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**MODELING STUDY ON THE CHARACTERISTICS OF OZONE POLLUTION IN TOKYO
METROPOLITAN AREA**

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Abstract: In Japan, a photochemical smog warning is issued when Ox concentration for hourly value is over 0.12ppm. The photochemical smog warnings had been issued during the period from 20th to 25th July 2010 in a row all over Tokyo Metropolitan area. In order to clarify the mechanism of the severe Ox pollution, numerical simulation analysis based on WRF/CMAQ was applied. It was found from the simulation results that there were two types of mechanism which caused the severe Ox pollution in the target period. It was suggested that high concentrations of Ox appeared around inland area were caused by the retention of precursors (NO₂ and VOCs) around the sea-breeze front. On the other hand, when the high concentrations appeared surrounding the coastal area of Tokyo Bay, it was found that the high concentration of O₃ were transported from the southwestern part of the simulation domain passed over the sea surface toward the coastal area of Tokyo Bay. Though the model showed a reasonable agreement with the diurnal variation of observed concentrations, model generally underestimated the daily maximum of observed Ox. In order to understand the main factor of the underestimation, it seems to be required to confirm the model performance of the vertical distribution and the sea surface concentration of Ox as well as the horizontal distribution. To do that, it is necessary to implement the observation on three dimensional basis in future.

Key words: *Air Quality Model, Oxidant, Ozone, Urban air pollution, Precursors*

1 INTRODUCTION

In Japan, the environmental air quality standard for photochemical oxidant (Ox) states that the hourly values should not exceed 0.06ppm (established in 1973). However, the achievement rate has been remaining at almost 0% nationwide during past a few decades. When the oxidant concentration for hourly value is over 0.12ppm, a photochemical smog warning is issued, and when over 0.24ppm, a photochemical smog alarm is issued. In this decade, Ox warnings were issued for the first time in several prefectures of Japan (e.g., 2006: Nagasaki and Kumamoto, 2007: Niigata and Oita, 2008: Nagano and Saga, 2009: Yamagata and Kagoshima, 2011: Kochi). This situation suggests that Ox pollution has been expanded rapidly. Generally speaking, the expansion of Ox pollution in whole of Japan seems to be caused by the transboundary air pollution from the continental, which occurs during the spring season. Air mass which contains relatively high concentrations of O₃ is transported easily from the continental when the low-pressure moves from the western part of Japan toward the east direction, or the seasonal rain front stays around south of Japanese archipelago. Thus, more attention has been paid to the risk of Oxidant exposure for human health and vegetation.

On the other hand, urban areas including Tokyo metropolitan also have severe Ox pollutions in summer season. Since a Pacific high covers the Japanese Islands in a usual summer, transboundary air pollutions from the continental cannot be a reason for the severe Ox pollution during summer season. Some previous studies have pointed out that the severe Ox pollutions were originated in the accumulation of the precursors (NO_x and VOCs) which were carried on the sea breeze from the southern part toward the inland of Tokyo Metropolitan area (e.g., Yoshikado, 2015). Accumulated precursors result in the severe Ox pollutions under the condition of strong UV and gentle wind in the daytime. It was also suggested that the taking of high Ox concentration existing in upper layer, which was created in the previous day, into mixing layer caused the increase of Ox background concentration in the daytime (Kiriya et al., 2015).

Especially, Tokyo metropolitan area had a severe Ox pollution in summer 2010. Ox warnings were issued for six consecutive days from 20th July in Tokyo. In addition, more than 200ppb of Ox was observed at many places in and around Tokyo during the period. In this context, this study aims to evaluate and clarify the mechanism of the severe Ox pollution observed in summer 2010 by means of modeling analysis.

2 METHODOLOGY

2.1 Model description

In this study, a modeling analysis was carried out using CMAQ (Community Multiscale Air Quality) version 4.7.1. The selected chemical reaction scheme was the same as Sakurai et al. (2015). Meteorological data in the three-dimensional space was calculated by WRF (Weather Research Forecast model) version 3.7.1. The global objective analysis data (FNL) of National Centers for Environmental Prediction (NCEP) was used in WRF simulation as the initial and boundary condition. In addition, RGT_SST of NCEP was also used for the sea surface temperature. The simulation period was 31 days in July 2010. As shown in Figure 1, modeling domain had a nesting system of 45 km (Domain1), 15 km (Domain2) and 5 km (Domain3) grid resolution, respectively. The domain sizes were 3,825 x 4,950 km² for Domain1, 1,095 x 1,200 km² for Domain2 and 335 x 335 km² for Domain3 with the domain center at 36N and 140E. The vertical layers consisted of 30 sigma-pressure layers from the surface to 100 hPa with the top height of the lowest layer being approximately 22 m.

