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Photochemical roles of rapid economic growth and potential abatement strategies on tropospheric ozone over South and East Asia in 2030

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Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Abstract

A regional air quality simulation framework including the Weather Research and Forecasting modelling system (WRF), the Community Multi-scale Air Quality modeling system (CMAQ), and precursor emissions to simulate tropospheric ozone over South and East Asia is introduced. Concentrations of tropospheric ozone and related species simulated by the framework are validated by comparing with observation data of surface monitorings, ozone zondes, and satellites obtained in 2010. The simulation demonstrates acceptable performance on tropospheric ozone over South and East Asia at regional scale. Future energy consumption, carbon dioxide (CO₂), nitrogen oxides (NO_x), and volatile organic compound (VOC) emissions in 2030 under three future scenarios are estimated. One of the scenarios assumes a business-as-usual (BAU) pathway, and other two scenarios consider implementation of additional energy and environmental strategies to reduce energy consumption, CO₂, NO_x, and VOC emissions in China and India. Future surface ozone under these three scenarios is predicted by the simulation. The simulation indicates future surface ozone significantly increases around India for a whole year and around north eastern China in summer. NO_x is a main driver on significant seasonal increase of surface ozone, whereas VOC as well as increasing background ozone and methane is also an important factor on annual average of surface ozone in East Asia. Warmer weather around India is also preferable for significant increase of surface ozone. Additional energy and environmental strategies assumed in future scenarios are expected to be effective to reduce future surface ozone over South and East Asia.

1 Introduction

China, India, and other developing countries in Asia are accomplishing rapid economic growth. On the other hand, expanding economic activities have caused significant increase of energy consumption and carbon dioxide (CO₂) emissions as well as heavy

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Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

air pollution. One of key air pollutants is tropospheric ozone. Rising trends of tropospheric ozone have been observed at least in Japan (Kurokawa et al., 2009; Tanimoto, 2009), China (Xu et al., 2008; Tang et al., 2009; Wang et al., 2009), and India (Kulkarni et al., 2010). Ozone has adverse effects on human health (WHO, 2006) and vegetation (Mauzerall and Wang, 2001). In addition, tropospheric ozone is getting more attentions as one of short-lived climate pollutants (SLCPs). Reduction of tropospheric ozone may achieve co-benefits which would save human health and vegetation, and simultaneously mitigate near-term climate change (Shindell et al., 2012). Tropospheric ozone could be transported across countries as its lifetime in the atmosphere is relatively long among air pollutants (Akimoto, 2003). Therefore, multilateral strategies would be desired to reduce tropospheric ozone at regional scale.

Ozone is not directly emitted from emission sources. It is formed in the atmosphere from precursors including nitrogen oxides (NO_x) and volatile organic compounds (VOCs) via photochemical reactions. Numerical simulation models which represent photochemical reactions in the atmosphere are frequently applied to consider how to control precursor emissions in order to reduce tropospheric ozone. The community of global models has conducted several studies to predict future tropospheric ozone. For example, the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) conducted the intercomparison of the tropospheric ozone (Young et al., 2013) and its radiative forcing (Stevenson et al., 2013) in past and future years simulated by the participating global models. However, there are limitations in these studies. One of them is coarse resolutions applied in global models. They may not be suitable to simulate tropospheric ozone over densely polluted regions such as megacities in Asia. Another limitation is future scenarios. Global future scenarios originally for green house gases (GHGs) are often used in their studies. However, superficial extrapolations of GHGs to air pollutants would lead to misleading conclusions (Amann et al., 2013). ACCMIP utilized four Representative Concentration Pathways (RCPs) (Van Vuuren et al., 2011). RCPs have been developed on the basis of emissions and associated concentrations of greenhouse gases. Although emissions of atmospheric

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

air pollutants are also estimated in RCPs, Van Vuuren et al. (2011) mentioned that there are limitations in the use of RCPs for specific air pollution applications because they assumed that air pollution control becomes more stringent just as a result of rising income levels. That means they imply an endogenous strengthening of legislation and compliance beyond what is currently agreed (Amann et al., 2013). Moreover, each RCP has been developed by different modelling groups. Their assumptions on socio-economic drivers and climate policies are not necessarily consistent. Therefore, differences among RCPs cannot be directly interpreted as effects of specific policies. They are not suitable to consider what, when, and how strategies should be implemented in individual countries to suppress tropospheric ozone.

The purpose of this study is to predict tropospheric ozone over South and East Asia in 2030 under future scenarios by using three-dimensional regional air quality simulations of which the resolution is much finer than global models. Yamaji et al. (2008) conducted a similar study, but there are some notable differences. Our target domain covered India, which is one of key developing countries besides China in Asia. Three future scenarios were developed in this study. The first scenario assumed a business-as-usual pathway. Future changes in NO_x and VOC emissions not only in China but also all the other Asian countries including India were estimated. The second scenario assumed implementation of additional energy strategies aiming at reducing energy consumption and CO_2 emissions in China and India. They also resulted in reduction of NO_x and VOC emissions. The third scenario assumed implementation of additional environmental strategies aiming at reducing NO_x and VOC emissions in China and India in addition to energy strategies. Simulation results for these future scenarios made possible to evaluate effects of potential energy and environmental strategies implemented in China and India on tropospheric ozone over South and East Asia. Abatement costs required to implement these strategies were also estimated. The information regarding to effects and associated abatement costs of various energy and environmental strategies in China and India obtained in this study would be helpful to consider effec-

60km. 41 vertical layers are set from the ground to 5639 Pa above (approximately 19.5 km) in WRF-ARW, and they are collapsed into 28 layers in CMAQ. The bottom layer height is approximately 34 m in both models.

Boundary concentrations of CMAQ were retrieved from the results of Model for Ozone and Related chemical Tracers, version 4 (MOZART-4) (Emmons et al., 2010) which was driven by the meteorological fields simulated by the Goddard Earth Observing System Model, version 5 (GEOS-5). They were obtained from the National Center for Atmospheric Research (NCAR, 2013). They were updated every 6 h, and were temporally and spatially interpolated to the boundaries of the domain.

All the simulations discussed in this paper were performed for fourteen months from November 2009 to December 2010 whereas results for the first month were discarded as a spin-up period. Future changes in meteorological fields were not considered in this study.

2.2 Energy and emission data

We estimated energy consumption and emissions of various species including CO₂ in 2010 (referred as BASE) in 22 Asian countries (Bangladesh, Bhutan, Brunei, Cambodia, China, India, Indonesia, Japan, North Korea, South Korea, Laos, Malaysia, Mongolia, Myanmar, Nepal, Pakistan, Philipinnes, Singapore, Sri Lanka, Taiwan, Thailand, and Vietnam). We also developed three future scenarios (BAU0, PC0, and PC1) which were designed following the concept of Xing et al. (2011) for 2030. BAU0 assumed a business as usual pathway. The energy and environmental legislations which have been currently determined were reflected in BAU0. PC0 assumed additional legislations and technological developments to suppress energy consumption and CO₂ emissions in China and India. PC1 assumed additional legislations and technological developments to improve air quality in China and India besides PC0. Socioeconomic drivers like population and gross domestic product (GDP) were consistent in all the scenarios. Differences between BAU0 and PC0, and PC0 and PC1 correspond to effects of additional energy and environmental strategies implemented in China and India, re-

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



spectively. It should be noted that future changes in emissions of species except for CO₂, NO_x, and VOC were not considered in this study. PC0 and PC1 considered additional energy and environmental strategies implemented only in China and India. Energy consumption and emissions in the countries except for China and India are the same in BAU0, PC0, and PC1. Following subsections briefly introduce them whereas detailed descriptions of methodologies are found in references shown therein.

