FUTURE PREDICTION OF SURFACE OZONE OVER EAST ASIA UP TO 2020

Kazuyo Yamaji* and Hajime Akimoto

Frontier Research Center for Global Change, Japan Agency for Marine Science and Technology, Yokohama, Japan

> Toshimasa Ohara and Jun-ichi Kurokawa National Institute for Environmental Studies, Tsukuba, Ibaraki, Japan

Itsushi Uno Research Institute for Applied Mechanics, Kyushu University, Kasuga, Fukuoka, Japan

1. INTRODUCTION

Tropospheric ozone (O₃) produced through photochemical reactions involvina primary pollutants such as nitrogen oxides (NOx) and nonmethane volatile organic compounds (NMVOC) is a major secondary pollutant. These O₃ precursors, which are mainly emitted due to fossil fuel uses, have rapidly increased over Asia during the last decade. Ohara et al. (2007) reported a NOx emission increase from 10.7 Tg in 1980 to 25.1 Tg in 2000 over Asia as shown in Figure 1. Enhanced NOx emissions in Asia have a great potential for increasing the amount of tropospheric O_3 not only in the emission source region but also on a hemispheric scale (Wild and Akimoto, 2001).

Over East Asia, O_3 concentration was enhanced from spring to autumn by chemicallyproduced O_3 by regional emissions (Yamaji et al., 2006). This means that the future O_3 over East Asia would be significantly affected by changes of O_3 precursor emissions depending on future economic growth and environmental policies in this area. In particular, China with its large population and extensive land mass that sustains high economic development is of great importance. Therefore we tried to predict the future surface O_3 using a regional chemical transport model and a regional emission inventory for Asia.

A regional emission inventory in Asia for anthropogenic air pollutants with a 0.5°×0.5° spatial resolution that employs specific information of regional, country, and province levels has been completed by the Frontier Research Center for Global Change (FRCGC) in Japan (Yan et al., 2003a,b; Yamaji et al., 2003,2004; Ohara et al.,

*Corresponding author: Kazuyo Yamaji,

2007). This emission inventory, named Regional Emission inventory in ASia (REAS), is available at http://www.jamstec.go.jp/frcgc/research/p3/emissi on.htm and provides a sequence of gridded emission data from the past to the future, which has been built under a consistent concept. Especially for future emissions, REAS employed three emission scenarios (the reference (REF), the policy succeed case (PSC), and the policy failure case (PFC) scenarios) for China, and the moderate scenario (the REF) for other countries.

This study used a regional chemical transport model, the Models-3 Community Multi-scale Air Quality Modeling System (CMAQ) (Byun and Ching, 1999) and REAS to prediction the future O_3 change caused by anthropogenic emission changes over East Asia.



Figure 1. NOx emission trends in Asia including South, Southeast, and East Asia. ^aIPCC, http://sres.ciesin.columbia.edu/ ^bOhara et al. (2007) ^cAmann (2006)

Frontier Research Center for Global Change, Japan Agency for Marine Science and Technology, 3173-25 Showa-machi, Kanazawa-ku, Yokohama 236-0001 Japan; e-mail: kazuyo@jamstec.go.jp; Web address: http://www.jamstec.go.jp/frcgc

2. DESCRIPTION FOR MODEL EXPERIMENTS

2.1 Model Descriptions

We used CMAQ version 4.4 driven by meteorological fields calculated by the Regional Atmospheric Modeling System (RAMS) Version 4.3 (Pielke et al., 1992; Cotton et al., 2003). The three-dimensional RAMS simulation used meteorological fields from NCEP/NCAR reanalysis data sets (2.5°×2.5°) with 6-hour intervals in 2000. Spatial domains for CMAQ and RAMS (shown in Figure 2) are $6240 \times 5440 \text{ km}^2$ (inside domain) and 8000 × 5600 km² (outside domain) centered at 25°N and 115°E, respectively, with 80 × 80 km² For vertical resolution, both arid resolutions. model systems have the same model height of 23 km and employ a hybrid sigma-pressure coordinate. CMAQ and RAMS have 14 and 23 vertical layers, respectively.

