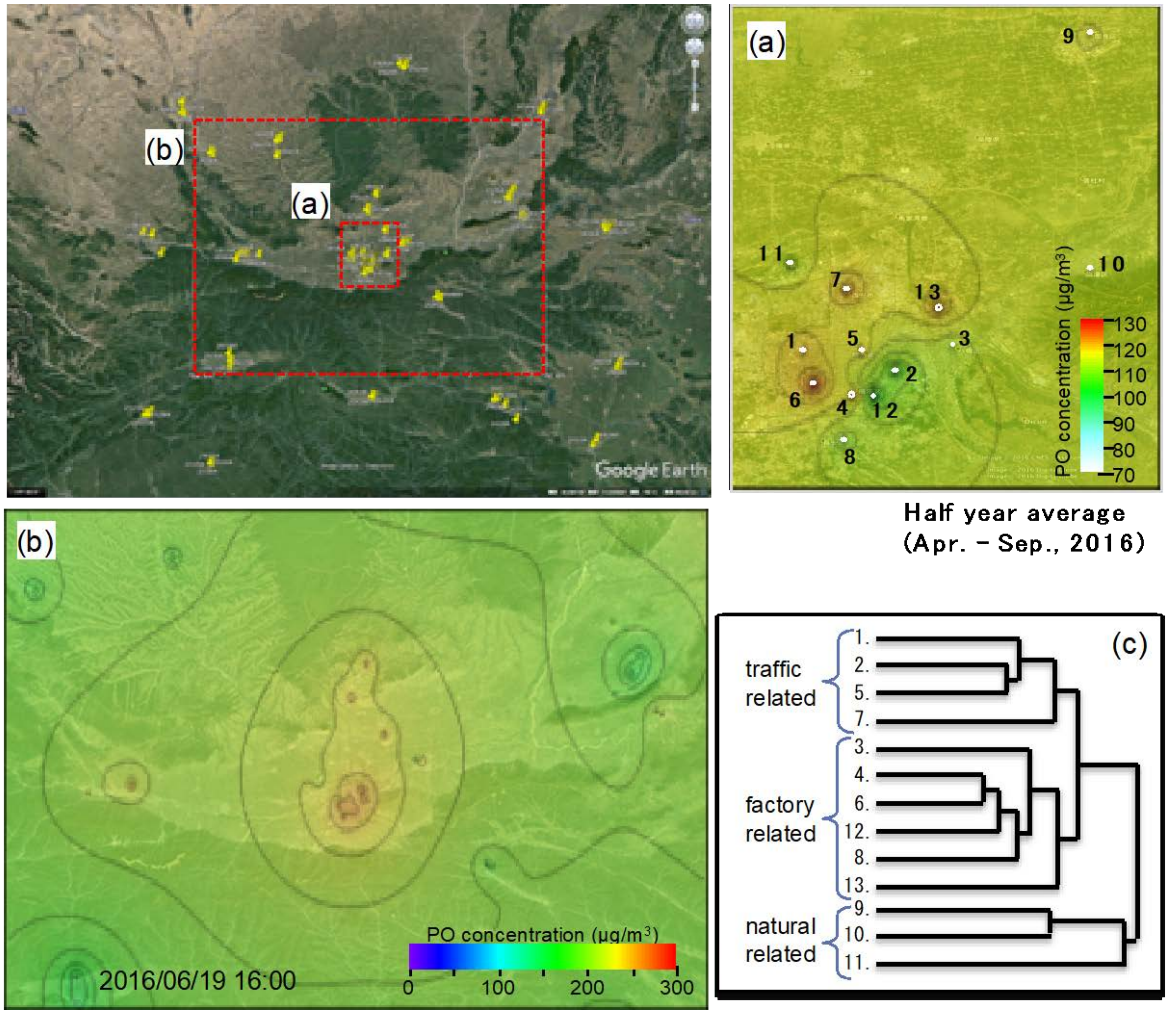

Figure 2. Analysis result of one hour data, (a) a half year average of Potential ozone (PO) concentration distribution in Xi'an city, (b) one hour PO distribution around Xi'an basin using data from 108 monitoring station, (c) Cluster classification of 13 stations in Xi'an obtained from ozone concentration variation of one hour data for a half year

In order to understand the spatial distribution of PO concentration in one hour data, the analysis range was expanded and 103 monitoring stations data around Xi'an city were used. Figure 2b shows the PO concentration distribution at the time when high

ozone concentration was observed in Xi'an. PO concentration distribution centered on Xi'an city was observed. It was found that the center of the PO distribution generated from the previous day moved from an eastern area to Xi'an city. Air pollutants such as PO was found to circulate in Xi'an and eastern regions due to local winds such as mountain breeze. This indicates that even if Xi'an City implements emission control, the ozone concentration may not decrease due to pollution in the surrounding area, and it became clear that pollution counter measures must be implemented not only in Xi'an city area but also in Shanxi province and a wide region. Furthermore, for future counter measures against wide-area pollution, it is considered that the prediction of the emission source contributions in a wide area using an air quality simulation model is effective. For that reason, the US EPA's atmospheric simulation model CMAQ (Byun and Schere, 2006) has introduced in the computer of the Xi'an City Environmental Science Institute to calculate the contribution of emission sources.

One method of using hourly data is to classify the pattern of concentration change and grasp the relationship with the emission source. As described above, ozone undergoes a concentration change due to NO. The power spectrum is obtained by Fourier transforming the concentration fluctuations for about half a year using one hour data of ozone concentration. A spectrum with high periodicity is obtained by Fourier analysis, and a common change is obtained by classifying the spectrum at each station. Figure 2c shows the cluster analysis result as a tree-like relationship for 13 stations. The monitoring stations could be roughly classified into three categories. The first group is a vehicle traffic related group that is affected by morning and night traffic rush. The next group is the group that is affected by factory emissions which showed the suspension of activities during the lunch break. The last group is strongly influenced by biogenic VOCs and solar radiation, and less influenced by anthropogenic NO.

3.3 Xiamen

Xiamen is a city with a population of 4 million, and is the second most environmentally friendly city in 2018 among 169 major cities in China. Xiamen's main facilities are concentrated on the Xiamen island and are connected to the mainland by four bridges. The number of registered cars is 1.57 million, which flows into the island from the surrounding mainland in the morning. NOx derived from automobiles is a big problem in Xiamen. The purpose of the research is to examine exhaust gas regulations using big data of automobile exhaust gas monitoring by remote sensing devices and actual (real-world) driving emission data by on-board vehicle NOx/PM meter.

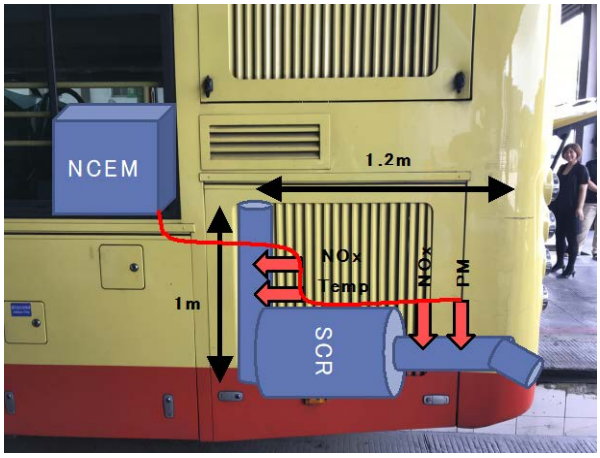

Figure 3. Sensors set-up

The remote sensing device (RSD) is a device that projects ultraviolet rays and infrared rays so as to cross the road and measures the exhaust gas concentration immediately after passing

through the vehicle from the light transmission amount. The Xiamen Environmental Monitoring center installed the RSD in all four bridges. The vehicle type and owner of a vehicle are specified from a license plate that is simultaneously photographed. Since there are multiple lanes, it becomes invalid when the vehicles passed simultaneously. However, hundreds of thousands of exhaust gas data measured are accumulated at the Xiamen Traffic Center. As a result of analyzing these big data, it became clear that the NOx concentration from diesel vehicles is high, but the NOx concentration of bi-fuel taxis and natural gas vehicles is also high. Since the NOx concentration increases in proportion to the vehicle speed, NOx concentration measured by the RSD should be considered its speed. The vehicle inspection station conducts an exhaust gas test using a chassis dynamometer, and there is exhaust gas concentration data for each vehicle speed that has been certified. Using the exhaust gas concentration data by vehicle speed collected in large quantities at the inspection station, it is possible to make a pass / fail judgment for the measured vehicle using the vehicle speed and NOx concentration measured by RSD.

4459 diesel city buses were registered in 2018, and electrification is under consideration by 2020 to reduce NOx emissions. For the purpose of grasping the NOx emission reduction effect, NOx emission amount of city bus in real-world driving was measured. The NOx sensor was installed before and after the selective reduction catalyst (SCR) of the 2017 model year 8.5 liter diesel vehicle shown in Figure 3. The PM sensor was installed after the SCR. In order to monitor the activity of SCR, a temperature sensor was attached upstream. The main device (NTK Compact Emission Monitor; NCEM) was installed at the rear of the cabin and was driven by a battery. The driving state of the city bus was recorded by the output of the on-board diagnostic device (OBD).

Continuous NOx, PM, exhaust temperature, and OBD measurement were conducted about 8 hours a day for 8 days in October 2018, including morning rush. The bus to be measured carried out a commercial operation that goes back and forth in Route 80 in approximately one hour. Passengers get on and off at 15 stops. The maximum passenger number was about 60. The mileage during the measurement period was 387km, and the average vehicle speed was 16.98km h^{-1}. The Relative Positive Acceleration (RPA; Tutuianu, et. al., 2013) indicating the state of acceleration of the vehicle was 0.200 m s^{-2}, which was close to the Japanese vehicle exhaust test cycle of 10 mode (RPA=0.198 m s^{-2} at average speed of 17.57 km h^{-1}). However, since the 10 mode is a test cycle for a light duty vehicle, the bus operation showed large acceleration / deceleration in consideration of the bus weight.

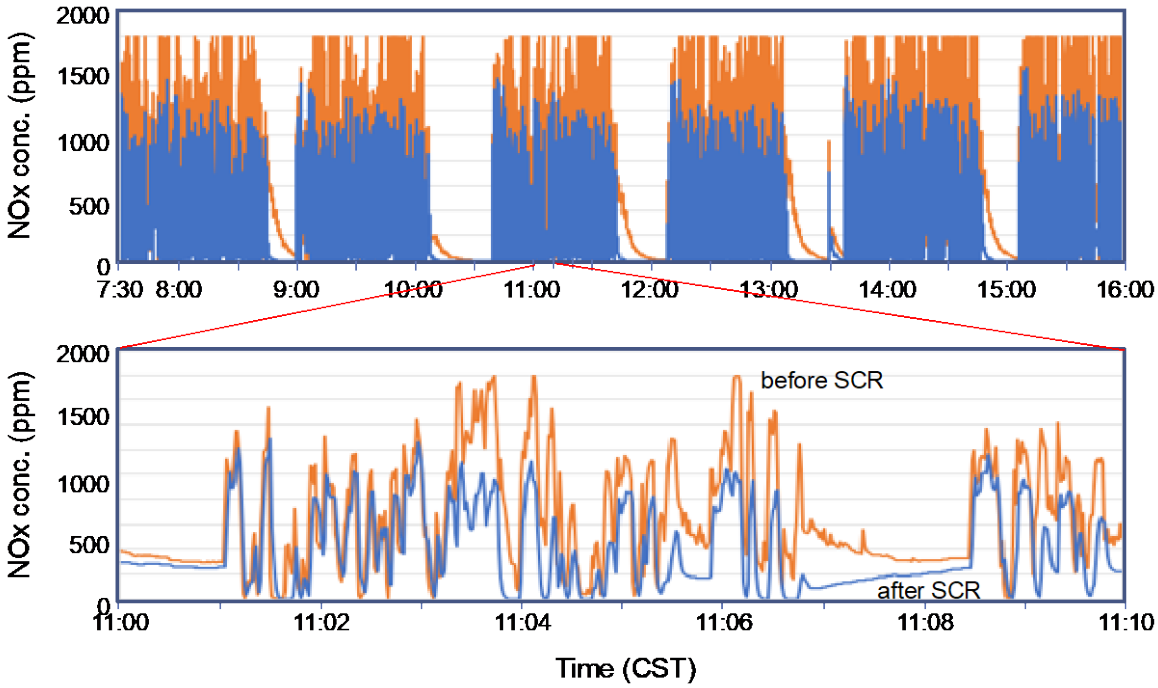

Figure 4. Diesel bus NOx exhaust (orange line; NOx concentration before SCR, blue line; NOx concentration after SCR)

The NOx concentration during real-world driving is shown in Figure 4. NCEM is a device designed for NOx concentration measurement for new regulated vehicles used in Japan, U.S. and Europe, with a maximum measurement range of 1800ppm. From Figure 4, it can be seen that the concentration of NOx before SCR exceeds 1800 ppm, and it decreased to about 1000 ppm after SCR. From the figure below, which shows the detailed time change, it became clear that the control with the SCR that injects urea is not always operating, but is operating when the inlet NOx concentration exceeds 1000 ppm. Although the exhaust NOx concentration was reduced by SCR, it was confirmed that fairly high NOx was discharged. The activity of SCR is affected by the exhaust temperature. The relationship between the exhaust temperature at the SCR inlet and the NOx purification rate (outlet concentration / inlet concentration) by SCR was investigated. However, there was no clear relationship, and the influence of urea injection control by inlet NOx concentration was larger.

A positive correlation between exhaust NOx concentration and vehicle speed was observed at speeds of 20 kmh^{-1} or higher. Even when the speed was 20 kmh^{-1} or less, the cases where the NOx concentration was high was observed sometime. Thus, the high NOx concentration observed at low vehicle speed which suggested due to no urea injection control has a great adverse effect on the atmospheric environment. The NOx emission amount for each travel section can be calculated from the emission concentration and the exhaust amount. The exhaust amount depends on the time that the vehicle stays in that section. The NOx concentration during high-speed driving is high, but because the residence time is short, the section emissions becomes small.

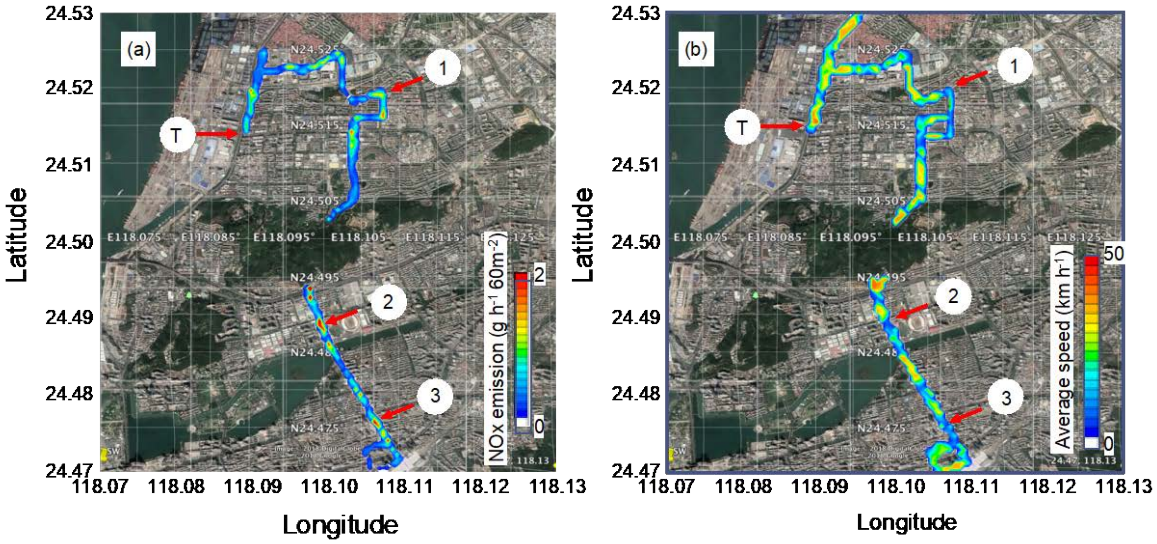

Figure 5. Spatial distribution of (a) NOx emissions and (b) average vehicle speed on bus routes (average value of all measured data)

Figure 5 shows (a) the distribution of section NOx emissions as a unit of $gh^{-1}60m^{-2}$ and (b) average vehicle speed, obtained using all measured data for 8 days. Route 80 has a part that passes through the tunnel shown in the center of the figure, and GPS data at this point could not be acquired, so the section display is missing. The city bus departs from the bus terminal marked T in the figure, travels south, returns at the end point, back to the terminal, and adjusts the time for about 30 minutes. The corner at the point marked 1 in the figure is always congested and the vehicle speed decreases, while NOx emissions increase. Similarly, the point marked 2 in the figure is the entrance to the bridge, and the point marked 3 is the downtown area, where traffic is constantly congested and the vehicle speed decreases while NOx emissions increase. At these points, OBD data showed that the frequency of acceleration / deceleration was high. At this congestion time, the load on the diesel engine of the city bus is large, and the NOx concentration becomes very high compared to steady driving.

Through this measurement, NOx and PM emission factors as a function of vehicle speed were obtained, and it was proved that they showed high values on the low speed side unlike the emission factor graph obtained using a conventional test cycle. The bus used for this measurement emits 77g of NOx and 111mg of PM per hour. By multiplying these values by the actual number of buses, the total amount of bus emissions can be obtained.

In order to reduce PM2.5 emission, the introduction of natural gas fueled buses has been implemented. Natural gas vehicles are said to have very little PM emissions and low NOx emissions. For liquefied natural gas (LNG) vehicles, the same measurements using NCEM as diesel buses measurement was conducted on November 2018. The NOx concentration during idling and unloaded racing was about 400 ppm and low enough. However, in actual driving with a load, the NOx concentration exceeded the upper limit of measurement range of NCEM (> 2000ppm). By using a chassis dynamometer and fixed NOx meter (Nanhua NHA-501A), it was found that running exhaust NOx concentration increased to about 4000 ppm, higher than that of diesel vehicles. This is

probably because the air fuel ratio (A/F) range where LNG is ideally burned is narrow and fuel control is not sufficient. This suggests that maintenance is even more important with the introduction of state-of-the-art vehicles.

3.4 Zhuhai

The purpose of the research is to support the establishment of a comprehensive volatile organic compound (VOC) countermeasure system for ozone suppression. In order to promote effective reduction measures for VOC, which is a precursor of ozone, development of the reliability of VOC monitoring is essential and contribution to the construction of a VOC countermeasure system through data analysis were conducted.

Through local inspections and discussions, detailed procedures for environmental air and source monitoring in accordance with Chinese national environmental protection standards (HJ759-2015, etc.) were prepared, and a standard operating procedure (SOP) was developed to improve the reliability of the data obtained. Furthermore, a VOC source profile and VOC emission inventory were created based on the results of VOC monitoring conducted by Zhuhai City. Based on the relationship between VOC, NOx concentration and ozone generation in Zhuhai City, which was calculated from air quality simulation using monitoring data, the knowledge about the effective reduction of the precursor for ozone was obtained.

4. Results of Intercity Cooperation

As a result of joint research with 4 cities for 4 years from 2015, the outcomes and new issues were obtained as below;

4.1 Chongqing

a) Impact on policy decisions

When instructing factories to control VOC emissions, it became clear that it was necessary to provide guidance not only for measures in the production process that uses VOCs, but also for measures to control non-organized emissions.

b) Expected effect

Chongqing will be able to reduce the amount of electricity required for VOC exhaust gas treatment in addition to the reduction of VOC emissions derived from industrial activities by strengthening control guidance including non-organized emissions.

c) Remaining issues

(1) The non-organized source could not be identified because the non-organized release was not fully assumed at the time of the survey. In addition, since the number of days and the number of times of measurement are small, there remains a problem in the accuracy of the survey. In the future, it is necessary to further improve the accuracy of the survey in order to grasp the effects of emission control measures and to identify the sources of non-organized emissions.

(2) In order for Chongqing City to provide guidance on restraint of non-organized emissions, it is necessary to utilize the prepared evaluation sheet and maintain technical manuals.

4.2 Xi’an

a) Impact on policy decisions

(1) Provided scientific basis for reviewing redevelopment and city planning that takes into account the reduction of pollution caused by ozone.
(2) In order to suppress the generation of high-concentration ozone in Xi'an City, etc., a scientific basis was provided for the need to establish a mechanism for implementing measures in cooperation with neighboring provinces.

b) Expected effect

A wide-area cooperation mechanism including the surrounding area of Xi'an City has been established, and the reduction of the ozone concentration in Xi'an City and the surrounding area can be realized by implementing the measures.

c) Remaining issues

It is required to understand the mechanism construction and implement the measures with each city in Shaanxi provinces by sharing the results of analysis.

4.3 Xiamen

a) Impact on policy decisions

(1) In the past, buses and taxis have been encouraged to introduce vehicles that use natural gas as the main fuel. Based on these results, the policy has been reviewed and was decided to convert to electric vehicles from encouraging natural gas vehicles.
(2) Similar high-concentration NOx emissions during traffic jams was suggested for large diesel trucks, and new traffic flow measures can be proposed with reference to the time and place measured by the on-board vehicle device.

b) Expected effect

When electric vehicles become more widespread through a new incentive policy, it will greatly contribute to reducing carbon dioxide emissions as well as air pollutants. From the on-board measurement results, the traffic environment with a large amount of fuel injection plays a big role, and more specific measures can be proposed.

c) Remaining issues

It is necessary to consider the introduction of efficient measures in terms of management and regulation by analyzing both the actual state of exhaust gas for each type of vehicles using the remote sensing devices, and the actual state of exhaust gas according to the traffic environment by on-board equipment.

4.4 Zhuhai

a) Impact on policy decisions

By clarifying the relationship between VOC and NOx concentrations and ozone generation sensitivity, it became possible to set VOC and NOx reduction targets for effective reduction of ozone based on scientific evidence.

b) Expected effect

By spreading the SOPs created this time, VOC monitoring and source inventory creation in China will be carried out in a unified manner, and a foundation for comprehensive countermeasure formulation will be established.

c) Remaining issues

In order to spread the created SOPs, it is important that the China Environmental Monitoring Bureau review the contents and authorize the SOPs. In addition, it is necessary to create SOPs at various locations based on the same concept and to obtain data of the same quality at the national level through the implementation of comparative tests between laboratories.

5. Conclusion

According to a preliminary report from the Ministry of Ecology and Environment (MEE), in 2018, the average concentration of PM2.5 in 338 cities nationwide improved (decreased) by 9.3% and the average concentration of PM10 improved by 5.3% compared to the previous year. The environment in China has been improving year by year, with the exception of ozone, due to the end-of-pipe emission control technology, including energy conversion from coal-fired power to natural gas. Up to now, measures for large-scale emission sources have contributed to environmental improvement. However, it is now necessary to take measures against various emission sources with small scales, including non-organized emission source. In addition, household emissions such as cooking emissions, automobiles, ships and off-road countermeasures are also important. Chongqing City started taking measures against VOCs from restaurants. Since MEE has positioned the Fenwei Plain as a base for environmental improvement, Xi'an City has started to estimate the source contribution using air quality model in cooperation with cities around Shaanxi Province. Xiamen City is also considering measures for NOx for diesel trucks and VOC for ships.

In this research cooperation project, the importance of measurement including frequency and duration considering surrounding situations and measurement target has been discussed, and at the same time, cooperation focused on analysis and utilization of measurement big data has been implemented. Support will be continued after the project is completed so that a large amount of measurement data can be used effectively in the future.

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Report of the joint research project on catchment analysis in Thailand

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Abstract

The Network Center for the EANET conducted the joint research project on catchment analysis in cooperation with Royal Forest Department and Environmental Research and Training Centre, Thailand. The study plot was established in a forested catchment in Sakaerat Silvicultural Research Station, Nakhon Ratchasima Province, northeastern Thailand. The surveys in Sakaerat site were conducted for the period from 2005 to 2015. Based on data from the surveys and other relevant studies, several scientific papers have been published in international journals. The scientific outputs will contribute to understanding atmospheric deposition and its effects in the region.

Keywords: Catchment analysis, Atmospheric deposition, Stream water chemistry, Forest area

1. Introduction

Promotion of catchment analysis is one of the subjects described in 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*", which was adopted at SAC14 in 2014. Asia Center for Air Pollution Research (ACAP) as the Network Center for the EANET conducted the joint research project on catchment analysis in cooperation with Royal Forest Department and Environmental Research and Training Centre, Thailand. In this report, we summarize major outputs from the project.

2. Project outline

2.1 Study site

The study catchment plot (35 ha) was established in a dry evergreen forest (DEF) in Sakaerat Silvicultural Research Station, Nakhon Ratchasima Province, northeastern Thailand. The area is within a tropical savanna climate under the Köppen climate

classification, which has distinct dry and wet seasons. In the Sakaerat site, the period between December and February is extremely dry with monthly precipitation less than 50 mm, whereas the period from April to October has a perhumid condition with monthly precipitation exceeding 100 mm (Sase et al. 2012).

2.2 Field surveys

The field surveys in Sakaerat site were conducted for the period from October 2005 to April 2015. Atmospheric deposition via rainfall outside forest canopy (RF) and throughfall and stemflow (TF+SF) and stream water chemistry were surveyed regularly through the observational period (Sase et al. 2017). Additionally, soil/soil solution physico-chemistry (Murata et al. 2009, 2012; Tominaga et al. 2009; Yamashita et al. 2011), nutrient fluxes in soil-plant system (Yamashita et al. 2010), elemental carbon fluxes by atmospheric deposition (Matsuda et al. 2012; Sase et al. 2012), and dynamics of sulfur in ecosystems using isotopic analysis (Sase et al. 2015) were intensively surveyed. Some of them were conducted in cooperation with other research projects in Sakaerat area.

3. Major outputs

Based on the data from the surveys, several scientific papers have been published in international journals. Most of them are ISI journals. The major topics presented by the papers can be summarized as follows:

- Atmospheric deposition: its clear seasonality under tropical savanna climate and importance of dry deposition for elemental carbon (Matsuda et al. 2012; Sase et al. 2012, 2017).
- Soil: Spatial variation of soil chemical properties and its controlling factors (Yamashita et al. 2011), clear seasonality of vertical nitrogen fluxes under tropical savanna climate (Yamashita et al. 2010) and comparison of soil physiological/chemical properties between deciduous dry forest and DEF (Murata et al. 2009,2012; Tominaga et al. 2009).
- Stream water chemistry: its sensitive reactions to seasonality of atmospheric deposition and retention-release cycle of sulfur in the forest ecosystem under tropical savanna climate (Sase et al. 2017).

Through the project above, one Japanese graduate student got Ph.D. He became an active scientist on soil chemistry and has been contributing the EANET activities as a resource person.

4. Conclusion

Based on the collaboration among scientists from the EANET community, aforementioned scientific outputs were produced. These papers will contribute to understanding atmospheric deposition and its effects in the region. In particular, some of the papers noted importance of seasonality under tropical savanna climate and possibility of changing climate. We should carefully assess effects of climate change even in context of atmospheric deposition.

We are going to assess remaining data. It can be expected that some more ISI journal papers will be produced soon.

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Report of the joint research project on catchment analysis in Japan

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Abstract

The Network Center for the EANET conducted the joint research project on catchment analysis in cooperation with Niigata Prefecture and Niigata University. The study plot was established in a forested catchment in Shibata City (former Kajikawa Village), Niigata Prefecture. The surveys in Kajikawa site have been conducted since 2002. Based on data from the surveys and other relevant studies, several scientific papers have been published in international journals. The scientific outputs will contribute to understanding atmospheric deposition and its effects in the region.

Keywords: Catchment analysis, Atmospheric deposition, Stream water chemistry, Forest area

1. Introduction

Promotion of catchment analysis is one of the subjects described in 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*", which was adopted at SAC14 in 2014. Asia Center for Air Pollution Research (ACAP) as the Network Center for the EANET conducted the joint research project on catchment analysis in cooperation with Niigata Prefecture and Niigata University. In this report, we summarize major outputs from the project.

2. Project outline

2.1 Study site

The study catchment plot (3.84 ha) was established in a Japanese cedar forest in

Shibata City (former Kajikawa Village), in the norther part (Kaetsu area) of Niigata Prefecture, Japan. The area is within a humid subtropical climate (Cfa) under the Köppen climate classification, which is in a perhumid condition through a year (Sase et al., 2012). The annual precipitation and annual mean temperature at Nakajo AMeDAS, the nearest meteorological station, were 2242 mm and 13.4 C, respectively.