2.2.1 China

The database of anthropogenic energy consumption and emissions in China used in this study has been originally developed by Wang et al. (2011) and Xing et al. (2011) for current and future years. This study used the database recently updated by Zhao et al. (2013a, b, c). Emissions of sulphur dioxide, ammonia, PM₁₀, PM_{2.5}, black carbon, and organic carbon as well as CO₂, NO_x, and VOC have been estimated for current years. VOC emissions have been speciated into CB05 species groups (Wei et al., 2008). The horizontal resolution of the data used in this study is 36 km × 36 km.

The annual energy consumption and CO₂, NO_x, and VOC emissions estimated for 2010 (BASE) and 2030 under the three future scenarios in China are shown in Table 1. The VOC/NO_x emission ratios are also shown. The energy consumption increases by 64 % from BASE to BAU0 due to growing economic activities. The CO₂ emissions increase in similar magnitude by 66 %. Increases of the NO_x and VOC emissions are 35 % and 27 %, which are relatively lower than that of energy consumption. Current environmental legislations effectively reduce NO_x and VOC emissions per energy consumption. The additional energy strategies assumed in PC0 realizes 22 %, 30 %, 29 %, and 16 % decreases of the energy consumption and CO₂, NO_x, and VOC emissions, respectively. They make the NO_x and VOC emissions comparable to those in BASE. The additional environmental strategies assumed in PC1 further reduce the NO_x and VOC emissions by 55 % and 31 %, which become significantly lower than those in BASE. The VOC/NO_x ratio increases from 0.82 in BAU0 to 1.47 in PC1 as NO_x is

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



more effectively reduced than VOC by the assumed energy and environmental strategies.

2.2.2 India

The database of anthropogenic energy consumption and emissions in India used in this study has been developed by Sharma et al. (2014). Emissions of carbon monoxide, sulphur dioxide, and total suspended particulates (TSP) as well as CO₂, NO_x, and VOC have been estimated for current years. VOC emissions have been speciated into CB05 species groups. The horizontal resolution of the data used in this study is 36 km × 36 km.

The annual energy consumption and emissions estimated for 2010 (BASE) and 2030 under the three future scenarios in India are shown in Table 1. The energy consumption and CO₂ emissions dramatically increase by 3.2 and 2.9 times from BASE to BAU0 due to rapid economic growth assumed in the scenarios (8% per year). The NO_x emissions also significantly increase by 3.5 times. Although legislations to reduce NO_x emissions have been already implemented, growing economy increases dependence on coal power plants, heavy industries, and vehicles, of which NO_x emissions per energy consumption is relatively high. On the other hand, magnitude of increase of the VOC emissions is below twice. The assumed decrease of dependence on biomass fuel in domestic use, which is the dominant VOC emission source in BASE, suppresses increase of VOC emissions. That also results in significant changes of the VOC/NO_x ratios. It is 1.82 in BASE, which is much higher than China, and decreases to 1.02 in BAU0. The additional energy strategies assumed in PC0 realizes 26%, 32%, 31%, and 20% decreases of the energy consumption and CO₂, NO_x, and VOC emissions, respectively. The additional environmental strategies assumed in PC1 further reduce the NO_x and VOC emissions by 49% and 29%, which become comparable to those in BASE. The VOC/NO_x ratio increases to 1.68 in PC1 as NO_x is effectively reduced than VOC by energy and environmental strategies as in the case of China.

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



2.2.3 Other Asian countries

The data of anthropogenic energy consumption and emissions in other Asian countries than China and India were obtained from the results of the Greenhouse Gas and Air Pollution Interactions and Synergies (GAINS)-Asia model (Klimont et al., 2009; Amann et al., 2011) under World Energy Outlook 2011 (IEA, 2011) energy projections. GAINS-Asia has a capability to estimate energy consumption and emissions in 99 regions in Asia. In addition, the gridded data of which the horizontal resolution is $0.5^\circ \times 0.5^\circ$ are also available (IIASA, 2013). The results of GAINS-Asia model for 99 regions were used to develop speciation factors for each sector, fuel and region based on SPE-CIATE 4.3 (USEPA, 2013) because the detailed information of sectors and fuels are available. Then, they were applied to the gridded data to convert VOC emissions into CB05 species groups.

The annual energy consumption and emissions estimated for 2010 (BASE) and 2030 (BAU0) in major Asian countries (Japan, South Korea, Thailand, and Indonesia) are shown in Table 1. Japan and South Korea indicate a different pathway from Thailand and Indonesia. The energy consumption and CO₂ emissions are comparable for BASE and BAU0 in Japan and South Korea. The NO_x and VOC emissions in BAU0 are lower than BASE in both countries. On the other hand, energy consumption and emissions of CO₂ and NO_x significantly increase from BASE to BAU0 in Thailand and Indonesia. Decrease of the VOC emissions in Thailand and Indonesia in BAU0 is mainly due to decrease of 2-wheel motorcycles in vehicles fleets.

2.2.4 Other emissions

Monthly values in the Global Fire Emissions Database (GFED) (Van der Werf et al., 2010) version 3.1 were applied for biomass burning emissions. Emissions from agricultural burning in GFED were ignored because they are included in the anthropogenic emissions described in previous subsections. Emission Database for Global Atmospheric Research (EDGAR) version 4.1 (European Commission, 2010) was applied

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



for international shipping emissions. Hourly biogenic emissions were estimated by the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006) version 2.04. Future changes in these emissions were not considered in this study.

3 Model performance

A simulation for the case using emissions for 2010 (BASE) was performed. Horizontal distributions of the simulated seasonal mean surface ozone concentration are shown in Fig. 2a. Note that averaged values of daily 8 h maximum ozone concentration within the bottom 3 layers (approximately 134 m) are discussed as mean surface ozone concentration hereafter in this paper because they are the most relevant to consider adverse effects on human health and vegetation. Notable seasonal features are found in horizontal distributions. A zone with high concentration encompasses mid-latitude regions from India to Japan in spring and autumn. It is shifted northward and the highest concentration appears around north eastern China in summer. By the contrary, the concentration significantly decreases around the same region in winter. The concentration is low for a whole year around the equator while some hot spots are found around megacities. Excited oxygen atoms, which are dissociated from ozone, are likely to be scavenged by abundant water vapour instead of reproducing ozone.

The simulated results of ozone and related species were compared with observations to validate them as discussed in following subsections.

3.1 Surface ozone

Surface ozone concentration is continuously observed at the Acid Deposition Monitoring Network in East Asia (EANET) monitoring sites. The monthly mean surface ozone concentration observed at the ten EANET monitoring sites which are classified as “Rural” and “Remote” (Network Center for EANET, 2012) and the corresponding

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

values simulated in the bottom layer are compared as shown in Fig. 3. The results of MOZART-4 in its bottom layer are also shown in it. The locations of the twelve EANET monitoring sites are indicated in Fig. 1. Most of them are located in Japan and South Korea.