The CMAQ chemical-transport model (CCTM) requires information for initial and boundary chemical concentrations. In this study, the initial conditions were chosen to reflect the East Asian situation and the validity of these was examined in a previous study by Zhang et al. (2002). The boundary conditions of O_3 and its precursors, NO_2 , HNO_3 , CO, PAN, PAN2 Ethene, Isoprene, Formaldehyde, Alkanes, and Alkenes were obtained from monthly-averaged outputs from the CHemical AGCM for Study of Atmospheric Environment and Radiative forcing, CHASER

(Sudo et al., 2002), which used Emission Database for Global Atmospheric Research (EDGAR) version 3.2. As for the gas-phase atmospheric chemical mechanism and the aerosol module, we used SAPRC–99 and AERO3, respectively. Chemical concentrations used in this study are instant CCTM outputs every three hours starting at 0 UTC for each day.



Figure 2 Model domains for CMAQ (inside) and RAMS (outside) simulations. The numerical characters indicate the locations of Japanese remote monitoring sites of EANET (Tappi, Happo, Oki and Hedo)

	NOx							NMVOC						
	2000 -	2010			2020			2000	2010			2020		
		PSC	REF	PFC	PSC	REF	PFC	2000 -	PSC	REF	PFC	PSC	REF	PFC
China BB ^b	11.2 0,8	12.2(9)	14.0(25) 1	6.9(51)	11.0(-2)1	.5.6(39) 25	.5(128)	14.7 20 2.7	0.8(41) 2	2.5(53) 2	3.6(61)	29.0(97)3	5.2(139) 38	7(163)
Japan BB ^b	2.0 < 0.1		1.8(-10)			1.8(-10)		1.9 < 0.1		2.2(16)			2.5(32)	
Other East Asi BB ^b	a° 2.5 0.2		3.1(24)			3.7(48)		1.9 0.5		2.7(42)		:	3.8(100)	
Southeast Asia BB ^b	^d 3.8 1.1		4.8(26)			5.8(53)		11.1 5.7	1	4.7(32)			19.1(72)	
China ^e (IIASA CLE)	11.7				1	3.8								

Table 1 Annual emissions of major O_3 precursors (NOx, Tgyr⁻¹ and NMVOC, Tgyr⁻¹) from anthropogenic sources for the years 2000, 2010, and 2020 (PSC, REF, and PFC for China and REF for the other regions)^a.

^aValues were obtained from REAS v1.1 (http://www.jamstec.go.jp/frcgc/research/p3/emission.htm; Ohara et al., 2006). ^bBB means emissions from biomass burning obtained from ACESS final version (http://www.cgrer.uiowa.edu/ACESS/acess_index.htm; Streets et al., 2003). ^cOther East Asia is Mongolia, North Korea, South Korea, and Taiwan. ^dSouthast Asia is Brunei, Cambodia, Indonesia, Indonesia, Laos, Malaysia, Myanmar, Philippines, Singapore, Thailand, and Viet Nam. ^eAmann (2006).

2.2 Emission Inventories

Past (2000) and future (2010 and 2020) emissions from anthropogenic sectors except biomass burning were obtained from REAS (Ohara et al., 2007). Future projection for the Asian emissions was performed on the basis of emission scenarios and emissions for the year 2000. Three emission scenarios, REF, PSC, and PFC for China, were developed by Zhou et al. For the other countries, the future (2003). emissions were on the basis of reference scenario projections in the World energy outlook (IEA, 2002). The REF is a sustainable scenario with increases moderate emission caused bv suppression of energy consumption through energy conservation, clean energy strategy, and moderate deployment of new energy technologies and new emission control technologies. The PSC is an optimistic scenario with low emission increase due to the implementation of strong energy and environmental policies and fast deployment of new energy technologies and new emission control technologies. As shown in Figure 1, NOx emission changes based on the REF and PSC are similar to IIASA CLE (Amann, 2006). The PFC is a pessimistic scenario with high emission increases caused by continuation of current energy supply structure, increased energy consumption, and slow deployments of new energy and emission control technologies. NOx emission changes based on the PFC is close to IPCC A2.

