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NUMERICAL ANALYSIS OF BIOGENIC EMISSION SOURCES CONTRIBUTING TO OZONE CONCENTRATION IN OSAKA, JAPAN

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Abstract: This study investigated biogenic volatile organic compounds (BVOC) emission sources contributing to ozone (O_3) concentration in Osaka, Japan in summer 2010 by using the Weather Research and Forecasting model (WRF) version 3.5.1 and the Community Multiscale Air Quality model (CMAQ) version 5.0.1. The contributions of 10 source regions surrounding Osaka were estimated by comparing the baseline case and the zero-out BVOC emission cases for each source region. In the baseline case, WRF/CMAQ captured well the temporal variations including the occurrence of the O_3 concentration peaks. The zero-out emission runs showed that the BVOC emissions substantially contributed to high O_3 concentration in Osaka with day-by-day variation of contributing source regions. Although O_3 concentrations were especially high on 23 July and 2 August, the contribution of BVOC on 23 July was much larger than that on 2 August. To investigate this difference, the backward trajectory analysis and another zero-out emission run for anthropogenic VOC (AVOC) in Osaka were performed. On 23 July, the O_3 concentration in Osaka was dominantly increased by the advection from the northwestern region outside Osaka, with the particularly large contributions of BVOC emissions from Hyogo (west from Osaka) and Kyoto (north from Osaka). On 2 August, the concentration was dominantly increased by the local photochemical production inside Osaka under weak wind condition, with the particularly large contribution of AVOC emissions from Osaka.

Key words: Photochemical ozone, BVOC, Source contribution, Zero-out emission, WRF/CMAQ

INTRODUCTION
Tropospheric ozone (O_3) adversely affects human health and vegetation. In Japan, the environmental standard of photochemical oxidants has not achieved at almost all the monitoring stations. In the last decade, anthropogenic emissions and concentrations of O_3 precursors (nitrogen oxides (NO_X) and volatile organic compounds (VOC)) have been decreasing. However, mean O_3 concentration has been gradually increasing. By increasing of ozone concentration in recent years, photochemical oxidant warnings are often issued. To solve the photochemical oxidant pollution problems, quantitative evaluations of the contribution of the oxidant precursors using a numerical model is important.

One of the causes of the increase in O_3 concentration may be the increase of the background concentration because of the long-range transport. Temperature rise due to global warming and urban heat island is also considered as one of its causes. Furthermore, biogenic VOC (BVOC) emission increase with the rise in temperature is also considerd as another cause because BVOC plays an important role in the O_3 formation. However, the behaviour of BVOC is still uncertain. To control the increase of O_3concentration, it is necessary to reveal the relationship between BVOC and ozone concentration.

High O_3 concentrations exceeding 100 ppb are often observed during summer in Osaka, which includes the third largest megacity in Japan. Osaka Prefecture has the smallest forest area in Japan, but is surrounded by several prefectures with large forest areas. Therefore BVOC emitted in the surrounding areas may substantially affect O_3 concentration in Osaka. In this study, in order to specify BVOC sources that affect O_3 concentration in Osaka, sensitivity analyses of BVOC emissions on ozone concentration were performed with the Community Multiscale Air Quality model (CMAQ) (Byun and Ching, 1999).

METHODOLOGY

This study utilized CMAQ version 5.0.1 driven with meteorological fields produced by the Weather Research and Forecasting model (WRF) (Skamarock et al., 2009) version 3.5.1. The WRF/CMAQ modelling system was run for 6 July to 9 August during 2010 with an initial spin-up period of 1 to 5 July 2010. In addition, the Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT) (Draxler and Hess, 1997) version 4 was used for backward trajectory analyses.

Figure 1 shows modelling domains for air quality simulations and locations of Kokusetsuosaka station used for model evaluation. The Kokusetsuosaka station is located at (135.535° E, 34.680° N) in the centre of Osaka Prefecture. The modelling domains consist of three domains: domain 1 (D1) covering a wide area of Northeast Asia, domain 2 (D2) covering the main land of Japan, domain 3 (D3) covering of the Kinki Region. The horizontal resolutions and the number of grid cells are 64, 16 and 4 km, and 108×96, 68×68 and 76×76 for D1, D2 and D3, respectively. The vertical layers consist of 30 sigma-pressure coordinated layers from the surface to 100 hPa.