5 The observed values are high in spring and low in summer. Peak values and seasonal variations are reasonably reproduced by the simulation. One of the problems is that the low values observed in summer are overestimated in the simulation at several sites. Such a tendency is more evident in the results of MOZART-4. The simulation in this study makes effectively the simulated values closer to the observed ones as realized at Rishiri and Ochiishi. However, declines of the observed values in summer are still not fully reproduced at Sado-seki, Oki, Cheju, and Imsil, which are located in or around Japan Sea. It appears that the simulation in this study and MOZART-4 share common difficulties in reproducing low values around Japan Sea in summer. Southerly winds are dominant but westerly winds also appear for some days in summer around Japan and South Korea. Relationships between the hourly surface ozone and wind direction at Sado-seki and Oki were investigated (not shown). It turned out that westerly winds mainly cause overestimation at both sites. The simulated values are the highest around upwind regions of westerly winds in summer as shown in Fig. 2a. Lin et al. (2009) and Chatani and Sudo (2011) have showed that surface ozone was overestimated around corresponding regions in their simulations. Excess ozone may be transported downwind to South Korea and Japan on westerly winds and cause overestimation in this study, too. Lin et al. (2009) implied surface ozone is highly sensitive to cloud cover and monsoonal rainfall over these regions. Chatani and Sudo (2011) mentioned uncertainties in emission inventory and coarse resolution as well as potential missing pathways in the chemical mechanisms embedded in the model as possible reasons for overestimation. The simulation in this study as well as MOZART-4 may still have similar difficulties in accurately representing these factors.

25 There is no monitoring network similar to EANET in South Asia. Kumar et al. (2012) validated their simulation by comparing the simulated monthly mean values of surface

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

ozone with the observed data at the seven sites in India which had been published in the past literatures. The target years of the observed data were not necessarily consistent with their simulation. We follow Kumar et al. (2012), though a part of the data is substituted with the data published in more recent literatures. As a result, the observed data of surface ozone at Ahmedabad (Lal et al., 2000), Gadanki (Naja and Lal, 2002), Mt. Abu (Naja et al., 2003), Pune (Beig et al., 2007), Anantapur (Reddy et al., 2012), Nainital (Kumar et al., 2010), and Thumba (David and Nair, 2011) were picked up in this study. The locations of these seven sites are indicated in Fig. 1. The monthly mean surface ozone concentration observed at them and the corresponding values simulated in the bottom layer are compared as shown in Fig. 4. The results of MOZART-4 in its bottom layer are also shown in it.

The observed values are high in late winter or spring, and low in summer. Such seasonal variations are generally reproduced in most sites including Anantapur, where the target year of the observed values is coincident with the simulation. The model performance at all the sites is comparable to Kumar et al. (2012). One of notable differences from them is amplitudes of seasonal variations. Kumar et al. (2012) showed better performance on low values in summer, though peak values in winter were significantly overestimated. The performance of the simulation in this study is opposite with them. Peak values in winter are reasonably reproduced whereas low values in summer is slightly overestimated. The reason is probably due to seasonal variations in emissions. Information regarding to seasonal variations in emissions in India have not been available in the database described in Sect. 2.2.2, whereas Kumar et al. (2012) applied seasonal variations represented in the Reanalysis of Tropospheric Chemical Composition (RETRO) database. The model performance in this study could be improved if any seasonal variations in emissions are applied. In addition, the total amount of NO_x emissions developed in this study for India is less than the Intercontinental Chemical Transport Experiment – Phase B (INTEX-B) inventory (Zhang et al., 2009) which was used in Kumar et al. (2012). It may prevent significant overestimation of surface ozone in winter in this study.

3.2 Vertical ozone profile

Vertical concentration profiles of the simulated ozone were compared with those of the ozone observed by ozonesonde at three stations (Sapporo, Tsukuba, and Naha) in Japan (JMA, 2012), one station in Hong Kong (Hong Kong Observatory, 2012), and two stations (Delhi and Thiruvananthapuram) in India (India Meteorological Department, 2013) which are archived by World Ozone and Ultraviolet Radiation Data Centre (WOUDC). The locations of the six stations are shown in Fig. 1. Vertical profiles of the observed and simulated values at them are compared as shown in Fig. 5. Measurements have been conducted several times in each season at Sapporo, Tsukuba, Naha, and Hong Kong. Both of the observed and simulated values at these four stations are those averaged over all the measurements conducted within each season. On the other hand, any values observed at Delhi and Thiruvananthapuram in 2010 are not available. Therefore, the simulated seasonal mean values are compared with the observed values averaged over all the available measurements in each season during 2000–2011 in Fig. 5 following Kumar et al. (2012). High gradients of the values observed in the upper troposphere are well reproduced by the simulation. The older versions of CMAQ had a problem in simulating them adequately when results of global scale models were utilized as boundary concentrations (Lam and Fu, 2009). The latest version of CMAQ used in this study has updated the representation of turbulent mixing during stable conditions and updates to the vertical advection scheme to reduce numerical diffusion in the upper model layers. It appears this update has contributed to significant improvement of the performance on vertical profile. The profiles in the lower and middle troposphere are also well reproduced, but the simulated values tend to be slightly higher around the surface and lower in the middle troposphere than the observed values except for Thiruvananthapuram. The simulated values are almost constant from the surface up to 200 hPa whereas a weak gradient are found in the values observed at Thiruvananthapuram. Uncertainties in vertical diffusions may result in devi-

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



ations between the observed and simulated values, and cause excessive accumulation of ozone in summer in the lower layers of the simulation shown in Figs. 3 and 4.

3.3 Tropospheric column NO₂

Tropospheric column NO₂, which is one of major precursors of ozone, has been retrieved from the observations of the Ozone Monitoring Instrument (OMI) mounted on the Earth Observing System (EOS) Aura satellite. The second release of collection 3 OMI/Aura Level-2 NO₂ data product OMNO2 (Bucsela et al., 2013) available at the National Aeronautics and Space Administration (NASA) Goddard Earth Sciences Data and Information Services Center (2012) was used in this study. The retrieved data of tropospheric column NO₂ within the target domain without any problems were selected based on their quality flags. The corresponding simulated value at the same location and timing were also picked up. Horizontal distributions of their seasonal mean values are shown in Fig. 6. The observed and simulated monthly mean values averaged over the regions shown in Fig. 1 in China and India as well as other major countries (Japan, South Korea, Thailand, and Indonesia) are shown in Fig. S1 in the Supplement.

The observed values are high around populated regions including North China Plain (Huabei), Yangtze River Delta (Huadong), Pearl River Delta (Huanan), Seoul (South Korea), and Tokyo (Japan) in East Asia. They indicate a clear seasonal variation with high values in winter and low values in summer. These features are well reproduced by the simulation as shown by Wang et al. (2011) which used the consistent emissions with this study. The values are systematically lower in South and Southeast Asia because NO_x emissions are significantly lower than East Asia. Note that the ranges of tropospheric column NO₂ in Fig. S1 in the Supplement are different in East and South/Southeast Asia. The values are underestimated by the simulation except for summer in South and Southeast Asia. As discussed in Sect. 3.1, possible reasons of underestimated NO₂ are the total amount and seasonal variation of NO_x emissions developed in this study. Ghude et al. (2013) derived 1.9 TgNyear⁻¹ as the optimized total amount of NO_x emissions for India in 2005 by using an inverse technique and iterative

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



procedure to minimize gaps between their simulated values and the observed values derived from OMI. The total amount of NO_x emissions developed in this study for India discussed in Sect. 2.2.2 is equivalent to 1.7 TgNyear⁻¹. If economic growth during 2005–2010 is taken into account, the NO_x emissions developed in this study seems to be underestimated. Kumar et al. (2012) showed their simulation overestimated tropospheric column NO₂ over Indo-Gangetic Plain (Center) in winter, and underestimated it in rest of seasons and regions. Such a spatial difference in the performance is not found in this study. The horizontal distribution of the NO_x emissions developed in this study may prevent accumulating NO_x in Indo-Gangetic Plain.