Inland anthropogenic emissions were derived from REAS Ver. 2.1 (Kurokawa et al., 2013) for Domain1 and EAGrid-Japan 2010 (Fukui et al., 2014) for Domain2&3. Ship emissions were derived from REAS Ver. 2.1 for Domain1 and emission inventories developed by Ocean Policy Research Foundation (OPRF, 2013) for Domain 2&3. Since the estimated period for REAS Ver. 2.1 was from 2000 to 2008, the emissions in 2008 were applied as they were for the simulation in 2010 in this study. In addition, GEIA (Global Emission Initiative database) and GFED Ver. 3.1 were applied for vegetable origin VOCs and biomass burning origin, respectively. The volcanic origin SO₂ emissions were also taken into account in the same way as Sakurai et al. (2015).

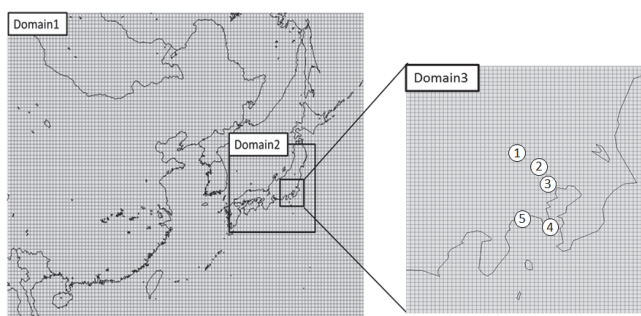


Figure 1 Model domains and locations of 5 monitoring sites to be compared with the calculated concentration.

(1:Fujimi, 2:Nerima-Shakujii, 3:Tokyo, 4:Kurihama, 5:Hiratsuka)

2.2 Analysis area of severe Ox pollution and observation data used in this study

In order to grasp the horizontal distribution of Ox concentration, hourly concentrations observed at the monitoring stations, which are managed by Ministry of the Environment, were analyzed by means of Geographical Information System (GIS). Target prefectures of this study were Tokyo, Kanagawa, Chiba and Saitama which constituted the metropolitan area. The number of monitoring stations in July 2010 was Tokyo: 47, Kanagawa: 58, Chiba: 93 and Saitama: 58. Of the total 256 monitoring stations, 5 stations shown in Figure 1 were selected for the model validation. Fujimi, Nerima-Shakujii and Tokyo were selected in order to confirm the model accuracy for the diurnal variation of Ox concentration. Kurihama and Hiratsuka were selected in order to validate the inflow of Ox carried by the sea breeze into Tokyo metropolitan area.

3 RESULTS AND DISCUSSION

Figure 2 shows the horizontal distributions of Ox concentrations observed at 2PM on each day through 21st to 24th July 2010 in Tokyo (47 sites), Kanagawa (58 sites), Chiba (93 sites) and Saitama (58 sites). The number of the stations where more than 120ppb of Ox concentration appeared was 66 sites on 21st, 53 sites on 22nd, 78 sites on 23rd and 105 sites on 24th in whole of the area.

3.1 Analysis for the horizontal distribution on 21st and 22nd July 2010

As shown in Figure 2, relatively higher concentrations appeared around the eastern part of both of Tokyo and Kanagawa at 2PM on July 21st. Moreover, over 160ppb of Ox concentrations were observed at 7 sites in the region. On the other hand, higher concentrations appeared around the northern part of Tokyo and the eastern part of Saitama at 2PM on July 22nd. Over 160ppb of Ox concentrations were observed at 3 sites only in Saitama.

Figure 3 shows the wind directions and the wind speeds observed at the meteorological stations of Nerima (Tokyo) and Tateyama (southern part of Chiba). On 21st July, the wind direction was south-west at Tateyama through the dawn. In this case, it is suggested that more precursors could be gathered and condensed around the inland area of Tokyo metropolitan by sea breeze, which passed over Kanagawa prefecture where population density and production activities were high. Figure 4 shows Ox and NMHC (Non Methane Hydrocarbons) concentrations observed at Nerima-Shakujii during the analysis period. It is obvious that higher NMHC concentrations were observed in the morning on 21st compared to the latter half of the period. Looking at the wind conditions at Nerima shown in Figure 3, south winds (sea breeze) of 1.1 m s^{-1} and 1.2 m s^{-1} were observed at 1PM on 21st and 22nd, respectively. However, though sea breeze did not prevailed on 21st, it continued to blow until the nighttime on 22nd.

As a result, it was suggested that more Ox was generated and 160ppb of concentrations appeared around the eastern part of Tokyo and Kanagawa on July 21st because precursors were carried and accumulated in the limited area south of Tokyo by a weak sea breeze. On the other hand, it is likely that higher concentrations appeared around the northern part of Tokyo and the eastern part of Saitama on July 22nd because the precursors were carried toward the wide region north of Tokyo by a prevailing south wind.