Emissions from biomass burning and volcanoes were obtained from ACE-Asia and TRACE-P Modeling and Emission Support System (ACESS) final version and Streets et al. (2003). Biogenic NMVOC, isoprene and terpene, emissions were obtained from the Global Emission Inventory Activity (GEIA) monthly global inventory (Guenther et al., 1995). Emissions from natural sources, e.g., soil and lightning NOx are not considered in our model simulations.

Table 1 shows major O_3 precursors, annual NOx (TgNO₂ yr⁻¹, hereafter Tg yr⁻¹ is used) and NMVOC (Tg yr⁻¹), emissions from anthropogenic sources including biomass burning (described as 'BB' in Table 1). Under the REF in 2020 (2020REF), NOx emission increases from 11.2 Tg yr⁻¹ in 2000 to 14.0 Tg yr⁻¹ in 2010 and 15.6 Tg yr⁻¹ in 2020 over China. The 2020REF causes a little NOx decrease (10%) over Japan in 2000–2020. An increase in NOx emissions in other East Asia is predicted as 48 % during 2000–2020. As for NMVOC emissions, the 2020REF brings about large increases. For example, NMVOC emission

in China increases by 139% from 14.7 Tg yr⁻¹ in 2000 to 35.2 Tg yr⁻¹ in 2020, and that is much higher than the NOx case. The PSC brings about a little NOx decrease (-2%) but a large NMVOC increase (139%) during 2000-2020 over China. Meanwhile, the PFC brings about large increases in both NOx (128%) and NMVOC (163%) during 2000-2020 over China. Full details, emission spatial distributions, and comparisons with the other inventories were reported by Ohara et al. (2007).

We have to mention here that all input parameters (boundary conditions, meteorological conditions, and biomass burning, volcanic, and natural emissions) except REAS emissions were held constant in these model experiments, because this work focuses on surface O_3 change caused by future anthropogenic emission changes on the basis of REAS.

3. RESULTS AND DISCUSSIONS

3.1 Validation of Modeled O₃ Concentrations by Observations

 O_3 concentrations by CMAQ reproduce observed annual O_3 variations at 4 Japan remote monitoring sites, Tappi (42°N, 139°E), Happo (38°N, 136°E, 1840 m asl), Oki (37°N, 132°E), and Hedo (28°N, 127°E) of the Acid Deposition Monitoring Network in East Asia (EANET) as shown in Figure 2. Figure 3 compares dailyaveraged simulated O_3 concentrations with dailyaveraged observed O_3 concentrations at the four monitoring sites. These daily-averaged concentrations were obtained from data taken every three hours. This model system can capture



Figure 3 Observed (blue lines) and simulated (black lines) daily-averaged O_3 concentrations at the Japanese monitoring sites of EANET (Tappi, Happo, Oki, and Hedo) for the year 2000. The correlation coefficients between observed and simulated concentrations are shown in the graphs.

the observed O_3 concentration levels with correlation coefficients ranging between 0.61 and 0.85 based on daily-averaged O_3 . More detailed comprehensive and systematic validations have been conducted in Yamaji et al. (2006 and 2007).

3.2 Future Surface O₃ Changes over East Asia under Three Different Emission Scenarios

This section compares surface O_3 concentration changes (below 2 km) over East Asia from the year 2000 to the years 2010 and 2020 under the REF, PSC, and PFC. Figure 4 shows the yearly-averaged increases from 2000 to 2010 and 2020 for the three future emission scenarios, respectively.

Under the REF scenario, yearly-averaged O_3 increases in 2-4 ppbv up to 2010 (see 10REF-2000) and 3-7 ppbv up to 2020 (see 20REF-2000) from central China to the Korean peninsula. Even over south Japan (Kyushu area), yearly-averaged O_3 increases in approximately 3 ppbv during 2000-2020 though NOx emission is projected to decrease there. The PSC causes a little O_3 changes during 2000-2010 (see 10PSC-2000) and 2000-2020 (see 20PSC-2000). It is noted that



Figure 5 Yearly-averaged O_3 changes (below 2 km) from the year 2000 to the years 2010 and 2020 under three scenarios (PSC, REF, PFC).