3.4 Total column CO

CO is also an important precursor of ozone as well as a product in photochemical reactions involving VOCs. Therefore, validations for CO are valuable in terms of ozone formation and VOC emissions. Total column CO has been retrieved from the observations of the Measurements Of Pollution In The Troposphere (MOPITT) mounted on the Earth Observing System (EOS) Terra satellite. The MOPITT Version 4 (Deeter et al., 2010) Level 2 product was used in this study. The retrieved data of total column CO within the target domain without any problems were selected based on their quality flags. The corresponding simulated value at the same location and timing were also picked up. Horizontal distributions of their seasonal mean values are shown in Fig. 7. The observed and simulated monthly mean values averaged over the regions are shown in Fig. S2 in the Supplement.

The observed values are high from winter to spring in east China, Indochina peninsula, and India. The values in Indochina peninsula and India become much lower in summer and autumn. Such a seasonal variation is well reproduced by the simulation, though the higher values from winter to spring are slightly underestimated in this study. Biomass burning is active in the corresponding regions and the seasons in which the observed values are high. It implies that the GFED biomass burning emissions used in this study are underestimated. Pechony et al. (2013) also indicated that the GFED

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



emissions are significantly underestimated in South and Southeast Asia in their inverse modelling using MOPITT and Tropospheric Emission Spectrometer (TES) satellite data.

3.5 Tropospheric column ozone

Ziemke et al. (2006) has derived tropospheric column ozone by subtracting stratospheric column ozone observed by Microwave Limb Sounder (MLS) mounted on EOS from total column ozone observed by OMI. The data of their monthly mean tropospheric column ozone was obtained from the NASA Goddard Space Flight Center (2012). Horizontal distributions of their seasonal mean values are shown in Fig. 8. The simulated values at the locations and timings from the surface up to the tropopause at which total column ozone was retrieved by OMI without any problems were picked up based on quality flags of the version 3 of the OMI/Aura Ozone Total Column Level-2 data product (OMTO3). The tropopause was determined as the height at which a gradient of temperature becomes lower than 2 K km^{-1} in the values simulated by WRF (Kumar et al., 2012) above 316 hPa, which is the lowest altitude retrieved from MLS. Horizontal distributions of their seasonal mean values are also shown in Fig. 8. The observed and simulated monthly mean values averaged over the regions are shown in Fig. S3 in the Supplement.

The observed values are high over the mid latitude in spring. The zone with high values is shifted northward in summer, and causes elevated values over northeast China. Such features in horizontal distribution are well reproduced in the simulation. The observed values are highest in summer in most of East Asia as shown in Fig. S3 in the Supplement whereas surface ozone shown in Fig. 3 in corresponding regions is lowest in summer. The observed and simulated values in all months agree well in Huabei where surface ozone is highest in summer. Therefore, uncertainties in vertical profiles of ozone shown in Fig. 5 may be a major factor causing overestimation of surface ozone in East Asia as discussed in Sect. 3.3.

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Although some issues which should be solved to improve the model performance on ozone still remain, we have judged the regional air quality simulation framework constructed in this study has acceptable performance to predict tropospheric ozone at regional scale based on validations described in this section.

4 Future prediction

Simulations for the nine cases shown in Table 2 were performed to examine various factors on surface ozone in 2030. Future changes of surface ozone from 2010 to 2030 were evaluated in BASE and BAU0 cases. Only NO_x and VOC emissions were changed from BASE to BAU0 in BAU0nox and BAU0voc cases, respectively, to evaluate their effects separately. Boundary ozone concentration was homogeneously increased by 5 ppb in BAU0o3 to evaluate potential effects of increasing background ozone on tropospheric ozone within the target domain. Similarly, homogeneous methane concentration was increased by 400 ppb from the value used in the default CMAQ (1850 ppb) in BAU0ch4 to evaluate potential effects of increasing background methane. The ranges of increasing background ozone and methane were set based on the maximum changes predicted by Dentener et al. (2006). Temperature was homogeneously decreased by 5 °C in BAU0ta to see sensitivities of temperature on surface ozone. Effects of the additional energy and environmental strategies on surface ozone in 2030 were evaluated in PC0 and PC1.

4.1 Future surface ozone in the BAU scenario

Horizontal distributions of the seasonal mean surface ozone concentration simulated in BASE and BAU0 are shown in Fig. 2a and b, respectively. Significant increase of surface ozone around India in BAU0 is evident in all seasons. Dentener et al. (2006) also indicated significant increase of future ozone around India under their current leg-

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

isolation (CLE) scenario. Surface ozone around north eastern China in BAU0 is similarly higher in summer whereas it is slightly lower in winter than BASE.

Horizontal distributions of differences in the simulated seasonal mean surface ozone concentrations between BAU0nox and BASE, BAU0voc and BASE, and BAU0 and BASE are shown in Fig. 9. Increase of surface ozone around India and surrounding regions including south western China and Indochina in all seasons is exclusively affected by increasing NO_x . The effects of VOC are negligible or slightly negative around those regions. Increase of surface ozone over most of China in summer is also much affected by increasing NO_x except for megacities like Yangtze River Delta and Pearl River Delta while its magnitude is less than India. Ozone chemistry in India for whole year and China in summer seems to be in NO_x -sensitive regime (Sillman, 1999). Kumar et al. (2012) also implied that the ozone chemistry over India is NO_x -limited except for a part of Indo-Gangetic Plain in winter. On the other hand, negative effects of increasing NO_x appear around north eastern China in spring and autumn, and they expand to whole eastern China in winter. Increasing VOC has positive effects around the corresponding regions. Ozone chemistry in eastern China in winter seems to be in VOC-sensitive regime. Liu et al. (2010) indicated that ozone chemistry in eastern China is in VOC-sensitive regime in January and NO_x -sensitive regime in July. They also indicated that it is in NO_x -sensitive regime in central and western China and in VOC-sensitive regime in major Chinese megacities throughout the year. Itahashi et al. (2013) suggested that regime of ozone chemistry in East Asia is NO_x -sensitive in summer, VOC-sensitive in winter, and either NO_x - or VOC-sensitive during spring and autumn. The results obtained in this study are consistent with their findings. Factors that affect NO_x - and VOC-sensitive regimes include VOC/ NO_x ratios as well as meteorological conditions (Sillman, 1999). Table 1 shows VOC/ NO_x ratios in BASE are much higher in India than China. They may result in stronger sensitivities of NO_x around India. Effects of NO_x are negative in all seasons, and those of VOC are also negative in summer around Japan and South Korea. Although a part of them may be af-

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



NO_x emissions are increased in both countries in BAU0. It turns out that higher temperature is one of factors which cause higher ozone concentration. The weather condition around India is warm throughout the year. Therefore, increase of precursor emissions has larger importance in terms of ozone formation in India.

4.3 Effects of energy and environmental strategies on future surface ozone

Horizontal distributions of the seasonal mean surface ozone concentration simulated in PC1 are shown in Fig. 2c. Surface ozone concentration in PC1 is comparable or slightly lower than BASE. Horizontal distributions of differences in the simulated seasonal mean surface ozone concentrations between PC0 and BAU0 and PC1 and BAU0 are shown in Fig. 12. Surface ozone is effectively reduced by the additional energy and environmental strategies around the regions in which it significantly increases from BASE to BAU0. Though, the regions with increasing ozone are found around eastern China where ozone chemistry is in VOC-sensitive regime. They do not mean that energy and environmental strategies are not preferable to mitigate air pollution. Ozone isopleths against NO_x and VOC have non-linear shapes (Sillman, 1999). If NO_x is reduced in VOC-sensitive regime, ozone should increase until it goes into NO_x-sensitive regime. It is essential to reduce NO_x to pass over the ridge which divides VOC- and NO_x-sensitive regimes and go into NO_x-sensitive regime where ozone is effectively reduced. As shown in Fig. 12, the regions with increasing ozone are shrunk in spring and autumn. They imply that regime of ozone chemistry changes from VOC-sensitive to NO_x-sensitive as VOC/NO_x ratios increase due to more effective reduction of NO_x than VOC as shown in Table 1. However, it is difficult to reduce ozone in winter around eastern China. In fact, surface ozone is lower than surrounding regions as shown in Fig. 2b because titration by NO_x overwhelms ozone formation. Reduction of NO_x brings surface ozone back to its background level. Nonetheless, it is still necessary to reduce NO_x because it products like NO₂ and nitrate are also important air pollutants.