2.2 Field surveys

The field surveys in Kajikawa site have been conducted since 2002. Atmospheric deposition via rainfall outside forest canopy (RF) and throughfall and stemflow (TF+SF) and stream water chemistry were surveyed regularly (Kamisako et al., 2008; Sase et al., 2008). Additionally, canopy interactions of deposited ions (Sase et al., 2008), elemental carbon fluxes by atmospheric deposition (Sase et al., 2012), changes in nitrogen leaching during heavy rain events (Kamisako et al., 2008), dynamics of sulfur and nitrogen in ecosystems using isotopic analysis (Sase et al., 2015; Nakagawa et al., 2018; Inomata et al. 2019), and river water chemistry in Niigata Prefecture (Matsubara et al., 2009; Kobayashi et al., 2013) were intensively surveyed. Some of them were conducted in cooperation with other institutions, including Nagoya University and Kanazawa University.

3. Major outputs

Based on the data from the surveys, several scientific papers have been published in international journals. Most of them are ISI journals. The major topics presented by the papers can be summarized as follows:

- Atmospheric deposition: Clear seasonality in SO_4^{2-} and other ions because of winter westerly winds (Kamisako et al., 2008; Sase et al., 2008; 2012), evaluation of transboundary air pollution effects using sulfur isotopic analysis (Inomata et al., 2019)
- Canopy interactions: Importance of leaf surface wettability for uptake/leaching of ions (Sase et al., 2008), accumulation of particulate matters, such as elemental carbon, on leaf surface (Sase et al., 2012)
- Stream water chemistry: Increase of NO_3^- concentrations during heavy rain events and possibility of nitrogen saturation (Kamisako et al., 2008), estimate of exporting ratio of unprocessed atmospheric NO_3^- to the stream based on the ^{17}O excess data (Nakagawa et al., 2018)
- River water chemistry in Niigata Prefecture: Long-term pH declining trends in Kaetsu area for the period from 1980s to the early 2000s (Matsubara et al., 2009), acid shock during snow-melting events and sulfur budget in an acid-sensitive river (Kobayashi et al., 2013)

Through the project above, more than 10 students from Niigata University got academic degrees, such as bachelor/master of agriculture. Moreover, in the near future, a few young scientists will obtain their Ph.D. degrees. The project largely contributed to development of environmental sciences.

4. Conclusion

Based on the collaboration with local scientists from Niigata Prefecture, in which the Network Center is located, aforementioned scientific outputs were produced. These papers will contribute to understanding atmospheric deposition and its effects in the region. In particular, some of the papers pointed out that the site has been strongly affected by long-range transboundary air pollution. However, as recent emission inventories and satellite observations suggested, both SO_2 and NO_X emissions in North East Asia have started decreasing (e.g., Lu et al., 2011; Zheng et al., 2018). Therefore, we should carefully monitor whether reactions of the ecosystem to a changing atmospheric environment, such as “recovery from acidification”, can be found. Currently, a few papers are under submission or preparation. It can be expected that some more ISI journal papers will be produced soon.

Acknowledgements

This study was conducted as one of the Network Center research activities for the EANET. Part of the data in this study was obtained from the Long-term Monitoring on Transboundary Air Pollution and Acid Deposition in the Ministry of the Environment of Japan. We thank previous colleagues worked for the Kajikawa catchment project, namely, Kamisako M, Kobayashi R, Matsui T, Oida T, Saito M, Takahashi A, Take N, Uchiyama S, and Yagoh, H, for their contribution to the field surveys and chemical analyses. We also thank the local government of Niigata Prefecture and the previous and current land managers, namely Mr. Kohei Funayama, Mr. Takeo Funayama, and Mr. Takanori Funayama, for their permission and assistance to use the selected forest area in Kajikawa as a study site.

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The Model Inter-Comparison Study for Asia (MICS-Asia): Phase III and next steps

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Abstract

Activities of the model intercomparison study for Asia (MICS-Asia) Phase III (2010-2019) are introduced in this report. MICS-Asia Phase III has following 4 topics: 1. Intercomparison of air quality model, 2. Development of reliable emission inventories in Asia, 3: Model intercomparison studies for air quality and climate change, 4: Studies for Southeast Asia. The MIX inventory (a mosaic Asian anthropogenic emission inventory) was developed by Topic 2 as common input emission data. Results of Topic 1 were summarized by analysis teams such as for ozone, $PM_{2.5}$, and deposition. Topic 3 examined how online coupled air quality models perform in simulating high aerosol

pollution in the North China Plain region during wintertime haze events and evaluate the importance of aerosol radiative and microphysical feedbacks. Southeast Asian Group was initiated as new topic during Phase III period. The Special issue of MICS-Asia Phase III: (Regional assessment of air pollution and climate change over East and Southeast Asia: results from MICS-Asia Phase III) is opened in Atmospheric Chemistry and Physics: https://www.atmos-chem-phys.net/special_issue987.html

Keywords: Air quality model, Climate model, Emission inventory, Inter-comparison study

1. Introduction

Air pollution along with demand of energy, motorization, industrial and agricultural products is a serious problem in Asia especially for its impact on public human health. In addition, huge growth of anthropogenic emissions in Asia are considered to affect not only local air pollutions, but also regional, inter-continental, and global air quality and climate change. Air quality and climate models are effective scientific tools for understanding status of atmospheric environment and evaluating effects of mitigation measures. However, current modeling systems have problems not only in reproducibility of monitoring results, but also differences among simulated results by different models. Therefore, common understanding of current model performances and uncertainties in Asia and efforts to improve the modeling system are essential for utilizing models especially for international problems on air pollution and climate change.

Under such backgrounds, MICS-Asia Phase I (1998-2000) for sulfur compound and Phase II (2004-2009) including nitrogen compounds, ozone and aerosols were carried out as international initiatives of model inter-comparison study. The findings of the Phase II activities were published in Atmospheric Environment in 2008 (Carmichael and Ueda, 2008).

The initiative was succeeded to MICS-Asia Phase III from the First International Workshop on Atmospheric Modeling in East Asia organized by Asia Center for Air Pollution Research and Chinese Academy of Sciences/Institute of Atmospheric Physics in Dalian in March 2010, which aimed to give an opportunity for more Asian scientists to participate in future international collaboration in atmospheric modeling and related atmospheric chemistry studies and at the Second International Workshop on Atmospheric Modeling in East Asia, three topics of MICS-Asia Phase III were accepted as follows.

Topic 1: Intercomparison of air quality model (Chief leader: Zifa Wang, Institute of Atmospheric Physics/Chinese Academy of Sciences)

Topic 2: Development of reliable emission inventories in Asia (Chief leader: Jung-Hun Woo, Konkuk University)

Topic 3: Model inter-comparison studies for air quality and climate change (Chief leader: Gregory Carmichael, The University of Iowa)

At the Third International Workshop on Atmospheric Modeling in East Asia, all the participants had group discussion to materialize more concrete activity plans for each

of the three topics. Based on the discussion, the draft work plan document was prepared and further discussed at the additional Workshop held at Beijing in July 2012 to finalize the Work Plans for the MICS-Asia Phase III. At the 6th International Workshop on Atmospheric Modeling in East Asia in 2014, Southeast Asian Group was established for new Topic of MICS-Asia Phase III where participants were working and reporting for their own project that could support the improvement of MICS-Asia.

In this report, basic framework and major results of MICS-Asia Phase III are introduced and next steps of MICS-Asia are discussed.

2. Framework of MICS-Asia Phase III

2.1 Topic 1: Intercomparison of air quality model

Figure 1 illustrates the basic framework of Topic 1. Major objectives of Topic 1 are to evaluate strengths and weaknesses of current air quality models for air quality prediction, and provide insights to reduce uncertainty and improve performance in Asia by inter-comparing simulated results of different air quality models. For these purpose, important points are common modeling domains and input data are used by all participant models as much as possible. In addition, the list of data sets which must be submitted from simulated results by all participants need to be prepared. Therefore, a lot of discussions and works were done before inviting researchers who will participate in actual works for model inter-comparison studies.

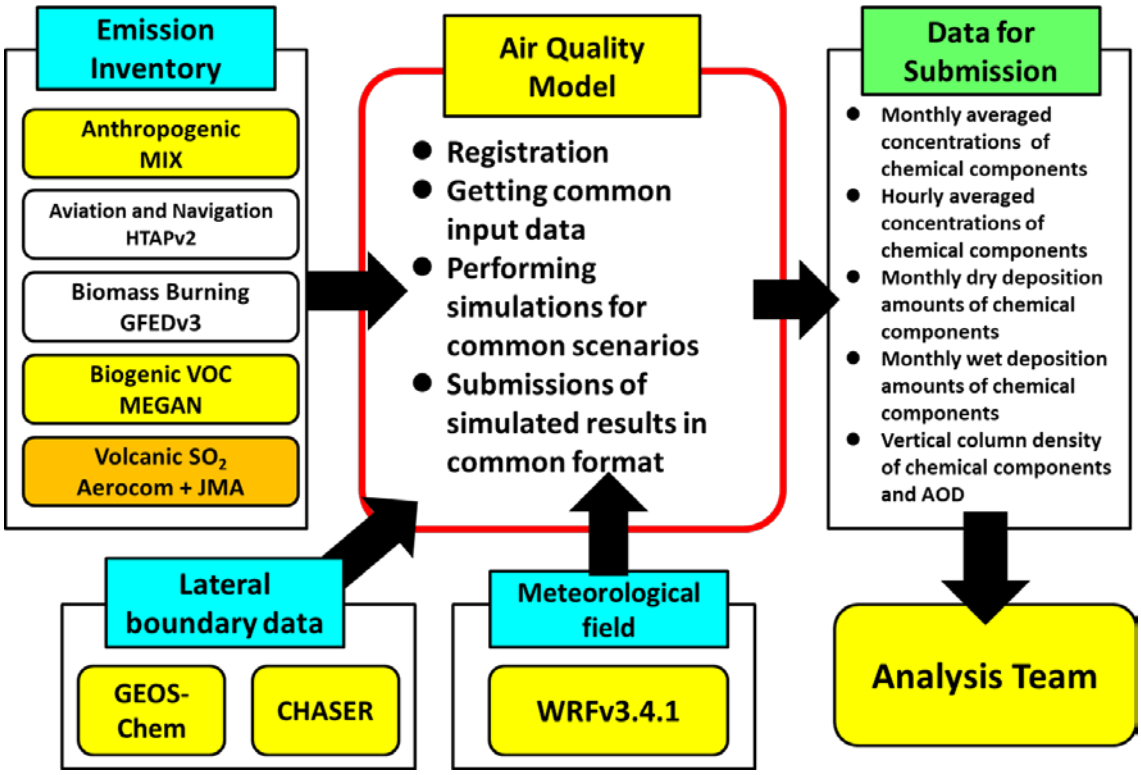

Figure 1. Basic framework of Topic 1

Figure 2 shows the setting of three model domains of Topic 1. The first (largest) domain (d01) covers whole Asian region including East, Southeast, South, and Central Asia. Horizontal resolution of d01 is 45 km by 45 km. In MICS-Asia Phase III, two

more domains with finer horizontal resolution were defined for nesting simulations. The second domain (d02: 15 km by 15 km) covers northeast Asia including northeast and southeast China, Korea, and Japan. The third domain (d03: 5 km by 5 km) is around Jing-jin-ji region (Beijing, Tianjin, and Hebei province).

For the input data, the standard meteorological fields, lateral boundary conditions, and emission inventories were provided for all participants. The standard meteorological data sets were created by the Weather Research and Forecasting model (WRF) version 3.4.1 with a four-dimensional data assimilation nudging toward the final analysis dataset from the National Center for Environmental Prediction (NCEP). Majority of participants used the standard data, but several models used their own meteorological models and WRF-Chem considers the feedback of pollutants to the meteorological fields. For lateral boundary conditions, two types of data sets were provided from global models CHASER (the Chemical AGCM for Study of Atmospheric Environment and Radiative Forcing) and GEOS-Chem for considering conveniences for settings of each participant model. It was briefly confirmed that two global models provide generally consistent results. For emission inventories, data set for anthropogenic sources, aviation and navigation, biomass burning, biogenic VOC, and volcanic SO_2 were prepared by Topic 2 (see section 2.2).

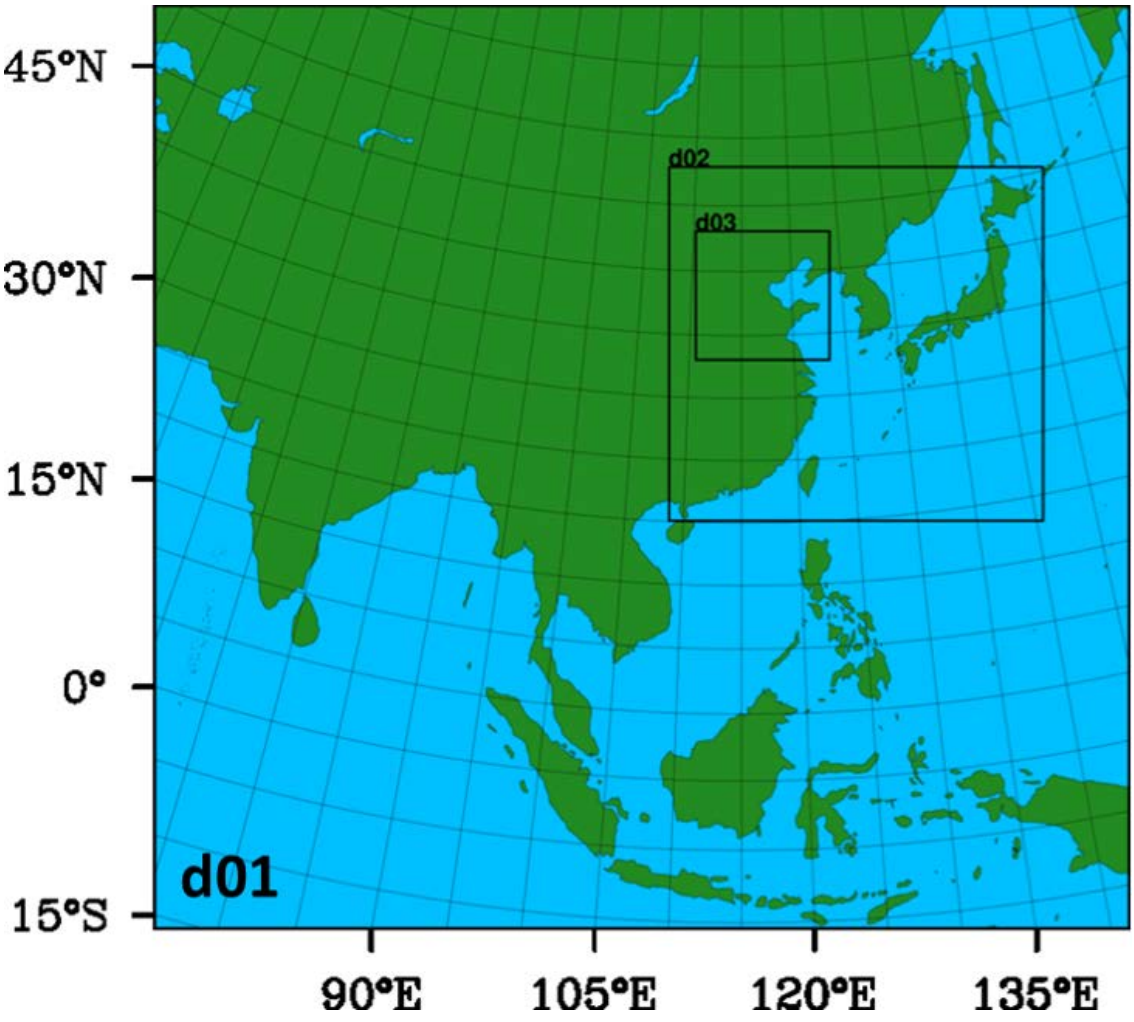

Figure 2. Model domains of Topic 1

In Topic 1, all participants performed the standard simulation for the year 2010 in d01 and submitted their simulated results according to the standard submission list where mandatory components for inter-comparison studies were indicated. Table 1 summarizes basic configurations of participating models (see Table 1 of Li et al., 2019 for detail.). Fourteen models participated in the standard inter-comparison studies of air quality models for MICS-Asia Phase III. In Topic 1, analysis teams preparing for scientific papers were established for major themes such as ozone, deposition, and $PM_{2.5}$. Leaders of each analysis team requested participants to submit additional components necessary for further analysis of each theme. Unfortunately, not all participants could submit all required results. Therefore, numbers of participating models are different among analysis themes.

Table 1. Configurations of participating models (see Table 1 of Li et al. (2019) for more details)

No.	Model	Gas-Chemistry	Advection	Vertical Diffusion	Dry Deposition	Wet Deposition
M1	WRF-CMAQ	SAPRC99	Yamo	ACM2	M3DRY	Foley
M2	WRF-CMAQ	SAPRC99	Yamo	ACM2	M3DRY	Foley
M3	WRF-CMAQ	CBM05	Yamo	ACM2	M3DRY	Foley
M4	WRF-CMAQ	SAPRC99	PPM	ACM2	M3DRY	Foley
M5	WRF-CMAQ	SAPRC99	PPM	ACM2	M3DRY	Foley
M6	WRF-CMAQ	SAPRC99	Yamo	ACM2	M3DRY	Foley
M7	WRF-Chem	RACM	5^{th}-order monotonic	3^{rd}-order monotonic	Wesely	Easter
M8	WRF-Chem	RACM	5^{th}-order monotonic	3^{rd}-order monotonic	Wesely	Easter
M9	WRF-Chem	RADM2	5^{th}-order monotonic	YSU	Wesely	Easter
M10	WRF-Chem	RADM2	5^{th}-order monotonic	YSU	Wesely	Easter
M11	WRF-NAQPMS	CBMZ	Walcek and Aleksic	Byun and Dennis	Wesely	Ge
M12	NHM-Chem	SAPRC99	Walcek and Aleksic	MYJ	Kajino	Kajino
M13	GEOS-Chem	Bey et al.	PPM	Holtslag and Bovill	Wesely, Wang	Liu
M14	RAMS-CMAQ	SAPRC99	PPM	ACM2	M3DRY	Foley

2.2 Topic 2: Development of reliable emission inventories in Asia

Major roles of Topic 2 are to provide reliable input emission data sets to air quality and climate models for the standard simulations of Topics 1 and 3. The most important outcome of Topic 2 is the MIX inventory for anthropogenic sources developed by harmonizing different local emission inventories with the mosaic approach. Figure 3 illustrates the components and methodology of the MIX emission inventory. Five emission inventories are incorporated into the mosaic inventory as follows: Regional Emission inventory in ASia (REAS) version 2.1 for the whole of Asia, the Multi-resolution Emission Inventory for China (MEIC) developed by Tsinghua University, a high resolution NH_3 emission inventory by Peking University, an Indian emission inventory developed by Argonne National Laboratory, and the official Korean emission inventory for the Clean Air Policy Support System (CAPSS). The MIX inventory has been incorporated into the official global emission inventory of the Task Force on Hemispheric Transport of Air Pollution (TF HTAP), HTAP v2.2 (Janssens-Maenhout et al., 2015). See Li et al. (2017) for details of development and results of the MIX inventory.

Topic 2 also prepared emission data for all other sources. Biogenic emissions data were developed by Konkuk University using MEGAN (Model of Emissions of Gases and Aerosols from Nature) version 2.0.4 with WRF meteorological field which were commonly used in air quality model simulations of Topic 1. For biomass burning such as forest and savanna fires and agricultural waste burning, emissions data were obtained from the Global Fire Emissions Database version 3 (GFEDv3). For air and ship sources, emissions data were taken from HTAP v2.2 global emission inventory. Volcanic SO_2 emissions were based on the AEROCOM hindcast emission inventory and reports of Japan Meteorological Agency.

All developed or collected emission data by Topic 2 were uploaded to the file exchange server of MICS-Asia Phase III and were downloaded by all participants of Topics 1 and 3 for their standard simulations.

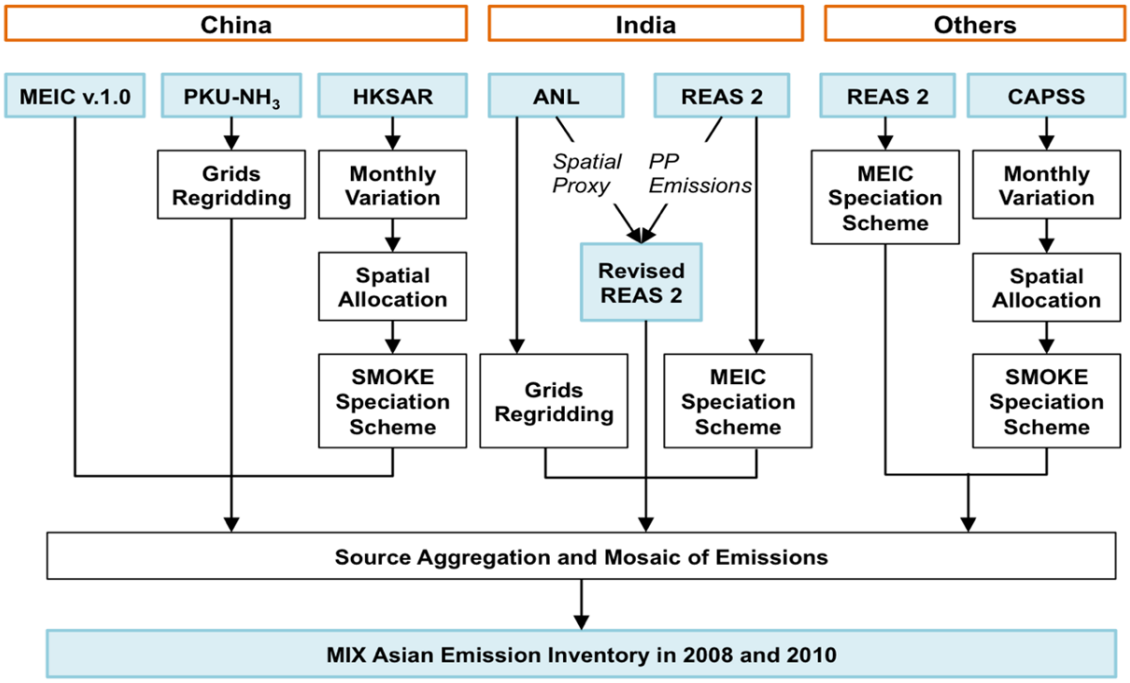

Figure 3. Schematic methodology of the MIX emission inventory development

2.3 Topic 3: Model inter-comparison studies for air quality and climate change

Relationships between air pollution and climate change are becoming important scientific topics for atmospheric environment. From MICS-Asia Phase III, new studies that the inter-comparisons of multiple online coupled chemistry-meteorology models to investigate aerosol-weather-climate interactions were included as the major target of Topic 3. In MICS-Asia Phase III, Topic 3 examined how online coupled air quality models perform in simulating high aerosol pollution in the North China Plain region during wintertime haze events and evaluates the importance of aerosol radiative and microphysical feedbacks. Compared to Topic 1, number of participating models were small. Figure 4 show modeling domains of seven participating models of Topic 3. In Topic 1, as shown in Figure 2, common modeling domain was defined for all participants. However, current studies in Topic 3, definitions of modeling domains were different among models as shown in Figure 4. In addition, although common input emission data sets (the same as those for Topic 1) were used for all models, different models used different boundary conditions of meteorological fields and chemical compounds. Therefore, in Topic 3, inter-comparisons were done for different modeling systems. See Table 1 of Gao et al. (2018) for detailed configurations of seven participant models.

All participants were requested to simulate meteorology, air quality, radiative forcing and effects of aerosols over the Beijing-Tianjin-Hebei region of east China during two periods: January 2010 and January 2013. Each participant was requested to submit hourly meteorological fields, concentrations, aerosol optical depths, aerosol direct radiative forcing at the surface, topic of the atmosphere, and inside the atmosphere, hourly mean integrated liquid water and cloud optical depth. Also, changes in air temperature and water vapor mixing ratio at 2m above the ground, wind speed at 10m above the ground and $PM_{2.5}$ concentrations at the surface due to both direct and

indirect aerosol effects.

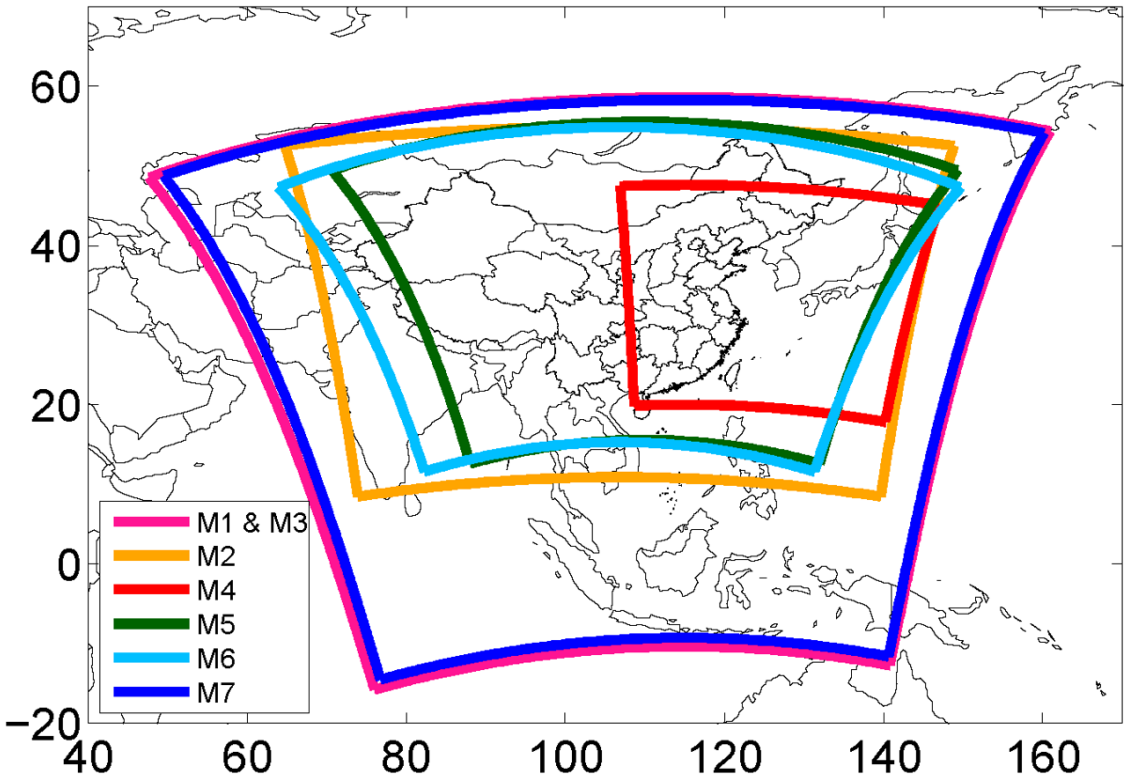

Figure 4. Domains of participating models of Topic 3 (Gao et al., 2018)

2.4 Topic 4: Study for Southeast Asia

The new movement in the MICS-Asia Phase III is establishment of Southeast Asia Group launched at the 6th International Workshop on Atmospheric Modeling in East Asia, in 2014. Actual model inter-comparison studies in modeling domains defined in Southeast Asian region have not been performed in Phase III, but are considered to be one of major activities in MICS-Asia Phase IV. In MICS-Asia Phase III, members of Southeast Asian Group were working on their own project and provide results and discussions at the International Workshop on Atmospheric Modeling in East Asia which could stimulate research activities of the MICS-Asia communities in Southeast Asian regions. For example, emission inventories of air and climate pollutants for anthropogenic sources in Thailand and Vietnam especially for road transport, and those of biogenic and biomass burning sources in Southeast Asia were studied and reported. For modeling studies, analysis of air quality in Malaysia, Thailand, Bangkok Metropolitan Region, Vietnam, Hanoi, Taiwan, and Hong Kong using air quality models were also reported. During the International Workshop on Atmospheric Modeling in East Asia, work plans for next steps of Southeast Asian Group were discussed and modeling domains were tentatively defined as shown in Figure 5. In MICS-Asia Phase IV, actual model inter-comparison studies will be performed not only by members of Southeast Asian Group, but also all participants interested in studies of these regions.