yearly-averaged O₃ over northeast China (but excluding central and east China plane) shows a little decrease (-2ppbv) from 2000 to 2020 under the PSC with a little NOx reduction and a large NMVOC emission increase from 2000 to 2020. As shown in 10PFC-2000 and 20PFC-2000 in Figure 4, the PFC leads to much higher O₃ increases compared to the PSC and the REF cases. Especially for central and east China, yearlyaveraged O₃ increases reach 3-7 ppbv in 2000-2010 and more than 10 ppbv in 2000-2020. During 2000-2020, moreover, the predicted yearly O₃ increases, which are only 0-2 ppbv over most of Northeast Asia under the PSC, are enhanced by 6-8 ppbv over the Korean peninsula and by 2-6 ppbv in Japan under the PFC scenario, though there are no differences between scenarios over Japan and the Korean peninsula. These results suggest that the PSC scenario can help lower the potential increase in O₃ concentration and that the PFC scenario promotes a substantial increase in O₃ concentration, not only over China but also over the Korean peninsula and Japan.

3.2 Surface O₃ in Early Summer

Early summer, especially June is the highest O_3 month over Northeast Asia (Yamaji 2006 and 2007), thus this section focus on surface O_3 concentrations in June. Figure 5 shows the monthly-averaged surface O_3 concentrations (below 2 km) in 2000 and 2020 (20REF, 20PSC, 20PFC).

The highest O₃ area, from the North China Plain to the Korean peninsula is covered with 70-80 ppbv of O_3 in the year 2000. The projected 2020REF emissions increase the O_3 concentrations to 80-90 ppbv in the central North China Plain and north the Korean peninsula. Over Northeast Asia, from central eastern China to the predicted monthly-averaged O₃ Japan, concentrations are increased by around 5-10 ppby during 2000-2020 by anthropogenic emission Meanwhile, the PSC is helpful to increases. moderate the O₃ increase in June. In large parts of northeastern and central China, the monthlyaveraged O_3 decreases up to 2 ppbv. On the other hand, a little O3 increase (2-4 ppbv) is confirmed at a few points near mega-cities at northeastern and central China where NOx emission increase are projected under the PSC (Ohara et al., 2007; Yamaji et al., 2007). The 2020PFC leads to much higher O₃ increases compared to the 2020REF case. Especially, monthly-averaged O₃ concentrations increase to

90-105 ppbv over the North China Plain and a part of the Korean Peninsula.



Figure 6 Monthly-averaged O_3 concentration (below 2 km) in June for the year 2000 and the years 2020 under three scenarios (PSC, REF, PFC).

4. SUMMARY

Tropospheric O₃ over East Asia in the future has been simulated by CMAQ coupled REAS emission inventory to predict the surface O_3 changes caused by future anthropogenic emission changes. For future predictions, REAS provides three emission scenarios for China, the REF, PSC, and PFC and one emission scenario, the REF for the other countries. Yearly-averaged O₃ increases in 3-7 ppbv up to 2020 under the REF. The PSC brings about a little O_3 changes in 2000-2020. Yearly-averaged O3 over the North China Plain increases in more than 10 ppbv in 2000-2020. June, monthly-averaged Especially in O₃ concentrations increase to 90-105 ppby over the North China Plain and a part of the Korean Peninsula. These model results suggest that the PSC scenario can help lower the potential increase in O3 concentration and that the PFC scenario promotes a substantial increase in O₃ concentration, not only in China but also over northeast Asia.

5. ACKNOWLEDGMENTS

We would like to thank EANET for providing ozone measurement data. This study has been supported by Global Environment Research Fund of the Ministry of the Environment, Japan (B-051).

6. REFERENCES

- Amann, M., 2006: Scenarios of world anthropogenic emissions of air pollutants and methane up to 2030, IIASA report (IR-06-023).
- Byun, D.W. and J.K.S. Ching (Eds.), 1999: Science algorithms of the EPA Models-3 community multi-scale air quality (CMAQ) modeling system. *NERL*, Research Triangle Park, NC. EPA/600/R-99/030.
- Cotton, W.R., R.A. Pielke, and R.L. Walko, 2003: RAMS 2001: Current status and future directionds. *Meteorol. Atmos. Phys.* **82**, 5-29
- Guenther, A., C.N. Hewitt, D. Erickson, R. Fall, C. Geron, T. Graedel, P. Harley, L. Klinger, M. Lerdau, W.A. Mckay, T. Pierce, B. Scholes, R. Steinbrecher, R. Tallamraju, J. Taylor, and P. Zimmerman, 1995: A global model of natural volatile organic compound emissions. *J. Geophys. Res.* **100**, 8873–8892.
- IEA, 2002: World Energy Investment Outlook 2002 [CD-ROM].