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



4.4 Regional summary

Figure 13 shows differences in the simulated monthly mean surface ozone concentrations among cases which are averaged over regions shown in Fig. 1 in China and India as well as other major countries (Japan, South Korea, Thailand, and Indonesia). As discussed in previous sections, different characteristics are found in eastern (Huabei, Dongbei, Huadong, and Huanan) and western (Xinan and Xibei) China. Surface ozone increases from BASE to BAU0, and is effectively reduced in PC0 and PC1 for whole year in western China as well as in India. Although similar responses are found in eastern China in summer, increase of NO_x from BASE to BAU0 results in decrease of surface ozone in winter. The additional energy and environmental strategies implemented in China and India assumed in PC0 and PC1 are effective to reduce surface ozone even in South Korea and Japan in summer. Decreasing domestic NO_x in BAU0 also contributes to decrease of surface ozone in summer in Japan. Whereas the difference between BAU0voc and BASE is negligible in India, BAU0-BASE* (the difference between BAU0 and BASE in which the difference between BAU0nox and BASE and the difference between BAU0voc and BASE are excluded) is evident. It means that sum of the individual effects of increasing NO_x and increasing VOC does not coincident with the combined effects of increasing NO_x and VOC. It implies that VOC has some effects on surface ozone in BAU0 in which the VOC/ NO_x ratio is significantly lowered than BASE. The additional energy and environmental strategies implemented in India and China assumed in PC0 and PC1 are effective to reduce surface ozone in late autumn in Thailand where surface ozone increases due to increasing NO_x in BAU0. Changes of surface ozone are negligible in Indonesia while the situation may be different around megacities locally.

Figure 14 shows the differences in the simulated annual mean surface ozone concentrations among cases which are averaged over regions in China and India as well as other major countries. Note that the ranges of vertical axis are different in East Asia (left) and South and Southeast Asia (right). The response in each case is simple and

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



increased in BAU0. The situation is a bit complex for annual mean surface ozone in East Asia. Increasing VOC as well as increasing background ozone and methane are also important for it. Various energy and environmental strategies are assumed in PC0 and PC1. It is desired to implement them in order of increasing abatement cost to suppress tropospheric ozone not only in China and India but also surrounding regions. Energy strategies assumed in PC0 could simultaneously suppress energy consumption and CO₂ emissions.

The simulation framework constructed in this study can be applied to not only tropospheric ozone but also other air quality issues like acid rain and haze. Especially, air pollution by particulate matter is also one of major environmental issues in Asia. Ambient particulate matter includes various primary and secondary components. Although this study estimated future NO_x and VOC emissions, it is necessary to estimate emissions of other components and precursors to predict future concentrations of particulate matter. In addition, strategies implemented in other Asian countries than China and India should affect the air quality over Asia whereas this study mainly focused on China and India. Future energy and emission scenarios in each country are required to evaluate their effects. We hope the outcome of this study will contribute to mitigate energy and environmental issues in Asia as a basis for considering desirable future pathways.

Supplementary material related to this article is available online at <http://www.atmos-chem-phys-discuss.net/14/9517/2014/acpd-14-9517-2014-supplement.pdf>.

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Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

- Xing, J., Wang, S. X., Chatani, S., Zhang, C. Y., Wei, W., Hao, J. M., Klimont, Z., Cofala, J., and Amann, M.: Projections of air pollutant emissions and its impacts on regional air quality in China in 2020, *Atmos. Chem. Phys.*, 11, 3119–3136, doi:10.5194/acp-11-3119-2011, 2011.
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Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Table 1. Annual energy consumption and CO₂, NO_x, and VOC emissions estimated for 2010 (BASE) and 2030 under three future scenarios. VOC/NO_x emission ratios are also shown.

	Country	BASE	BAU ¹	PC0 ²	PC1 ³
Energy consumption (EJyear ⁻¹)	China	121.7	199.5	155.0	
	India	26.4	83.8	62.2	
	Japan	21.4	21.5		
	South Korea	10.6	12.2		
	Thailand	5.3	7.7		
	Indonesia	8.2	13.5		
CO ₂ emission (Gtyear ⁻¹)	China	8.50	14.10	9.90	
	India	1.71	6.63	4.51	
	Japan	1.25	1.17		
	South Korea	0.63	0.62		
	Thailand	0.28	0.44		
	Indonesia	0.43	0.81		
NO _x emission (Mtyear ⁻¹)	China	26.1	35.4	25.2	11.5
	India	5.6	19.5	13.4	6.8
	Japan	1.70	0.86		
	South Korea	1.18	0.84		
	Thailand	0.86	1.16		
	Indonesia	1.58	1.88		
VOC emission (Mtyear ⁻¹)	China	22.8	29.0	24.4	16.8
	India	10.2	19.8	16.0	11.4
	Japan	1.41	1.11		
	South Korea	0.67	0.51		
	Thailand	0.91	0.81		
	Indonesia	3.98	3.68		
VOC/NO _x emission ratio	China	0.87	0.82	0.97	1.47
	India	1.82	1.02	1.19	1.68
	Japan	0.83	1.29		
	South Korea	0.57	0.61		
	Thailand	1.06	0.70		
	Indonesia	2.52	1.96		

¹ BAU0 scenario is based on current legislations and implementation status for energy saving policies and end-of-pipe control strategies.

² PC0 scenario assumes that new energy-saving policies will be released and enforced more stringently. End-of-pipe control strategies are the same as BAU0.

³ PC1 scenario assumes that new pollution control policies would be released and implemented. Energy-saving policies are the same as PC0.

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Table 2. Overview of simulation cases performed in this study.

Case	Emission	Other change
BASE	BASE	
BAU0nox	BAU0 (NO _x only) + BASE	
BAU0voc	BAU0 (VOC only) + BASE	
BAU0	BAU0	
BAU0o3	BAU0	+5 ppb boundary ozone
BAU0ch4	BAU0	+400 ppb background methane
BAU0ta	BAU0	−5 °C temperature
PC0	PC0	
PC1	PC1	