Table 1 shows the model configurations and datasets used in this study. The WRF/CMAQ modeling system was configured with the same physics and chemistry options as those used by Shimadera et al. (2014). Analysis data obtained from the U.S. National Centers for Environmental Prediction (NCEP) and the Japan Meteorological Agency (JMA) were used for Initial and lateral boundary conditions and grid nudging in the WRF simulation. The hourly WRF results were processed by the Meteorology-Chemistry Interface Processor (MCIP) version 4.1 for the CMAQ simulations. The MCIP-processed meteorological data were also used for biogenic emission estimates and backward trajectory analyses. Emission data for the CMAQ simulations include anthropogenic, biogenic, biomass burning and volcanic emissions. CMAQ was configured with the SAPRC99 mechanism for the gas-phase chemistry and the fifth generation CMAQ aerosol module for the aerosol process.

The BVOC emission sources contributing to ozone concentration in Osaka were identified by conducting zero-out BVOC emission runs. Figure 2 shows BVOC source regions and mean NMVOC emission rates in D3. The land areas in D3 were divided into 10 source regions: Mie Prefecture (Reg1), Shiga Prefecture (Reg2), Kyoto Prefecture (Reg3), Osaka Prefecture (Reg4), Hyogo Prefecture (Reg5), Nara Prefecture (Reg6), Wakayama Prefecture (Reg7), Chubu-Hokuriku region in D3 (Reg8), Chugoku Region in D3 (Reg9), Shikoku Region in D3 (Reg10). The zero-out BVOC emission runs were conducted for each source region. In this study, the contribution of a source region is defined as the difference of ozone concentration in the baseline case and a zero-out case for the region.

Biogenic isoprene and monoterpene emissions in Reg1-7 were estimated by using the experimentally-derived standard emission rates of the dominant tree species and the forest database in the regions (Bao et al., 2008). The other biogenic emissions were estimated with the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006) version 2.04. In Figure 2b, while large emissions in the coastal lowland areas are dominated by anthropogenic VOC (AVOC) from urban areas, large emissions in the mountainous areas are dominated by BVOC from forest vegetation. The total NMVOC emission rates in D3 was 3703 molC s^{-1}. Isoprene and monoterpene emissions accounted for 57% of the
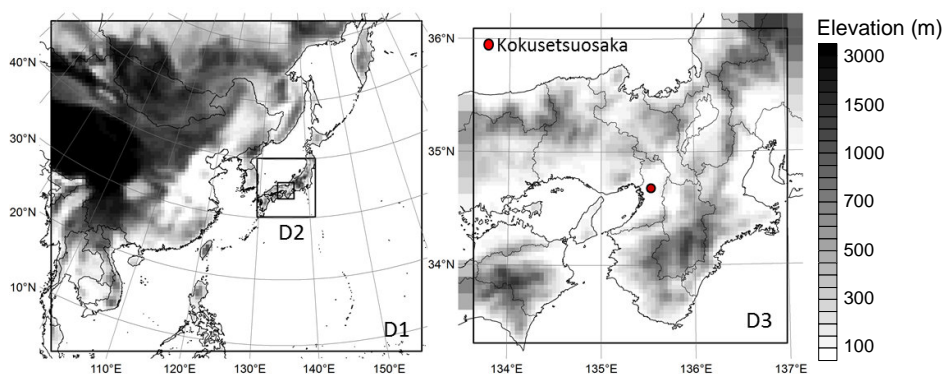

Figure 1. Modelling domains for air quality simulations and location of Kokusetsuosaka station