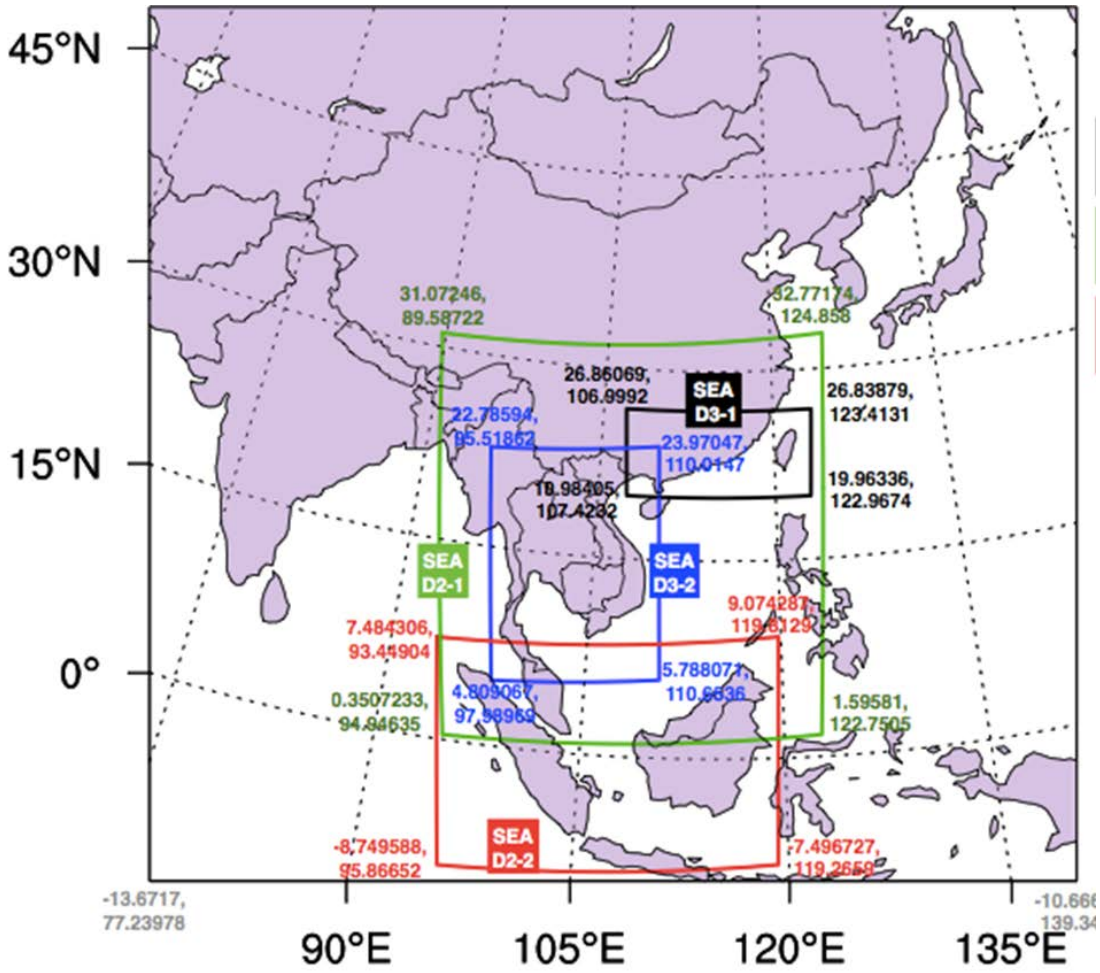

Figure 5. Tentative definition of modeling domains for Southeast Asian Group

3. Special Issues of MICS-Asia Phase III

MICS-Asia Phase III launched the special issue "Regional assessment of air pollution and climate change over East and Southeast Asia: results from MICS-Asia Phase III" on 1st August 2018. The research foci in the MICS-Asia III study include the following aspects: development of reliable emission inventories in Asia; ability and uncertainty of current air quality models in Asia in predicting ozone, nitrogen oxides, VOC, sulfur oxide, $PM_{2.5}$, etc.; ability of online coupled models to simulate aerosol radiative forcing and ozone and aerosol–weather/climate interaction through aerosol–radiation–cloud feedback; and prediction of future air pollution and climate. In this section, overview papers for ozone, $PM_{2.5}$, deposition, and air quality and climate change submitted to the special issue were introduced briefly based on abstract and summary of each paper. For details and other submitted papers, visit the special issue site: https://www.atmos-chem-phys.net/special_issue987.html

3.1 Ozone

Li et al. (2019) intercompared spatiotemporal variations of O_3 and NO_x mixing ratios from 14 state-of-the-art chemical transport models and evaluated with O_3 observations in East Asia. Most models captured the key patterns of monthly and diurnal

variation of surface O_3 and its precursors in the North China Plain and western Pacific Rim, but failed in the Pearl River Delta. A significant overestimation of surface O_3 was found from May to September/October and from January to May over the North China Plain, the western Pacific Rim and the Pearl River Delta. Comparison with observation indicated that the considerable diversity in O_3 photochemical production partly contributed to this overestimation and to high levels of inter-model variability in O_3 for North China. The ensemble average of participants models did not always exhibit superior performance compared with certain individual models which suggests that the spread of ensemble-model values does not represent all of the uncertainties of O_3 or that most MICS-Asia III models missed key processes. Compared with MICS-Asia II, MICS-Asia III was less prone to underestimating surface O_3 in March for Japanese site. However, it predicted excessively high surface O_3 concentrations for western Japan in July, which was not the case for MICS-Asia II. The ensemble average of models reproduced the vertical structure of O_3 for the western Pacific, but overestimated O_3 levels below 800hPa in the summer. For the industrialized Pearl River Delta, the ensemble average presented an overestimation of O_3 levels for the lower troposphere and underestimations in the middle troposphere. Large inter-model variabilities of O_3 were considered to be caused by the internal parameterizations of chemistry, dry deposition and vertical mixing of models, even though the native schemes in models are similar. Major challenges still remain with regard to identifying the sources of bias in surface O_3 over East Asia in chemical transport models.

Akimoto et al. (2019) compared surface O_3 simulation among selected regional models, CMAQ v5.0.2, CMAQ v4.7.1, and NAQPMS to clarify the causes of variability among the model outputs. Three causes of the differences have been identified and discussed. The first is that the chemistry mechanism sub-module SAPRC99 used in the CMAQ was found to give higher net ozone production values than CBM-Z in NAQM, agreeing with previous studies. The second is that higher NO concentrations have been predicted by NAQM than by CMAQ, possibly due to the inclusion of a heterogeneous renoxification reaction of HNO_3 (on soot surface) $\rightarrow$ $NO+NO_2$, which gave a better agreement with observational concentration, particularly for nighttime NO and O_3. The last one is that a vertical downward O_3 flux was found to substantially affect the diurnal pattern and mixing ratios of O_3.

3.2 $PM_{2.5}$

Chen et al. (2019) analyzed the following objectives: (1) provide a comprehensive evaluation of current air quality models against observations, (2) analyze the diversity of simulated aerosols among participant models, and (3) reveal the characteristics of aerosols components in large cities over East Asia. In general, participated models could well reproduce the spatial-temporal distributions of aerosols in East Asia during the year 2010. The multi-model ensemble mean showed better performance than most single-model predictions. In comparison with results of MICS-Asia Phase II, frequent updates of chemical mechanisms in chemical transport models during recent years make the inter-model variability of simulated aerosols concentrations smaller, and better performance can be found in producing the temporal variations of observations. However, a large variation in the ratios of sulfate, nitrate, and ammonium to $PM_{2.5}$ is

calculated among participant models. A more intense secondary formation of SO_4^{2-} is simulated by the Community Multi-scale Air Quality (CMAQ) models, because of the higher sulfur oxidation ratio than other models. The nitric oxide ratio calculated by all models has large values than the observations, indicating that overmuch NO_3^- is simulated by current models. NH_3-limited condition (the mole ratio of ammonium to sulfate and nitrate is smaller than 1) can be successfully reproduced by all models, which indicates that a small reduction in ammonia may improve the air quality. A large coefficient variation is shown in simulated coarse particles (subtract $PM_{2.5}$ from PM_{10}). The poor consistency, especially over the arid and semi-arid regions, is mainly caused by dust aerosols, which means current chemical transport models have difficulty reproducing similar dust emissions by using different dust schemes. Analyzing the ratios of each composition to $PM_{2.5}$, NO_3^- is the major component in Beijing and Delhi, SO_4^{2-} is the major one in Guangzhou, and similar contributions of SO_4^{2-} and NO_3^- are calculated in Shanghai, Seoul, and Tokyo, which suggest that different air-pollution control plans should be taken in different cities.

3.3 Deposition

Itahashi et al. (2019) analyzed the simulated results of participant models focusing on deposition. Dry and wet depositions of S (sulfate aerosol, sulfur dioxide, and sulfuric acid, N (nitrate aerosols, nitrogen monoxide, nitrogen dioxide, and nitric acid), and A (ammonium aerosols and ammonia) were analyzed. Simulated results of wet deposition were analyzed and evaluated using ground observation data from the Acid Deposition Monitoring Network in East Asia (EANET). In MICS-Asia Phase III, the number of observation sites was increased to 54 from 37 in MICS-Asia Phase II, and data of Southeast Asian countries were newly added. In addition, whereas the analysis period was limited to representative months of each season in the Phase II, the Phase III analyzed the full year of 2010. In general, models can capture the observed wet deposition over Asia, but underestimate the wet deposition of S and A and show large differences in the wet deposition of N. For the ratio of wet deposition to the total deposition (the sum of dry and wet deposition), the general dominance of wet deposition over Asia and attributions from dry deposition over land were consistently found in all models. For the balance between deposition and anthropogenic emissions, excesses of deposition, rather than of anthropogenic emissions, were found over Japan, North Asia, and Southeast Asia. These results indicate that the possibility of long-range transport within and outside Asia, as well as other emission sources. The simple ensemble and weighted ensemble approaches (weight is correlation coefficient between model and observation) were confirmed to be better way to improve the model performance, allowing the elimination of extreme performance. On the other hand, the precipitation-adjustment approach revealed a potential way to improve the simulation of wet deposition. Therefore, model performances for precipitation and related parameters such as water vapor mixing ratio should be refined as the key input data to chemical transport models.

3.4 Air quality and climate change

Gao et al. (2018) provided the detailed information linked to Topic 3 simulation and

comparisons. Basic study design including description of participating models and model inputs, the experimental designs, and results of model evaluation are presented. Simulated results of participants models are compared to meteorology and air quality managements, including data from the Campaign on Atmospheric Aerosol Research Network of China (CARE-China) and EANET. All models could capture the observed near-surface temperature and water vapor mixing ratio, but near-surface wind speeds were overestimated by all models to varying degrees. The observed daily maximum downward shortwave radiation and particularly low values during haze days were represented in the participating models. For air pollutants, the main features of the accumulation of air pollutants are generally reproduced in the current generation of on-line coupled air quality models. The correlation coefficients between the multi-model ensemble mean and the CARE-China observed near-surface air pollutants range from 0.51 to 0.94 (0.51 for ozone and 0.94 for $PM_{2.5}$) for January 2010. However, large discrepancies exist between simulated aerosol chemical compositions from different models. The coefficient of variation can reach above 1.3 for sulfate in Beijing and above 1.6 for nitrate and organic aerosols in coastal regions. During clean periods, simulated aerosol optical depth from different models are similar, but peak values differ during severe haze events, which can be explained by the differences in simulated inorganic aerosol concentrations and hygroscopic growth efficiency. However, it was found that using the ensemble mean of the models produced the best prediction skill. Based on the experiments results, Gao et al. (2019) discussed the estimates of aerosol radiative forcing, aerosol feedbacks, and possible causes for the differences among the models. Visit the special issue site for details of Gao et al. (2019).

4. Next steps

At the 10th International Workshop on Atmospheric Modeling in East Asia in 2019, problems and new ideas which should be taken into considered in the next step were discussed. Major discussion points are as follows:

- One of key tasks of Phase IV is to organize a group working for collecting and qualifying observation data for modeling studies. In addition to collect available high quality observation data, it is proposed that MICS-Asia Phase IV will perform own measurements and establish new monitoring sites, especially to evaluate transboundary air pollution.
- Development of a new emission inventory (upgrade of MIX inventory) is one of important issues in Phase IV.
- It is proposed to consider a group working for model improvements in Asian region.
- For specific topics, one idea is to organize small groups who are interested in the topics. This way does not have to wait for more than 10 model runs, but requires hard works of participants.
- Impacts are one of potential new themes in Phase IV which related to policy making.
- Training (capacity building) is one of important roles of MICS-Asia Phase IV to improve quality of modeling studies in Asia.
- Enhancement of MICS-Asia community by involving potential scientists outside the current community is important in Phase IV.

Detailed work plan on MICS-Asia Phase IV is planned to be discussed at the 11th International Workshop on Atmospheric Modeling in East Asia.

Acknowledgements

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Joint Research Project with Asian Institute of Technology, Pollution Control Department and JICA Research Institute: Source analysis and reduction strategy of fine particulate matter in Bangkok Metropolitan Region, Thailand

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Abstract

Bangkok Metropolitan Region (BMR) is a region consisting of populated central area of Bangkok and surrounding provinces, sharing industry, infrastructure, and housing. BMR 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 government operates national monitoring network of gaseous and particulate pollutants as well as precipitation. There were several previous studies focusing on the PM pollution in BMR with the detail PM compositions for source apportionment studies. However, there are lack of long term monitoring data of PM and precipitation, which are necessary to analyze source apportionment of PM. The two joint projects between Japan and Thailand have implemented monitoring of chemical components of $PM_{2.5}$, coarse particles (>2.5 μm) and precipitation at the selected sites in BMR. By using these monitoring data and receptor models, vehicle exhaust, biomass burning and secondary formation could be identified as the main sources of $PM_{2.5}$ in BMR. These results can be compared with the simulation results by the air quality models and will provide information on the sources for which intensive measures should be taken. The projects proposed policy action plans based on air pollution countermeasures described in the national agenda and technical action plans based on the results of monitoring and modeling analysis.

Keywords: $PM_{2.5}$, ozone, Wet deposition, Source apportionment, Air quality modeling

1. Introduction

Many urban cities in Southeast Asian countries is faced to serious air pollution

issues, which will cause death and health damages and also affect economic growth. In rural areas of Southeast Asia, biomass burning is the most dominant source of air pollution followed by vehicle emission, and in urban areas, vehicle and industrial emissions are the most important sources of air pollution. Bangkok Metropolitan Region (BMR) covers an area of 7,762 km^2 and has 14.6 million population. BMR has been rapidly urbanized, and the region has spread to the neighboring provinces since the 1980s. On the other hand, heavy commuter traffic in BMR is one of major causes of air pollution, and the number of registered vehicles is increasing.

According to the "Thailand State of Pollution Report in 2018", the overall air quality in 2018 has been steady state (Pollution Control Department, 2019). However, $PM_{2.5}$, PM_{10} and ozone exceed the national air quality standard. The 24 hours average values of $PM_{2.5}$ was in the range of 3-133 $\mu g/m^3$ in countrywide Thailand, and some stations exceeded the standard value (50 $\mu g/m^3$). The annual average values of $PM_{2.5}$ were between 9-41 $\mu g/m^3$. $PM_{2.5}$ had a tendency to be decreasing since 2015, while in 2018 the annual average concentration of $PM_{2.5}$ in 2018 has increased. The 24 hours average value of PM_{10} was in the range of 2-303 $\mu g/m^3$ in countrywide Thailand, and some stations largely exceeded the standard value (120 $\mu g/m^3$). The annual average values of PM_{10} were between 23-120 $\mu g/m^3$.

BMR is also suffered high ozone pollution because of large emission of ozone precursors such as NOx and VOCs and year-round active photooxidation. The maximum 1 hour average of ozone at stations was 123 ppb as nationwide average and 193 ppb as the maximum value of stations. These values are exceeded the national standard (100 ppb). The maximum 8 hours average value from each station was 97 ppb as nationwide average and 149 ppb as the maximum value of stations. These values are also exceeded the national standard (70 ppb). Although ozone values still exceed the standard, they have been steady state over previous several years.

There were several previous studies focusing on the PM pollution in BMR with the detail PM compositions for source apportionment studies (Chuersuwan et al., 2008, Wimolwattanapun et al., 2011). However, the previous studies were mainly based on intermittent PM sampling two to three times of 24-hour sampling per week and mainly focusing on selected months to represent the wet and dry seasons. In order to obtain the information on $PM_{2.5}$ levels and comprehensive compositional data based on a continuous monitoring throughout the year, 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), Asian Institute of Technology (AIT) and Japan International Cooperation Agency Research Institute (JICA-RI) since 2014. The project has been also supported by Thai national research counterparts including Pollution Control Department (PCD), Environmental Research and Training Center (ERTC), King Mongkut's University of Technology Thonburi (KMUTT) and Ladkrabang (KMITL). The key activities of the research project include (i) Monitor chemical components of $PM_{2.5}$, coarse particles (>2.5 μm) and precipitation at the selected sites in Bangkok over a year, (ii) Clarification of temporal variations of chemical components of $PM_{2.5}$, coarse particles and precipitation at selected sites in Bangkok, (iii) Emission inventory development and model simulations (a receptor model and/or a chemical transport model) to identify the sources of $PM_{2.5}$ and O_3 in Bangkok, and (iv) Socioeconomic

analysis of $PM_{2.5}$ and O_3 by using model results and input on air pollution mitigation measures in BMR.

The continuous monitoring was performed through a weekly sampling using the filterpack samplers (one sample was collected over seven days of a week) from September 2015 to March 2017. The monitoring was done in two locations of BMR, a suburban site (in the AIT campus) and a city center site (at the Pollution Control Department office). Then, subsequent analysis of $PM_{2.5}$ mass and compositions of water soluble ions, trace elements, and elemental carbon (EC)/organic carbon (OC). Because deposition processes are important to determine the lifetime of PM and atmospheric burden on the ground, the rain-water chemical compositions were analyzed to determine the wet acid deposition, while the dry deposition was determined by the inferential method.

The Ministry of the Environment, Japan (MOEJ) and the Ministry of Natural Resources and Environment, Thailand (MONRE) have been promoting collaboration in many fields of the environment. To formalize collaboration between the two countries in a comprehensive way, the both ministers signed a Memorandum of Cooperation (MOC) in 2018 to strengthen, facilitate and develop mutual cooperation in the field of environment. The Japan Thailand Clean Air Partnership (JTCAP) for Particulate Matter Reduction Strategy and Measures Development launched to implement analysis of emission inventory and identify source characteristic of $PM_{2.5}$ in Thailand. The project's timeframe is set for 2 years (2018-2020). Preliminary study area is designed for BMR to tackle the upcoming $PM_{2.5}$ pollution episode in December 2018 to April 2019. ACAP, PCD, National Institute for Environmental Studies (NIES), KMUTT, Thammasat University (TU), Suranaree University of Technology (SUT) are participated in JTCAP.

Besides the research activities, the joint workshop was held to enhance the further cooperation and discussions. The workshop invited all stakeholders involved in tackle air pollution problems. JTCAP will also be a platform for academics and researchers to share ideas and views on air quality research issues, gain active discussions and extend networking for the benefit of future research collaboration.

In this article, the outcomes of the "A Study on Urban Air Pollution Improvement in Asia" project and JTCAP are presented.

2. Outcomes of "A Study on Urban Air Pollution Improvement in Asia" project

2.1 PM and Acid Deposition monitoring in BMR

Long term monitoring of PM and acid deposition was conducted at the two sites which represent urban and suburb area of Bangkok (Narita et al., 2019). One is located at the rooftop of the PCD in urban area of Bangkok. The rooftop of PCD building is located at 13.8º (N) and 100.5º (E) that is situated of 64 meters 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. A Sky train line is located above this road. The other site is at the rooftop of the ambient laboratory of AIT in Pathumtani, suburb area of Bangkok, which is located at 14.1º N and 100.6º E and 6 meters 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. The monitoring were conducted simultaneously at these two sites during the period from September 2015 to March 2017, that yielded a total of 78 weekly samples at each site. (A weekly sample was collected over a week period, from Monday to Monday of next week).

One five-stage and one two-stage filter pack sampler were installed at each site to collect weekly ambient samples for PM mass and composition measurement, as well as the acidic and basic gases for dry deposition analysis. A wet deposition sampler was also installed at each site to collect the rain samples for elucidation of PM removal processes. The five-stage filter pack collected air samples on two types of filters on the first and second stages: a quartz filter for the coarse PM (PM with size >2.5 µm, FC) and a Teflon filter for the fine PM ($PM_{2.5}$, F0) which were used for mass and ionic composition analyses. A $PM_{2.5}$ impactor controlled at the flow rate of 2.0 L min^{-1} was used to collect fine particle ($PM_{2.5}$) on the F0 stage. The remaining 3 stages collected the gases (SO_2, HCl, HNO_3 and NH_3) on the impregnated filters. To collect additional PM samples for other analysis, a 2-stage filter pack was used in parallel which collected coarse and fine PM on quartz filters for subsequent EC/OC analysis using the Thermal Optical Reflectance (TOR) method. The sampling pumping rate was controlled at 2.0 L/min continuously over a one-week sampling period by using a mass flow controller. The PM mass was quantified using a microbalance. Water soluble ions (SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, Na^+, K^+, Ca^{2+}, Mg^{2+}) of PM and wet deposition samples were analyzed by ion chromatography. Selected key elements in $PM_{2.5}$ were also analyzed by ICP-MS. The sampling and analyses were performed by following technical manuals and QA/QC of EANET to ensure required data quality (EANET 2010a, 2010b, 2013).

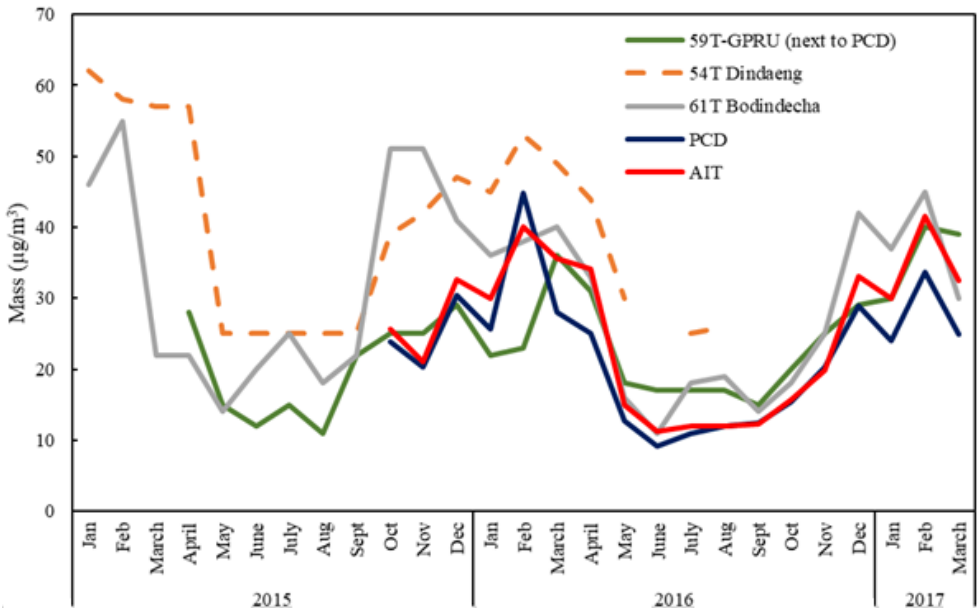

Figure 1. Monthly average $PM_{2.5}$ concentrations at the sites in BMR (2015-2017)

Figure 1 shows the monthly average $PM_{2.5}$ mass concentrations calculated from weekly sampling results as well as the hourly data at Thai government network stations (GRPU, Dindaeng and Bodindecha) (Narita et al., 2019). The levels of $PM_{2.5}$ at AIT and PCD had similar seasonal variation patterns, higher in the dry season and lower in the wet season. The levels measured at AIT and PCD stations were in the similar range of those measured at a Thai government network station. The annual average of $PM_{2.5}$ concentrations in 2016 was 20.6 $\mu g/m^3$ at PCD and 22.5 $\mu g/m^3$ at AIT that were below the National Air Quality standard (25 $\mu g/m^3$). The seasonal averages of $PM_{2.5}$ concentrations in the dry season; 29.0 $\mu g/m^3$ at PCD and 32.6 $\mu g/m^3$ at AIT, were significantly higher than those in the wet season; 14.7 $\mu g/m^3$ at PCD and 15.2 $\mu g/m^3$ at AIT.

2.2 Source contribution of PM in BMR by the receptor modeling and emission inventory

The source contributions to PM pollution in BMR was determined by 2 methods: the receptor modeling and emission inventory (EI). By using Chemical Mass Balance receptor modeling, the weekly monitoring data of $PM_{2.5}$ mass and composition at 2 sites was analyzed to the identify the key $PM_{2.5}$ sources and estimate their quantitative contributions to the ambient $PM_{2.5}$ mass (Narita et al., 2019, Kim Oanh, 2017). The major contributing sources to the $PM_{2.5}$ mass at both AIT and PCD sites were traffic and biomass burning, accounting for 23% to 41%, respectively. Both sites had almost the same relative contributions to $PM_{2.5}$ mass during wet season at each site, whereas during the dry season, biomass burning had significantly higher relative contribution especially at the suburban AIT site. The most significant biomass open burning in the study area is the rice straw field burning, and thus AIT surrounded by the rice paddy field will be expected to have more influence from this source. Regional and transboundary transport of biomass burning smoke would bring the emissions from the outside BMR could be considered, but current results could not quantify the contribution from outside BMR.

The EI for air quality simulation in BMR was updated on the base year of 2016. The EI domain included the Bangkok city and surrounding provinces within coordinates of 13°27'32.4"-14°05'27.6"N and 101°05'31.2"-100°09'43.2"E. The detailed procedure to update EI is described in the literature (Narita et al., 2019). The EI results show that primary $PM_{2.5}$ in BMR was emitted from three main sources; on-road mobile (accounted for 59%), biomass open burning (20%), and industry (19%). The emissions of other gaseous species were also mainly contributed from these sources. 70% of NOx, 48% of CO, and 22% of NMVOC were emitted from on-road traffic, while 29% of CO and 4% of NMVOC were from biomass open burning. It should be noted that the street cooking emissions may be significant in Bangkok but was not included in this estimation.

Emission shares of different on-road vehicle types in the BMR were also calculated. The large pick-up trucks had the highest share of $PM_{2.5}$ emission followed by the normal trucks. The pick-up trucks were also the major contributor to NOx, followed by other diesel-fueled vehicles of truck and bus. The major contributors to NMVOC were the gasoline-fueled private cars and motorcycles, but the diesel-fueled vehicles also contributed significantly. It should be noted that off road mobile sources such as boats, construction machines and agricultural machineries may contribute significantly to the

PM pollution in BMR but these have not been included in the EI.

2.3 Levels of acid deposition and potential risk in BMR

Figure 2 shows monthly variations of precipitation pH at AIT and PCD sites in order to discuss acidity of rain water. The ranges of monthly pH values at PCD and AIT from September 2015 to February 2017 were 4.59 – 7.16 and 4.69-7.03 (Narita et al., 2019), respectively. Approximately 40% at PCD and 20% at AIT of rain samples were acidic (pH ≤ 5.6). The average pH values at PCD and AIT sites in the wet period was 5.59 ± 0.73 and 5.77 ± 0.62, respectively, and the average pH values in dry period were 4.91 ± 0.82 and 5.18 ± 0.79, respectively. The pH values were lower in the dry period, and this trend was similar to that observed in Bangkok and Pathumthani EANET sites (EANET, 2017). The data analysis of rain water components and the relationship with PM components will be reported in out forthcoming paper.