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

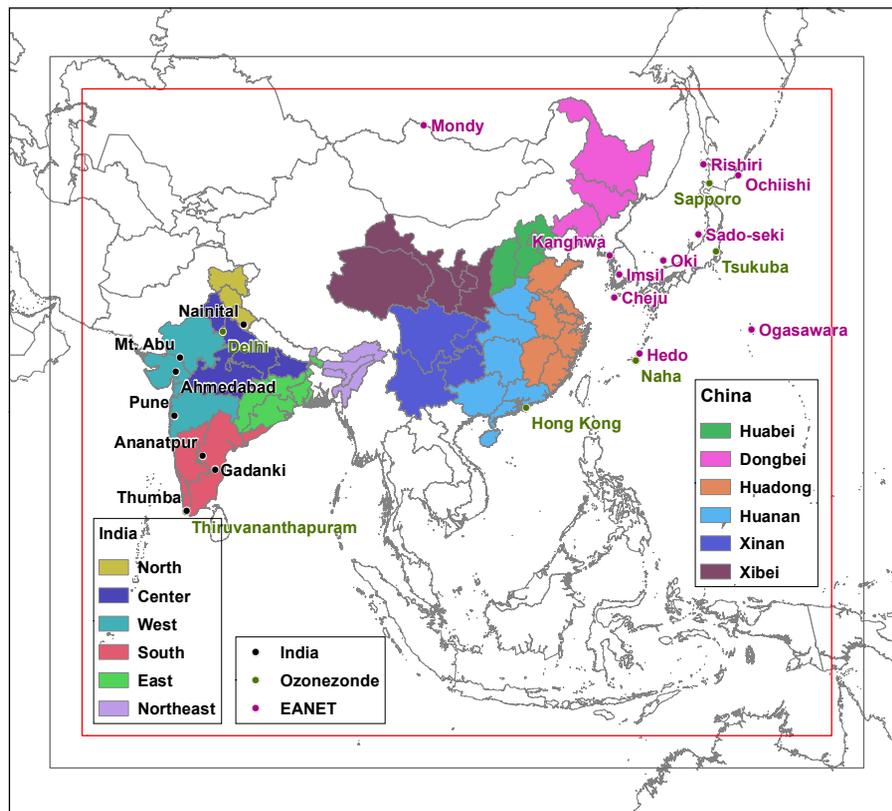


Fig. 1. Target domains of WRF-ARW (gray) and CMAQ (red). Monitoring sites of EANET, ozonezone, and India are indicated. Regions in China and India are color-coded.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

⏪ ⏩

⏴ ⏵

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

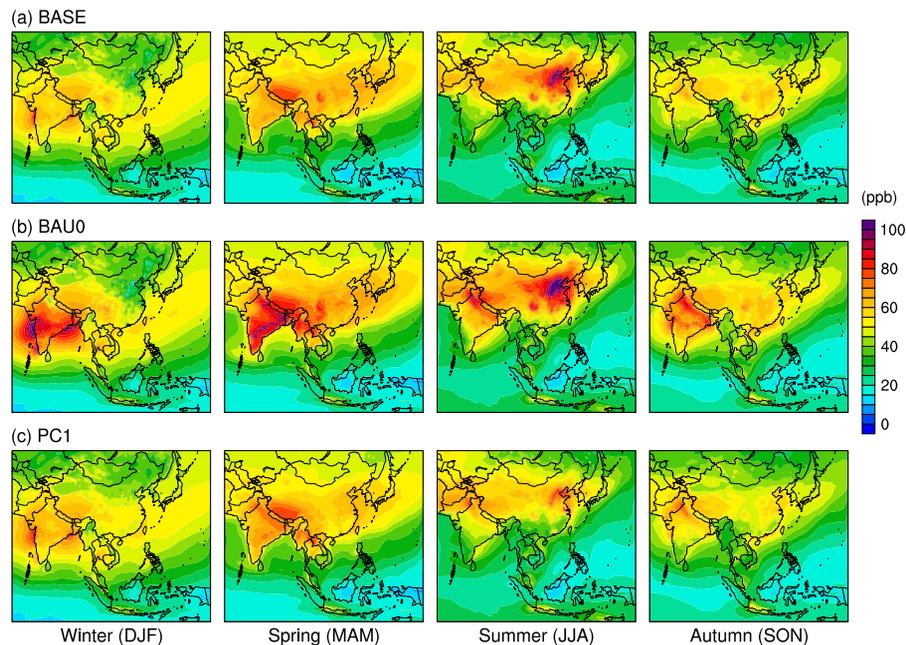


Fig. 2. Horizontal distributions of simulated seasonal mean surface ozone concentration in **(a)** BASE, **(b)** BAU0, and **(c)** PC1.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

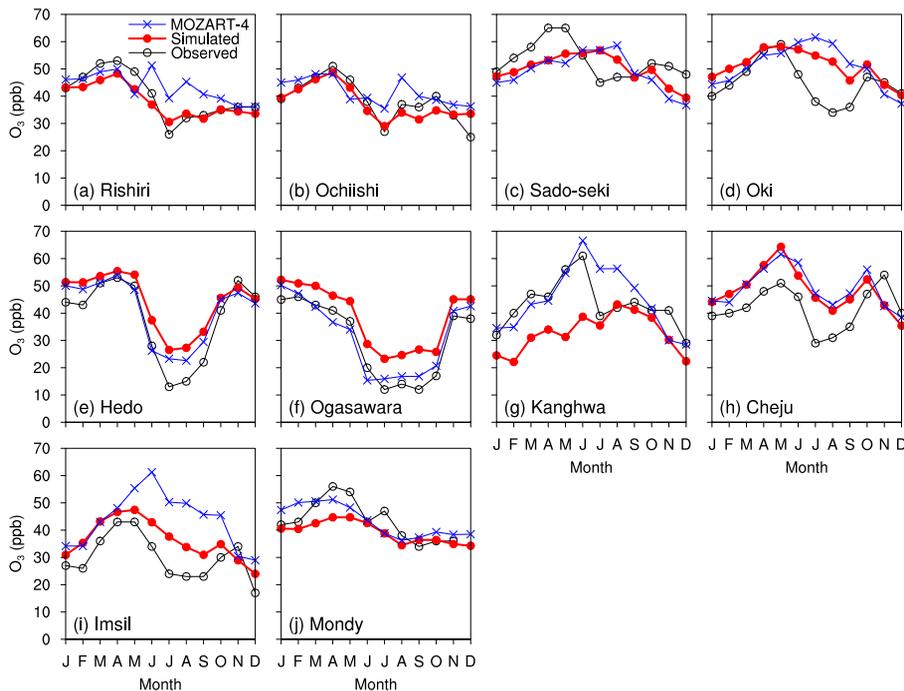


Fig. 3. Monthly mean observed and simulated surface ozone concentration at ten EANET monitoring sites. Results of MOZART-4 are also shown.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

⏪ ⏩

⏴ ⏵

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

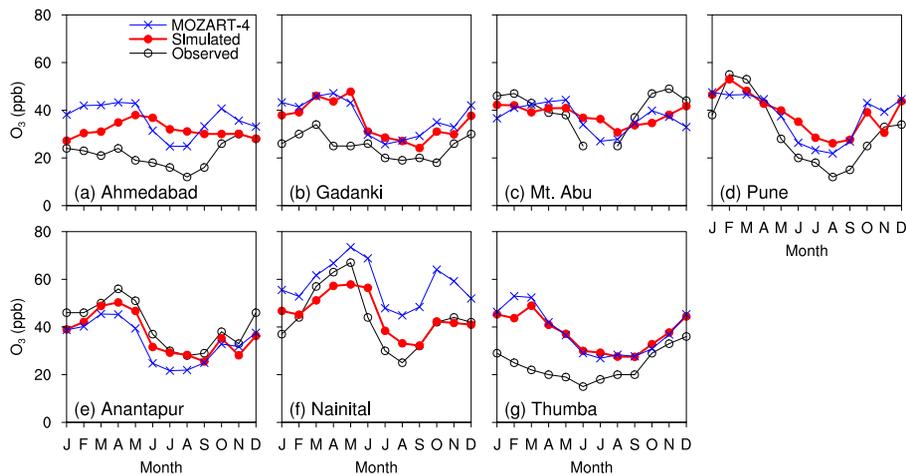


Fig. 4. Monthly mean observed and simulated surface ozone concentration at seven sites in India. Results of MOZART-4 are also shown.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

