Research Report

Development of a Framework for a High-resolution, Three-dimensional Regional Air Quality Simulation and its Application to Predicting Future Air Quality over Japan

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ABSTRACT We have developed a framework for a three-dimensional regional air quality simulation that is applicable to various air quality studies over Japan. The framework consists of the following simulation model systems: the Weather Research and Forecasting (WRF) model to simulate meteorological fields; the Community Multi-scale Air Quality (CMAQ) modeling system to simulate pollutant concentrations; emissions estimate models; and emission databases. Motor vehicle emissions in Japan are estimated using the Japan Auto-Oil Program (JATOP) vehicle emissions estimate model; anthropogenic emissions from sources other than motor vehicles in Japan are estimated using the Georeference-Based Emission Activity Modeling System (G-BEAMS); and biogenic emissions are estimated using the Model of Emissions of Gases and Aerosols from Nature (MEGAN). The Regional Emission inventory in Asia (REAS) is used for emissions in Asian countries except for Japan. The most prominent feature of our framework is its ability to simulate multi-scale air quality. The framework allows for the simulation of emissions and the dynamic transport of pollutants in heavily polluted urban areas with a maximum resolution of 1 × 1 km, and the long-range transport of pollutants is also taken into account. This framework is used to analyze the impact of future emissions from anthropogenic sources on air quality over the Tokyo metropolitan area. NOx, NMVOC and primary $PM_{2.5}$ emissions over the Tokyo metropolitan area are estimated to be reduced by 44.5%, 18.1% and 41.7%, respectively, from 2005 to 2020. The simulation predicts that concentrations of NO_2 and $PM_{2.5}$ over the Tokyo metropolitan area will decrease by approximately 30-40% and 15-20%, respectively, during the above period. O_3 concentrations significantly increase in winter due to decreased titration by NO, whereas no significant variations are observed in spring and summer. In addition, we analyzed the impact of future long-range transport projected under three emission scenarios provided by REAS. The simulation indicates that future long-range transport will affect concentrations of O_3 and $PM_{2.5}$ over the Tokyo metropolitan area; however, its impact is small compared with that of future anthropogenic emissions in Japan.

KEYWORDS Three-dimensional Regional Air Quality Simulation, High Resolution, Tokyo Metropolitan Area, Future Prediction, Long-range Transport

1. Introduction

Although air quality has been gradually improving in Japan, there is still concern over concentrations of nitrogen dioxide (NO_2), ozone (O_3) and fine particulate matter ($PM_{2.5}$). The Japanese government has two air pollution monitoring networks. One network comprises ambient air pollution monitoring stations (APMSs), and the other comprises roadside air pollution monitoring stations (RAPMSs). RAPMSs are located along major roadways, and APMSs are designed to monitor the general air quality and are not affected by specific emission sources. NO_2 concentrations have decreased overall; however, there are still concentrations higher than the Environmental Quality Standard (EQS) at some RAPMSs located in urban areas. O_3 concentrations have gradually increased over the last twenty years. Consequently, most of the APMSs show nonattainment of the EQS for O_3.(1) The Japanese government has recently issued the new EQS for $PM_{2.5}$; however, the current $PM_{2.5}$ concentrations likely exceed the new EQSs in most areas in Japan. To develop measures to meet the EQSs for these pollutants, the prediction of future air quality is required. Various external factors should be considered as well, including meteorological conditions, emissions from natural sources, and the

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transport of pollutants from outside of the country.

It can be said that, in Japan, issues related to primary pollutants have been mitigated, while those related to secondary pollutants remain. The reduction in concentrations of NO_2, O_3 and secondary aerosol components has been relatively small.[2] These are typical secondary pollutants that are mutually related through photochemical reaction chains in the atmosphere.[3] Nonlinear relationships between precursor emissions and ambient concentrations must be considered to develop effective measures to reduce secondary pollutants. Three-dimensional regional air quality simulation models have been developed that incorporate complex photochemical reaction chains and aerosol formation in the atmosphere, and they have become essential tools for investigating nonlinear relationships between precursor emissions and ambient concentrations of secondary pollutants, including aerosols. Chatani et al.[4] conducted sensitivity analyses using a three-dimensional air quality simulation. The model results indicated that ambient concentrations of various chemical species are nonlinearly affected when precursor emissions are changed.

Studies have been performed in which three-dimensional regional air quality simulations were applied to Japan. An example of recent simulation studies is that of Yamaji et al.[5], in which future concentrations of surface ozone were predicted over East Asia. The prediction revealed that surface ozone concentrations over Japan would increase from the years 2000 to 2020 under the three future emission scenarios. Kurokawa et al.[6] analyzed the impact of meteorological variability on interannual variations of springtime boundary layer ozone over Japan. They indicated that a gradual increase of ozone concentrations in Japan could be explained by the increase of emissions in East Asia, while interannual variations of ozone concentrations are explained by meteorological variability that is seen in the wind patterns and surface pressures over the Pacific Ocean. Both studies focused on the long-range transport of pollutants, and both used a coarse-resolution configuration (80 × 80 km) in their simulations. Our concern, however, is that high concentrations of pollutants are mainly observed in urban areas. Coarse-resolution simulations have difficulty reproducing detailed spatial distributions of high concentrations of pollutants in urban areas. A much higher resolution, on the order of ten kilometers or less, is necessary. The long-range transport of pollutants should be taken into account simultaneously. Few studies have used such high resolutions for regional air quality simulations for Japan.[7,8] A database suitable for high-resolution simulations is not readily available in Japan. The absence of essential data is an obstacle to performing high-resolution simulations.

One of the objectives of this study is to develop a three-dimensional regional air quality simulation framework in which a meteorological model, an air quality model, emissions estimate models and databases are coupled. The meteorological and air quality models incorporated in the framework were developed in the United States; however, the main features of the framework are the emissions estimate models and the databases, which have been developed to reflect the situations in Japan in detail with high temporal and spatial resolution. These emissions are suitable for simulating the fine-scale spatial distribution of pollutants concentrated in Japan's urban areas.

The simulation framework is applied to predict the impacts of future anthropogenic emissions and the long-range transport of pollutants on air quality over the Tokyo metropolitan area. According to the definition of the United Nations, the Tokyo metropolitan area is the world's largest urban agglomeration, or "megacity".[9] Air quality over the Tokyo metropolitan area has been the worst in Japan in terms of most pollutants, so the analysis results could be useful for developing effective measures to improve air quality over Japan, especially over the Tokyo metropolitan area. The results may also be helpful in projecting future directions and developing measures to improve air quality over other megacities, the number of which is rapidly increasing, especially in developing countries.

Section 2 describes the three-dimensional regional air quality simulation framework developed in this study. Section 3 explains the performance of the simulation framework when reproducing current concentrations of pollutants over the Tokyo metropolitan area. Section 4 describes the results of the analyses to investigate the impacts future anthropogenic emissions and the long-range transport of pollutants will have on air quality over the Tokyo metropolitan area. These impacts are predicted using the simulation framework developed. Finally, Section 5 summarizes the outcomes of this study.

2. Description of the Three-dimensional Regional Air Quality Simulation Framework

Figure 1 is a schematic of the three-dimensional regional air quality simulation framework developed under this study. The pollutant concentrations were calculated using the Community Multi-scale Air Quality (CMAQ) modeling system. The meteorological fields were obtained from the Weather Research and Forecasting (WRF) model. The emissions estimate models and the database shown in Fig. 1 were used to estimate precursor emissions. Details of each component incorporated in the three-dimensional regional air quality simulation framework are described in this section. The target domains and settings used in the simulations (to be described in Sections 3 and 4) are also explained here.