Table 1. Model configurations and datasets

	Settings
Meteorology (WRF v3.5.1)	Topography: 30-sec USGS
	Land-use: 30-sec USGS (D1, D2) and 100-m Japan National Land Numerical Information (D3)
	Analysis data: NCEP FNL, JMA MSM and NCEP/NOAA RTG_SST_HR
	Physics option: Kain-Fritsch (D1, D2), WSM6, YSU PBL, Noah LSM and RRTM/Dudhia
	Grid nudging: $G_{t, q, uv} = 3.0 \times 10^{-4}$ s^{-1} (D1, D2) and $G_{uv} = 7.5 \times 10^{-5}$ s^{-1} (D3)
Emission	Anthropogenic outside Japan: INTEX-B v1.2 and REAS v1.11
	Anthropogenic in Japan: JEI-DB (vehicle in 2010) and EAGrid2000-JAPAN, OPRF (ship)
	Biogenic: MEGAN v2.04 and Bao et al. (2008)
	Biomass burning: FINN v1; Volcanic SO_2: JMA and Aerocom
Air quality (CMAQ v5.0.1)	Initial and boundary: CMAQ default concentration profile
	Horizontal/vertical advection: Yamartino/WRF; Horizontal/vertical diffusion: Multiscale/ACM2
	Photolysis rate: In-line module; Gas/aerosol/cloud phase: SAPRC99/AERO5/ACM
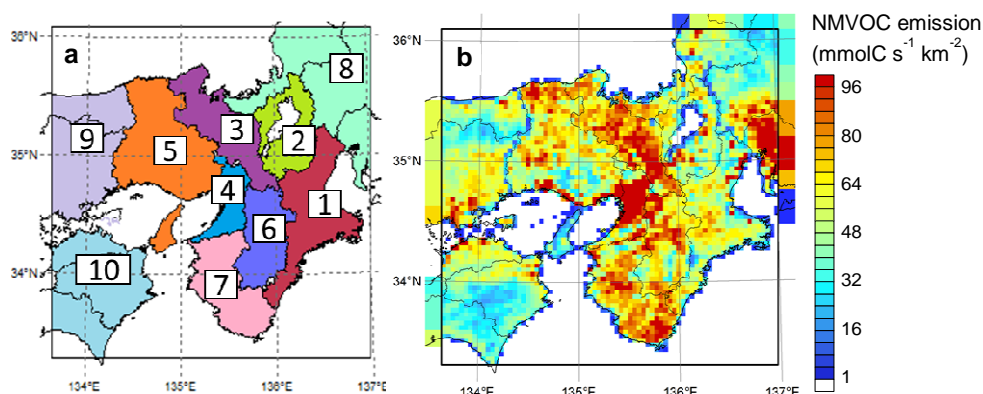
Figure 2. BVOC source regions (a) and horizontal distribution of mean NMVOC emission rates (b) in D3

total NMVOC emission in D3. Among the 10 source regions, the BVOC emission from Hyogo was the largest and that from Osaka was the smallest.

RESULTS AND DISCUSSION
Model performance for O$_3$ concentration
Figure 3 shows hourly time series of observed and simulated O$_3$ concentrations at the Kokusetsuosaka station from 6 July to 9 August 2010. High O$_3$ concentration peaks exceeding 100 ppb were observed on several days under clear sky and high temperature conditions. The baseline CMAQ simulation captured well the temporal variations including the occurrence of the O$_3$ concentration peaks. Consequently, the Pearson's correlation coefficient between the observation and the baseline simulation was 0.87, and the mean simulated daily maximum concentration (64.5 ppb) was close to the observed value (60.5 ppb). The results indicated that CMAQ showed fairly good performance for O$_3$ simulation, and therefore was applicable to estimates of the contribution of BVOC emissions to O$_3$ concentration.