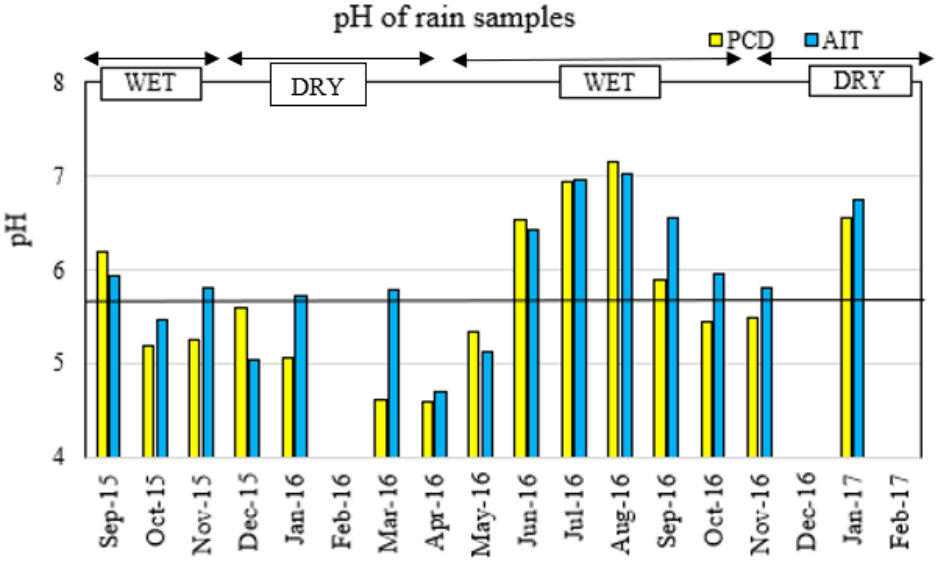

Figure 2. Monthly variations of precipitation pH at AIT and PCD sites (2015-2017)

The total deposition amounts at the AIT and PCD sites were estimated by the sum of dry deposition and wet deposition fluxes. Higher total deposition amounts of sulfur and nitrogen compounds were observed in the wet period (e.g. 140 kgS/km^2/month and 375 kgN/km^2/month at the AIT site in July 2016). The wet deposition was dominant during the wet period which showed that wet deposition played an important role to remove sulfur and nitrogen species from the atmosphere. The total deposition of sulfur compounds during September 2015 – February 2016 (over 18 months) at PCD and AIT sites were 837 and 821 kgS/km2, respectively, while the total nitrogen compounds were 3,132 and 3,043 kgN/km^2, respectively.

The critical loads of sulfur and nitrogen in Thailand obtained by previous studies (Bouwman et al., 2002, Milindalekha et al., 2001) were compared to the above total deposition amounts. The sulfur deposition at AIT in 2016 was 586 kgS/km^2/yr that was lower than the critical load (3,000 – 5,000 kgS/km^2/yr). Likewise, the nitrogen deposition was 2,235 kgN/km^2/yr that was also lower than the critical load (6,090 -

9,030 kgN/km^2/yr). This implies that the soil in Pathumthani province still has buffering capacity to neutralize the current acid deposition rate. However, the adverse effects such as oversaturation of acidic species in soil and leaching of metallic components from the soil may be occurred, if the emissions of SO_2, NOx and precursors of PM are not controlled in the near future.

2.4 Policy implications

The obtained scientific results of BMR measurements and data analysis confirm the general picture of air pollution in BMR. The levels of $PM_{2.5}$ are generally alarming in the context of the WHO guidelines (10 µg/m^3 for daily mean and 25 µg/m^3 for annual mean). The observational data also reveal more precise characteristics of the problem, such as the diversity of pollution emission sources composed of traffic, biomass burning and those emitting precursors of the secondary particles. These clarifications of major pollution sources in the Bangkok metropolitan area provide important scientific knowledge for air pollution control policy in Thailand. The air pollution issues faced by Thailand are also common for many other the emergent nations in East Asia.

Thailand is one of the newly industrialized countries a medium-developed country and has established environmental regulations similar to those of developed countries. The national air quality standards is set to the equitant levels of developed countries, and vehicle emissions standards are based on the EU's emission standards (Euro 5 and 6 by 2023). Furthermore, a national air quality monitoring network has already covered nationwide. Therefore, it is important to observe current regulations before considering a review of the regulatory standards. It is important to consider measures against newly regulated pollutants, such as $PM_{2.5}$, are difficult even for developed countries that introduce advanced control technologies at the emission sources because formation mechanism of air pollutant is complicated. The “A Study on Urban Air Pollution Improvement in Asia” project proposed the below measures to reduce more air pollution in Thailand (JICA research institute, 2020).

(1) Promotion of comprehensive measures against air pollution, including energy policy, agricultural policy, urban planning and land use policy, etc.
(2) Incentives and innovation are key components for comprehensive measures including not only air pollution measure but energy policy, etc.
(3) Deep analysis of air pollution data and current emission control policy for each source, and promotion on $PM_{2.5}$ pollution mitigation policy based on the analysis results

3. Outcomes of Japan Thailand Clean Air Partnership (JTCAP) project

3.1 Source analysis of $PM_{2.5}$ based on observations in BMR

During severe pollution period of December 2018 to April 2019, 24 hour sampling of $PM_{2.5}$ was conducted at 3 sites in BMR once a two days. The monitoring sites were selected to cover various site characteristics. One is located in the industrial area of Bang Na district, the second one is in the residential area of Phaya Thai district, and third one is at the roadside of Din Daeng district. The collected $PM_{2.5}$ samples were analyzed by sent to Japan for ionic and metallic components by PCD, and for carbon components for analysis by ACAP.

Figure 3 shows daily average $PM_{2.5}$ concentrations at 3 sites in BMR. The $PM_{2.5}$ concentrations at 3 sites exceeded the national standard of 24 hour average (50 $\mu g/m^3$) from late December 2018 to late January 2019. The air quality index (AQI) levels on January 30 in Bangkok reached 175, which is classified as unhealthy. The Ministry of Education decided to close more than 400 public schools during the severe pollution period. Thereafter, the $PM_{2.5}$ concentration was lowered in February and the concentration increased again in April and May. In this period, air pollution caused by biomass burning in northern Thailand and neighboring countries is significant, so it may be affected by long range transportation. By comparison among the sites, the roadside site was the highest, followed by the industrial site and the residential site.

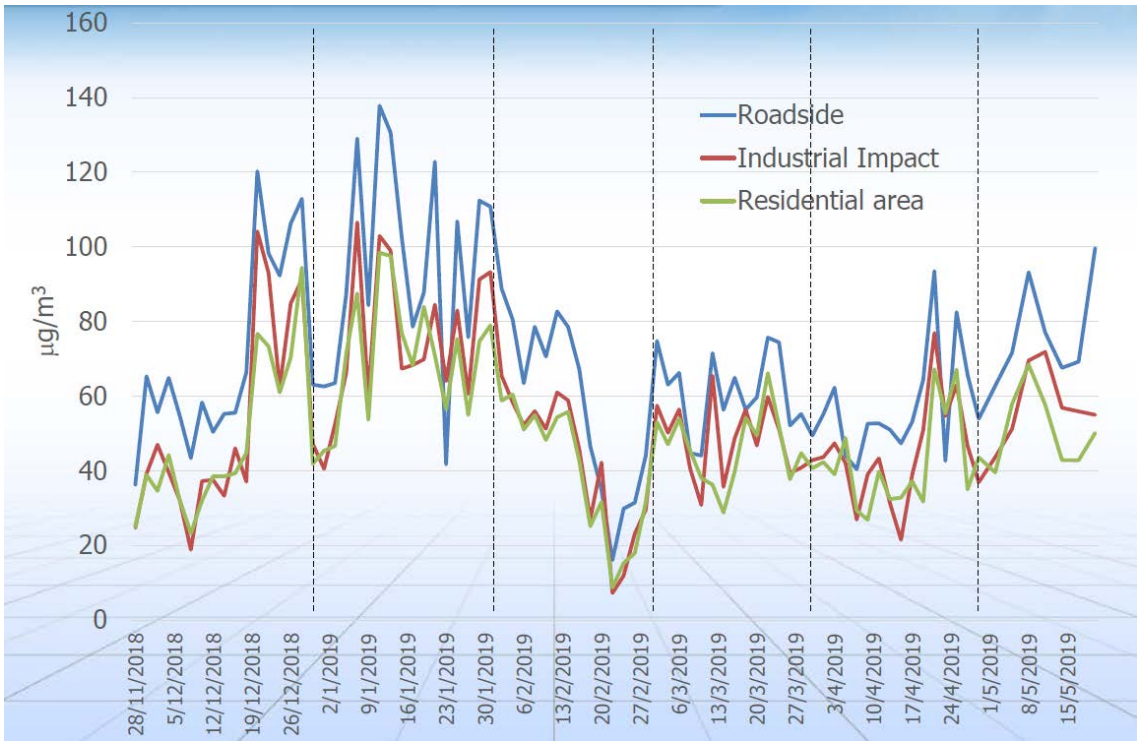

Figure 3. Daily average $PM_{2.5}$ concentrations at the sites in BMR (2018-2019)

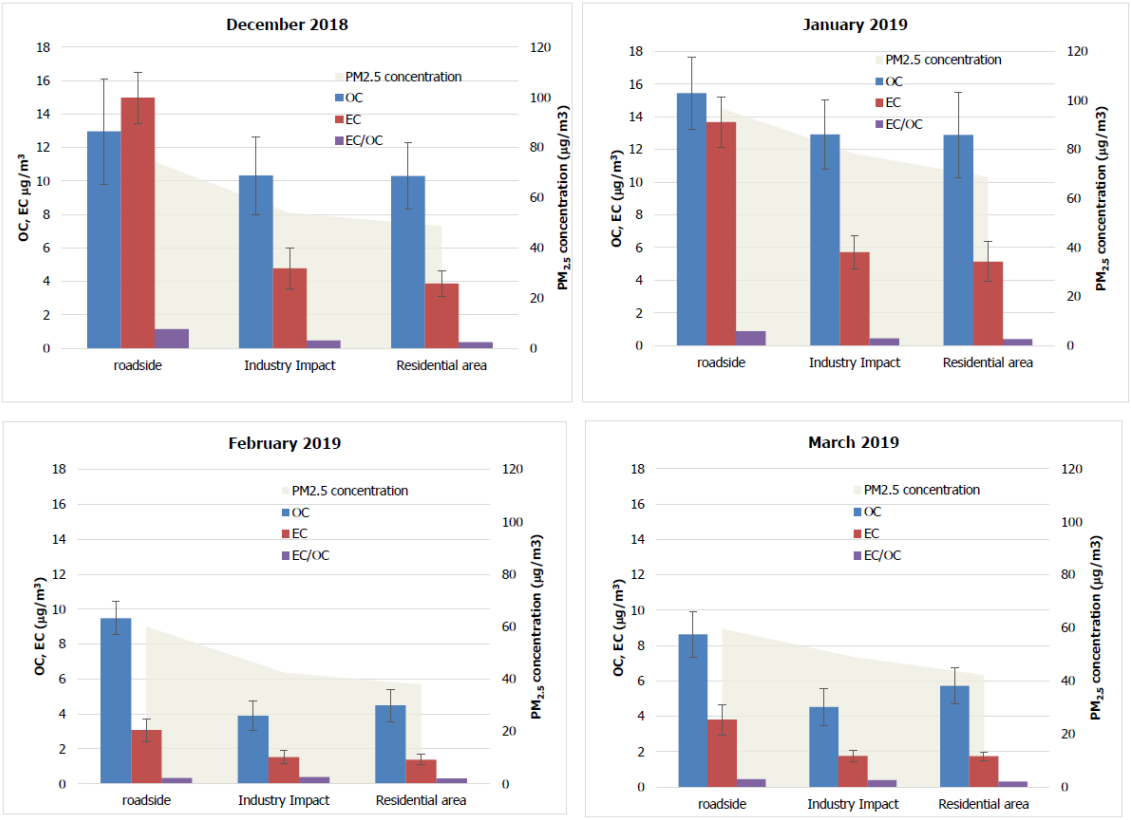

Figure 4. Monthly average of EC/OC concentrations in $PM_{2.5}$ at 3 sites in BMR

Figure 4 shows monthly average of elementary carbon (EC) and organic carbon (OC) concentrations in $PM_{2.5}$ at 3 sites in BMR from December 2018 to March 2019. Both EC and OC concentrations were higher in December and January than the other months. The EC/OC ratio was highest at the roadside site, which implied that the contribution of EC emitted from automobile exhaust was high. The average EC concentrations from December 2018 to March 2019 were 9.11 ± 5.94 µg/m^3 (roadside), 3.55 ± 2.37 µg/m^3 (industrial) and 3.12 ± 2.06 µg/m^3 (residential, and the average OC concentrations were 11.81 ± 4.61 µg/m^3 (roadside), 8.15 ± 4.93 µg/m^3 (industrial) and 8.55 ± 4.71 µg/m^3 (residential). Compared with the annual average concentrations in BMR investigated by the former JICA-RI project from 2015 to 2017 (OC: 2.75 ± 1.44 µg/m^3, EC: 4.29 ± 3.44 µg/m^3), the OC concentration in this study was remarkably higher. The results suggest that the contribution of OC originated open burning was high during severe pollution period.

Regarding the ionic component concentrations, Figure 5 demonstrates the concentrations tended to be high in January when the $PM_{2.5}$ concentration was high, and particularly the NO_3^- concentration was high. It is considered that NO_3^- is originated from biomass combustions and nitrogen oxides emitted from automobiles. The overall trend shows that SO_4^{2-} concentration was highest, followed by NH_4^+ and K^+. The origin of SO_4^{2-} is assumed to be mainly due to sulfur content in automobile fuels. No distinctive differences were found for comparisons among monitoring sites.

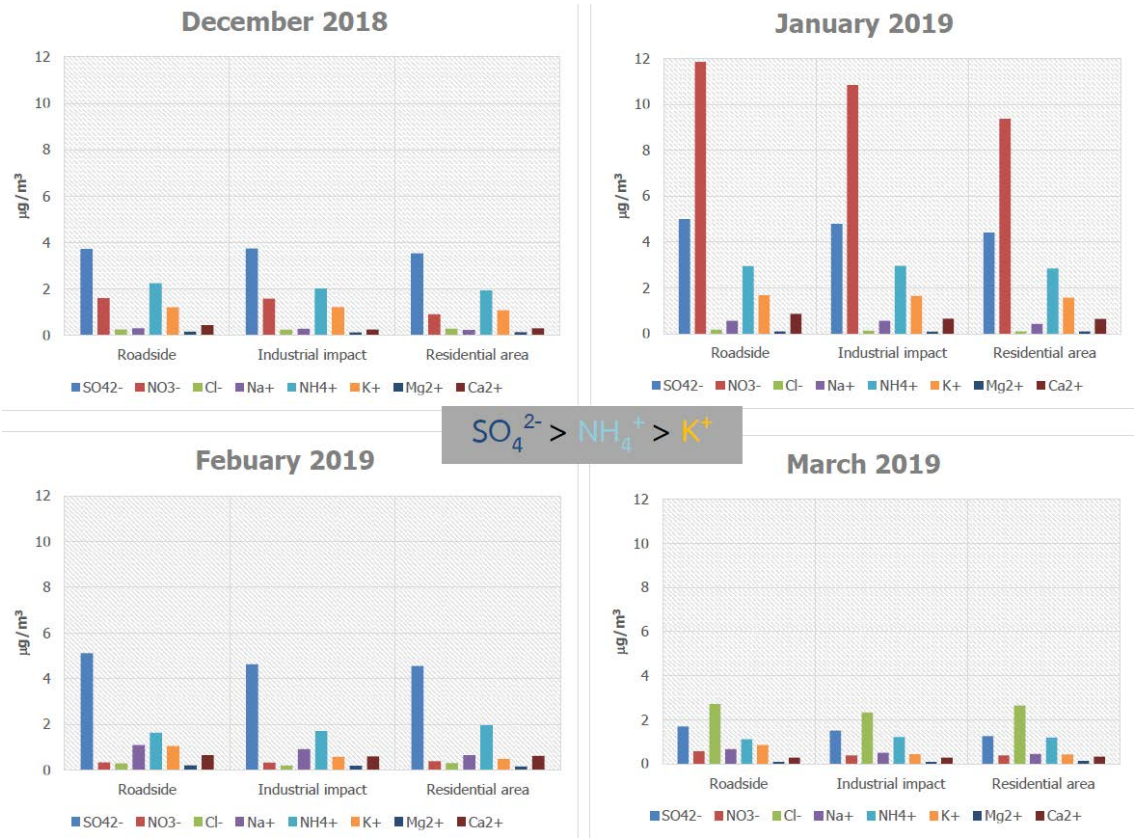

Figure 5. Monthly average of ionic component concentrations in $PM_{2.5}$ at 3 sites in BMR

The observed data of $PM_{2.5}$ components were applied to a receptor model, a positive matrix factorization (PMF) model, and the source analysis was performed. There were 6 significant factors identified. The Factor 1 can be related to road dust origin because of the high contribution of Cu and Sb in brake pads and Zn used as an additive in tires. Similarly, the Factor 3 was considered to be sea salt origin because of the high contributions of Na and Cl, the Factor 4 was considered to be secondary particle formation and diesel exhaust particles because of high contribution of NO_3^- and EC, and the Factor 6 was considered to be mixed origin of secondary particles, biomass combustion and diesel exhaust particles because of the high contribution of Zn, SO_4^{2-}, NO_3^-, NH_4^+, K^+, OC and EC.

Figure 6 shows the contribution rate to $PM_{2.5}$ mass concentration for each factor identified by PMF. It was difficult to estimate the contribution of each source to the $PM_{2.5}$ mass concentration because multiple sources were assigned to the same factor. These results demonstrated that secondary particles, biomass combustion and diesel exhaust particles were the major sources of $PM_{2.5}$ in the BMR as with the case of source apportionment by the JICA-RI project (Narita et al., 2019, Kim Oanh, 2017).

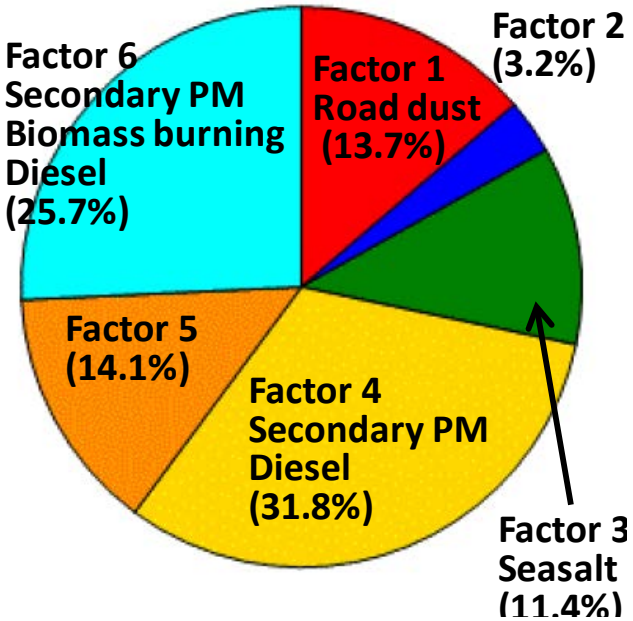

Figure 6. Contributions of source factor identified by PMF to $PM_{2.5}$ Mass (Roadside site of Din Daeng district)

3.2 Update of the latest emission inventory and clarification of PM2.5 pollution status in BMR by air quality model

For the purposes of understanding the actual state of air pollutant emissions in Thailand and preparing the emission data to be input into the air quality model, PCD invited Dr. Savitri Garivait. of KMUTT was invited as a collaborator to update the latest emission inventory. The outline of updated inventory is shown as follows.

(Anthropogenic emissions data)

- In Thailand: Update of the base year 2015 data prepared by KMUTT and provided to PCD
- Outside Thailand: Use ECLIPSE_V5a from the International Institute for Applied Systems Analysis (IIASA) or EDGARv4 from the European Commission's Joint Research Center (JRC)

(Biomass combustion emissions data)

- In Thailand: Updated the data for full-year 2018 and January-March 2019 by KMUTT and provided to PCD
- Outside of Thailand: Use GFEDv4 from research institutes in Europe and USA

(Natural emissions data)

- Outside of Thailand: Use MEGANv2, created by the National Center for Atmospheric Research (NCAR)

Figure 7 shows the distribution of major anthropogenic pollutant emissions in Thailand. NOx and VOC emissions were higher in the BMR, where the traffic volume was significantly higher and a number of industrial estates were located. While NOx emissions from liquefied petroleum gas (LPG) use in households were high in BMR, PM_{10} emissions from wood fuel combustion are high in rural areas. This difference suggests that household energy use patterns are largely different between urban and

rural areas. The monthly $PM_{2.5}$ emissions from agricultural fires in Thailand showed a peak in February and were higher during the dry season from December to April, while the $PM_{2.5}$ emissions from forest fires were highest in March. These data showed that the emissions from agricultural land are about five times higher than those from forests, and control measures for the burning of agricultural residues is necessary. It was pointed out that the frequency of small-scale biomass burning is high in BMR, so it is necessary to validate this emission data to be used for the air quality model.

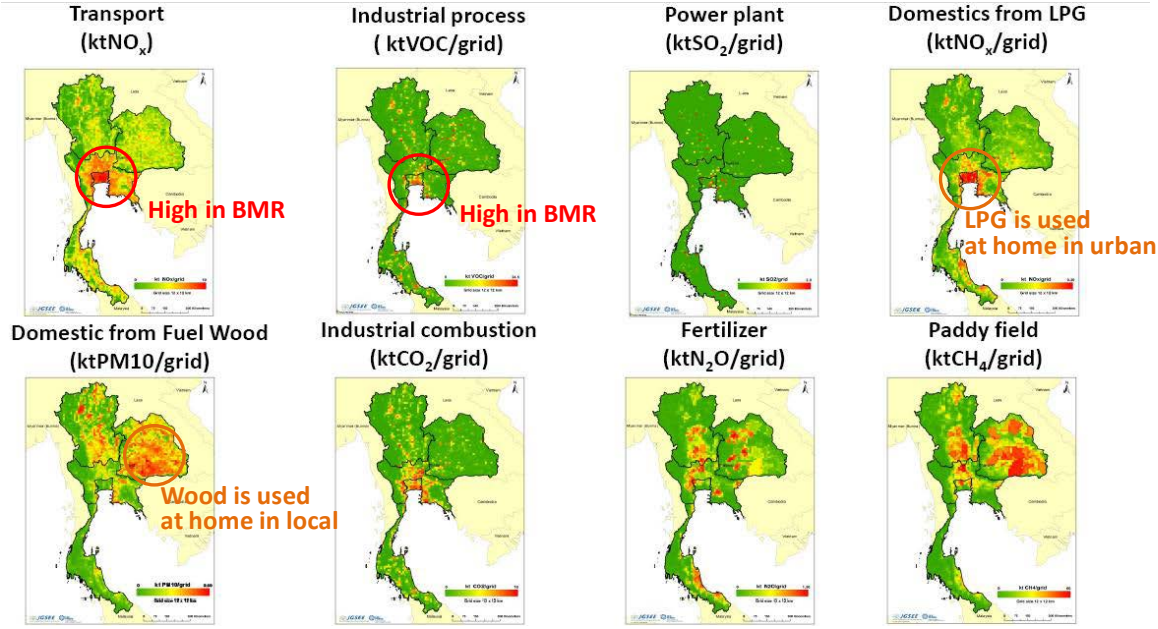

Figure 7. Distribution of major anthropogenic pollutant emissions in Thailand (2015, Spatial resolution 12x12km)

Using the emission inventories shown above as the input data, the air quality model simulation in BMR was performed. When the simulated and observed $PM_{2.5}$ concentrations in November 2019 were compared, the high concentrations area in $PM_{2.5}$ were consistent, indicating that the air quality model has good reproducibility in the period when the influence of biomass burning is small. In addition, JTCAP discussed the development of emission inventory guidelines to create a system for continuous development of Thailand's national emission inventory, and the PCD will prepare the outline of the emission inventory guidelines or platform, the basic structure, and the draft development plan by March 2020.

3.3 Development of air pollution policies and appropriate measures with building relationships with stakeholders

Based on the results of the monitoring and modeling analyses conducted by JTCAP, the JTCAP participants have compiled effective policy recommendations to reduce $PM_{2.5}$ concentrations, including public awareness, health benefits, and economic benefits, based on the knowledge of the air pollution status and the major $PM_{2.5}$ sources in BMR. Information on experiences of air pollution control in Japan has been provided to PCD, and PCD reviewed the information on domestic and international policy measures in

Thailand.

In August 2019, the cabinet of Thailand approved the National Agenda for Solving the Dust Pollution Problem, an action plan for air pollution measures. This agenda presents short-term targets to be achieved by 2021 and long-term targets to be achieved by 2024 shown as follows.

(Short-term target)

- Reduce and control emission from vehicles making use of incentive measures
- Reduce and control emission from open burning; Encouragement of the cultivation of perennial plants; Promote use of agricultural residues
- Reduce and control emission from construction and urban planning projects
- Reduce and control emission from industrial origins by setting emission standards with load factor as an indicator
- Reduce and control emission from households by promoting the use of clean energy

(Long-term target)

- Strengthen emission reduction and pollution control for vehicles; Enforce the air pollution standards that meet the new Euro 6 standards by 2022; Enforce the fuel standards that do not exceed 10 ppm of sulfur content by 2024
- Control and reduce pollutants from open burning by using agricultural residues instead of burning; Blanket ban on open burning; Ban on burning any sugar cane by 2022
- Strengthen pollution control from construction and urban planning projects; Development of measures and standards to control dust from construction by setting regulations
- Control and reduce pollutants from industrial sources by reviewing and modifying standards so that the air pollution standards from industrial plants are consistent with international standards
- Control and reduce pollutants from household by supporting use of clean energy at home

Among the air pollution measures listed in the National Agenda, the JTCAP selected control of vehicle source, stationary sources, biomass burning sources, and upgrade of observation networks and reporting systems as priority issues. For each issue, the JTCAP summarized positive social involvement measures, laws and regulations, new technologies, and regulation and local authority regulation/responsibility.

For measures not listed in in the National Agenda, the JTCAP proposed the policy recommendations as technical action plans. The items are listed below.

(1) Further investigation on the $PM_{2.5}$ sources by observation and modeling
(2) Clarification of vehicle emissions by vehicle type and age
(3) Consideration of possible exhaust gas inspections and regulations to be introduced in the future
(4) Elucidation of the relationship between meteorological conditions and the origin of $PM_{2.5}$ during pollution and normal periods
(5) Promote public awareness, health impact analysis of $PM_{2.5}$ and cost benefit analysis
(6) Restrictions on the entry of high-load diesel vehicles into BMR
(7) Elucidation of the secondary formation mechanism of $PM_{2.5}$ including literature review

The intensive discussions on vehicle measures were also hold. It is important to reduce the sulfur content of fuels used for vehicles which satisfy EURO 5 and 6 emission standards because use of high sulfur fuels may lead to the failure of emission treatment systems. It was pointed out that measures should be taken to prevent high sulfur fuels from being used for EURO 5 and 6 vehicles.

4. Summary and future plans

The projects of JICA-RI and JTCAP demonstrated that vehicle exhaust, biomass burning and secondary formation could be identified as the main sources of $PM_{2.5}$ in BMR by receptor model analysis. These results can be compared with the simulation results by the air quality models and will provide information on the sources for which intensive measures should be taken. Air quality model results in BMR showed good reproducibility when the effect of biomass combustion is small. At the next step, air quality model calculations will be conducted for high and low $PM_{2.5}$ concentration periods to analyze the characteristics of $PM_{2.5}$ concentration distribution during high pollution period in BMR. It will also evaluate the impact of emissions from outside BMR, mainly from open burning, during high $PM_{2.5}$ concentration period.

JTCAP reviewed the current method and extracted issues of emission inventory in Thailand. JTCAP also summarized future prospective of the emission inventory guidelines development and the role of government to the development. The summary results will lead to create a system for the continuous development of Thailand's national emission inventory.

The projects of JICA-RI and JTCAP proposed policy action plans based on air pollution countermeasures described in the national agenda and technical action plans based on the results of monitoring and modeling analysis. In the future, we should review existing laws, environmental standards and guidelines and improve as appropriate, For countermeasures for major $PM_{2.5}$ sources, we should consider tangible measures for open burning, create emission factor data by vehicle type and age, and control emissions of used vehicles based on the obtained emission factor data.

The 2nd Japan-Thailand Environment Policy Dialogue was held in January, 2020, and the continuation of JTCAP has been agreed. Now the participants of JTCAP is preparing the report summarizing the Phase 1 activities and a proposal for the Phase 2 activities that will start in April 2020.

Acknowledgements

The collaborative research project of “A Study on Urban Air Pollution Improvement in Asia” was funded by JICA Japan International Cooperation Agency Research Institute (JICA-RI). Besides ACAP researchers, Prof. Nguyen Thi Kim Oanh and Dr. Didin Agustian Permadi at AIT and Dr. Daiju Narita at JICA-RI are major researchers of the project. The monitoring and analysis operation is supported by Thai national research counterparts such as PCD, Environmental Research and Training Center (ERTC), King Mongkut’s University of Technology Thonburi (KMUTT) and Ladkrabang (KMITL).