2. 1 Target Domains

Maps of the target domains used in this study are shown in **Fig. 2**. Three levels of nested domains (East Asia, Japan and Kanto) were used. The Tokyo metropolitan area, which lies in the Kanto region, consists of Tokyo's twenty-three special wards and its surrounding cities. In 2001, the government issued a law concerning special measures for the total emission reduction of nitrogen oxides and particulate matter from automobiles in specified areas (referred to as the "Automobile NOx-PM law"). Figure 2 illustrates Tokyo's twenty-three special wards (in black) and the specified area for the Automobile NOx-PM law (in gray). The grid cell size for the Kanto domain is 4 × 4 km.

The air quality over the Tokyo metropolitan area appears to be affected by the long-range transport of pollutants from outside the Kanto domain in addition to the local emissions. Considerations were taken regarding the long-range transport of pollutants as follows. The Japan and East Asia domains are located outside the Kanto domain. The Japan domain covers most of Japan except for the southern islands (this domain also covers a part of the Asian continent). The grid cell size for the Japan domain is 16 × 16 km. The

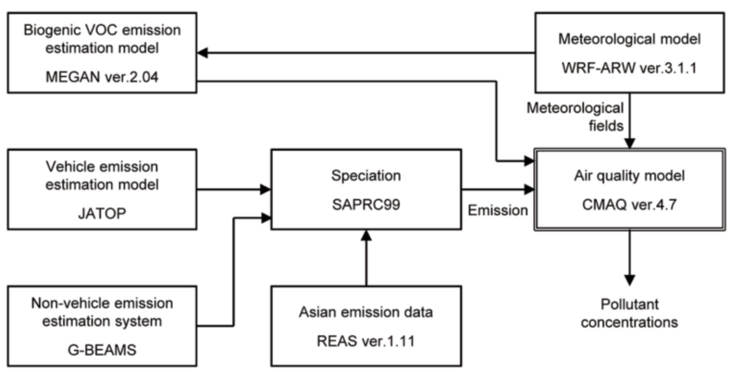

Fig. 1 Schematic of the three-dimensional regional air quality simulation framework developed under this study.

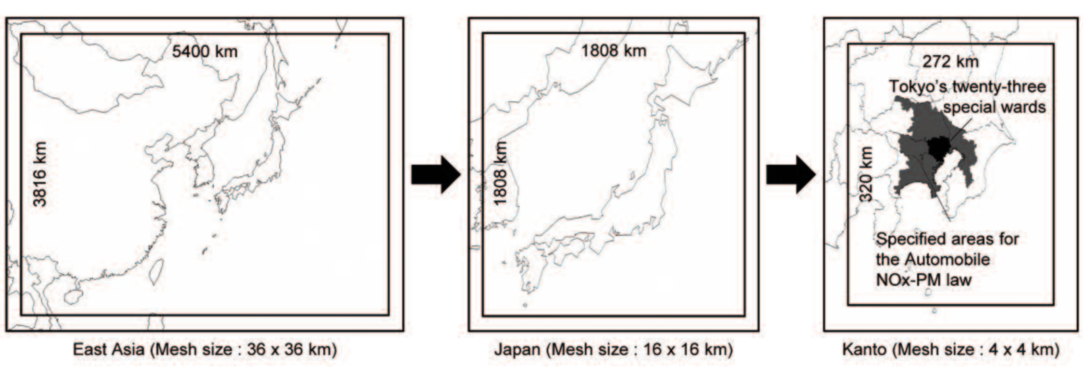

Fig. 2 Maps of the target domains used in this study. Outer thick lines indicate the WRF domains, while inner thin lines indicate the CMAQ domains. Tokyo's twenty-three special wards are shown in black, while the specified area for the Automobile NOx-PM law is in gray.

East Asia domain covers the PRC, Mongolia, North Korea, South Korea and Taiwan, as well as Japan, which allows consideration of the long-range transport of pollutants from outside Japan. The grid cell size for the East Asia domain is 36 × 36 km.

Number of vertical layers in all domains was 29 in WRF and 19 in CMAQ from the surface to 100 hPa. The height of the lowest layer was 31 m in all domains.

2. 2 Air Quality Model

CMAQ version 4.7[10] was used to calculate pollutant concentrations. The chemical mechanism employed in this study was the SAPRC99.[11] The input emissions and initial and boundary concentrations were prepared based on species groups used in the SAPRC99. A default database provided with CMAQ was used for the initial concentrations for all domains and boundary concentrations for the East Asia domain. The impact of initial concentrations was eliminated by beginning the simulation a month prior to the target period, which was considered a spin-up period. The impact of boundary concentrations was expected to be small because the most important regions that appear to influence on air quality over Japan are included in the East Asia domain. One exception is O_3. Some studies[6] have used the results of global chemical transport models as boundary concentrations. However, Lin et al.[12] reported that the artificial downward transport of O_3 from the lower stratosphere causes overestimation of O_3 in CMAQ simulations. How to set the appropriate boundary concentrations of O_3 is one of the remaining issues. The simulations were performed consecutively in the descending order of domain size: East Asia, Japan and the Kanto region. The boundary concentrations for the inner domain were provided from the simulation results of the outer domain (1-Way nesting). The aerosol module used in this study was aero5. The sea salt emissions were calculated using this module within CMAQ.

2. 3 Meteorological Model

The Advanced Research WRF (WRF-ARW) version 3.1.1[13] was used to calculate the meteorological fields. The National Centers for Environmental Prediction (NCEP) Final Analysis data (1-degree resolution, every 6 h) were used for initial and boundary conditions and grid nudging. For sea surface temperatures, data from the real-time global sea surface temperature (RTG_SST) analysis developed by the NCEP/Marine Modeling and Analysis Branch (MMAB) (0.5-degree resolution, every 24 h) were used. The United States Geological Survey (USGS) land use data provided with the WRF model were used for the Asian countries, while detailed plant information provided in the Japan Integrated Biodiversity Information System (J-IBIS) (approximately 1 × 1 km resolution) were used for Japan. Simulations of the East Asia domain were performed separately from those of the Japan and Kanto domains, which were performed simultaneously (2-Way nesting). The hourly results were fed to CMAQ. This frequency may be too coarse for high wind speeds, but it is expected to resolve high concentrations in 4 × 4 km meshes caused by low wind speeds on the order of 1 m/s^{-1}.

2. 4 Biogenic VOC Emissions

Biogenic VOC emissions were estimated using the Model of Emissions of Gases and Aerosols from Nature (MEGAN)[14] version 2.04. The gridded databases of leaf area index, plant functional types and emission factors with a resolution of 150 seconds, which were provided with the model, were used. Hourly outputs from the WRF simulations were used to consider the dependence of emissions on the meteorological conditions. The model was slightly modified to generate biogenic sesquiterpene emissions, which were input into the aero5 aerosol module of CMAQ, which considers the formation of secondary organic aerosols from sesquiterpenes.