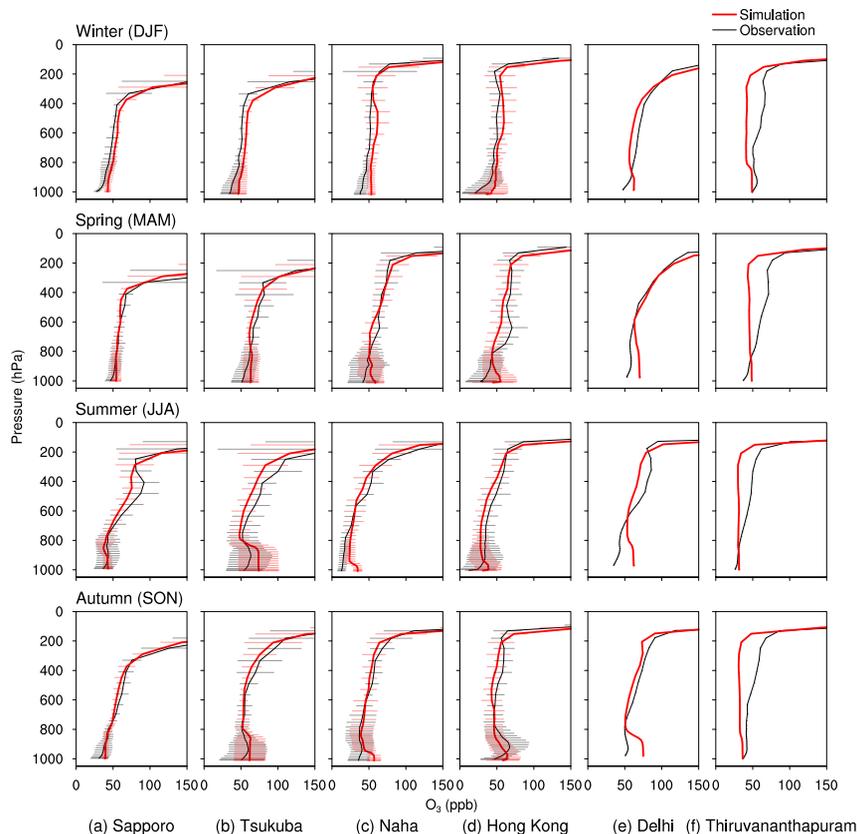


Fig. 5. Vertical profiles of observed and simulated ozone concentration at six monitoring stations. Values are averaged over all ozonesonde measurements within each season at Sapporo, Tsukuba, Naha, and Hong Kong. Error bars represent standard deviations. Simulated seasonal mean values are compared with values averaged over all available ozonesonde measurements within each season during 2000–2011 at Delhi and Thiruvananthapuram.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

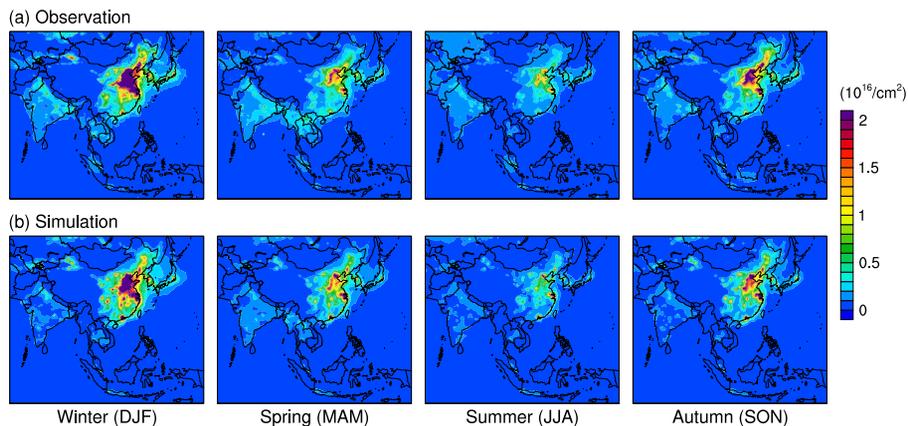


Fig. 6. Horizontal distributions of observed and simulated seasonal mean tropospheric column NO_2 .

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

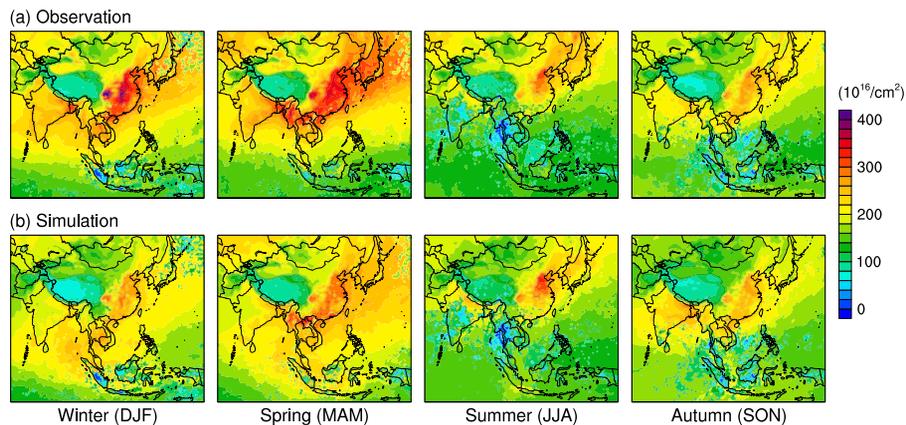


Fig. 7. Horizontal distributions of observed and simulated seasonal mean total column CO.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

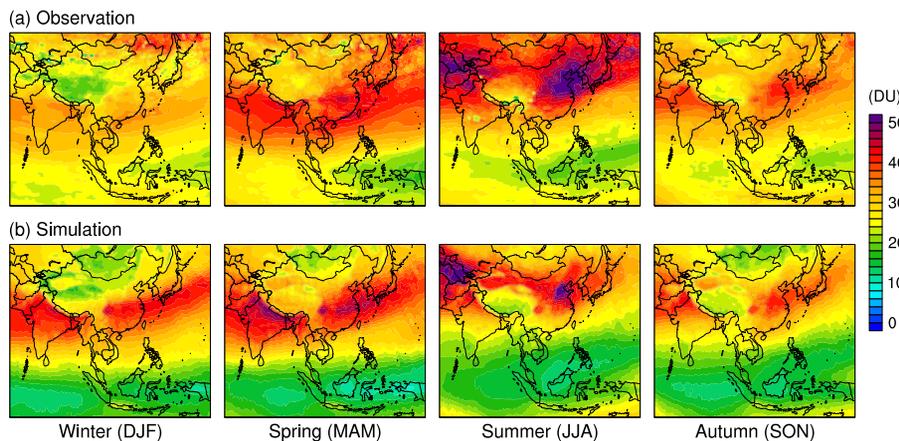


Fig. 8. Horizontal distributions of observed and simulated seasonal mean tropospheric column ozone.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[⏴](#)[⏵](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