3.2 Analysis for the horizontal distribution on 23rd and 24th July 2010

Unlike 21st and 22nd July as mentioned above, it was found that over 160ppb of Ox concentrations appeared surrounding the coastal area of Tokyo Bay including Chiba prefecture. Over 160ppb of Ox concentrations were observed at 8 sites on 23rd and 17 sites on 24th in whole of the area. Looking at the wind conditions at Nerima shown in Figure 3, on 23rd July, the north-east wind blew from noon and 0.8 m s^{-1} of weak south wind was finally observed at 3PM. On 24th July, weak wind conditions continued during the morning and 0.8 m s^{-1} of weak south wind was finally observed at 1PM. Those wind conditions on 23rd and 24th are similar to 21st July and it was discussed above that such condition could cause severe Ox pollution in the limited area south of Tokyo. However, NMHC concentrations in the early



Figure 2 Horizontal distributions of Ox concentrations at 2PM through 21st to 24th July 2010 in Tokyo, Kanagawa, Saitama and Chiba.

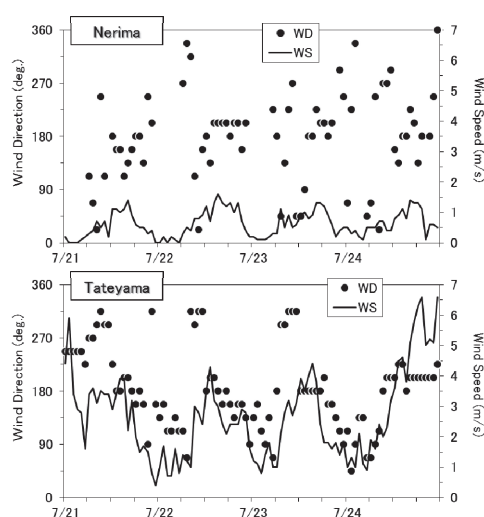


Figure 3 Wind directions and wind speeds observed at Tateyama and Nerima during the analysis period.

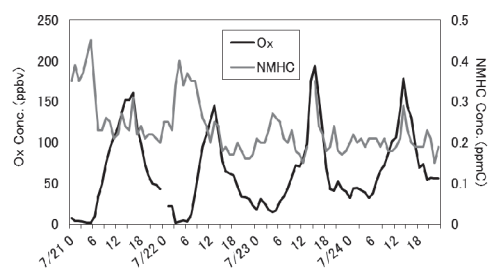


Figure 4 Ox and NMHC concentrations observed at Nerima-Shakujii during the analysis period.

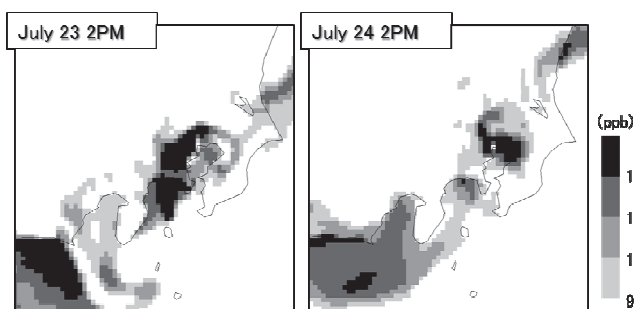


Figure 5 Horizontal distribution of simulated O₃ at 2PM on 23rd and 24th July 2010.

morning observed at Nerima-Shakujji didn't become as high as 21st because the wind directions in the dawn at Tateyama weren't from south-west but closer to east. As a result, the horizontal distributions of Ox on 23rd and 24th could be different from that on 21st regardless of the similar weak wind condition at Nerima. Shown in Figure 4, daily maximum of NMHC appeared at the same time with Ox concentration on 23rd and 24th July, which was a different diurnal variation on 21st and 22nd. Thus, the mechanism for the severe Ox pollution on 23rd and 24th July seems to be different from the previous 2 days.

In order to evaluate the reason why high concentrations appeared surrounding the coastal area of Tokyo Bay on 23rd and 24th July, horizontal distributions of ground level O₃ concentration at 2PM, simulated by WRF/CMAQ, were shown in Figure 5. It was found from the simulation results that the high concentration of O₃ were transported from the southwestern part of the simulation domain passed over the sea surface toward the coastal area of Tokyo Bay. It was also found from the simulation result in Domain2 that the transported O₃ was generated in and around Osaka and Nagoya, which were regarded as the major cities in Japan.

As a result, it was suggested that Ox high concentrations observed around the coastal area of Tokyo Bay and in the southern part of Kanagawa on 23rd and 24th were originated in the transported O₃ from western Japan passed over the sea surface in addition to the local Ox generation.