2. 5 Vehicle Emissions

The Japan Auto-Oil Program (JATOP) vehicle emissions estimate model[15] was used to estimate vehicle emissions in Japan. This model can estimate hot-running and cold-start emissions, evaporative emissions (running loss, hot soak loss and diurnal breathing loss), and emissions of tire wear and resuspended road dust originating from vehicles on the road. The species considered in the model are SO_2, nitrogen oxides (NOx), total hydrocarbon (THC), CO, ammonia (NH_3) and SPM. Hourly, weekday/weekend and monthly variations are taken into account. This model considers accelerated vehicle fleet turnover and the retrofitting of in-use vehicles with exhaust after-treatment systems required to comply with the latest emissions regulations, including the “Automobile NOx-PM law”. Some vehicle fleets are assumed to be “high emitter vehicles”, which have much higher emissions than well-maintained vehicles. The horizontal resolution of the estimated emissions is approximately 10 × 10 km for the whole of Japan, and 1 × 1 km for the Kanto region.

The NOx, THC and SPM emissions are speciated into NO and NO_2, the SAPRC99 VOC species, and aerosol components, respectively, using our speciation database. The ratio of NO_2 in NOx is 2% for gasoline vehicle emissions and 13% for diesel vehicles emissions; however, the higher value of 44% is allocated for hot-running emissions from newer diesel vehicles equipped with oxidation catalysts. Methane is removed from THC, and oxygenated compounds that are not identified as THC are added. SPM is divided into fine and coarse fractions. The fine fraction is further speciated into elemental carbon (EC), organic carbon (OC), sulfate, nitrate and other components. The SPM emissions from vehicles are assumed to be classified into the fine fraction exclusively.

2. 6 Other Anthropogenic Emissions

The Georeference-Based Emission Activity Modeling System (G-BEAMS)(16) was used to estimate emissions from anthropogenic sources except for motor vehicles in Japan. The species considered in the model are SO_2, NOx, non-methane volatile organic compounds (NMVOC), CO, NH_3, SPM and $PM_{2.5}$. The model estimates annual emissions, and splitting factors that are specified for each source category have been prepared to consider monthly and hourly variations. The horizontal resolution of the estimated emissions is approximately 10 × 10 km for the whole of Japan and 1 × 1 km for the Kanto region.

The Ministry of the Environment of Japan (MOE) has developed the VOC emissions inventory in Japan. The total amounts of VOC emissions estimated using G-BEAMS were corrected to those in the MOE emissions inventory for source categories that are included in it. Emissions from source categories that are included in the MOE inventory but not in G-BEAMS were ignored in this study because their spatial and temporal distributions are not known. Emissions from ships and large point sources had not yet been incorporated in G-BEAMS when the simulations described in Sections 3 and 4 were conducted, so a database similar to the East Asian Air Pollutant Emissions Grid Database (EAGrid-2000)(17) was used. This database has stack information for each large point source; therefore, plume rise was calculated from it and the meteorological fields.

2. 7 Emissions in Asian Countries

The Regional Emission inventory in Asia (REAS)(18) ver.1.11 was used for emissions of anthropogenic sources and ships in Asian countries out of Japan. The horizontal resolution is 0.5 × 0.5 degrees. REAS only has annual emissions, and any temporal variations were ignored in this study. REAS was used in the East Asia domain and outside of the Japanese islands in the Japan domain. Emissions from biomass burnings in Asian countries except for Japan were not taken into account in this study.

3. Simulation Performance

Simulations were conducted to evaluate the performance of the three-dimensional regional air quality simulation framework developed under this study. The current target of the framework is to simulate daily concentrations of NO_2 and $PM_{2.5}$ accurately because the EQSs for NO_2 and $PM_{2.5}$ have been set in terms of daily concentrations. However, for $PM_{2.5}$, there were limited observation data available for Japan. Therefore, in this section, the simulation performance of suspended particulate matter (SPM), which approximately corresponds to PM_7, is discussed instead of $PM_{2.5}$. Daily and monthly concentrations were mainly used in the evaluation. The simulation performance of daily and monthly concentrations of O_3 is also shown to emphasize the strong relationship between O_3 and NO_2. The target periods were set to April, June and November 2005.

The correlation coefficient, mean normalized bias (MNB) and mean normalized error (MNE) were calculated from the daily observed and simulated concentrations at 26 APMSs located in Tokyo’s twenty-three special wards. These factors were calculated from the simulation results in the East Asia, Japan and Kanto domains, respectively, as shown in **Table 1**. These statistical indicators, horizontal distributions, time series variations, and the correlation between observed and simulated concentrations are discussed in this section.

In fact, most of the monitoring stations in Japan observe photochemical oxidants, which are not limited to O_3. However, the difference between concentrations of photochemical oxidants and O_3 is small. Therefore, the observed concentrations of photochemical oxidants are treated as the observed concentrations of O_3 in this study.

Table 1 Correlation coefficient, MNB and MNE calculatedfrom observed and simulated concentrations at 26 APMSs located in Tokyo's twenty-three special wards.

Species	Month	Domain	Correlation coefficient	MNB	MNE
NO_2	April	East Asia	0.463	−30.0%	35.0%
		Japan	0.737	−29.5%	32.5%
		Kanto	0.741	−29.7%	33.0%
	June	East Asia	0.345	−24.6%	34.1%
		Japan	0.577	−17.4%	29.1%
		Kanto	0.665	−23.1%	30.2%
	November	East Asia	0.521	−16.7%	24.5%
		Japan	0.805	−6.73%	14.8%
		Kanto	0.833	−6.52%	15.2%
O_3	April	East Asia	0.457	+41.6%	44.9%
		Japan	0.720	+32.1%	34.5%
		Kanto	0.727	+30.4%	33.0%
	June	East Asia	0.496	+67.5%	73.6%
		Japan	0.581	+33.1%	49.4%
		Kanto	0.692	+36.4%	47.3%
	November	East Asia	0.329	+139%	142%
		Japan	0.510	+95.0%	102%
		Kanto	0.532	+87.1%	94.2%
SPM	April	East Asia	0.702	−51.1%	51.3%
		Japan	0.706	−55.9%	56.0%
		Kanto	0.701	−55.7%	55.7%
	June	East Asia	0.490	−58.5%	58.7%
		Japan	0.507	−58.6%	58.7%
		Kanto	0.460	−61.0%	61.2%
	November	East Asia	0.793	−45.3%	46.0%
		Japan	0.847	−37.6%	39.1%
		Kanto	0.842	−37.2%	38.5%

Note: Underlined values indicate the highest value amaong the three domains.

3. 1 Horizontal Distribution

Figure 3 shows horizontal distributions of monthly observed and simulated concentrations of NO_2 in the Kanto domain. The observed concentrations at the APMSs are allocated to the corresponding 4 × 4 km grid cells, and the grid cells in gray represent the absence of APMSs. High concentrations are observed over the heart of Tokyo, and the simulations reproduce them well. The simulations also reproduce high concentrations spreading to the southwest of Tokyo in November, while an underestimation is found in the surrounding regions of Tokyo in April and June. NOx emissions from the surrounding suburb cities may be underestimated. These simulations also show that high concentrations occur over Tokyo Bay. These high concentrations are considered to result from ship emissions. The validity of this supposition, however, cannot be verified because of the absence of monitoring stations in the sea.