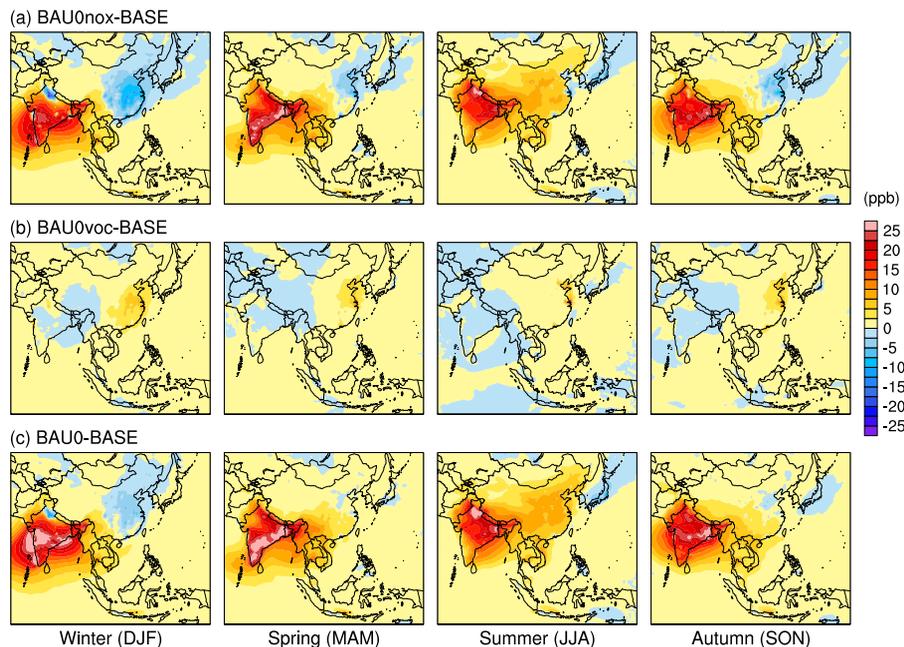


Fig. 9. Horizontal distributions of differences in simulated seasonal mean surface ozone concentrations between (a) BAU0nox and BASE, (b) BAU0voc and BASE, and (c) BAU0 and BASE.

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

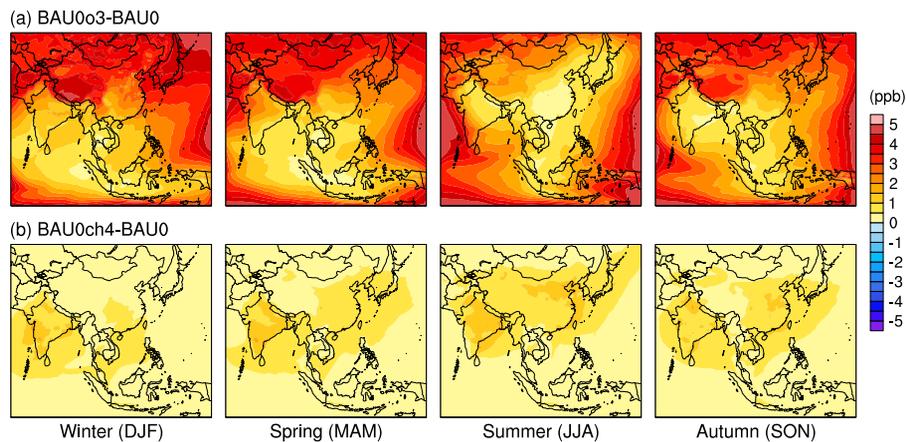


Fig. 10. Horizontal distributions of differences in simulated seasonal mean surface ozone concentrations between **(a)** BAU0o3 and BAU0, and **(b)** BAU0ch4 and BAU0.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[⏴](#)[⏵](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

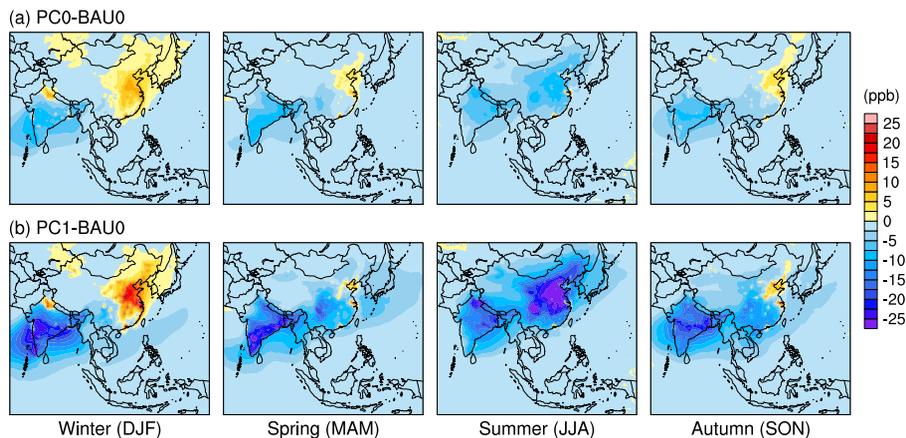


Fig. 12. Horizontal distributions of differences in simulated seasonal mean surface ozone concentrations between **(a)** PC0 and BAU0, and **(b)** PC1 and BAU0.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[⏴](#)[⏵](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Photochemical roles of rapid economic growth and potential abatement strategies

S. Chatani et al.

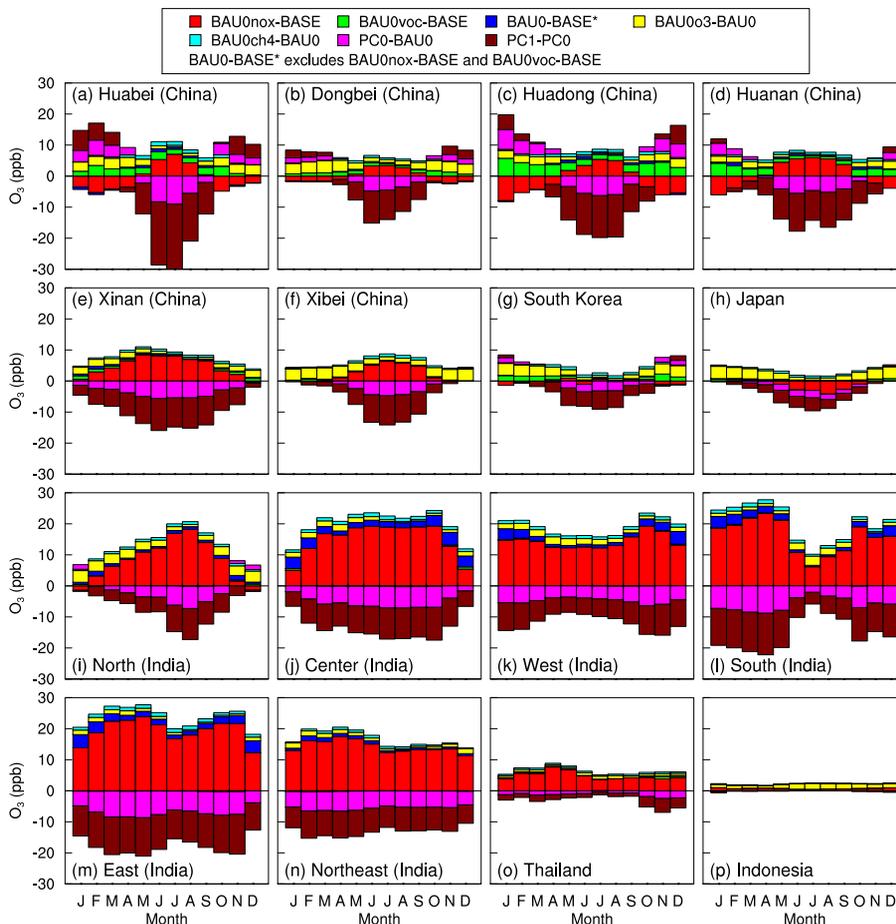


Fig. 13. Differences in simulated monthly mean surface ozone concentrations among cases which are averaged over regions in China and India as well as Japan, South Korea, Thailand, and Indonesia.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