Contribution of BVOC emission to O$_3$ concentration
Figure 4 shows estimated contributions of BVOC emissions from the 10 source regions in D3 to the daily maximum O$_3$ concentration at the Kokusetsuosaka station. The BVOC emissions in D3 substantially contributed to high O$_3$ concentration in Osaka with day-by-day variation of contributing source regions. The O$_3$ concentrations were especially high on 23 July and 2 August (Figure 3), with the values being respectively 134.0 and 147.3 ppb at 14 JST (when 1-h values were the daily maximum in the observation) in the baseline case. However, estimated BVOC contributions were quite different on the two days. Among the 10 BVOC source regions, Hyogo (Reg5) and Kyoto (Reg3) mainly contributed to high O$_3$ concentration in Osaka (Reg4) on the both days, whereas the contribution of BVOC on 23 July was much larger than that on 2 August. On 23 July, the contributions of BVOC from Hyogo and Kyoto were 18.95 ppb (14.1%) and 14.3 ppb (10.7%), respectively. On 2 August, the contributions of BVOC from Kyoto, Osaka and Hyogo were 7.5 ppb (5.1%), 4.8 ppb (3.3%) and 4.2 ppb (2.9%), respectively. The difference between the two days are further discussed in the following paragraphs.
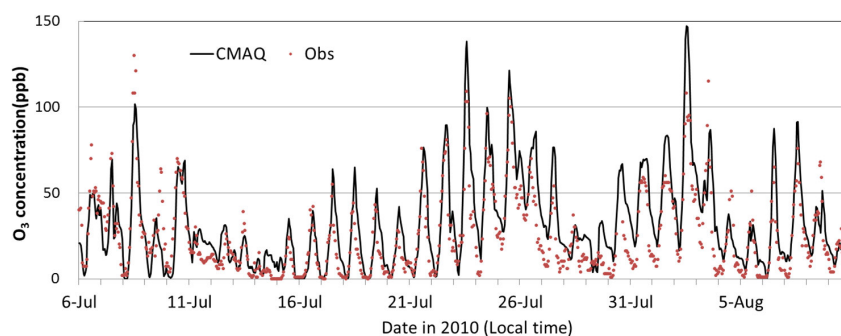
Figure 3. Hourly time series of observed and simulated O_3 concentration at Kokusetsuosaka station
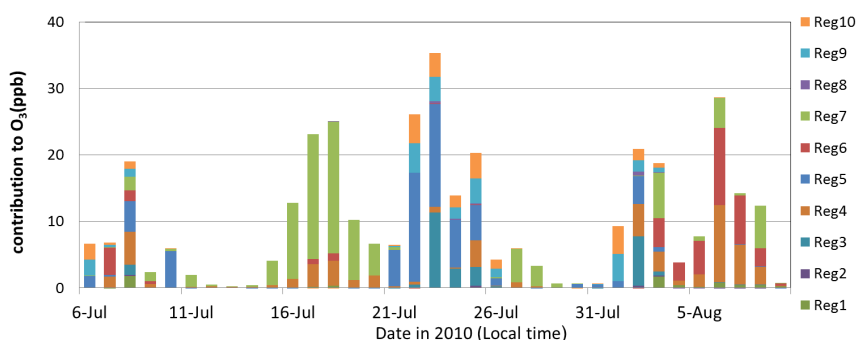

Figure 4. Estimated contributions of BVOC emissions from 10 source regions in D3 to daily maximum 1-h O_3 concentration at Kokusetsuosaka station

Figure 5 shows horizontal distributions of O_3 concentration at 14 JST on 23 July and 2 August with wind fields and backward trajectories arriving at the Kokusetsuosaka station. On 23 July, high O_3 concentration was widely spread in the coastal areas and in and around Osaka. On the other hand, high O_3 concentration was limited only in Osaka on 2 August. The trajectories indicated that air mass reaching Osaka was transported from the west of Osaka and the BVOC emissions from Hyogo and Kyoto mainly contributed to O_3 concentration in Osaka. As indicated by the transport distances on the trajectories, wind in morning to daytime was stronger on 23 July than on 2 August. The difference in the contributions of BVOC emissions on the two days was attributed to the difference in the wind fields.

To investigate the difference of the results on 23 July and 2 August, another zero-out emission run for AVOC in Osaka (Reg4) was performed. Figure 6 shows diurnal variations of the simulated O_3 concentration in the baseline case and the estimated contributions of VOC emissions to the O_3 concentration on the two days at the Kokusetsuosaka station. While the contribution of AVOC from Osaka was less than or comparable to that of BVOC from the source regions outside Osaka on 23 July, the contribution of AVOC from Osaka was much larger than the BVOC contributions. This finding indicated that the local photochemical reactions of VOC dominantly accelerated O_3 formation in Osaka under weak wind condition on 2 August.