Figure 4 shows horizontal distributions of monthly observed and simulated concentrations of O_3 in the Kanto domain. The features of the distributions of simulated concentrations of O_3 and NO_2 are clearly opposite: the concentrations are low at the center of Tokyo and over Tokyo Bay, but are high in the surrounding areas. These distributions indicate that monthly simulated concentrations are strongly affected

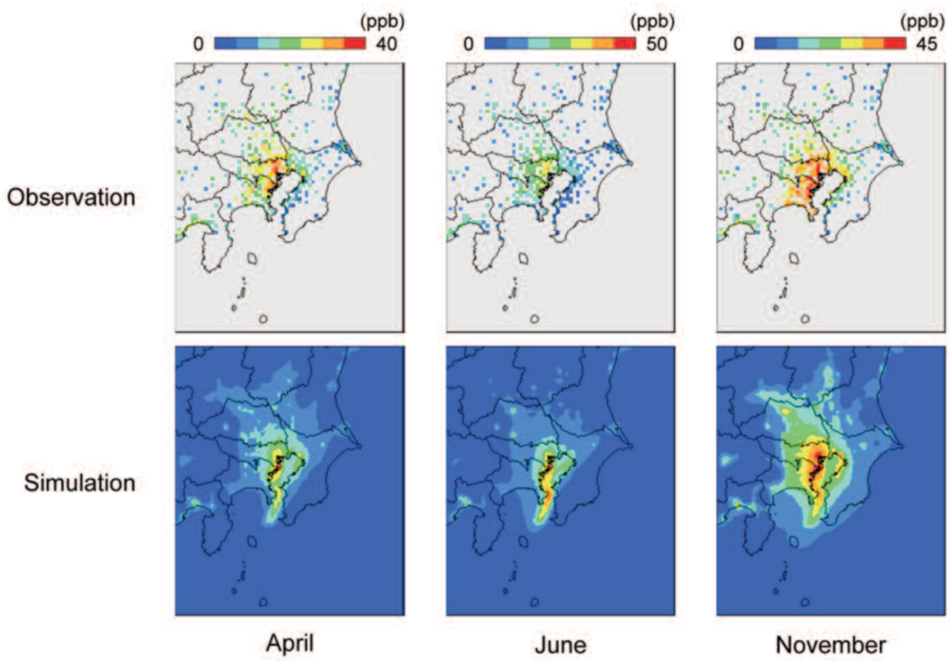

Fig. 3 Horizontal distributions of monthly observed and simulated concentrations of NO_2 in the Kanto domain in April, June and November in 2005. Observed concentrations at APMSs are allocated to corresponding 4 × 4 km grid cells, and grid cells in gray are those where there are no APMSs.

by titration due to NO emissions. Such features are also seen in the distribution of the observed concentration of O_3. The simulated background concentrations reach 60 ppb in April and June; the validity of this figure is unclear, however, because most of the APMSs established by local governments are usually not located in background areas. Overestimation is seen in the simulated concentrations of O_3 in the surrounding areas, especially in November, which implies an underestimation of NOx emissions in the surrounding suburbs.

Figure 5 shows horizontal distributions of monthly observed and simulated concentrations of SPM in the Kanto domain. The distributions of observed

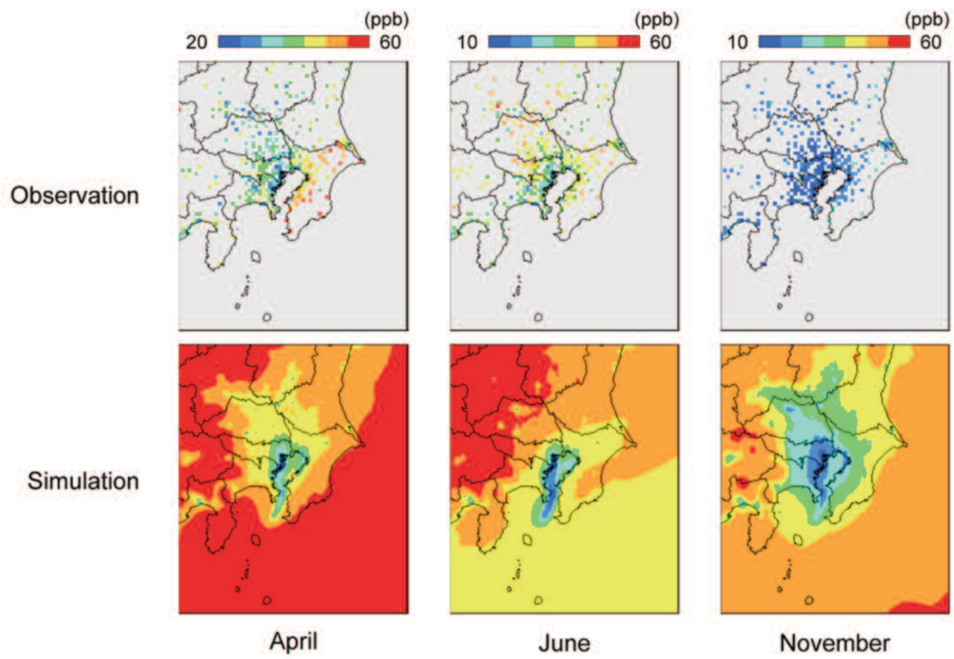

Fig. 4 Horizontal distributions of monthly observed and simulated concentrations of O_3 in the Kanto domain in April, June and November in 2005.

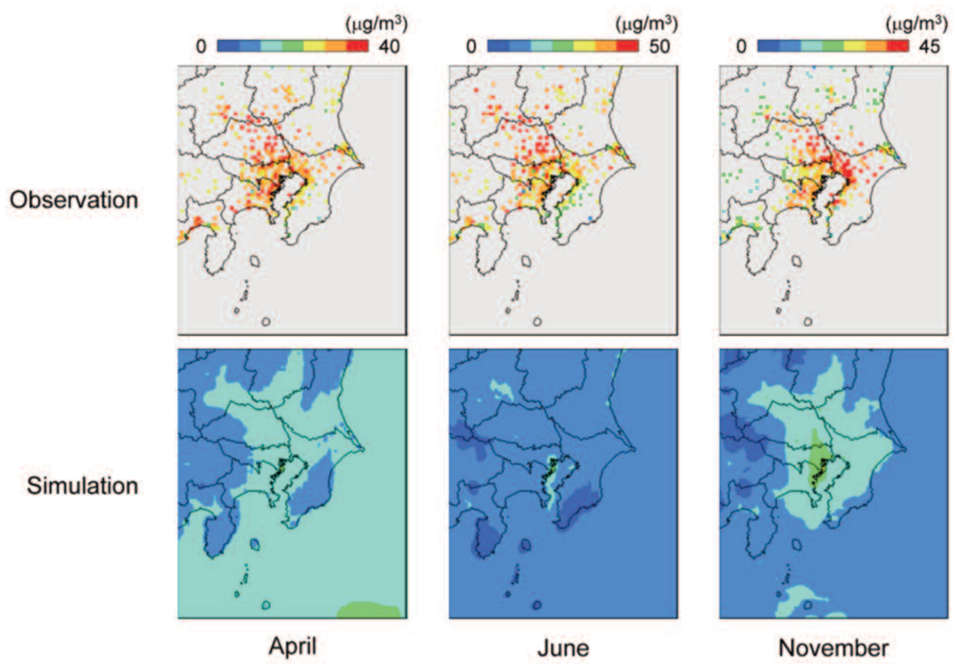

Fig. 5 Horizontal distributions of monthly observed and simulated concentrations of SPM in the Kanto domain in April, June and November in 2005.

concentrations of SPM show that high concentrations are not concentrated over the heart of the Tokyo metropolitan area, and they spread to the north, where emissions are relatively low. Therefore, they seem to be affected by the formation of secondary aerosol components from precursors during the transport from the Tokyo metropolitan area to the downwind areas. Similar spatial features are qualitatively captured in the distributions of the simulated concentrations of SPM in April and November, while the absolute values of the concentrations are significantly underestimated, especially in June.