IPCC, http://sres.ciesin.columbia.edu/

- Ohara, T., H. Akimoto, J. Kurokawa, N. Horii, K. Yamaji, X. Yan, and T. Hayasaka, 2007: An Asian emission inventory of anthropogenic emission sources for the period 1980–2020. *Atmos. Chem. Phys.* **7**, 4419-4444.
- Pielke, R.A., W. R. Cotton, R.L. Walko, C.J. Tremback, W.A. Lyons, L.D. Grasso, M.E. Nicholls, M.D. Moran, D.A. Wesley, T.J. Lee, and J.H. Copeland, 1992: A comprehensive meteorological modeling system–RAMS. *Meteorol. Atmos. Phys.* **49**, 69–91.
- Streets, D.G., K.F. Yarber, J.-H. Woo, and G.R. Carmichael, 2003: Biomass burning in Asia: Annual and seasonal estimates and atmospheric emissions. *Global Biogeochem. Cycles.* **17**, Art. No. 1099, doi:10.1029/2003GB002040.
- Sudo, K., M. Takahashi, J. Kurokawa, and H. Akimoto, 2002: CHASER: A global chemical model of the troposphere-1. Model description. *J. Geophys. Res.* **107**, Art. No. 4339, doi:10.1029/2001JD001113.
- van Aardenne, JA., GR. Carmichael, H. Levyll, D. Streets, and L. Hordijk, Anthropogenic NOx emissions in Asia in the period 1990–2020, Atmospheric Environment, 33(4), 633-646, 1999
- Wild, O. and H. Akimoto, 2001: Intercontinental transport of ozone and its precursors in a threedimensional global CTM. *J. Geophys. Res.* **106**, 27729-27744
- Yamaji, K., T. Ohara, I. Uno, H. Tanimoto, J. Kurokawa, and H. Akimoto, 2006: Analysis of the seasonal variation of ozone in the boundary

layer in East Asia using the Community Multiscale Air Quality model: What controls surface ozone levels over Japan? *Atmos. Environ.* **40**, 1856-1868.

- Yamaji, K., T. Ohara, I. Uno, J. Kurokawa, and H. Akimoto, 2007: Future Prediction of Surface Ozone over East Asia using the Models-3 Community Multi-scale Air Quality Modeling System (CMAQ) and the Regional Emission Inventory in Asia (REAS), *submitted to J. Geophys. Res.*
- Yamaji, K., T. Ohara, and H. Akimoto, 2003: A country-specific, high-resolution emission inventory for methane from livestock in Asia in 2000. *Atmos. Environ.* **37**, 4393-4406.
- Yamaji, K., T. Ohara, and H. Akimoto, 2004: Regional-specific emission inventory for NH_3 , N_2O , and CH_4 via animal farming in South, Southeast, and East Asia. *Atmos. Environ.* **38**, 7111-7121.
- Yan, X., Z.C. Cai, T. Ohara, and H. Akimoto, 2003a: Methane emission from rice fields in mainland China: Amount and seasonal and spatial distribution. J. Geophys. Res. **108**, Art. No. 4505, doi:10.1029/2002JD003182.
- Yan, X., H. Akimoto, and T. Ohara, 2003b: Estimation of nitrous oxide, nitric oxide, and ammonia emissions from croplands in East, Southeast, and South Asia. *Global Change Biology* **9**, 1080-1096.
- Zhang, M.G., I. Uno, S. Sugata, Z.F. Wang, D. Byun, and H. Akimoto, 2002: Numerical study of boundary layer ozone transport and photochemical production in east Asia in the wintertime. *Geophys. Res. Lett.* **29**, Art. No. 1545, doi:10.1029/2001GL014368.
- Zhao D., Y. Dai, C. Yu, Y. Guo, Y. Zhu, 2003: China's Sustainable Energy Scenarios in 2020. China Environment Science Press, Beijing, China.