The variation of O_3 concentration is mainly controlled by advection, diffusion, dry deposition, chemical (photochemical production and NO_X titration) processes. The O_3 concentration in Osaka was dominantly increased by the advection from the northwestern region outside Osaka on 23 July, and by the local photochemical production inside Osaka on 2 August.

CONCLUSION
The WRF/CMAQ modelling system was used to evaluate the contribution of BVOC emission sources in the Kinki Region of Japan to O_3 concentration in Osaka in summer 2010. The BVOC emissions substantially contributed to high O_3 concentration in Osaka with day-by-day variation of contributing source regions. Although O_3 concentrations were especially high on 23 July and 2 August, the contribution of BVOC emissions on 23 July was much larger than that on 2 August. The difference in the
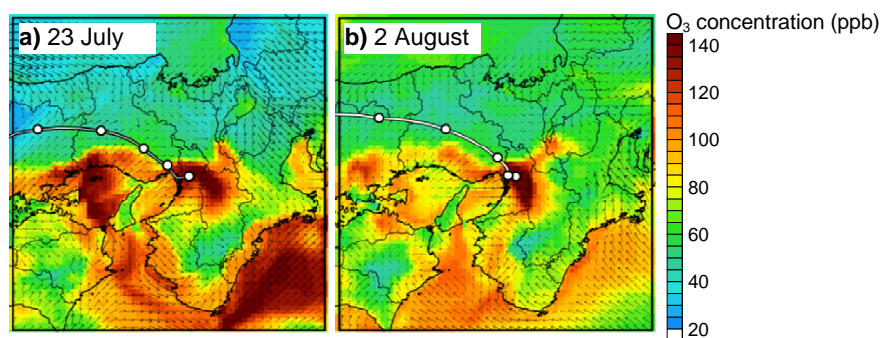
Figure 5. Horizontal distributions of O₃ concentrations and wind fields at the first layer and backward trajectories arriving at 300 m above Kokusetsuosaka station (circles represent 3-hour transport distances) at 14 JST on 23 July (a) and 14 JST on 2 August (b)
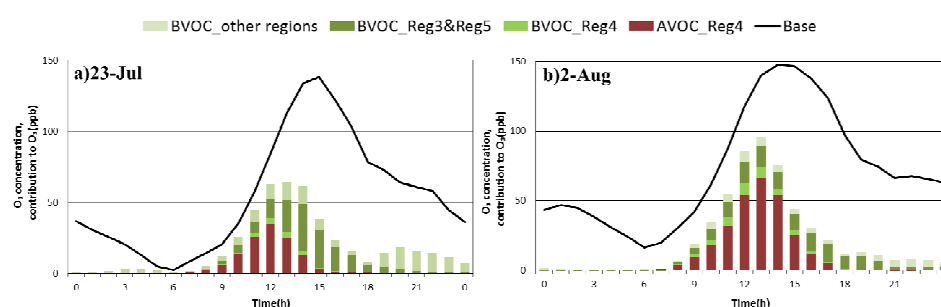

Figure 6. Simulated O₃ concentration and estimated contribution of AVOC emissions from Osaka (Reg4), BVOC from Osaka, from Hyogo (Reg5) and Kyoto (Reg3) and from the other 7 source regions in D3 to the O₃ concentration at Kokusetsuosaka station on 23 July : (a) and 2 August (b)

contributions of BVOC emissions on the two days was attributed to the difference in the wind fields. On 23 July, the O₃ concentration in Osaka was dominantly increased by the advection from the northwestern region outside Osaka, with the particularly large contributions of BVOC emissions from Hyogo (west from Osaka) and Kyoto (north from Osaka). On 2 August, the O₃ concentration in Osaka was dominantly increased by the local photochemical production inside Osaka under weak wind condition, with the particularly large contribution of AVOC emissions from Osaka.

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