3. 2 Time Series Variations

Figure 6 shows time series variations of the daily observed and simulated concentrations of NO_2, O_3 and SPM, which are averaged over 26 APMSs located in Tokyo's twenty-three special wards. The simulated concentrations over East Asia, Japan and Kanto domains are shown. The simulation captures part of the day-to-day variations of the three species. The NO_2 concentrations are slightly underestimated, while the O_3 concentrations are overestimated. This tendency is most evident for O_3 in the East Asia domain. Meanwhile, in the Japan and Kanto domain, the higher resolution tends to bring the simulations closer to the observations as reflected by the correlation coefficients, MNB and MNE. Obviously, the representation of the impacts of strong titration by NO are much better in the high-resolution simulations than the coarse-resolution simulations. However, the simulations significantly overestimate O_3 concentrations in November. More appropriate settings for the boundary concentrations of O_3 may be required. Moreover, the NO_2 underestimation and O_3 overestimation appear to be correlated in Fig. 6. The performance would be improved if more titration by NO occurred. G-BEAMS uses a top-down approach that allocates emissions to finer meshes using surrogates. If the appropriate surrogates are not used, the emissions tend to be concentrated over the heart of urban area, where social activities are high. Titration by NO would occur more effectively when the NO emissions are spatially diffused to the rural areas.

The simulations had difficulty in reproducing the

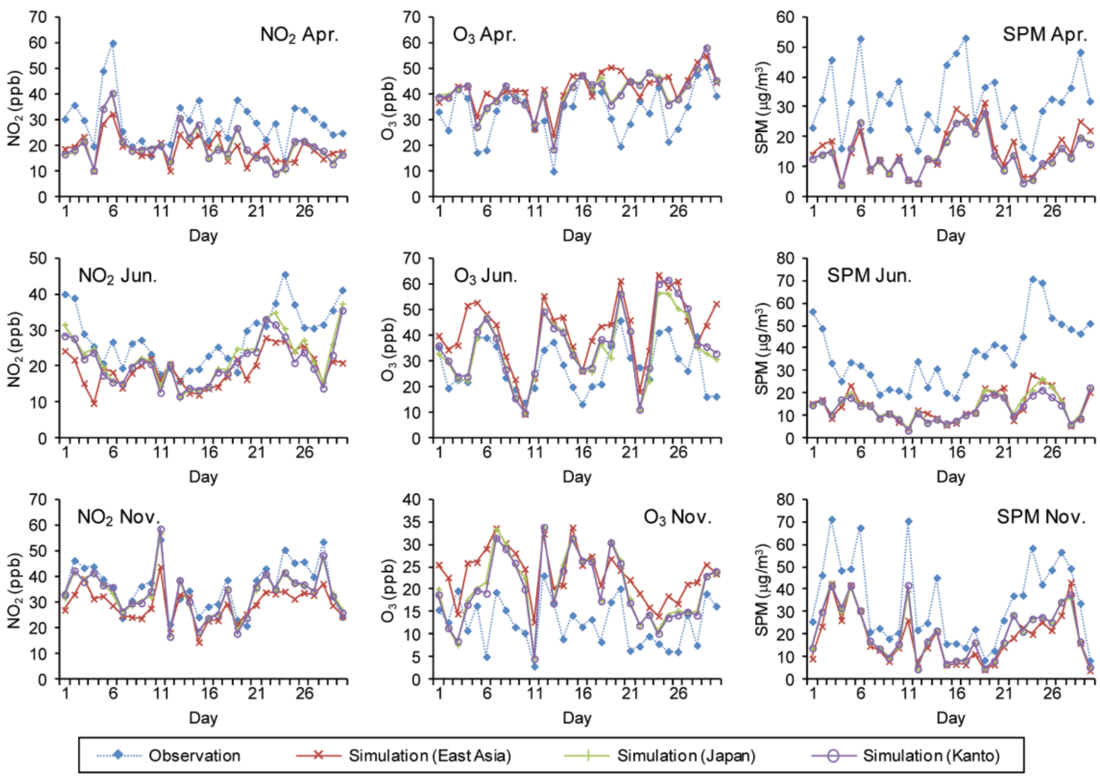

Fig. 6 Time series variations in daily observed and simulated concentrations of NO_2, O_3 and SPM, which are averaged over 26 APMSs located in Tokyo's twenty-three special wards. Simulated concentrations in East Asia, Japan and Kanto domains are shown.

concentrations of SPM. The absolute values of MNB are approximately coincident with MNE values, indicating that the SPM concentrations are underestimated for almost all days and over all of Tokyo's twenty-three special wards. In addition, the high resolution does not improve the performance in April and June. Generally, concentrations are more affected by stable meteorological conditions in November, while they are more affected by photochemical reactions in the atmosphere in April and June. One of the possible reasons for the underestimation in April and June is the formation of secondary aerosol components in and around the Tokyo metropolitan area. The simulations can, however, reproduce stagnant conditions in winter. There may also be problems with the observed concentrations of SPM. Most APMSs measure SPM concentrations using the beta absorption technique, which may have interference from water vapor, condensation and the evaporation of semi-volatile components. Such interference may be one of the reasons the simulation performance is worse under the high-temperature and humid conditions typically experienced in June. Rainfall amount may be another factor that affects the SPM performance.

In 2005, there was only one APMS in Tokyo's twenty-three special wards, where the simultaneous measurement of SPM and $PM_{2.5}$ was conducted. This APMS, called Ayase, is located 10 km northeast of the heart of the Tokyo metropolitan area. **Figure 7** shows time series variations of daily observed and simulated concentrations of SPM and $PM_{2.5}$ at the Ayase APMS. There are evident differences between the observed concentrations of SPM and $PM_{2.5}$. The average ratio of the observed concentrations of $PM_{2.5}$ to the observed concentrations of SPM is approximately 70%. Meanwhile, differences between the simulated concentrations of SPM and $PM_{2.5}$ are negligible. The above reveals that most of the underestimation of SPM comes from the SPM-$PM_{2.5}$ fraction. Coarse particles are mainly derived from mechanical processes, including soil dust, sea salt, and pollen, among others. There may be ignorance or underestimation of the emissions of these particles in the emission inventory. The simulation performance of $PM_{2.5}$ concentrations should be better than that of SPM concentrations. Extensive evaluations of $PM_{2.5}$ concentrations are required after the establishment of monitoring networks due to the implementation of new EQSs for $PM_{2.5}$ in Japan.

3. 3 Correlation

Figure 8 shows correlations and regression lines between daily observed and simulated concentrations of NO_2, O_3 and SPM at 26 APMSs located in Tokyo's twenty-three special wards. The simulated concentrations in the Kanto region are shown. The simulated concentrations of NO_2 have a good correlation with the observed concentrations, especially in November. However, some high concentrations of NO_2 could not be reproduced in the simulations in April and June. The correlation coefficients for NO_2 are the highest in the Kanto domain and the lowest in the East Asia domain. The correlation of observed and simulated concentrations of NO_2 is effectively improved by the high-resolution simulations. For O_3 concentrations, the slope of the regression line is almost 1.0 in June, while it is lower in April and November. Higher intercepts indicate that the simulations had difficulty reproducing low concentrations in the target months. The correlation coefficients for O_3 concentrations are the highest in the Kanto domain and the lowest in the East Asia domain.