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Kessinee Unapumnuk and Dr. Patcharawadee Suwanathada at PCD are major researchers of the project. The JTCAP activity is supported by other Japan and Thai research counterparts such as PCD, National Institute for Environmental Studies (NIES), KMUTT, Thammasat University (TU), Suranaree University of Technology (SUT).

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Development of Emission Inventory Manual for Mongolia

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Abstract

In reply to the request from Mongolia, the Emission Inventory Manual for Mongolia was developed by Asia Center for Air Pollution Research in cooperation with SUURI-KEIKAKU Co., Ltd, from JFY2016 to JFY2018 on the Integrated Programme for Better Air Quality in Asia (IBAQ Programme) of Clean Air Asia. The manual was written in Mongolian, English and Japanese for convenience of users, and will be used to develop the national emission inventory of Mongolia to assess the air pollution problem in Mongolia. The process of development of the manual is described in this paper.

Keywords: Emission inventory, Manual, Mongolia, Air pollution

1. Introduction

Mongolia is a coal rich country with heavy dependence on coal for energy sources. Since most of the coal used in Mongolia contains a great amount of water and ash, dust emission with coal combustion is large and air pollution in the winter season is severe when coal is used as heating. The coal consumption, especially in Ulaanbaatar (hereinafter UB) city, the capital city of Mongolia, is increasing and air pollution is much severe due to population concentration and economic development. The major emission sources are coal combustion at coal fired power plants for electricity and heat generation, Heat Only Boilers (hereinafter HOBs), small boilers such as Coal Fired Water Heaters (hereinafter CFWHs), and small stoves for heating and cooking at home. Additionally, the increase of traffic volume in recent years is leading to concerns about air pollution caused by vehicle exhaust gas.

According to the result of air quality monitoring, major air pollutants in UB city are sulfur dioxides (SO_2), particle matter (TSP, PM_{10}, $PM_{2.5}$), and nitrogen dioxides (NO_2). In the season using a heater in 2017, 50 percent of the total available measuring hours in these pollutants exceed the acceptable concentration in air quality standard MNS4585:2016.

Under these conditions, donor agencies such as World Bank, Asia Developing Bank, Japan International Cooperation Agency (hereinafter JICA) are implementing the project for air pollution control mitigation in UB city. JICA has been implementing the technical cooperation project for capacity development for air pollution control in UB city since March 2010. Clean Air Asia (hereinafter CAA) has been implementing Integrated Programme for Better Air Quality (hereinafter IBAQ), which is the comprehensive support on air pollution for the Ministry of Environment and Tourism of Mongolia (MET) and National Agency for Meteorology and Environmental Monitoring (NAMEM), since 2016. As part of this programme, this manual was elaborated to develop emission inventory to cover entire Mongolia.

Using this manual, the emission amount of air pollution in the entire Mongolia will be estimated, which will be the calculation evidence of Air Law and Air Payment Law as well as the scientific and basic data to discuss the regional air pollution control measures.

2. Method (general description)

In general, emissions of air pollutants are estimated by the following basic formula for each source category, when it is difficult to measure them directly. Details of emission estimation methods are explained in each other sector.

$$\text{Emission} = \underline{\text{Emission Factor (EF)}} \times \underline{\text{Active Data (AD)}}$$

2.1 Emission Factors (EF)

Emission factors are the average rate of emission of a pollutant per unit of activity data for a given category. Examples of emission factors are the following;

・SOx emission per the amount of fuel burnt, calculated based on the sulfur content of the fuel, the sulfur retained in the ash and the reduction achieved by emission control technology (fuel combustion)

・NOx emission per distance (exhaust gas emission from vehicles)

・SOx emission per the amount of copper smelted (copper smelting)

In this manual, the usage of emission factors reflecting the measurement data in Mongolia is given first priority. However, when there is no emission factor specific for Mongolia, default values from established manuals can be used, such as the *Guidelines for Developing Emission Inventory in East Asia* (Japan), *GAP Forum Manual* (Sweden), *EMEP* [1]*/EEA* [2] *Air Pollutant Emission Inventory Guidebook* (EU), *Compilation of Air Pollutant Emission Factors (AP-42)* (USEPA, 1995), and *COPERT* [3] (EU).

[1] The European Monitoring and Evaluation Programme

[2] European Environment Agency

[3] Computer programme to calculate emissions from road transport

When emission is calculated using emission factor of the above manuals, attention to different conditions in Mongolia, such as meteorology, altitude, property of fuel, state of facilities, and so forth, is paid.

2.2 Activity Data

Activity data indicates the extent of emission-causing activities. Required data can be basically collected from statistics and surveys. Examples of activity data are the amount of fuel burnt (fuel combustion), the distance of vehicle traveled (exhaust gas emissions from vehicles), and the rates of the production of the commodity (industrial process without combustion).

In this manual, the available or statistics data of Mongolia are prioritized as long as it exists. Otherwise, international statistical data are used.

2.3 Emission Calculation Tool

In order to calculate emission using the format table for collecting activity data by source, emission calculation tool is prepared in this manual. Emissions are calculated by inputting emission factors to the collected activity data sheet.

3. Process of development of the manual

(JFY2016)

These tasks are the review of existing emission inventory guideline, the discussion for elaborating Emission Inventory (EI) Manual (EIM) and the agreement of the specification for this manual, and the elaboration of EIM for preliminary draft.

Existing emission inventory guideline was developed by Ministry of Environment of Japan (MOEJ) and was provided to MET. By the interview survey for application of this guideline, this guideline was not used in MET and NAMEM. These reasons are;

1. Since developing emission inventory in UB city is based, it is difficult to apply in other counties/cities.
2. Content of guideline is too high level to developing emission inventory only using this guideline.
3. Translation from Japanese to Mongolia is very poor.

On the other hand, JICA technical cooperation project “Capacity Development Project for Air Pollution Control in UB City” is being conducted. Capacity building on the developing EI in UB and the conducting dispersion simulation for officer in APRD (Air Pollution Reducing Department of the Capital City) [4] and NAMEM is being conducted and the manual for the conduction of them is being elaborated. In this field survey, discussion with the relevant organizations such as NAMEM, MET, and so on

[4] This organization name has changed to DAAP (Department of Against Air Pollution of the Capital City)

was conducted and ger [5] area was visited.

NAMEM requested that air pollution emission in the entire Mongolia is obtained. To do so, the manual that NAMEM and the local office are able to calculate emission is needed. Then the elaborating the manual and calculation tool that the local staff is easily able to calculate emission using the statistical data is proposed and agreed with NAMEM. Then, emission in UB city is calculated using 'Update manual for emission inventory' elaborated in JICA project and the purpose of EIM is different from JICA's manual was understood to NAMEM.

The specification for elaborating EI was described based on the above agreement in this fiscal year. This included the list of necessary data and the schedule for elaborating EI. Based on this specification NAMEM is collecting the necessary activity data etc. In the next fiscal year, calculation of emission using collected data, the elaboration of EI Manual using this know-how, and the training using EIM will be conducted.

(JFY2017)

As one of the activities of IBAQ Programme of CAA, Emission Inventory Manual (EIM) and calculation tool of EI, based on the discussion with NAMEM and MET in 2016, was elaborated and trained the Mongolian side regarding the method for using.

Mongolian side has been requesting to match EIM to the condition of Mongolia. Therefore, preliminary version of EIM was prepared based on Manual for Development and Updating of Emission Inventory elaborated in "Capacity Development Project for Air Pollution Control in Ulaanbaatar City Phase 2 in Mongolia" and Guidelines for Developing Emission Inventory in East Asia elaborated by Asia Center for Air Pollution Research (ACAP). The deficiency in these documents was supplemented by the manual and guidelines of Europe and the United States. The purpose of the training is that the Mongolian side will be able to calculate the EI of the entire Mongolia on their own. In order to develop the capacity of developing EI of the Mongolian side, not only listening to the lectures of Japanese side unilaterally, but also the participants will actually calculate EI. Additionally, Japanese side directed NAMEM to collect the activity data for the training.

In September 2017, Japanese side discussed with NAMEM regarding the contents of EIM requested by Mongolian side and the necessary activity data to calculate EI and also requested NAMEM to collect the activity data. In December, preliminary version of EIM was presented to NAMEM and Japanese side confirmed the progress of collecting activity data that was requested to NAMEM in September.

Person in charge from not only the concerned agency in Ulaanbaatar city, but also from the local agency of NAMEM participated in the training held in May 2018. MET has commented this training was effective. Additionally, the Mongolian side and CAA commented on EIM, thus the activities of the next fiscal year are planned to revise based on the comments of EIM, to request NAMEM to collect activity data for 2017, and to hold training or workshop in Ulaanbaatar.

[5] "Ger" is traditional house in Mongolia.

(JFY2018)

The first draft of EIM was updated and training using the calculation tool of EI was implemented. This work's objective is to enable related agencies in Mongolian to calculate EI for the whole of Mongolia independently.

In May 2018, the training course on calculating EI was held using first draft of EIM and EI calculation tool by sources. The detail of the training and discussion is written in "Final Report for Survey of Developing Air Pollution Emission Inventory in Mongolia in fiscal 2017".

In September 2018, collection progress of data on stationary sources in the local area was confirmed to NAMEM, and NAMEM's comments for the first draft of EIM were collected.

In December 2018, the training course on the calculation of EI was implemented for the person in charge of 17 prefectures which didn't participate in the training in May. Also, the conference of revised EIM was held with the related agency in Ulaanbaatar city such as NAMEM and APRD. Based on the comments from Mongolian side at this conference, EIM was revised again.

Final seminar for EIM was held in March 2019. This seminar's contents were the summary of finalized EIM and the trial calculation result of EI in Mongolia. Ms. Bulgan, Director General of MET, commented that Mongolian side needs to implement the legislative preparations to enable to update EI continuously based on this manual. Additionally, making the operation procedure of calculation tool for Excel was agreed with Mongolian side by the end of May 2019.

4. Conclusion

The objective of this manual is to figure out the total emission of sources in Mongolia by calculating the emission amount using the source information and data in the capital city and local area. In the future, the emission factors are necessary to be updated considering the condition of Mongolia to improve the accuracy of emission amount, especially for the region with much severe air pollution.

When utilizing this manual, capacity development for implementing a measurement survey for setting emission factors and estimating emission is necessary.
Meanwhile, the manual for the developing emission inventory of some sources has been elaborated by JICA technical cooperation project in UB city, thus for more interest, please use the manual as an additional reference.

This manual is utilized in entire Mongolia. If the emission factor of a source does not exist, then guidelines, manuals, and data in Europe, US, Japan, and other countries shall be used. However, climate, environment, and social background technical basis are different from Mongolia. Thus, the appropriateness and uncertainty of data must be understood. In the future, if data of foreign countries do not represent the reality of Mongolia, measurement and field survey shall be implemented to obtain more information about the reality of Mongolia.

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technical cooperation project in UB city.

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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.

Temporal variation of atmospheric dry deposition at Kaba-aye site, Yangon in Myanmar

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Abstract

Atmospheric dry deposition of gaseous (SO_2, NH_3, HNO_3, HCl) and particulate ions (SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, Na^+, K^+, Mg^{2+}, Ca^{2+}) during 2012-2017 were investigated at Kaba-aye site, Yangon, Myanmar. The investigation of this study aimed to understand the temporal variations and behavior of atmospheric dry deposition at the Kaba-aye site. The study results observed that, the mean concentration of pollutant gaseous in a descending order were NH_3> SO_2> HNO_3>HCl. The concentration of cations in descending order were Ca^{2+}> NH_4^+> K^+> Na^+> Mg^{2+} and that of anions were SO_4^{2-} > NO_3^- > Cl^-. The dominant pollutants in the study area were observed as NH_3 and SO_4^{2-}. Temporal variation of gaseous and particulate ions species showed lowest concentration during wet season (June to October) and highest in dry season (November to May). All gaseous were decreasing trend in both dry and wet season except HNO_3, while both positive and negative trend in pollutant ions. This study found NH_3 had higher concentration than SO_2 that reflecting NH_3 emissions had a larger influence than SO_2 emissions during the study period.

Keywords: Dry deposition, Gaseous, Kaba-aye, Particulate ions, Temporal variation

1. Introduction

Environmental problem such as global warming, sea level rising, changes in precipitation patterns, increasing temperature, extreme climate, impacts on the ecosystem, and socio-economics (IPCC. 2007). Among the environmental issues, air pollution due to forest fires and its associated recurring transboundary haze pollution have become the most prominent and biggest environmental health risk particularly in the ASEAN region (UNEP. 2018). Urbanization, rapid growth of population, industrial activities and high fuel consumption in the East Asian countries have created to increase air pollution that can cause damaging effects on the regional environment (EANET. 2006).

Myanmar is the largest mainland country in Southeast Asia and rich in natural resources particularly forest, land and water resources in addition to biodiversity. Myanmar is facing environmental issues and climate change issues with growing

population, urban and industrial development. According to the hotspot information from ASEAN Specialised Meteorological Centre, increasing annual hotspot count due to forest fires and other types of fires during February to May was observed every year in Myanmar. Myanmar was the highest annual hotspot count among ASEAN countries from 2009 to 2018 (ASMC. 2019). During March, 2019, haze pollution was occurring at the boundary areas of Myanmar and Thailand. It effected to the ambient air quality worsening continuously that consequently impacting on the public health and tourism industry (ECD. 2019).

Emission from different sources into the atmosphere of large amount of pollutants, e.g. sulphur (S), nitrogen (N) oxides, and ammonia (NH_3) transformed in the atmosphere to sulphuric and nitric acid and to ammonium (NH_4) that become important components of the ionic content of atmospheric deposition (Rogora et al., 2016). An accurate estimation of atmospheric deposition (wet and dry) is needed to measure the concentration of atmospheric particles that contribute to acidification and eutrophication of ecosystems. Therefore, this study aimed to investigate temporal variations of dry deposition of gaseous and particulate ions as baseline information for future studies in Kaba-aye site, Yangon. The results will contribute basic information for future evaluation of the impacts of air pollution on environment and human health to find out appropriate measures for adaptive capacity.

2. Method

2.1 Study site

The study site is located at latitude 16°30’ N and longitude 96°07’ E, and 22 meter above MSL, Kaba-aye, Yangon city, Myanmar (Figure 1). The site was classified as urban site according to the Acid Deposition Monitoring Network in East Asia (EANET) criteria (EANET. 2013). The climate of Yangon is monsoon climate, that of average temperature range is about 21 – 33 ºC, and that of annual total precipitation is about 2783 mm (DMH. 2017). Yangon is the most populated density as well as the largest economic zone of Myanmar.

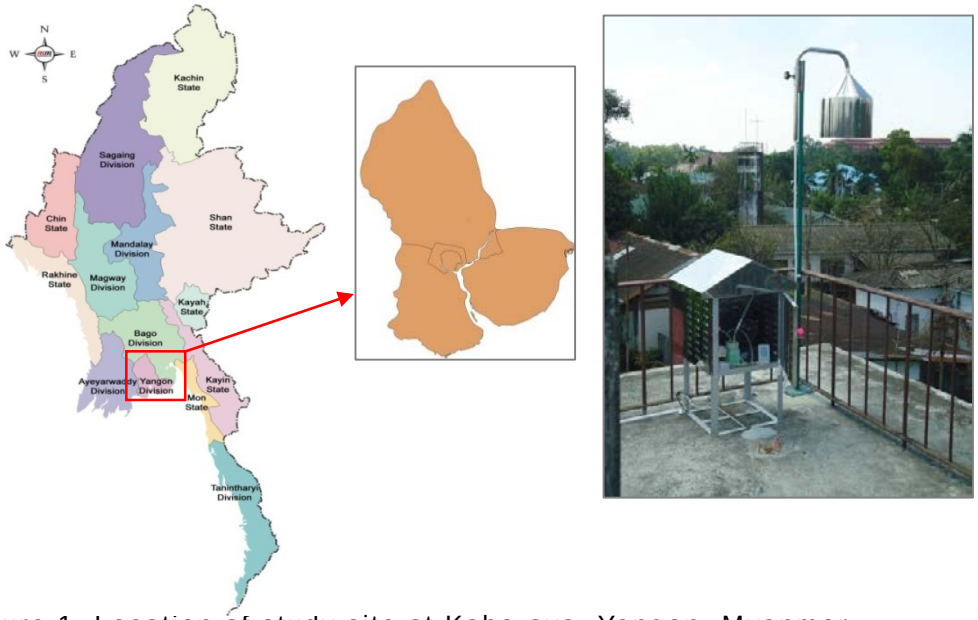


Figure 1. Location of study site at Kaba-aye, Yangon, Myanmar

2.2 Sampling data

The filter pack measurement data of air concentration of gaseous and particulate ions (SO_2, NH_3, HNO_3, HCl, SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, Na^+, K^+, Mg^{2+}, Ca^{2+}) of biweekly sampling data were collected from the annual data report of EANET during 2012 to 2017. During the study period, 12.75 % of the data was missing in all the gaseous and particulate ions.

2.3 Sampling method and analysis

A dry deposition sampling method is carried out to measure the level of the air pollution in atmosphere. A four-stage filter pack method was used to measure the main pollutants of gaseous (SO_2, NH_3, HNO_3, HCl) and particulate ions (SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, Na^+, K^+, Mg^{2+}, Ca^{2+}) which are identified as the first priority chemical species in EANET (EANET. 2013). The sampling procedure performs four stage, the first two layers are polytetrafluoro ethylene filter (PTFE), F0 and polyamide filter, F1. The other two layer filters are cellulose filter, F2 and F3, which impregnated with potassium carbonate, phosphoric acid and glycerin solution. The flow rate of air input was 1 liter/minute and sampling was operated for 14 days long, 2 times a month. F0, F1 and F3 filter papers were extracted with 20ml deionized water while F2 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, Ion chromatograph (IC S 1000) was used for determination of major air concentration of gaseous and particulate ions (EANET. 2013).

The linear trend analysis was performed for all the collected time series (2012-2017) annual mean data (for dry and wet period) of gaseous and ionic species to determine either increasing or decreasing. The magnitude of trends was calculated using Thiel-Sen nonparametric method based on Kendall's tau (τ) (Wang, 2008). The trend was considered to be statistically significant when significance level $p \leq 0.05$ using Mann–Kendall test (Sen, 1968; Mann, 1945). All analysis was conducted using the R-programming platform.

3. Results and discussion

3.1 Monthly variations of dry deposition

Figure 2 describes the monthly variation of all the species. Table 1 shows monthly mean concentration of gaseous (SO_2, NH_3, HNO_3, HCl) and particulate ions (SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, Na^+, K^+, Mg^{2+}, Ca^{2+}). The results showed that the higher concentration of all species observed during November-May (dry period) than the rest of the months June-October (wet period).

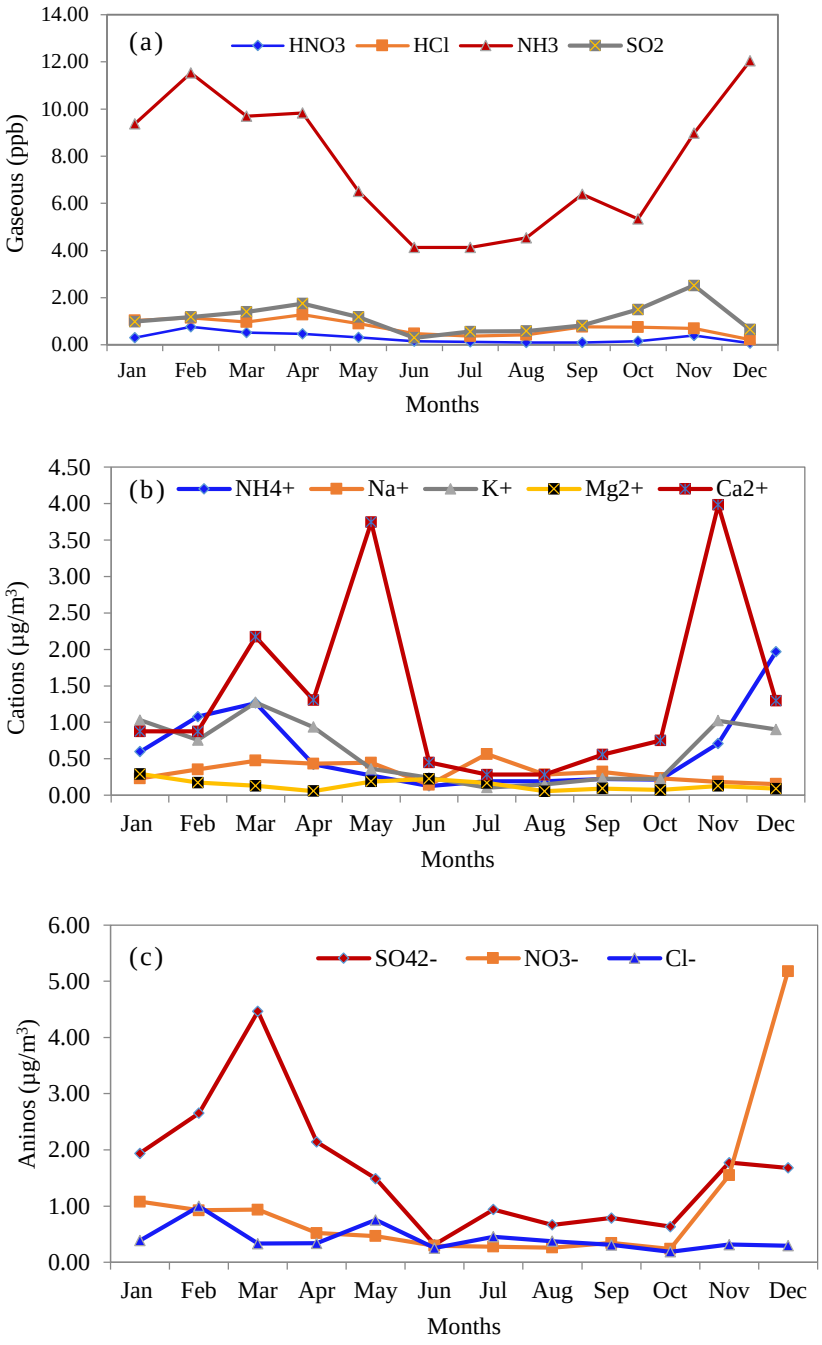


Figure 2. Monthly variations of the species (a) gaseous, (b) cations and (c) anions at Kaba-aye

Among the gaseous species, NH_3 was significant highest concentration particularly in February/December (Figure 2.a). Similarly, SO_4^{2-} and Ca^{2+} were highest concentration in particulate ions especially in May/November in Ca^{2+} and March in SO_4^{2-} (Figure 2.b, c). The concentrations in dry period were approximately 2 times higher than that of wet period.

Table 1. Monthly mean concentration of gaseous and particulate ions during 2012-2017

Species	Jan	Feb	Mar	Apr	May	Jun	July	Aug	Sep	Oct	Nov	Dec	Avg
SO_2 (ppb)	0.98	1.18	1.40	1.75	1.18	0.30	0.57	0.58	0.83	1.50	2.52	0.65	1.12
NH_3 (ppb)	9.37	11.53	9.70	9.83	6.50	4.13	4.13	4.54	6.38	5.34	8.98	12.05	7.71
HNO_3 (ppb)	0.30	0.76	0.52	0.46	0.32	0.16	0.13	0.10	0.10	0.15	0.40	0.07	0.29
HCl (ppb)	1.04	1.15	0.97	1.28	0.90	0.48	0.37	0.42	0.77	0.75	0.70	0.23	0.75
NH_4^+ ($\mu g/m^3$)	0.60	1.08	1.27	0.42	0.27	0.13	0.19	0.9	0.22	0.21	0.71	1.97	0.61
Na^+ ($\mu g/m^3$)	0.23	0.35	0.48	0.43	0.45	0.14	0.56	0.28	0.32	0.23	0.19	0.15	0.32
K^+ ($\mu g/m^3$)	1.03	0.76	1.27	0.93	0.36	0.24	0.11	0.15	0.23	0.22	1.02	0.90	0.60
Mg^{2+} ($\mu g/m^3$)	0.29	0.17	0.13	0.06	0.19	0.23	0.17	0.06	0.09	0.07	0.13	0.09	0.14
Ca^{2+} ($\mu g/m^3$)	0.88	0.88	2.17	1.31	3.74	0.45	0.28	0.28	0.56	0.75	3.98	1.30	1.38
SO_4^{2-} ($\mu g/m^3$)	1.94	2.65	4.47	2.14	1.49	0.32	0.94	0.67	0.79	0.63	1.77	1.68	1.62
NO_3^- ($\mu g/m^3$)	1.08	0.93	0.94	0.52	0.47	0.30	0.28	0.26	0.34	0.24	1.55	5.18	1.01
Cl^- ($\mu g/m^3$)	0.39	1.00	0.33	0.34	0.75	0.25	0.46	0.38	0.31	0.19	0.32	0.30	0.42

3.2 Annual concentration of dry deposition

The annual concentration of gaseous and particulate ions described in Figure 3. NH_3 has highest concentration in all years, followed by SO_2, HNO_3 and HCl. On the other hand, SO_4^{2-} has highest concentration followed by Ca^{2+}, NO_3^-, NH_4^+, K^+, Cl^-, Na^+ and Mg^{2+} during study period. Most of the species tend to decrease in all years although SO_4^{2-}, NO_3^-, and NH_4^+ likely to increase during 2017 (Figure 3. b).

The results prove that higher NH_3 emission reflect higher concentration of NO_3^- as NH_3 reacts with first H_2SO_4 then with HNO_3 and became SO_4^{2-} and NO_3^- in the air (Luo et al., 2016). This study shows that the higher SO_4^{2-} and NO_3^- which could be related with mobile sources for the particulates formation in study site (Luo et al., 2016). The higher deposition of SO_4^{2-}, NO_3^- and NH_4^+ at study site could be contributed potential acidification and eutrophication of ecosystems. Moreover, the higher concentration of SO_2 could be impact on buildings and monuments as well as deposition of base cations could be important for nutrient cycling in soils and ecosystems in the study area (Kumar et al., 2003).

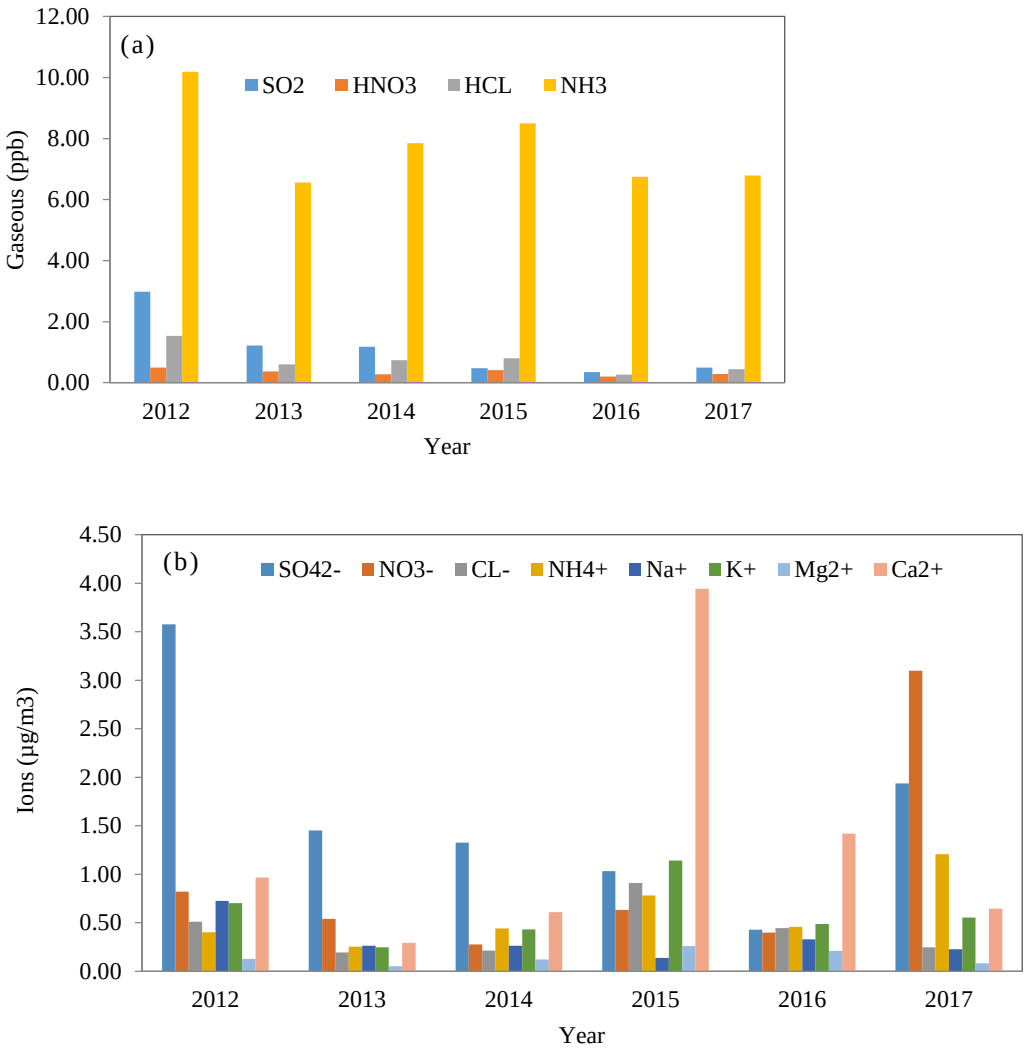


Figure 3. Annual concentration of dry deposition (a) gaseous and (b) particulate ions

Table 2 describes the 6-year averages of the concentration of dry deposition of gaseous (ppb) and particulate ions (µg/m^3) during the period 2012 to 2017. The highest concentration of gaseous in order of NH_3> SO_2> HNO_3>HCl while for the cations Ca^{2+} was the most abundant and Mg^{2+} the least abundant cations species. Consequently, the anions SO_4^{2-} was the most and Cl^- was the least abundant anions species. Other cations and anions were in the order of Ca^{2+}> NH_4^+> K^+> Na^+> Mg^{2+} and SO_4^{2-} > NO_3^- > Cl^- respectively.