3.3 Model validation based on the observation

In order to clarify the model performance for the severe Ox pollution in the summer 2010, Hourly comparisons of observed and simulated Ox were conducted at 5 sites shown in Figure 1. In this comparison, most of observed Ox were considered to be O₃ and it was compared directly with simulated O₃. In addition, simulated O₃ was derived from the output of Domain3. As shown in Figure 6, it was clarified that the simulated concentrations at Hiratsuka and Kurihama, which were selected in order to validate the inflow of Ox carried by the sea breeze into Tokyo metropolitan area, showed a reasonable agreement with the observation through the period. In the previous section, it was pointed out that the transport of O₃ passed over the sea surface seemed to be one of the reasons to cause the severe Ox pollution around Tokyo Bay area on 23rd and 24th July 2010. Since model successfully reproduced the increase of Ox observed at Kurihama in the dawn on 23rd and 24th, it is suggested that the air masses which contained O₃ had passed several times near Tokyo Bay area in the both days.

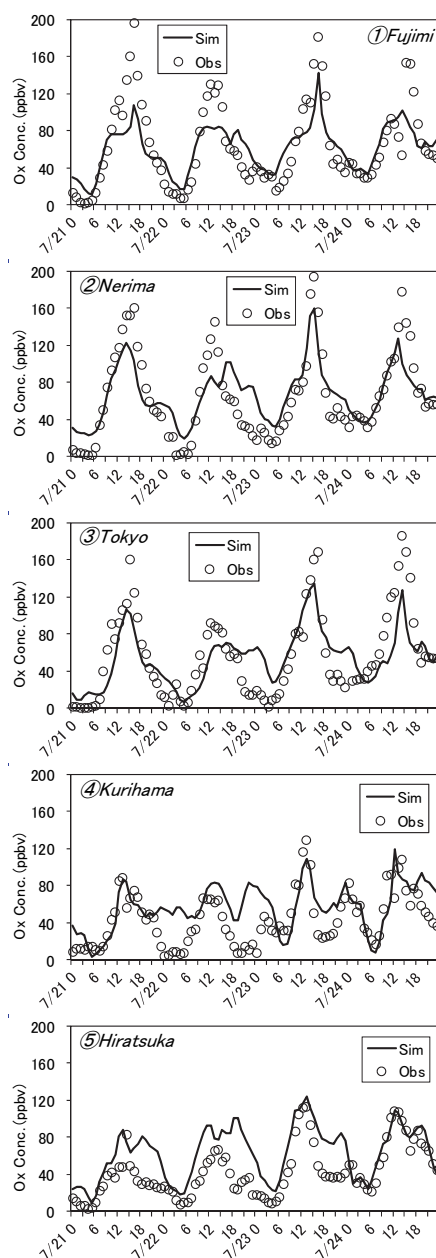


Figure 6 Hourly comparisons of observed Ox and simulated O₃ at 5 sites shown in Fig. 1.

On the other hand, though the model showed a reasonable agreement with the diurnal variation in the comparison with Fujimi, Nerima-Shakujii and Tokyo, model generally underestimated the daily maximum of observed Ox. In order to understand the main factor of the model underestimation, it seems to be necessary to clarify the model performance for the vertical distribution and the sea surface concentration of Ox as well as the horizontal distribution. To do that, it is required to obtain the observation data on three dimensional basis.

4 CONCLUSION

The photochemical smog warnings had been issued during the period from 20th to 25th July 2010 in a raw all over Tokyo Metropolitan area. In order to clarify the mechanism of the severe Ox pollution, numerical simulation analysis based on WRF/CMAQ was applied. It was found from the simulation results that there were two types of mechanism which caused the severe Ox pollution in the target period. It was suggested that high concentrations of Ox could appeared around the inland area were caused by a retention of precursors (NO₂ and VOCs) around the sea-breeze front. More Ox could be generated and 160ppb of concentrations appeared around the eastern part of both of Tokyo and Kanagawa on July 21st because precursors were carried and accumulated in the limited area south of Tokyo by a weak sea breeze. On the other hand, it is likely that higher concentrations could appeared around the northern part of Tokyo and the eastern part of Saitama on July 22nd because the precursors were carried toward the wide region north of Tokyo by a prevailing south wind.

On the other hand, when the high concentrations appeared surrounding the coastal area of Tokyo Bay, it was found from the simulation results that the high concentration of O₃ were transported from the southwestern part of the simulation domain passed over the sea surface toward the coastal area of Tokyo Bay. It was also found that the transported O₃ was generated in and around Osaka and Nagoya, which were regarded as the major cities in Japan.

Though the model showed a reasonable agreement with the diurnal variation of observed Ox concentrations, model generally underestimated the daily maximum of observed Ox. In order to understand the main factor of the underestimation, it seems to be required to clarify the model performance of the vertical distribution and the sea surface concentration as well as the horizontal distribution. To do that, it is necessary to implement the observation on three dimensional basis in future.

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