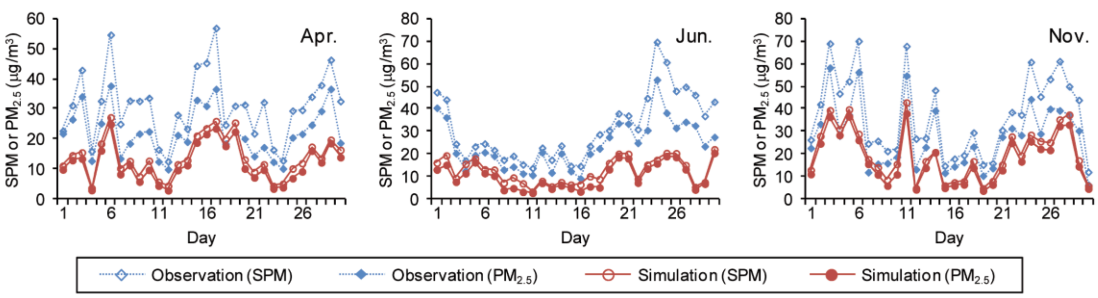

Fig. 7 Time series variations in daily observed and simulated concentrations of SPM and $PM_{2.5}$ at Ayase APMS, where simultaneous observations of SPM and $PM_{2.5}$ were made in the target months. Simulated concentrations in the Kanto domains are shown.

The high-resolution simulations are effective for the correlation of observed and simulated concentrations of O_3 and NO_2. Meanwhile, systematic problems are seen in the correlation of observed and simulated concentrations of SPM. The slope of the regression line is low, especially in June, where the slope is 0.16 but the correlation coefficients are close to those for O_3 and NO_2.

Based on the simulation performance in terms of horizontal distributions, time series variations and statistic indicators, the high-resolution simulations using the three-dimensional regional air quality simulation framework were able to simulate the air quality over the Tokyo metropolitan area more accurately, although there are still issues in reproducing SPM concentrations.

4. Case Studies

The three-dimensional regional air quality simulation framework described in the previous sections has been applied to various studies.(4) This section describes two case studies in which predictions of future air quality over the Tokyo metropolitan area were conducted. The monthly average concentrations over Tokyo's twenty-three special wards in April, June and November are considered as representative of the simulation case. The total emissions shown in this section are the averages of emissions in April, June and November, which are aggregated over the specified area for the Automobile NOx-PM law in the Kanto domain, which encloses Tokyo's twenty-three special wards.

4. 1 Impacts of Future Domestic Anthropogenic Emissions

Stringent emission regulations have been implemented in Japan. However, the impacts of these emission regulations on air quality do not immediately appear because motor vehicle fleet turnover takes several years. Consequently, the impacts of existing regulations on future air quality should be predicted to determine whether more stringent regulations are required. Regulations have also been implemented on anthropogenic sources other than motor vehicles. In

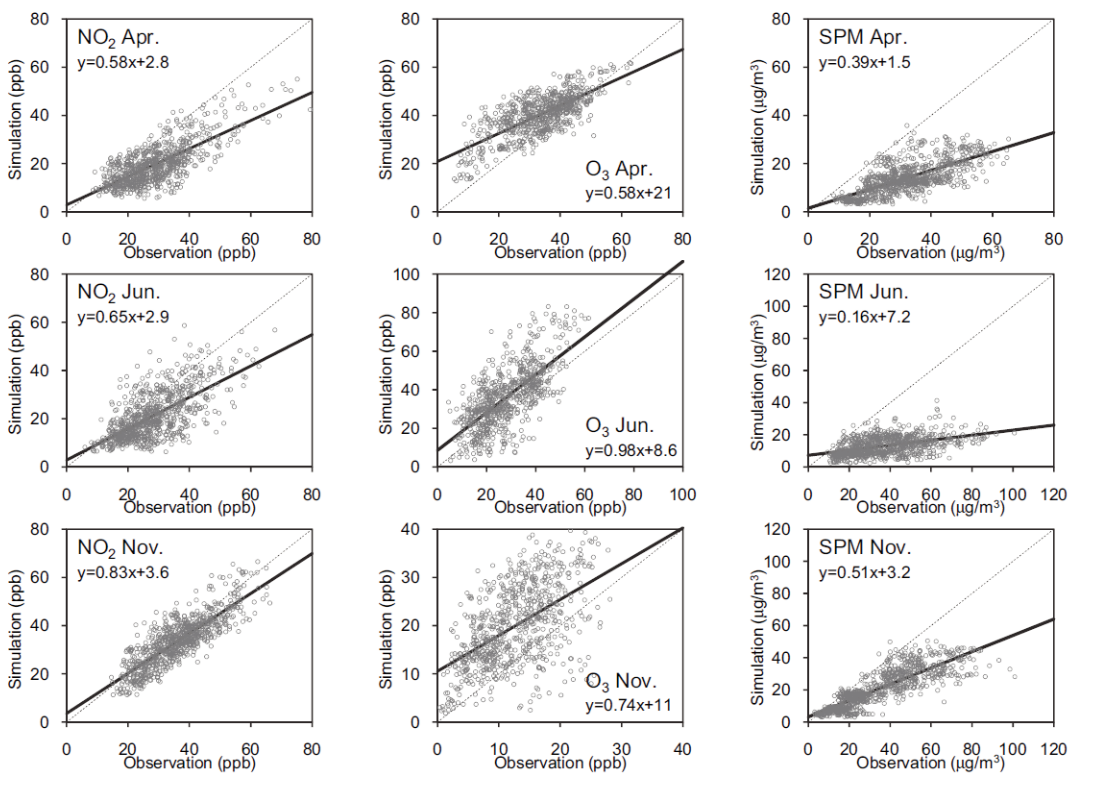

Fig. 8 Correlations and regression lines between daily observed and simulated concentrations of NO_2, O_3 and SPM at 26 APMSs located in Tokyo's twenty-three special wards. Simulated concentrations in the Kanto region are shown.

this case study, four simulation cases (All-05, Veh-15, Veh-20 and All-20), which are listed in **Table 2**, were executed to analyze the impacts of existing regulations on the future air quality over the Tokyo metropolitan area. Emissions in year 2005 were used in Case All-05. Estimated vehicle emissions in the years 2015 and 2020 were used in Case Veh-15 and Veh-20, respectively, to evaluate the impacts of existing regulations on vehicle emissions. When future vehicle emissions were estimated, the fleet turnover rate was assumed to maintain the same distribution of vehicle age. Future possible changes in vehicle population, vehicle mileage traveled and other related parameters were not taken into account. The emission reduction rates in 2020, which were estimated based on existing regulations on other anthropogenic sources, were applied in Case All-20 to analyze their impacts. Changes in emissions from Asian countries are not considered in this case study.

Figure 9 shows the estimated emissions of NOx, NMVOC and $PM_{2.5}$ for four cases in the specified area for the Automobile NOx-PM law in the Kanto domain. The NOx emissions are reduced by 30.6%, 34.3% and 44.5% in Cases Veh-15, Veh-20 and All-20, respectively, from Case All-05. The contribution of vehicles and off-road mobile sources becomes smaller, while large point sources show the largest contribution in Case All-20. NMVOC emissions are reduced by 10.2%, 10.6% and 18.1% in Cases Veh-15, Veh-20 and All-20, respectively, from Case All-05. Evaporative emissions, including emissions from solvent use, are reduced in Case All-20, though it still shows the largest contribution. The contribution of biogenic VOC emissions is relatively small because vegetation is limited in extent in the Tokyo metropolitan area. $PM_{2.5}$ emissions are reduced by 33.4%, 34.8% and 41.7% in Cases Veh-15, Veh-20 and All-20, respectively, from Case All-05. The contribution of vehicle exhaust becomes negligible, while the contribution of tire wear and resuspended road dust emissions becomes the largest in Case All-20.