Table 2. Average dry deposition of gaseous and particulate ions during 2012-2017

Species	Mean	Standard Deviation
SO_2(ppb)	1.12	± 0.99
HNO_3(ppb)	0.34	± 0.11
HCl(ppb)	0.73	± 0.44
NH_3(ppb)	7.77	± 1.41

NH_4^+ (µg/m^3)	0.59	± 0.35
Na^+(µg/m^3)	0.32	± 0.21
K^+(µg/m^3)	0.59	± 0.31
Mg^{2+}(µg/m^3)	0.14	± 0.08
Ca^{2+}(µg/m^3)	1.31	± 1.34
SO_4^{2-}(µg/m^3)	1.63	± 1.08
NO_3^-(µg/m^3)	0.96	± 1.06
Cl^-(µg/m^3)	0.42	± 0.27

3.3 Trend analysis

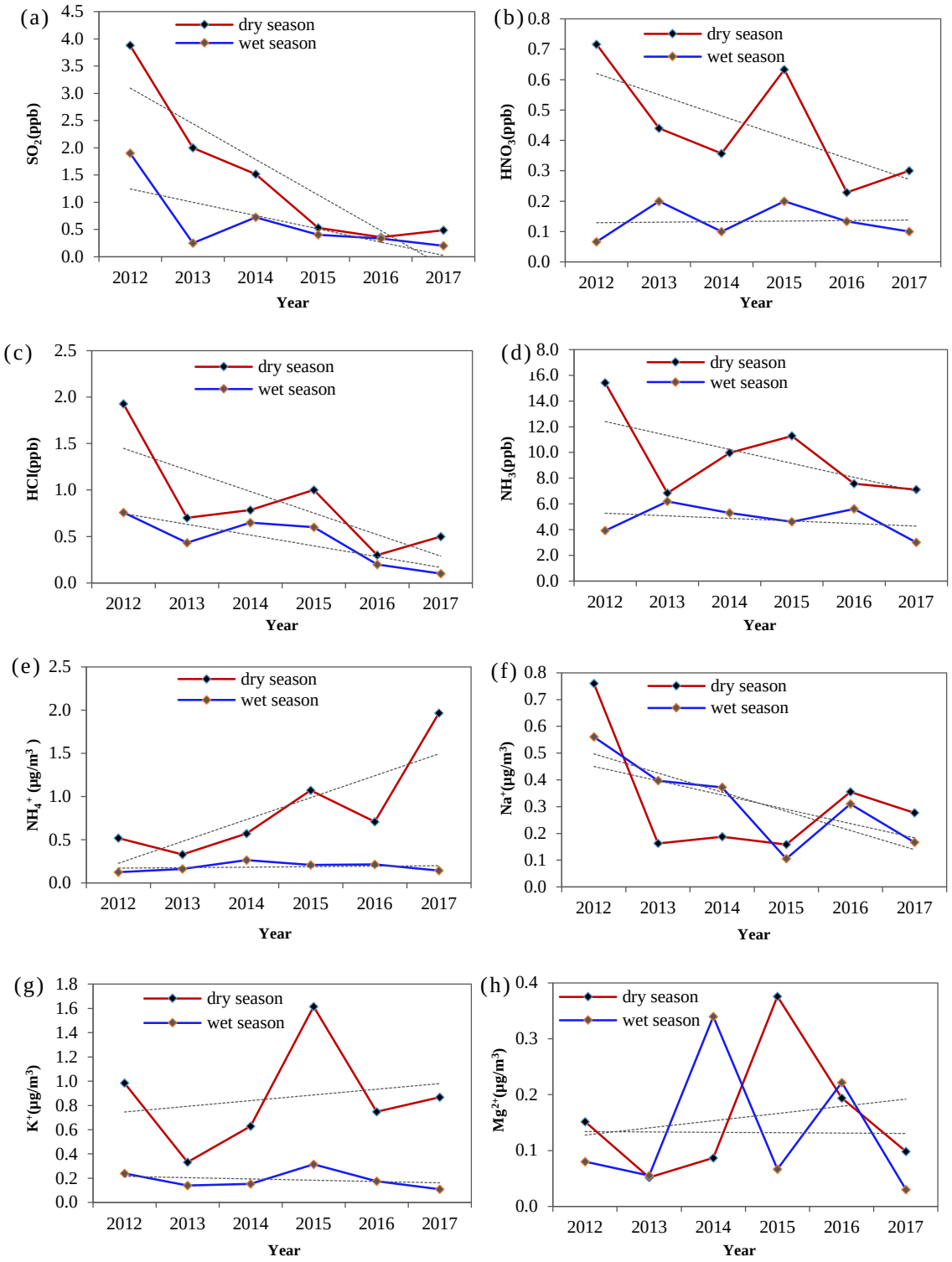
The simple linear regression model was used to identify trends of all gaseous and ions for dry season (November to May) and wet season (June to October). Statistically significance of temporal trends was assessed when $p \leq 0.05$ (95% confidence level) using Mann–Kendall test. Trends of yearly mean concentrations of all species at Kaba-aye site are shown in Figure 4. The magnitude of slope and *p* value of all the species are shown in Table 3.

The concentration of gaseous and particulate ions exhibited similar variations, with higher values observed in dry season than in wet season (Figure 4). The highest concentrations were identified as the year 2012 and 2015 during study period (2012-2017) in most of the gaseous and particulate ions.

All the gaseous (SO_2, HNO_3, HCl, NH_3) showed decreasing trend in both dry and wet season, except HNO_3 which showed increasing trend in wet season. Among them, SO_2 and HCl observed significant decreased (p=0.013 and p=0.043) in dry and wet season, respectively.

Ca^{2+}, K^+, NH_4^+ and Mg^{2+} showed insignificant increasing trend in dry season, while Na^+ showed decreasing trend. Trend of cations in wet season observed opposite of dry season with significant in Na^+, except NH_4^+. Among the cations, NH_4^+ is significant increasing trend in dry season (p=0.05) and insignificant in wet season. Increasing in dry and decreasing trend in wet season of both NO_3^-, Cl^- were observed. SO_4^{2-} is decreased in both dry and wet season.

In general, the decline of gaseous and particulate ions in study area over the 6-year period (2012-2017), could be favor to minimize further harm from acid rain and to promote ecosystem (American & America, 2016). However, dry deposition is a complex process which influenced by the chemical properties of aerosols and their sources, weather conditions, and surface characteristic features (Khan & Perlinger, 2017). Therefore, more research on dry deposition related with weather phenomena and their impacts on the environment and human health should be carried out to support decision makers for implementation of laws and policy to reduce emissions.


(a)
dry season
wet season
SO2(ppb)
Year
(b)
HNO3(ppb)
(c)
HCl(ppb)
(d)
NH3(ppb)
(e)
NH4+ (μg/m3)
(f)
Na+(μg/m3)
(g)
K+(μg/m3)
(h)
Mg2+(μg/m3)
2012
2013
2014
2015
2016
2017

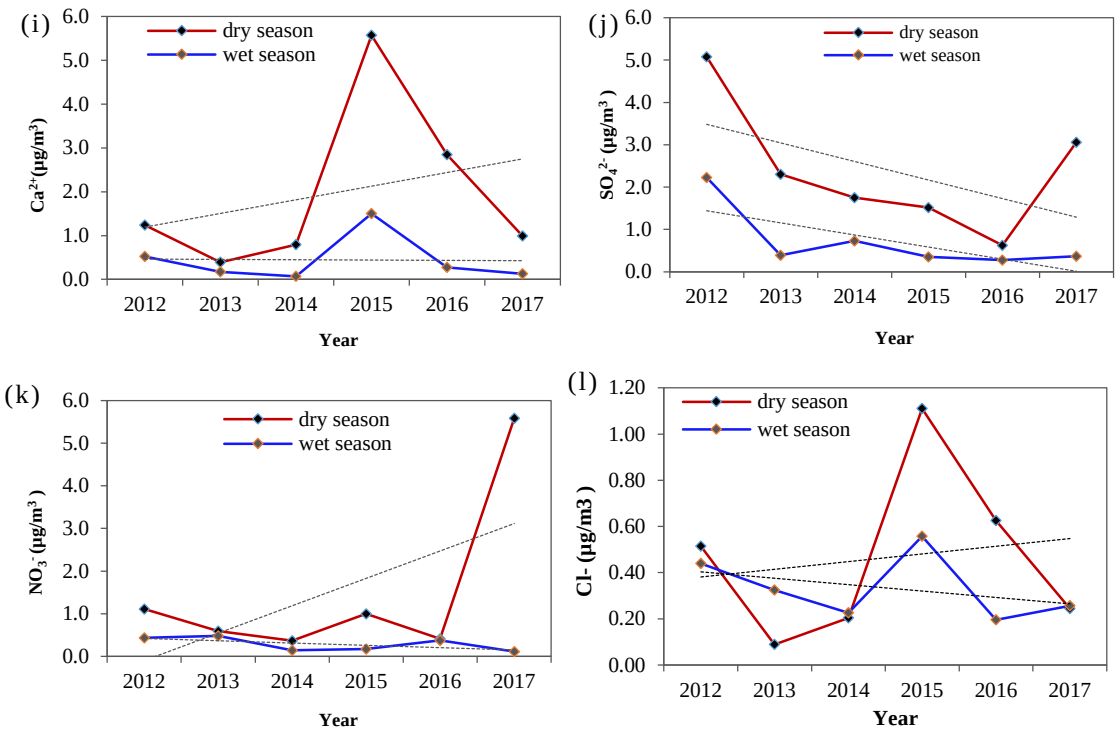


Figure 4. Trends of gaseous and ions for dry (November to May) and wet (June to October) season at Kaba-aye site

Table 3. Temporal trends of dry deposition of gaseous and particulate ions (2012-2017)

Species	Dry Season (November-May)			Wet Season (June-October)		
	Slope estimate	Slope error	*p* value	Slope estimate	Slope error	*p* value
SO_2(ppb)	-0.654	0.156	0.013	-0.245	0.123	0.115
HNO_3(ppb)	-0.070	0037	0.133	0.001	0.015	0.941
HCl(ppb)	-0.233	0.099	0.079	-0.115	0.039	0.043
NH_3(ppb)	-1.088	0.697	0.193	-0.203	0.297	0.532
NH_4^+ (µg/m^3)	0.254	0.096	0.058	0.004	0.013	0.762
Na^+(µg/m^3)	-0.523	0.056	0.401	-0.071	0.025	0.049
K^+(µg/m^3)	0.047	0.113	0.698	-0.010	0.020	0.635
Mg^{2+}(µg/m^3)	0.013	0.031	0.693	-0.001	0.032	0.973
Ca^{2+}(µg/m^3)	0.312	0.499	0.566	-0.006	0.144	0.967
SO_4^{2-}(µg/m^3)	-0.438	0.350	0.279	-0.286	0.142	0.114
NO_3^-(µg/m^3)	0.643	0.434	0.213	-0.054	0.033	0.170
CL^-(µg/m^3)	0.034	0.099	0.751	-0.027	0.035	0.471

4. Conclusion

This study investigates the behavior of temporal variations of atmospheric dry deposition during 2012 to 2017 at Kaba-aye site, Yangon in Myanmar. The key findings can be summarized as follows:

- Highest amount of gaseous and particulate ions was observed as NH_3, NH_4^+, and SO_4^{2-}.

- All the species showed highest concentration in 2012 and 2015, while the lowest concentration in 2013 and 2016.
- NH_3 and SO_4^{2-} were identified as dominant pollutants in the study area.
- All the species observed lowest concentration during wet season and highest in dry season.
- Decline of gaseous and particulate ions in study area over the 6-year period. Assuming that declination of gaseous and particulate ions could help to mitigate acid rain and better ecosystem in the study area.

Acknowledgements

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Airborne ammonia concentrations in areas of two EANET stations in Baikal region derived from EANET monitoring and satellite measurements

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Abstract

Satellite data allows to assess spatial distribution of air pollutants and to evaluate regions with higher and lower concentrations. Total amount of gaseous ammonia retrieved from satellite Infrared Atmospheric Sounding Interferometer (IASI) measurements was recalculated with help of modelling vertical profile to obtain its surface concentrations. The results were compared with EANET monitoring data for proper use and interpretation of remote sensing data. We found a good agreement between mean levels of two mentioned types of measurement results for both Listvyanka and Mondy station areas. NH3 pollution levels at Listvyanka are closer to the concentration values retrieved from remote sensing data above water more than above land. Mean seasonal ammonia concentrations from IASI measurements were calculated for the area surrounding the stations for the years from 2015 to 2017. It was recognized that mean summer and autumn concentrations of ammonia are very high at several places and reach up to 14 ppb. The locations of such regions coincide with places of forest fires happened during the years of interest. However mean concentrations at both EANET stations did not exceed 5 ppb and were at the same level of air pollution as for majority of considered region.

Keywords: Ammonia, Remote Sensing, Baikal, Filter Pack measurements

1. Introduction

Ammonia is a widely rife air pollutant and an important compound to form acid deposition (Galloway, 1995). The dry deposition of NH3 itself has also a negative effect on ecosystems for eutrophication and loss of biodiversity (Sheppard, 2011). It takes part in PM2.5 formation together with other compounds (Malm, 2004) that provides a proved negative impact on human health far from emission areas (WHO, 2013). The main sources of atmospheric ammonia are animal husbandry, nitrogen fertilizers and biomass

burning. Some papers provided the emission evaluation of the ammonia for the wide regions of Eurasia (Ryaboshapko, 2001).

Ammonia monitoring is performed by many international air quality monitoring networks such as EMEP (Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe), regional GAW-WMO (World Meteorological Organization) network (former BAPMoN), and others. The continuous monitoring of ammonia at EANET stations in Russia is carried out since 2000.

Ground-based measurements are performed by continuously sampling through the year and considered as precise and reliable. An application of additional type of observations such as satellite data may be used to get the more complete and detail picture of air pollution in the wider regions.

In this paper we assessed mean seasonal concentrations in Baikal region of Russia with satellite and ground-based measurements made at EANET stations. The standard deviations of values were calculated together with mean levels to understand whether the higher concentrations were constant or episodic.

2. Data and methods

Listvyanka and Mondy are rural and remote EANET monitoring stations respectively at Baikal region in Russia. Listvyanka (51° 50' 47"N, 104° 53' 34"E) station is located directly at Baikal shore, in the vicinity of the small settlement. Mondy (51° 37' 18"N, 100° 55' 10"E) is a high altitude remote station in mountains (around 2000 m) located at the Russian border with Mongolia (Figure 1).

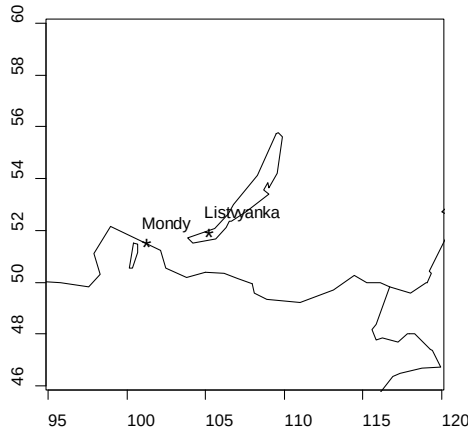


Figure 1. Location of Mondy and Listvyanka stations

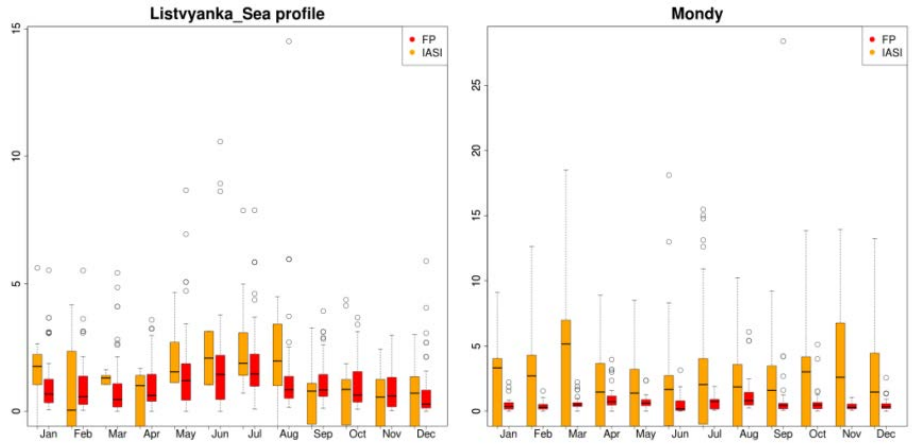
Ammonia measurements at Listvyanka and Mondy are performed by filterpack method. Filters are exposed for two weeks at Listvyanka and up to for four weeks at Mondy station.

Total column ammonia amount was retrieved from IASI (Infrared Atmospheric Sounding Interferometer) measurements, this instrument was onboard MetOp-A satellite (Clarisse et al, 2009). The satellite has a sun synchronous orbit, therefore global daytime and nighttime measurement data is available. Daytime measurements were used only in this paper because of their higher level of confidence. The calculated footprint

of the instrument is the circle area with 12 km diameter in nadir position. Two types of a priori profiles were used for the ammonia retrieval (Van Damme et al, 2017). The first supposes the maximum ammonia concentration near the surface (“land”) while the second is “sea” surface approach where maximum is at some height above the surface level. The total column amount of NH3 and the type of its profile used for the retrieval is proved together and that profile type was used further to recalculate total column value to surface concentrations. In spite of this recalculation may give an additional mistake, it makes possible to compare the remote sensing data with ground-based measurements and to assess the pollution level near the surface from the satellite data.

Ground-based measurements from EANET stations were compared with concentrations retrieved and recalculated from IASI measurements for years 2015 to 2017. For the comparison we distinguished the satellite measurements whose center gets into the square 0.5x0.5 degree around the station. Since Listvyanka is located at the shore of the lake, the data retrieved with both types of the profiles were included into the square around the station. Using all of those data gives not good coincidence of remote sensing and in situ measurements. However, by excluding “land” profile data, e.g. data over the land, we found that considering the data over the water body only leads to much better agreement (Figure 2a). Seasonal changes with maximum at the summer months are evident from filter pack and IASI data. The difference may be caused by uncertainties in remote sensing data as well as by unevenness in measurement period also. The measurements at EANET stations are performed by sampling for two or four weeks (filter exposing) and therefore mean concentration is obtained for period of several weeks. In opposite satellite measurements are instant and done once a day approximately at 10 a.m. Moreover, under some conditions an error of measurements is too large, and in this case retrieved data are not considered.

For Mondy station the remote sensing data is higher than from ground measurements (Figure 2b). It corresponds to the conclusion for other regions that over areas where ammonia amount is low, satellite data may overestimate real values (Whitburn et al, 2016). Also the retrieval results depend on the temperature contrast between surface and near-ground air that is low in this area for winter months. This circumstance explains an additional error and bias of results. So the concentrations calculated from satellite measurements above Mondy station are demonstrated to be higher than measured values near the surface.



a) b)

Figure 2. Comparison of ammonia concentration measured from EANET filter pack (FP) data (red boxes) and the retrieved values from IASI (orange boxes) for months of 2015-2017 period for Listvyanka (a) and Mondy (b). The circles show the highest values (outliers)

Performed comparison of ground based and remote sensing measurements provided the information how to make better interpretation of satellite data. Because of evident errors appeared for winter months due to the temperature contrast, next we consider to evaluate spring, summer and fall seasons only.

Measurements at EANET stations are more sensible to small variations of ammonia abundance. Since Mondy is a remote station, the concentrations there are lower and the seasonal changes are not so evident according to filterpack measurements. Due to very low ammonia amount at Mondy, the satellite data occur to be slightly overestimated.

The wide region from 95o to 120o East and from 51o to 65o North was chosen for the assessment of seasonal mean levels of ammonia concentrations obtained from IASI data. The single cell is chosen as the square with one degree side. Together with mean seasonal concentrations for years 2015 to 2017 the standard deviations were calculated. Low standard deviation in the region means that the observed levels of concentrations, being of low or high values, are stable during the season. In case of high standard deviation, high levels of ammonia amount registered in atmosphere are not caused by permanent emission sources, but by sporadic effects or cases of heavy pollutant events such as forest fires or fertilizer applications.

3. Results and discussion

The results of seasonal concentrations assessment from satellite measurements are presented at Figure 3. Left column is correspondent to mean values of ammonia abundance measured from the satellite at 10:00 a.m. every day for each season (3 months) from 2015 to 2017 while right one presents the standard deviation of the same sets of values.

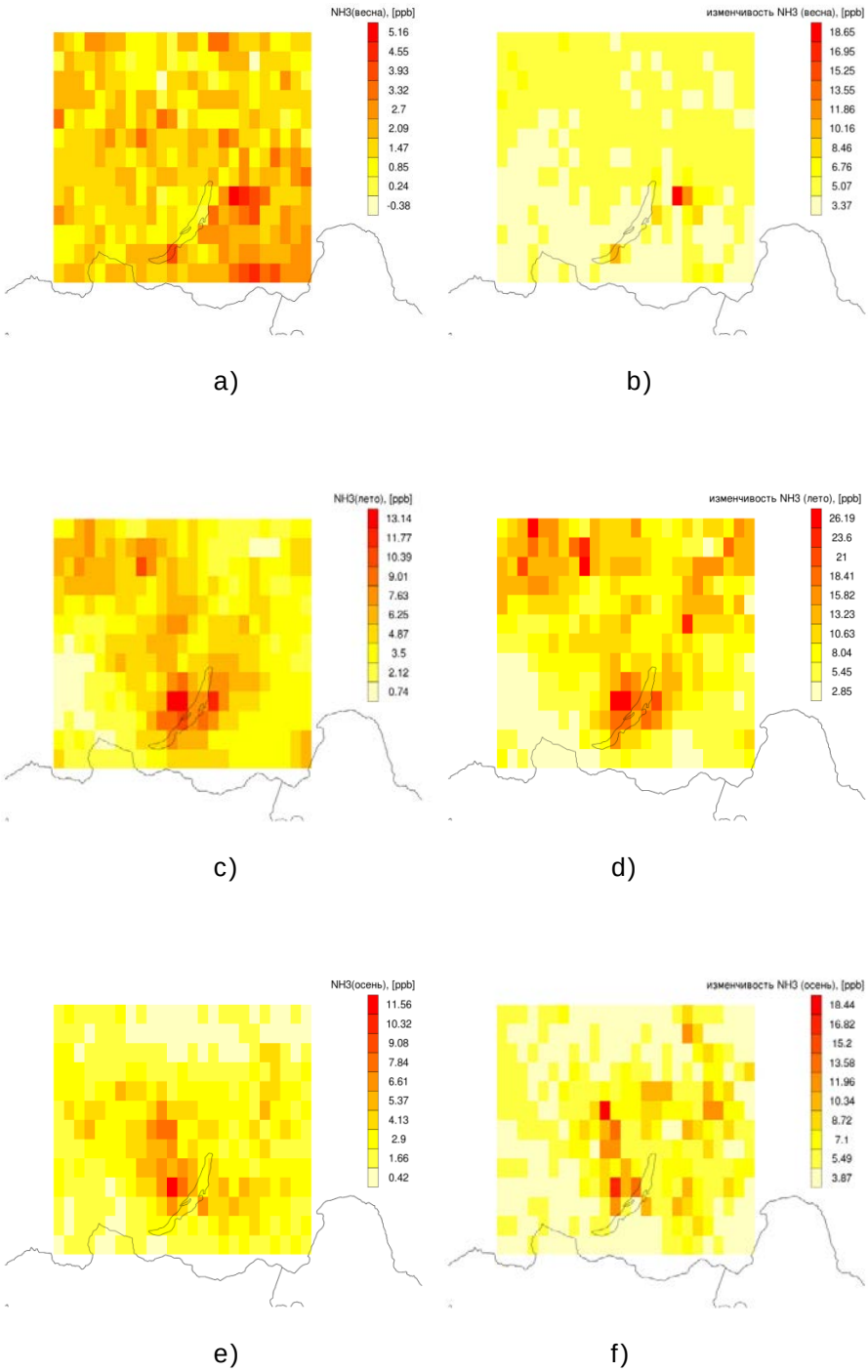


Figure 3. Mean concentrations of ammonia (left) recalculated from IASI measurements and standard deviation (right) for spring (a, b), summer (c, d) and fall (e, f)

For the spring season (March, April, May) the spatial distribution of the ammonia level over the region is homogeneous, mean concentrations are approximately the same and are not high. For some areas such as Mondy surrounds the real concentrations may even be lower than retrieved ones. However, during summer (June, July, August) and

fall (September, October, November) the areas with enormously higher NH3 amount are evidently recognized: some individual recalculated values may exceed 100 ppb. The standard deviation is also high for exactly same areas.

As for land use there are not many agricultural territories located in the region. The higher values of NH3 concentrations are registered partially even at national parks and may reach up to 14 ppb. One of the reasons of recognized high values might be forest fires, which occur in these regions often every year. Total column amount of ammonia including smoke from forest fires may exceed 1018 Dobson units retrieved from satellite measurements near Baikal (Trifonova-Yakovleva, Gromov, 2018), that corresponds to concentration of about 400 ppb near the surface.

However, it can be clearly seen from the pictures that EANET stations Listvyanka and Mondy are located in relatively clean regions. Ammonia concentrations at both of them do not exceed 5 ppb. Since the standard deviation of the recalculated IASI measurements of both sites showed small values, mean ammonia amount at the stations is not directly influenced by sporadic high concentrations events. The concentrations on the sites are of the regional level, especially taking into account that some overestimating of retrieved values could be above clean territory.

4. Conclusion

Remote sensing data provides information about spatial distribution of the airborne species and allows to distinguish the areas of similar pollutant levels. At Baikal region there are some areas located mainly around the lake where mean levels of ammonia concentration are very high. One of possible reason for higher contamination might be ammonia emission during the forest fires. The plumes are transported air pollutants over the large territory. However, the areas with highest concentration of ammonia do not include the places of Listvyanka and Mondy EANET stations. The concentrations observed at these sites are inherent to the territories not directly influenced by forest fires.

Acknowledgements

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Technical note on more precise approach to calculate the water discharge of small rain-feeding river at the temperate region of Far East

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Abstract

This study examines the possibility of estimating the surface water discharge of small stream through continuous measuring the river water level at non-artificial cross section site. A comparison was made of the variations or changes in water discharge and water level with the use of data obtained from daily measurements for the Komarovka River at EANET site in Russian Far East. The results proved that, in general, the dependence between the parameters is non-linear while any relationship for the cold period cannot be detected. Furthermore, it was found that for the warm period the dependence parameters shifted (varied) from year to year, which requires additional investigation both to analyze the dynamics of the transverse river profile and the role of other possible factors.

Keywords: Water discharge, Watershed, Chemical runoff, Small catchment

1. Introduction

The investigation of the river water regime is necessary to understand the balance of nutrients and dissolved pollutants in the catchment and to evaluate changes of its components. Daily data on surface water discharge being continuously recorded are often very important in assessing the removal of investigated chemicals from a river catchment because of it could be a main discharge part of watershed heavily dependent on rainwater supply. Such measurement data are difficult to be obtained for rivers at remote natural territories because they may be absent or inaccessible for a large number of small rivers outside towns.