Figure 10 shows the calculated average monthly concentrations of NO_2, O_3 and $PM_{2.5}$ over Tokyo's twenty-three special wards in April, June and November in the four cases. The upper figures show absolute concentrations, while the lower figures show the rate of change in the concentrations in each case with respect to that in Case All-05. The NO_2 concentrations are reduced by 31.6 to 41.2% in Case All-20 from Case All-05. The reduction range of NO_2 concentrations is similar to that of NOx emissions. In Japan, NO_2 concentrations have not been significantly reduced, even though NO concentrations have been effectively reduced in recent years. It is believed that this phenomenon is related to the nonlinear chemistry of

Table 2 Years and scenarios of emissions used in the case studies.

Case	Vehicle emissions in Japan	Other anthropogenic emissions in Japan	Emissions out of Japan
All-05	2005	2005	2005
Veh-15	2015	2005	2005
Veh-20	2020	2005	2005
All-20	2020	2020	2005
REF-20	2020	2020	2020 (REF)
PSC-20	2020	2020	2020 (PSC)
PFC-20	2020	2020	2020 (PFC)

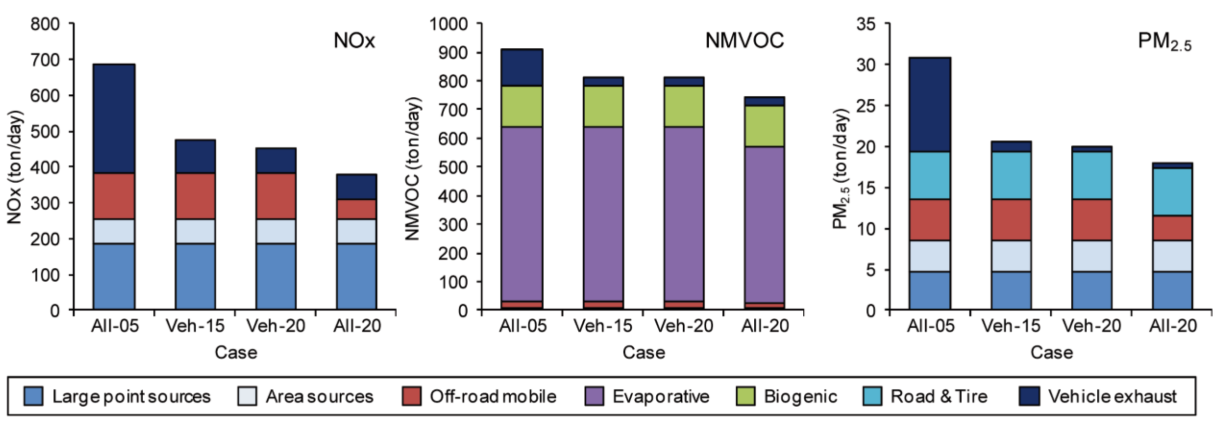

Fig. 9 NOx, NMVOC and SPM emissions estimated for Cases All-05, Veh-15, Veh-20 and All-20 in the specified area for the Automobile NOx-PM law in the Kanto domain.

NOx. Most NOx is emitted as NO. NO reacts with O_3 and is then converted to NO_2 in the atmosphere. Therefore, NO_2 formation is not limited by NO, but mostly by O_3 when NO concentrations are sufficiently higher than O_3 concentrations and vice versa. The result implies that the NO_2 formation regime may change from an O_3-limited regime to a NO-limited regime. NO_2 concentrations, then, may be effectively reduced in the near future. A decrease in NO_2 formation in the NO-limited regime is equal to a decreased titration of O_3 by NO, which is why O_3 concentrations are significantly increased by 56.4% in November in Case All-20 from Case All-05. It must be noted that the O_3 concentrations are initially very low in November due to the strong titration by NO. An increase in O_3 concentration corresponds to a return to the background level. O_3 concentrations do not significantly increase in April and June, because photochemical reactions are more active compared with those in November, and the suppression of O_3 formation occurs because of the reduced emissions of NOx and NMVOC. The $PM_{2.5}$ concentrations are reduced by 14.7 to 21.9% in Case All-20 from Case All-05. The reduction range of $PM_{2.5}$ concentrations is smaller than that of total $PM_{2.5}$ emissions. The reduction of secondary aerosol components seems smaller compared to that of the primary aerosol components, and it is presumable that long-range transport from outside Japan has an impact on $PM_{2.5}$ concentrations.

4. 2 Impacts of Future Emissions in Asian Countries

Emissions in Asian countries in 2005 were used in all cases of the case study described in the above subsection. However, emissions in Asian countries may certainly change in the future because of rapid development in these years. Analyzing the impacts of future emissions in Asian countries is essential for developing effective measures to improve the air quality over Japan. Future emissions in Asian countries estimated under three scenarios are provided in REAS[18]. The three scenarios include the Reference scenario (REF), the Policy Succeed Case scenario (PSC) and the Policy Failed Case scenario (PFC). In this case study, four simulation cases (All-20, REF-20, PSC-20 and PFC-20), as listed in Table 2, were executed to evaluate the impacts of future emissions in Asian countries on the future air quality over the Tokyo metropolitan area. REAS emissions estimated under the REF, PSC and PFC scenarios in 2020 were used in Cases REF-20, PSC-20 and PFC-20, respectively. The emissions in 2020 were used for domestic sources in all cases.

Figure 11 shows the calculated average monthly concentrations of NO_2, O_3 and $PM_{2.5}$ over Tokyo's twenty-three special wards in April, June and November in the four cases. The upper figures show

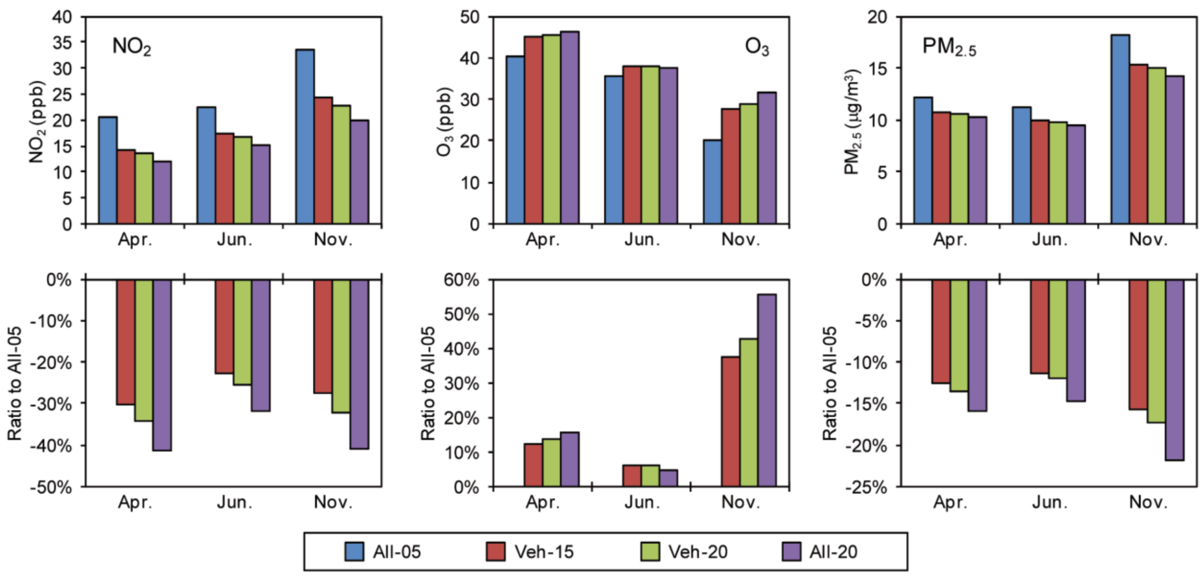

Fig. 10 Calculated average monthly concentrations of NO_2, O_3 and $PM_{2.5}$ over Tokyo's twenty-three special wards in April, June and November in Cases All-05, Veh-15, Veh-20 and All-20. Upper figures show absolute concentrations, while lower figures show the rate of change in the concentrations in each case with respect to that in Case All-05.

absolute concentrations, while the lower figures show the rate of change in the concentrations in each case with respect to that in Case All-20. The NO_2 concentrations were almost identical in the four cases, indicating that NO_2 concentrations are not affected by emissions in Asian countries. One of the reasons for this tendency is the short lifetime of NO_2 in the atmosphere, which prohibits it from being directly transported from outside Japan. Another possible reason is the NO_2 formation regime described in the previous section. The NO_2 formation regimes are NO-limited for all cases, and NO_2 formation is not affected by O_3. Meanwhile, the concentrations of O_3 and $PM_{2.5}$ are affected by emissions in Asian countries because their lifetimes in the atmosphere are longer than that of NO_2. O_3 concentrations are increased by 1.4 to 2.2%, while $PM_{2.5}$ concentrations are decreased by 4.0 to 9.4% in Case REF-20 from Case All-20. The concentrations of both species are decreased in Case PSC-20, while they are increased in Case PFC-20. The future trend of emissions in Asian countries may have an impact on the future air quality over the Tokyo metropolitan area in terms of O_3 and $PM_{2.5}$.

It must be noted that future emissions in Japan, as discussed in the previous case study, have a larger impact compared with future emissions in Asian countries. Monthly average concentrations of O_3 are predicted to increase mainly due to the decreased titration by NO, especially in winter. As described in the introduction, O_3 concentrations gradually increase, whereas concentrations of precursors decrease in Japan. Although the reasons for the tendency have not been definitively identified, the reduction in NOx emissions is likely to be one of the key factors increasing O_3 concentrations due to decreased titration by NO.

5. Summary

A three-dimensional regional air quality simulation framework was developed that allows high-resolution emissions to be estimated and that is suitable for simulations of air quality with a resolution of a few kilometers. In this study, the simulation performance for concentrations of NO_2, O_3 and SPM was evaluated. High-resolution simulations provided an improved simulation performance for NO_2 and O_3 concentrations. However, the SPM concentrations were significantly underestimated, so additional approaches should be considered to mitigate the underestimation. The simulation performance of $PM_{2.5}$ concentrations is expected to be better because the underestimation of SPM concentrations mainly occurs in the coarse fraction of SPM.

Two case studies were conducted to predict the impacts of future anthropogenic emissions in Japan and the long-range transport of pollutants on the air quality over the Tokyo metropolitan area. Future

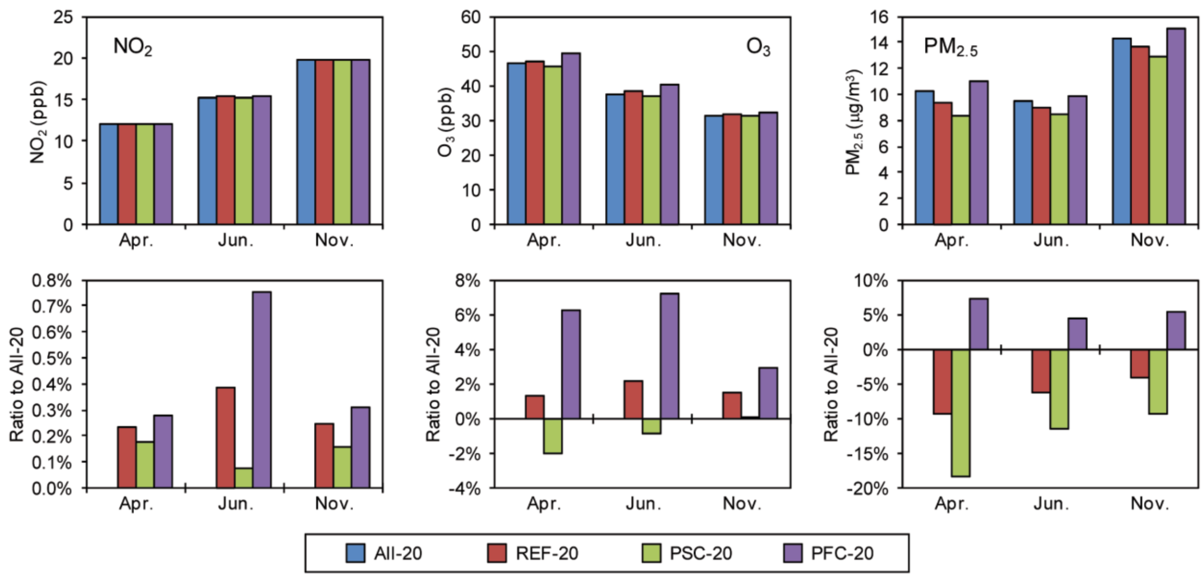

Fig. 11 Calculated average monthly concentrations of NO_2, O_3 and $PM_{2.5}$ over Tokyo's twenty-three special wards in April, June and November in Cases All-20, REF-20, PSC-20 and PFC-20. Upper figures show absolute concentrations, while lower figures show the rate of change in the concentrations in each case with respect to that in Case All-20.

anthropogenic emissions in Japan are estimated to be reduced due to existing emission regulations, which may result in an effective decrease in concentrations of NO_2 and $PM_{2.5}$. However, the O_3 concentrations are predicted to increase because titration by NO is decreased with decreasing NO emissions. The future trend of the long-range transport of pollutants is likely to affect the air quality over the Tokyo metropolitan area. Nevertheless, the emissions from Japan will have a greater impact than the long-range transport of pollutants.

This study reveals that the three-dimensional regional air quality simulation framework can provide valuable information useful for reducing concentrations of pollutants, especially secondary gaseous and aerosol species. Further evaluation is needed on individual components involved in photochemical reaction chains in the atmosphere as well as regulated pollutants such as NO_2, O_3 and PM to determine whether the simulation framework represents the formation of secondary pollutants. For $PM_{2.5}$, further evaluation is required because there are very limited observed data in Japan. A more extensive evaluation could be possible in the near future because a $PM_{2.5}$ monitoring network is scheduled to be built as a result of the introduction of new EQSs for $PM_{2.5}$ in Japan.

The simulation framework described in this paper is open to the public to provide opportunities for researchers to conduct studies on atmospheric processes over Japan and surrounding Asian countries. This framework may also be valuable for the policymakers of Japan to develop effective measures for reducing concentrations of pollutants, especially secondary pollutants.

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