If an absence of detailed data on water discharge is found in the end point section of a small catchment with natural water supply, some indirect methods can be applied with sufficient accuracy to obtain such necessary information. In order to quantify the hydrograph regime (time dependent water discharge) of Komarovka river at the western

slope of the Sikhote-Alin ridge (Far East) we used the solution by the way of identifying the dependence of stream flow rate on the daily water level.

2. Object of research

The Komarovka River is a small surface water stream with mainly rainwater feeding flowing in the Primorsky Kray in the vicinity of Ussuriysk city (Russian Far East). The beginning point of the river is located on the western spurs of the Sikhote-Alin ridge at the altitude of 380 m above sea level. Komarovka is a left tributary of the Razdolnaya River which belongs to the general drainage basin of the Sea of Japan.

There is a number of factors to make a choice to use Komarovka River as an object for research. The selected upper section of the river is a closed small catchment located in the natural forest zone. This area has been presumably stable in a quasi-natural state for more than half a century and located within the boundaries of the Ussuriysky Reserve protection zone with a strong restriction of land use and human activity.

Since the 1950s, research hydrological measurements had been conducted in the lower part of the catchment on a regular basis for the studies on river flood forecasting in Primorye, and long-term data on some hydrological parameters have been obtained and used. The EANET Primorskaya station was established at the non-artificial hydrological post Tsentralny on the Komarovka River, and since 2003 the observations on the chemical composition of atmospheric precipitation and surface water have begun within the framework of the EANET international program (Khodzher et al., 2011). Based on the data obtained, a qualitative and quantitative analysis of hydrological regimes can be carried out with the aim of further using information to assess the components of pollutant balance in the catchment.

3. Materials and methods

We used the available data on the discharge and surface water level of the Komarovka river at the hydrological post “Central” of the Primorsky UGMS for several reporting years (2001, 2005, 2010 and 2015) to assess their dependence. The stream observations were carried out in accordance with the regular measurement program of the 1st category (G-1) hydrological station, these results were published after statistical processing in the issues of the hydrological Yearbooks (SHI. 2003). To assess the data quality the graph-analysis method with smoothed curve application was used, and mentioned dependencies were constructed using standardized data processing tools of MS Excel spreadsheets.

4. Results and discussion

The graph sets were constructed on the dependence of daily water discharge (of catchment river outflow) on the correspondent recorded level of surface waters at the natural range transverse profile of Komarovka river using the available data rows (Figure 1). Analysis of the graphs showed that two characteristic regimes of dependence can be distinguished for each year:

- the first is corresponding to the cold season, with small flow rates close to zero; it may be associated with ice core formation and winter low-water river regime; as a result, an evident dependence was not found for the cold season due to the small runoff values and the almost complete absence of water supply by precipitation;

- the second mode, corresponding to the warm season, is characterized by significantly higher values of water flow; there are found also higher water level values during relatively fast increasing a runoff in the short periods of rain falls and floods. The changing ratio of investigated parameter values during these periods is described by graphs of a parabolic curve.

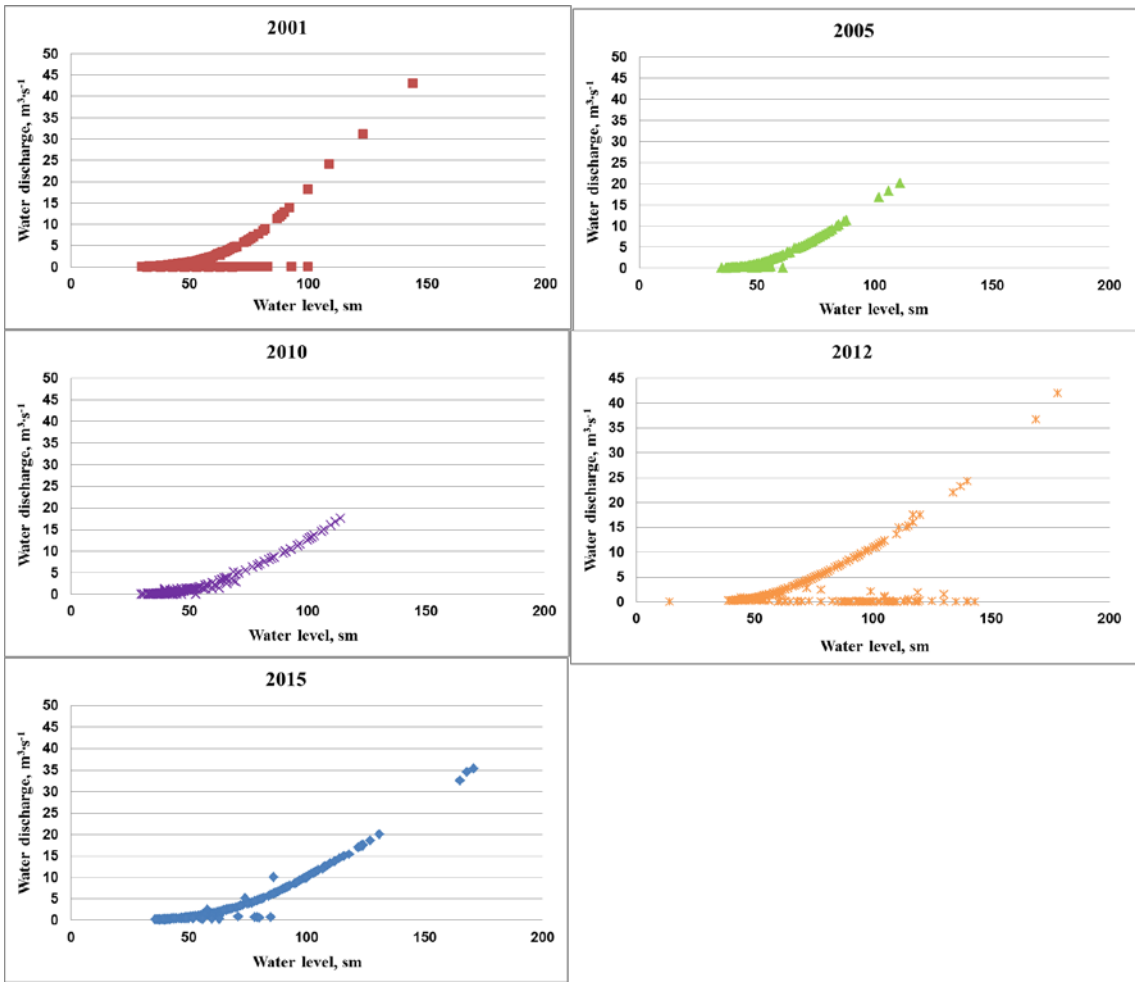

Figure 1. The dependence of the daily outflow on the water level of the river for some years

The estimates of the hydrograph curve of Komarovka River for 2015 are presented at Figure 2. It can be noted that the period of low daily water flows ("cold period") extends from the beginning of the calendar year to the beginning of April, and continues from mid-November to the end of the year. It should be evidently correspond to the establishing snow cover on the catchment area and freezing rivers from surface to bottom. In the warm period the significant fluctuations of flow rates were observed depending on the variation of hydrological mode (rainy or snow melting days *vs* smooth sunny weather).

The results of calculations of monthly average discharges and water levels are presented in Table 1. The highest values of surface water discharge were observed in April-May and for July-August, which are correlated with the higher values of water level during the warm period.

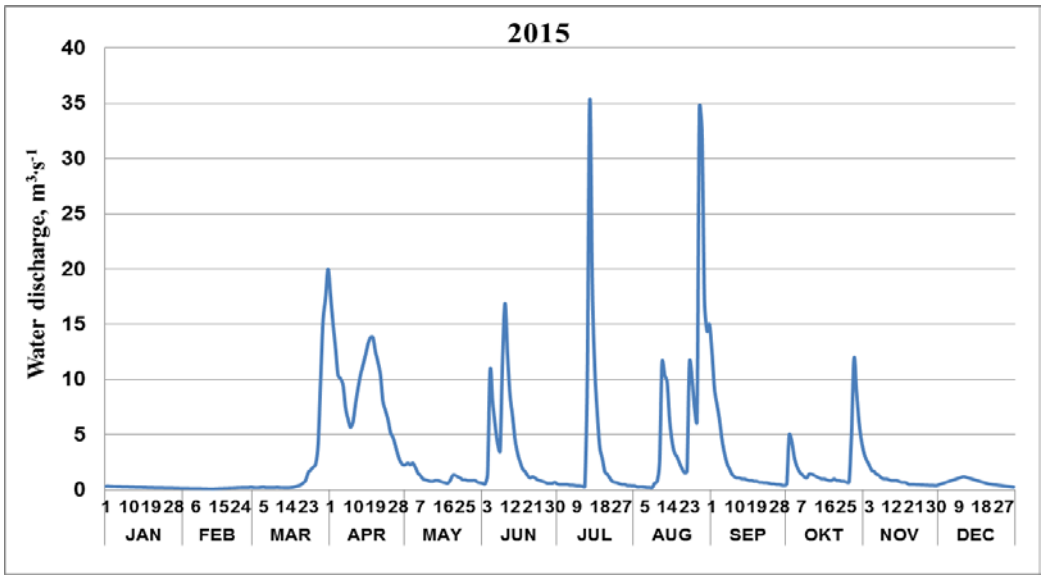

Figure 2. Annual hydrograph of water discharge for 2015

Table 1. Monthly average values of discharge flow rate (Q_w, m^3s^{-1}) and level (H_L, cm) of surface waters for 2001 - 2015

Water Parameter	I	II	III	IV	V	VI	VII	VIII	IX	X	XI	XII
2001 discharge (Q_w)	0.089	0.058	0.13	4.87	2.73	0.93	2.80	6.05	0.36	0.53	0.32	0.035
2001 level (H_L)	38	74	50	67	59	46	56	65	39	42	38	43
2005 discharge (Q_w)	0.263	0.147	0.201	5.89	7.74	0.89	3.19	3.07	0.87	0.68	1.10	0.23
2005 level (H_L)	41	37	39	70	77	49	60	60	47	47	50	44
2010 discharge (Q_w)	0.038	0.019	0.055	6.22	6.05	0.32	0.61	1.91	0.30	0.63	0.56	0.94
2010 level (H_L)	35	41	34	73	74	40	44	54	40	45	43	45
2012 discharge (Q_w)	0.06	0.018	0.017	3.29	2.15	0.48	0.38	4.41	8.83	9.59	3.41	0.38
2012 level (H_L)	80	103	97	103	60	45	44	68	87	93	68	51
2015 discharge (Q_w)	0.24	0.15	2.60	9.03	1.15	3.86	3.32	6.83	2.18	2.38	0.98	0.70
2015 level (H_L)	40	38	55	95	55	69	60	77	59	62	53	59

To calculate the values of average annual “surface runoff module” from the square of watershed, we used the catchment area upstream the observation point of 155 km^2 estimated by GIS tool from the maps in Internet resources (Table 2). This parameter approximates the values of surface water within watershed to be equal the precipitation amount if whole water discharge was originated from atmospheric fall-out.

Table 2. Average annual values of surface water discharge (m^3s^{-1}) and surface runoff modulus ($L \cdot s^{-1}$ km^{-2}) for 2001 - 2015

Year	Water discharge	Surface runoff modulus
2001	1.59	10.24
2005	2.04	13.14
2010	1.48	9.54
2012	2.75	17.76
2015	2.79	18.02

By examination of the result of regression analysis of the data, we concluded that a flow rate is evidently dependent on the individual transverse profile of the river (width of stream, bottom slopes, etc.). The dynamics of the inter-annual variability of the dependence of river discharge on water level for the warm seasons of different years were presented in Figure 3: there is a shift of the dependence curves towards a decrease in water discharge for the same level values from 2001 to 2015. This phenomenon is most likely associated with a change in the river transverse profile which should be investigated in more detail. We cannot also rule out other causes that could probably be identified in further studies.

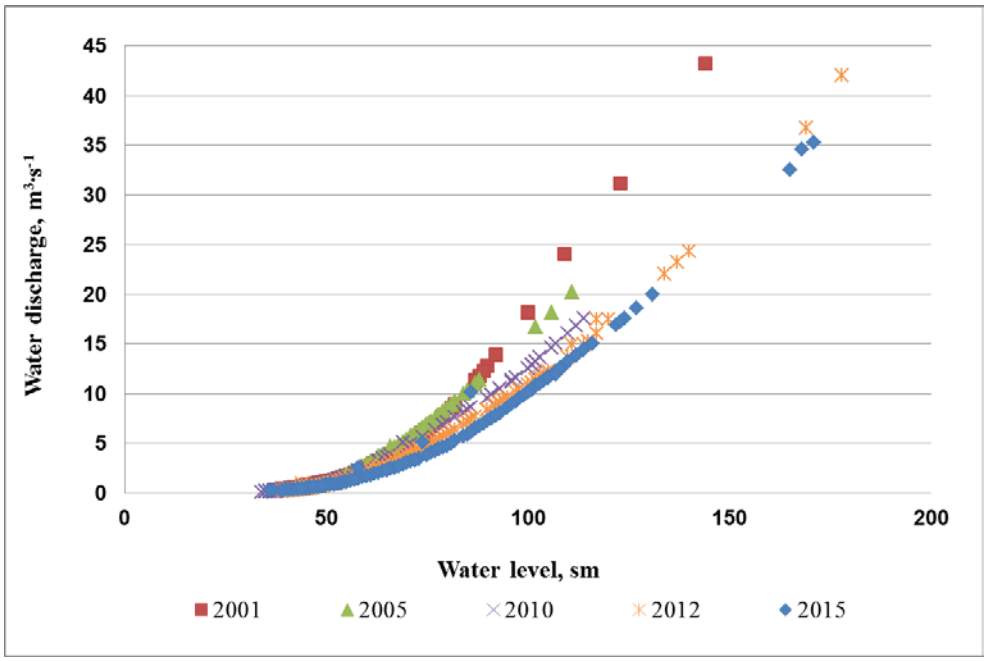

Figure 3. Dependence of river discharge on water level in the warm season for some years

5. Conclusion

A regression analysis of two hydrological parameters measured on Komarovka river has been applied to fill gaps of knowledge on water discharge variation between the dates of stream speed measurements and combined surface water sampling. The reconstructed hydrographs allow to estimate chemical runoff from natural catchment in more details as well as to take inter-annual fluctuations of river water feeding into

account. However, for the cold period, the dependence of the discharge flow rate on the water level cannot be precisely detected due to the low values of the water flow rate in freezing period of the year.

Before using the method of calculating the dependence of the discharge rate on surface water level for estimations, it is necessary to conduct additional investigation of the transverse profile of the river and its deformation. Also, other reasons of the change in dependency cannot be ruled out and they still need to be identified.

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Technical Note

QA/QC activities and Data Management in the Acid Deposition Monitoring Network in East Asia (EANET)

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Abstract

The roles of Network Center (NC) for Acid Deposition Monitoring Network in East Asia (EANET) include data management such as compile, evaluate, analyze and store the EANET monitoring data and related information, disseminate monitoring data and related information, implementation and coordinate quality assurance/quality control (QA/QC) activities in the EANET participating countries. This article presents some technical information on the internal and external QA/QC activities and data management in EANET. As for the internal QA/QC activities, the NC encouraged to prepare Standard Operating Procedures (SOPs) for each analysis laboratory in EANET. As for the external QA/QC activities, the NC have conducted the Inter-laboratory Comparison (ILC) project more than 20 years, and key factors that may cause large uncertainty and outlier for ionic component analysis were identified. As for data management, the new EANET data archive page and data request registration and download system were established in April 2019. It is important to disseminate EANET data to more data users so that many people will use EANET data for research, policy analysis, etc.

Keywords: Internal QA/QC, Standard Operating Procedures, External QA/QC, Inter-laboratory Comparison, Data management

1. Introduction

Acid Deposition Monitoring Network in East Asia (EANET) was established as a regional cooperative initiative to promote efforts for environmental sustainability. EANET was started the preparatory phase activities in April 1998, and this activity has been already implemented for more than 20 years. The cooperative activities of EANET participating countries based on developed national monitoring plans include the implementation of monitoring of wet deposition, dry deposition, inland aquatic environment, soil and vegetation and catchment scale monitoring in line with the guidelines and technical manuals with the conducting of quality assurance/quality control (QA/QC) programs as an important part of the monitoring activities. It is expected that the participating countries create a common understanding on the status

of the acid deposition problems through EANET activities, which will become a scientific basis for taking further steps to tackle the problems. EANET member counties recognized that improvements on monitoring and data quality are ones of the most important directions for EANET. EANET activities regarding QA/QC activities and data management are shown as follows.

(1) Each participating country develops and implements their national monitoring plans. Acid deposition monitoring is implemented in accordance with the monitoring guidelines, technical manuals and other technical documents adopted by EANET.
(2) The monitoring data and other information submitted by participating countries is compiled, evaluated and stored by the Network Center (NC).
(3) In order to obtain monitoring data of high quality, the quality assurance/quality control (QA/QC) programs are implemented in full collaboration among the participating countries.

QA/QC activities are classified into two categories, the internal and external QA/QC. The internal and external QA/QC activities complement each other. The internal QA/QC activities should be implemented in each laboratory and include preparing and disseminating the Standard Operating Procedures (SOPs), validation of analytical methods and reliability check of measurements. The internal QA/QC activities should be reexamined when the analysis method, the analytical equipment, the state of maintenance of the equipment, and the technical capabilities of the analyst are changed. In addition, the SOP should be specific for each analytical institution and easy-to-understand. EANET has published technical manuals and QA/QC Guidebook to ensure the Internal QA/QC (EANET 2000, 2010a, 2010b, 2010c, 2010d, 2013, 2016).

The external QA/QC activities should be implemented by the third-party institution and aims at evaluating whether the measured values by one laboratory are sufficiently identical to those by other laboratories. To participate in a proficiency test conducted by the third-party institution can check measurement techniques. If there is outlier of the measurement value from the mean value or standard value, there may be an uncertainty factor that is not found in the internal QA/QC. Then, the laboratory which has outlier data should examine the error factors and improve the analytical technique. EANET has conducted Inter-laboratory Comparison (ILC) Project for wet deposition, dry deposition, soil and inland aquatic environment samples among the analytical laboratories in EANET participating countries, based on the QA/QC programs of EANET (EANET, 2019a). In addition, some EANET participating countries have implemented audits of monitoring sites and analytical laboratories by the National Center or the third-party institution. The QA/QC Guidebook describes the recommended procedure of the audit (EANET, 2016).

Data management is also important component of EANET. Each local institution submits the analyzed data of the manual monitoring samples and automatic monitoring instruments data to the National Center. Then, each National Center submits the compiled data followed by the prescribed format to the Network Center for EANET (NC). All the data have been checked by the National QA/QC manager before submission, and the NC also checks the data internally and by external experts. The data compilation

and screen procedure are described in the technical manuals (EANET 2000, 2010a, 2010b, 2010c, 2010d, 2013). As the final product, the NC published annual data report (EANET, 2019b). The data user can freely download the data report and numerical data file. In April 2019, the NC developed new EANET data request registration and download system. In order to obtain the raw data such as hourly, weekly and bi-weekly data of wet and dry deposition monitoring, the data user should input registration information and submit on the website. After approval, the data user can download the raw data.

In this article, some technical information on the internal and external QA/QC activities and data management in EANET are presented.

2. Internal and external QA/QC activities

2.1 Internal QA/QC activities: Preparation of Standard Operating Procedures

Standard Operating Procedures (SOPs) are the procedures used in all the processes of the monitoring system, including field (sampling site) and laboratory operations and data management. The SOPs provide a method to ensure that all personnel follow the same procedures to avoid variance of data quality between personnel in charge, and ensure also that they conduct their works with scientific sound understanding of QA/QC. The preparation of SOPs is key component of Internal QA/QC activities. Each of the sampling and chemical analysis organizations or laboratories should make effort to prepare SOPs that meet the actual conditions of respective organizations in consideration of the Technical Manuals and the national QA/QC programmes. The SOPs should be prepared to be specifically and clearly addressed even for beginners by careful reviewing, and updated timely in accordance with the latest technical and administrative advances. The major items and recommended structures to be described in the SOPs are shown in the QA/QC Guidebook (EANET, 2016).

When the ILC project on wet deposition is conducted, the NC send the questionnaire condition of SOP preparation to the participating laboratory. Figure 1 shows annual variations of the number of laboratories that prepare the SOPs. In 2018, approximately 60% of laboratories have prepared SOPs. The percentage of laboratories that prepare the SOPs is increasing with the passing of the years. The Medium Term Plan for the EANET (2016-2020) describes one of the activities includes continuation of monitoring of all EANET monitoring items, improvement in monitoring methodologies and better instrument maintenance. The Updated SOP of each laboratory is indicator of this activity. The NC will provide guidance of the SOPs when the technical mission to each EANET participating country is dispatched. If the laboratory has not prepared, the NC instructs the importance of SOPs for the internal QA/QC activities.

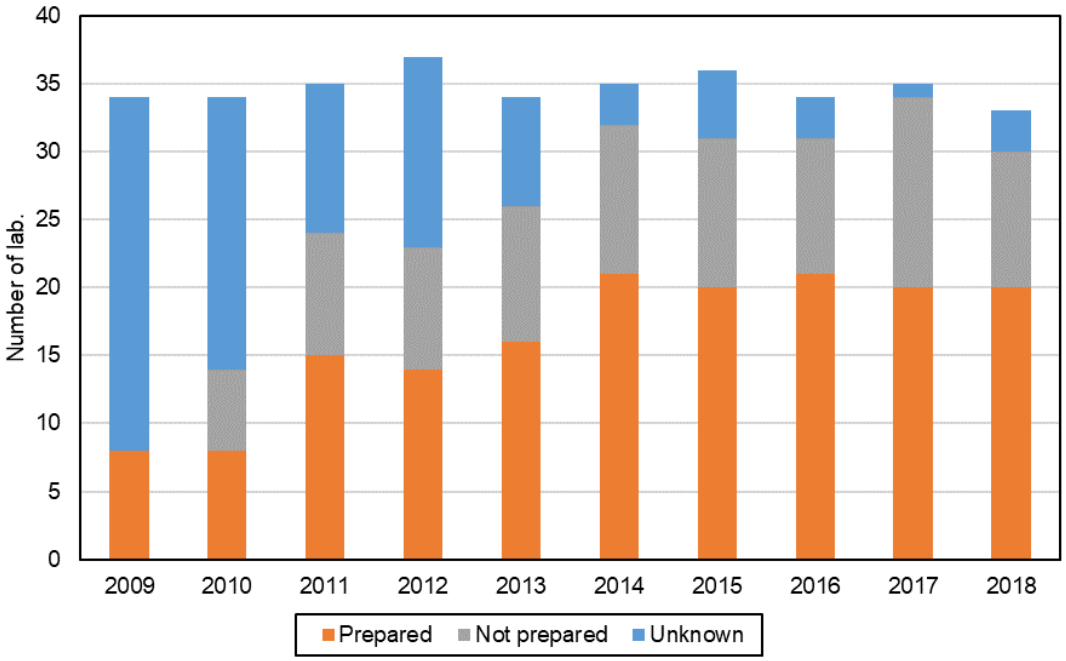

Figure 1. Annual variations of the number of laboratories that prepare the SOPs (2009-2018)

2.2 External QA/QC activities: Inter-laboratory Comparison Project

The Inter-laboratory Comparison (ILC) Project is key component of external QA/QC activities. It was conducted among the analytical laboratories in EANET participating countries, based on the QA/QC programs of EANET. The objectives of the ILC are shown below. It covers the evaluation of analytical results, analytical equipment, and its operating condition and other practices. The ILC project has been implemented by the NC annually for the items of wet deposition (artificial rain water sample), dry deposition (artificial filter samples for collecting gaseous substances), soil (homogenized soil sample at the selected site) and inland aquatic environment (artificial inland water sample). The annual results are published and opened to the public to demonstrate analytical accuracy for each laboratory (EANET, 2019a).

(1) To recognize the analytical precision and accuracy of the measurement in each participating laboratory,
(2) To give further opportunities to improve the quality of the analysis on wet deposition, dry deposition (filter pack method), soil and inland aquatic monitoring of EANET,
(3) To improve the reliability of analytical data through the assessment of suitable analytical methods and techniques.

The first ILC project was implemented in 1998, and there is long time record of the results. Figure 2, 6 and 7 show annual variations of ILC results for wet deposition, dry deposition and inland aquatic environment, respectively (EANET, 2019a). For the wet deposition results, the percentage of the data within the Data Quality Objectives (DQOs) was lower in the early years, and then the percentage increased with passing of

the years. This trend implies the overall analytical performance was improved by attention to contamination of analytical facility, laboratory wares and deionized water and analytical credibility of laboratory staffs. Over recent 10 years, the ratios of flagged data are relatively stable for both high (5-10%) and low (9-21%) concentration samples.

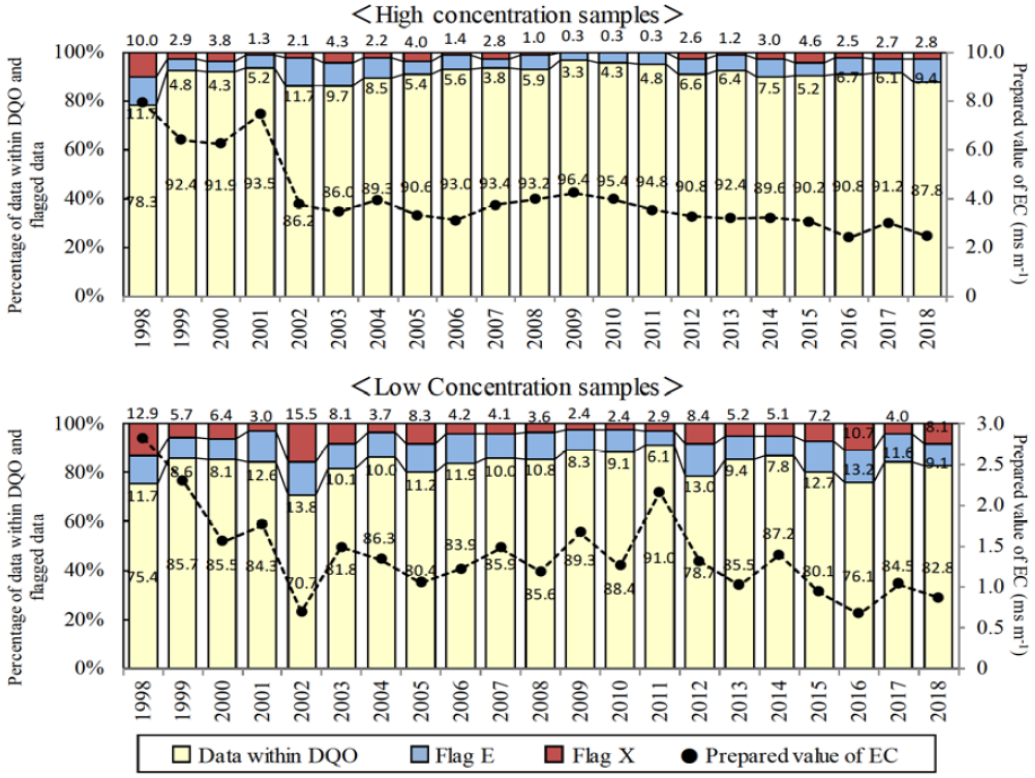

Figure 2. Annual variations of ILC results for wet deposition (1998-2018)

Figure 3 shows the annual variation of the percentage of the data within the DQOs for respective participating laboratories (EANET, 2019a). While all of the analytical data of 2 laboratories (JP10 and TH08*) were satisfied DQO, the percentage of the data within DQO in 7 laboratories was below 80%. These results show that there is significant discrepancy of analytical accuracy among the participating laboratories in EANET. It is hard to identify tangible reasons of this discrepancy, but according to site and laboratory audits for EANET sites, key factors that may cause large uncertainty and outlier for ion chromatography analysis were identified as follows.

(i) Improper calibration curve

It is necessary to make a calibration curve of an ion chromatography or other analytical instruments before measuring unknown samples. The calibration curve is usually determined by a linear relationship between the signal output of analytical instrument such as peak area and peak height and the concentration of the analyte ion. The ion concentrations are determined by linear interpolation between multiple calibration points. The range of measurement points on the calibration curve has large influence on accuracy and precision.

According to the ILC project report, the range of calibration curves in some laboratories does not include the concentration of the analyte (Namely, the prepared

values for the artificial rainwater). Many of such laboratories show the flagged data. It is recommended to set calibration points to include the concentration of the analyte. Especially, adding the lower level of calibration point may improve analytical performance.

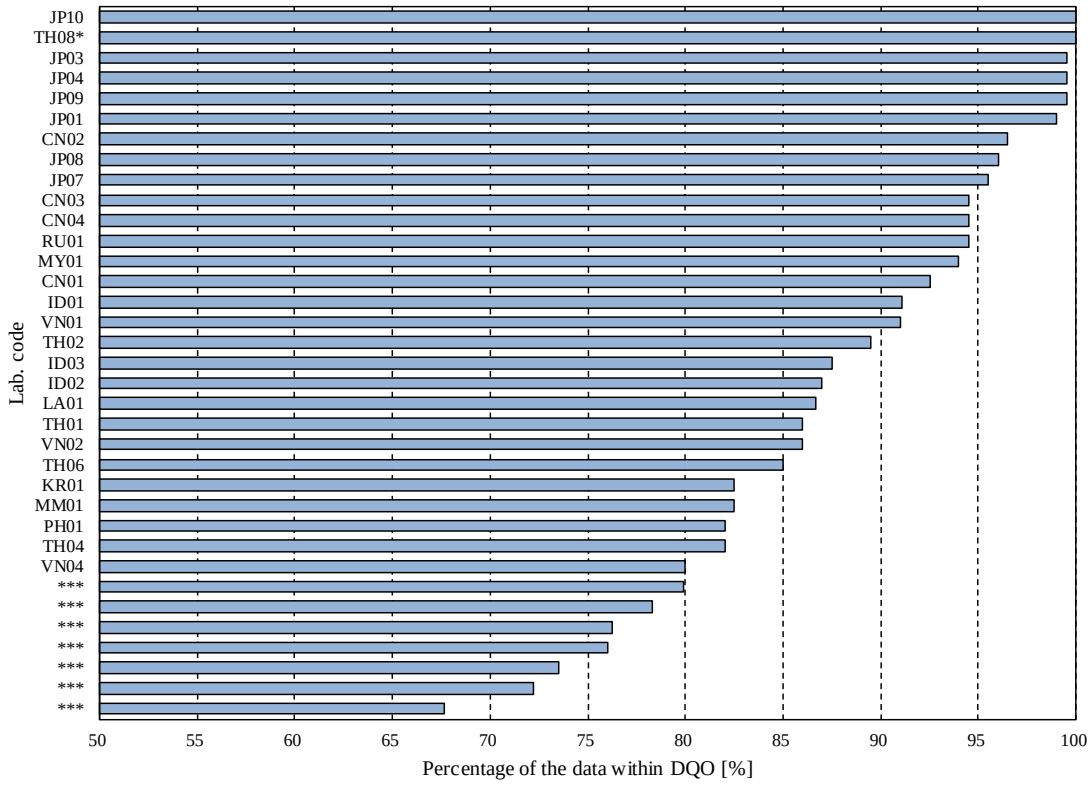

Figure 3. Percentage of the data within the DQOs for respective participating laboratories(Calculated from the data from 2009-2018)

Note: Lab code included as "***" in case that percentage of the data within DQO is below 80%.

TH08*: Ion parameters are not included in the percentage of TH08

(ii) Contamination of the samples, apparatus, analyzer, and autosampler

Eliminating contamination to perform highly accurate analysis requires careful precautions, including handling of the samples, apparatus, analyzer, and autosampler in a laboratory. Water obtained from laboratory ultrapure water systems should have a resistance over 18 MΩ·cm and contains almost no inorganic ions. Two main types of contamination and check items are shown as follows.

1) Sample contamination

- Keep clean all of the sample storage and analytical equipment. It is recommended to prepare the SOP of laboratory wares cleaning in each laboratory.
- A sample vial is one of the key components to cause contamination. To avoid contamination, do not touch the septum with bare hands. Rinse the vial interior with ultrapure water, and then rinse with the sample solution.

2) Carryover in the sample injection unit

- When the blank sample is analyzed after analysis of high concentration samples,

ions remaining in the sample injection unit may be detected. To avoid carryover, low concentration standards or samples should be analyzed prior to high concentration ones. The order of sample analysis must be carefully considered.

(iii) Analytical instability

Analytical instability during one analytical run will cause large uncertainty. The Technical Manual for Wet Deposition Monitoring in East Asia describes duplicate analysis of a sample should be conducted at least every 20 samples (EANET, 2010a). Analytical precision is defined as the standard deviation between the two analyses:

$$Si = (\Sigma di^2/2Ni)^{1/2}$$

where di denotes the difference between the two measurements, and Ni is the number of the sample pairs for the duplicate analysis in the reporting period. The DQO of analytical precision is determined as 15%. It is recommended to check analytical precision prior to the re-analysis of artificial rainwater samples.

The Technical Manual also describes that the analytical system should be checked by analyzing standard solutions of the high level concentration or additional QC samples at least once a day. (EANET, 2010a). After the ILC was done, artificial rainwater samples can be used as Standard Reference Material (SRM) for working standard measurement. The checking procedure by using the SRM is described in Technical Manual. The NC recommend each laboratory measure Standard Reference Materials in the analytical sample stream. Figure 4 and 5 show time variation of working standard measurement for SO_4^{2-} and NH_4^+ at Banryu site, respectively. All of the analysis results satisfied the DQOs of the ILC (± 15% difference from the setting values). If the deviation from SRM values are large, the laboratory staff should check the contamination shown above and instrumental condition.

(iv) Unstable analytical condition.

Baseline drift of ion chromatography is one of the major factors of large uncertainty. The Technical Manual for Wet Deposition Monitoring in East Asia describes the room temperature of the analytical laboratory should be constant (EANET, 2010a). It should be within ±3 ºC to minimize the baseline drift and detector response. The baseline should be monitored for a few hours after startup. After a stable baseline is achieved, the total conductivity of ion chromatography should be compared to the previous values to confirm normal conditions. If the baseline is still drifted, the drift should be corrected followed by the instruction of the Technical Manual (EANET, 2010a) or the operation manual of the manufacturer.

For the annual variations of dry deposition and the inland aquatic environment results over recent 10 years, the ratios of flagged data for dry deposition are also in certain range for both large (6-29%) and small (15-43%) quantity samples, and those for inland aquatic environment are relatively stable (9-16%). These results imply that the reasons of large uncertainty in some laboratory have not been identified. The possible causes of large uncertainty and outlier may be similar to wet deposition, but the other factors such as filter extraction procedure for dry deposition samples and pretreatment for inland aquatic environment sample may influence on analytical

accuracy.

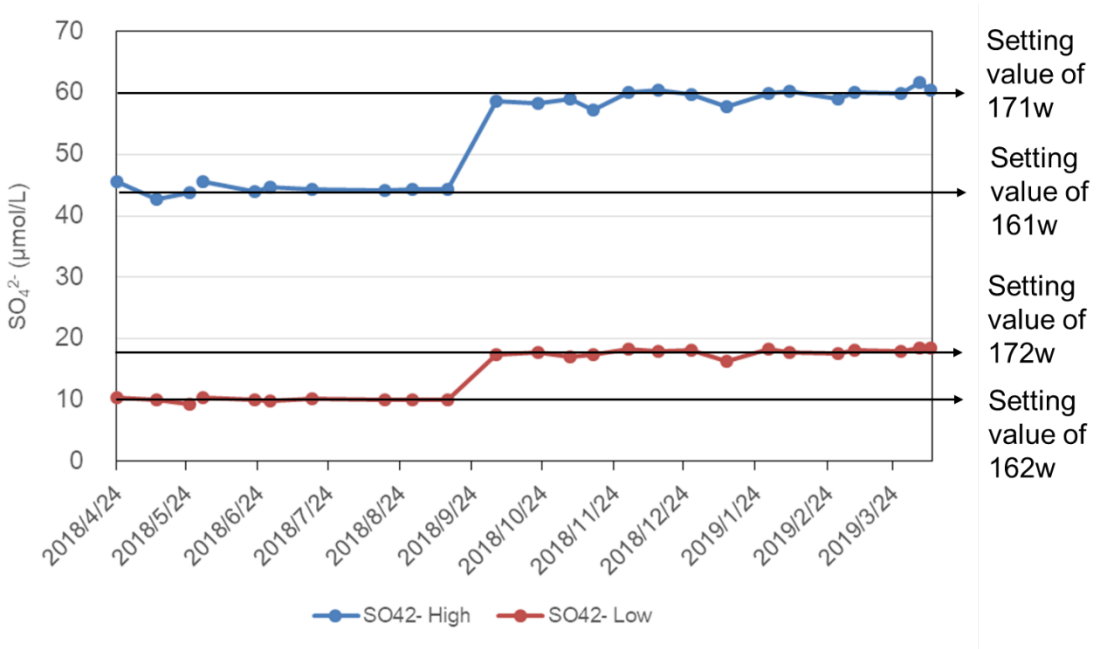

Figure 4. Time variation of working standard measurement for SO_4^{2-} at Banryu site

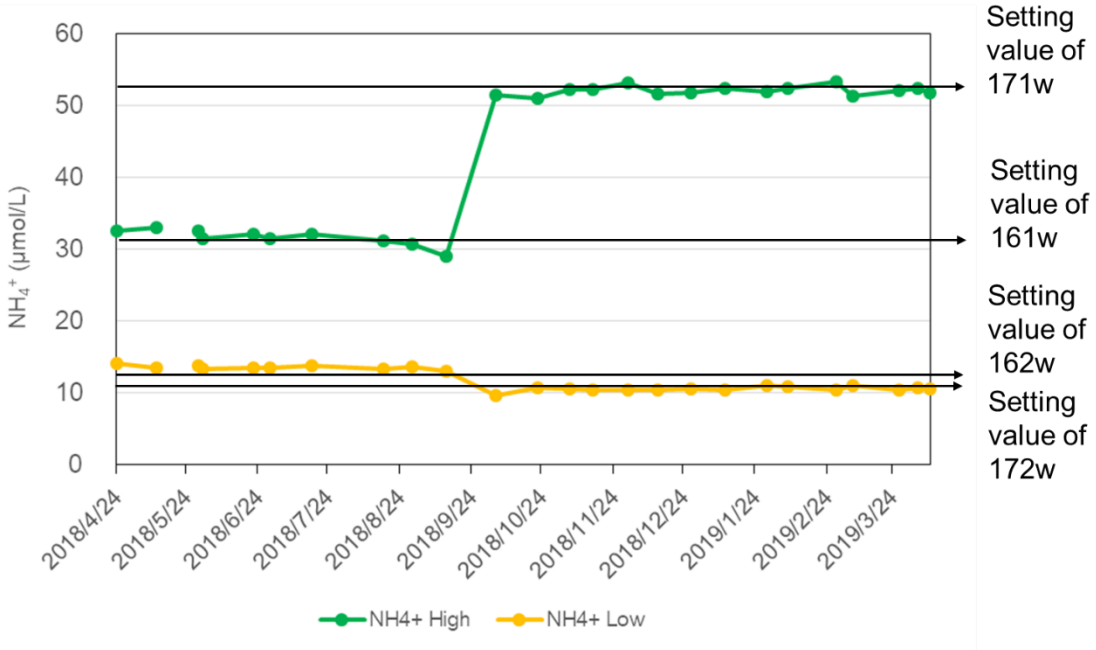

Figure 5. Time variation of working standard measurement for NH_4^+ at Banryu site

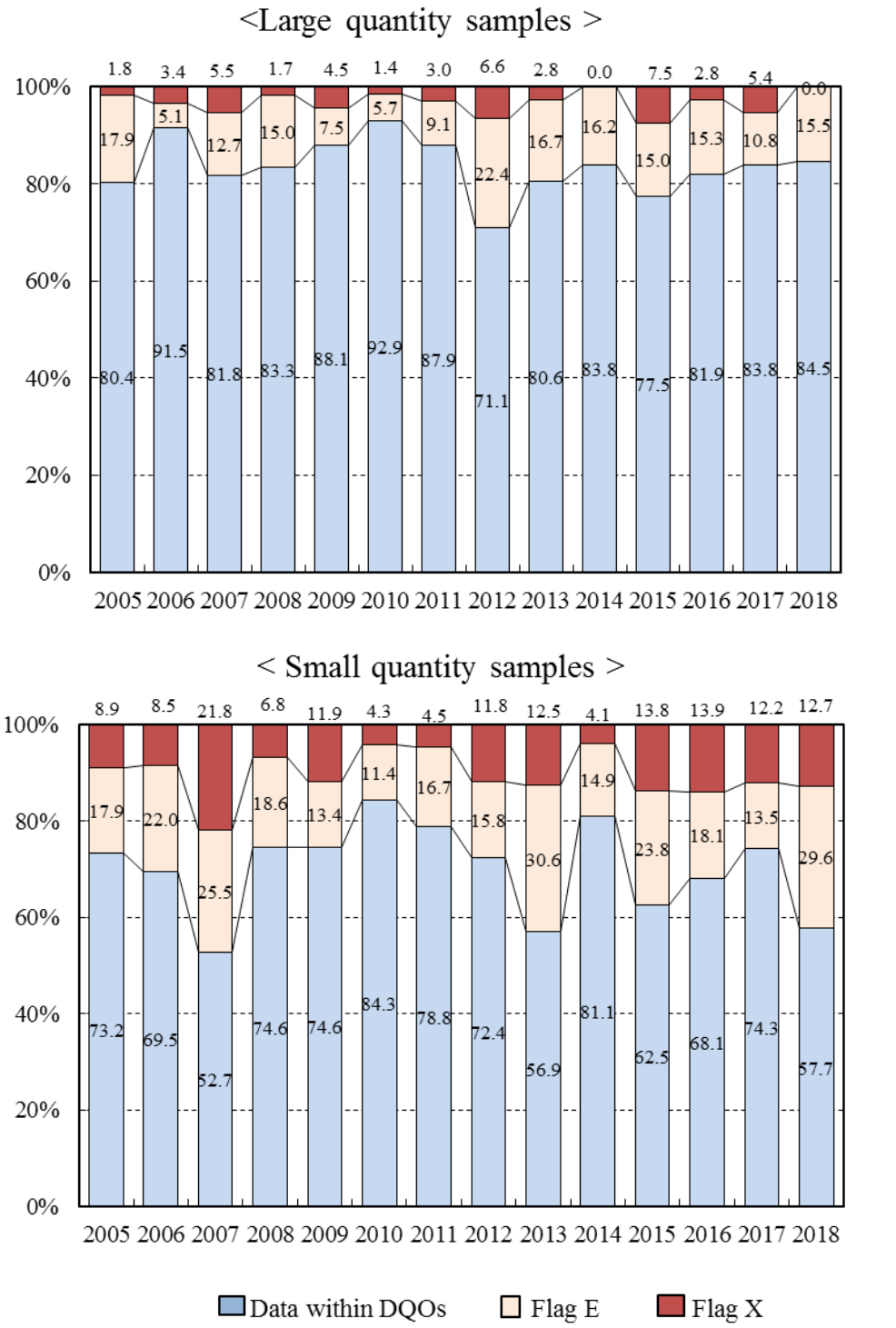

Figure 6. Annual variations of ILC results for dry deposition (2005-2018)

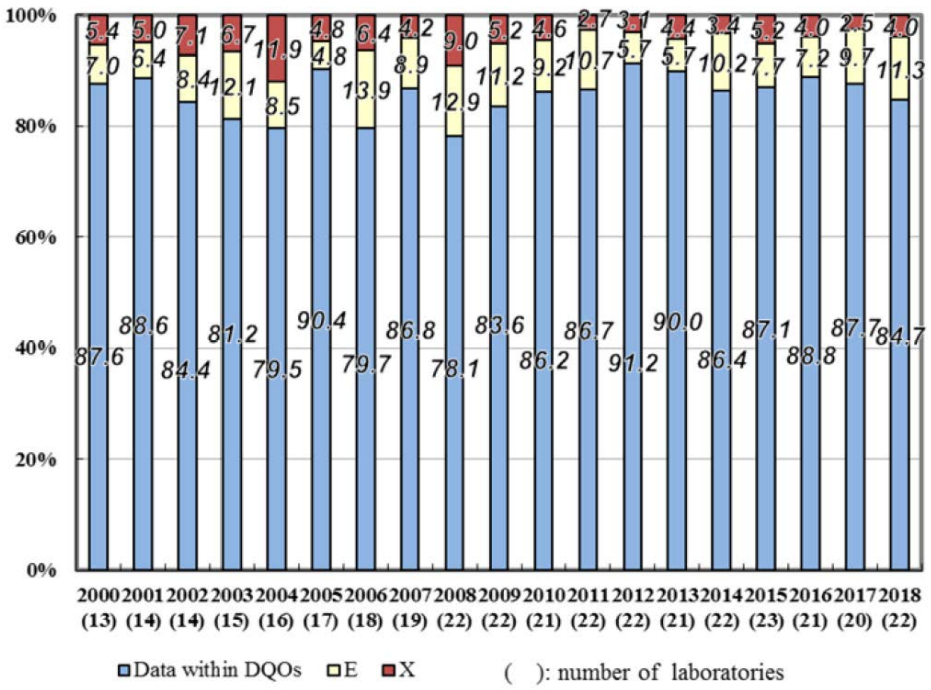

Figure 7. Annual variations of ILC results for inland aquatic environment (2000-2018)

3. Data management activities

As described in introduction, data management is important component of EANET to ensure reliable data to evaluate the status of acidic deposition and air pollution in East Asia. The data management procedure is different among respective monitoring items. For example, each laboratory submits the analyzed data of the precipitation samples to the National Center, and then each National Center submits the compiled data in the prescribed format to the NC in the following year of the monitoring. All the data have to be checked by calculating values of ion balance (R1) and conductivity agreement (R2). The National QA/QA manager of each country is responsible for finalizing validated data. If a sample or individual datum has problems including “insufficient sample volume” or “low precision”, the flags corresponding to the problems should be recorded onto the data sheet.

To disseminate the EANET monitoring data to the public users, annual data report, the data reports and the ILC Project reports in pdf format, monthly and annual data of wet and dry deposition and all data of soil and vegetation, inland and catchment in xlsx format have been uploaded to the EANET Web page (https://monitoring.eanet.asia/document/public/index). The EANET data archive page was redesigned at the same time with revamping of the EANET website in April 2019. Figures 8 and 9 show the number of downloads for each annual data report and ILC Project report, respectively. The total numbers of file downloads for data report and ILC Project report are 12300 and 1962, respectively. The numbers of downloads for 2017 report are higher than the other years reports, which suggests the public is interested in the most recent data. Encouragement for the data user to transfer information is important so that many people will use EANET data for research, policy analysis, etc.

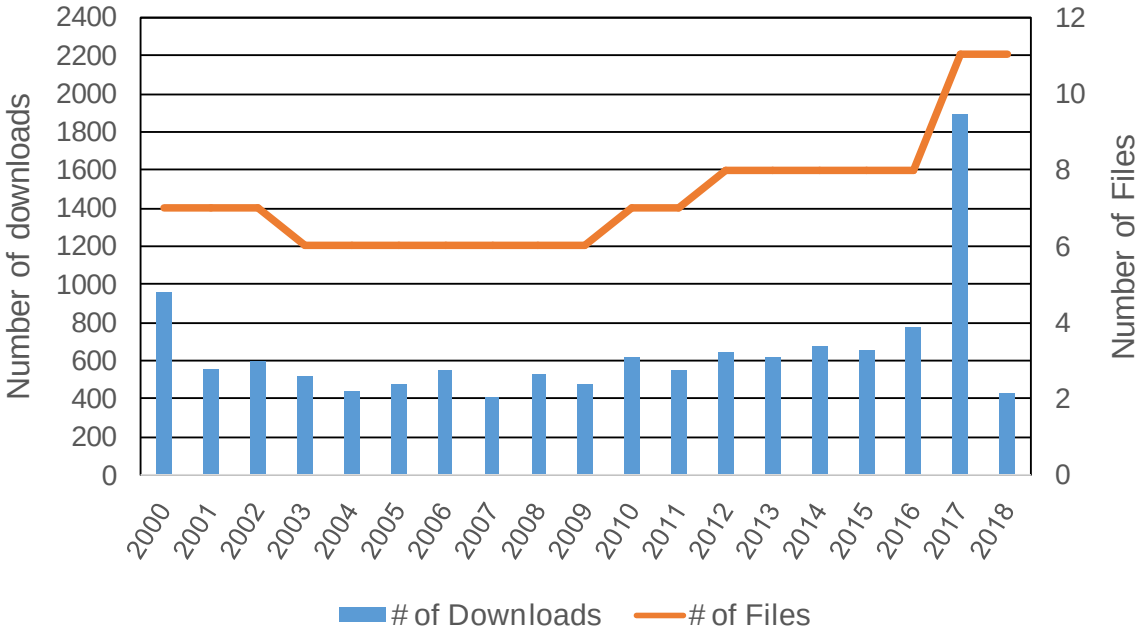

Figure 8. Number of file downloads for each annual data report (April 2019-March2020)

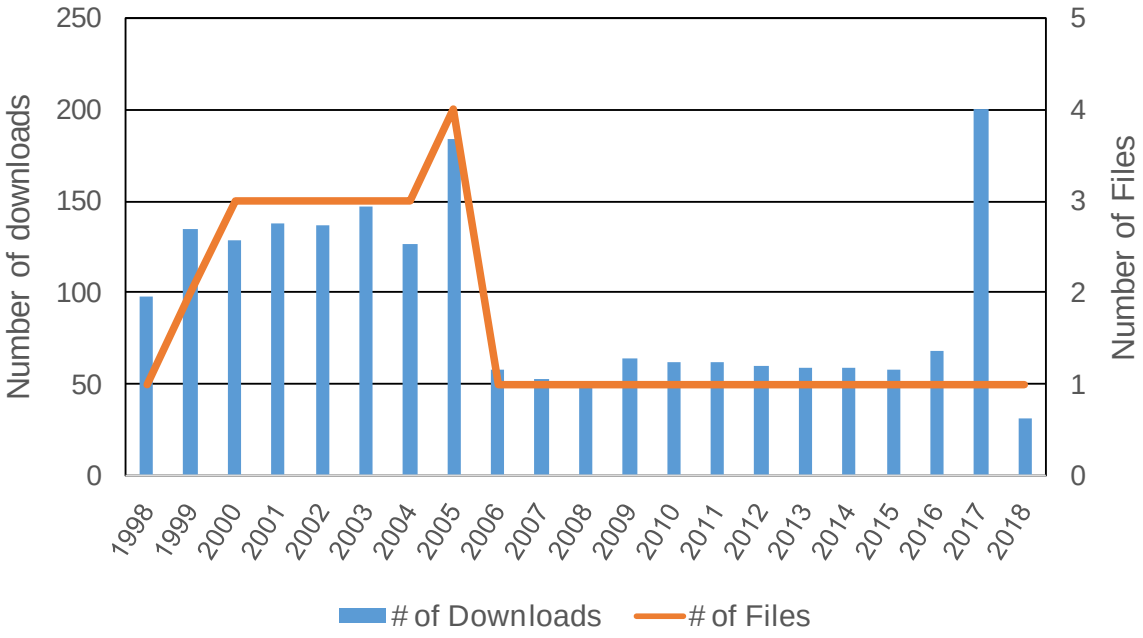

Figure 9. Number of file downloads for each annual ILC Project report (April 2019-March2020)

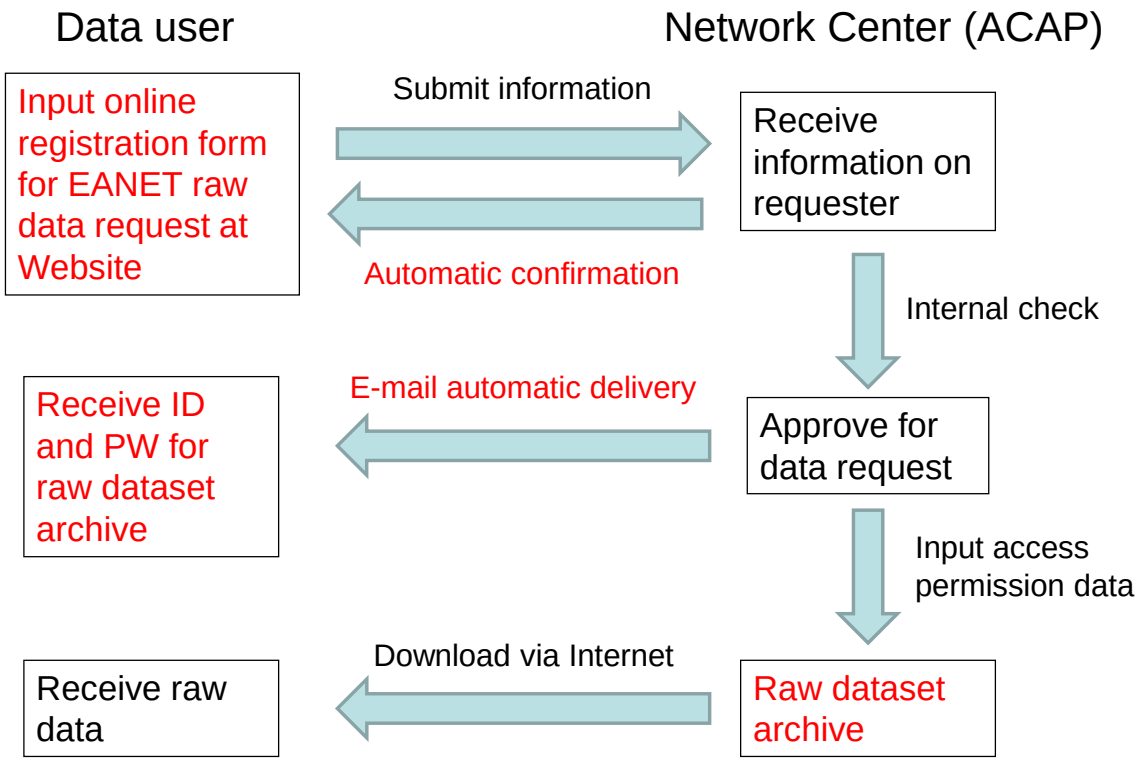

Figure 10. New EANET raw data request procedure

In order to obtain the raw data such as hourly, weekly and bi-weekly data of wet and dry deposition monitoring in csv format, the data user should input registration information and submit on the website. Figure 10 illustrates new EANET raw data request procedure since April 2019. After approval by the NC, ID and PW for data archive access will be sent by E-mail. The new system enables the data user to obtain the raw data in a shorter time. The numbers of enrollments and data downloads from April 2019 to March 2020 are 89 and 886, respectively. These numbers are significantly less than those of other observation networks such as the Global Atmosphere Watch Programme of the World Meteorological Organization (WMO/GAW) and European Monitoring and Evaluation Programme (EMEP). In August 2018, the Letter of Arrangement (LOA) for the recognition of EANET as a contributing network for WMO/GAW was signed by the EANET representative and the Deputy Secretary-General of WMO. The activities described in the LOA include establishing links from the WMO/GAW World Data Center to the data archive of the EANET. When the link is established, the EANET data will be disseminated to more users in the world and it will contribute integrated data assessment between EANET and other regional monitoring networks.

4. Summary

The NC has implemented data management such as compile, evaluate, analyze and store the EANET monitoring data and related information, disseminate monitoring data and related information, implementation and coordinate QA/QC activities. The NC has published EANET has published technical manuals and QA/QC Guidebook to ensure

the Internal QA/QC. To implement the activities described in Medium Term Plan for the EANET (2016-2020), the NC encouraged to prepare Standard Operating Procedures (SOPs) for each analysis laboratory in EANET when technical mission to each EANET participating country. The NC have conducted the Inter-laboratory Comparison (ILC) project more than 20 years, and the series of results showed that there is significant discrepancy of analytical accuracy among the participating laboratories in EANET. As the next step, it is important to conduct follow-up action to identify key factors that may cause large uncertainty and outlier. The NC requests re-analyze artificial rainwater for the laboratory which has flagged data for the artificial rainwater. After re-analysis, the NC will provide technical guidance on ionic component analysis. As for data management, the new EANET data archive page and data request registration and download system were established in April 2019. The numbers of downloads for 2017 report are higher than the other years reports, which suggests the public is interested in the most recent data. It is important to disseminate EANET data to more data users so that many people will use EANET data for research, policy analysis by collaborating other regional monitoring networks.

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