Effect of NOx and VOC Controls for Surface Ozone Concentration in Summertime in Kanto Region of Japan

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In recent years, the concentrations of precursors of ozone have been decreasing in Japan, but the ground-level ozone concentrations tend to increase. During the summertime in the Kanto region, anthropogenic emission control in Japan is an important factor for decreasing the ozone concentration. In this study, we conducted an air quality simulation using WRF and CMAQ with the anthropogenic emissions data in 2000 and 2005 to verify the effect of the emission controls between 2000 and 2005 on the ozone concentration in the Kanto region. The NOx and VOC reductions contribute to the increase in the daytime maximum ozone in the central Tokyo surrounding area but to the decrease in the other areas of the Kanto region. For the NOx reduction case, the daytime maximum ozone was increased in a wide area of the plains. On the other hand, the VOC reduction effectively works to decrease the ozone concentration in the entire Kanto area. Based on the results of the ozone sensitive analysis for a high concentration day, the NOx sensitive and mixed sensitive regime account for half of the number of occurrences in inland Kanto area. However, the VOC sensitive regime was the majority in the large city. These results show the effectiveness of the reduction of precursors to the decrease in the ozone concentration in each area of Kanto.

Key words: Ozone air pollution, WRF/CMAQ, Emission reduction, Sensitivity regime

1. Introduction

In recent years, the attainment ratio to the national standard for photochemical oxidant (or simply ozone; describing ozone in below sentence) has been quite low. As show by Wakamatsu et al. (2013) from monitoring results of air pollutants by national and local governments, concentrations of ozone precursors (i.e., nitrogen oxides, NOx, and non-methane hydro carbons, NMHC) have been decreasing, but ground-level ozone concentrations tend to increase from latter half of 1980s to the present. A possible cause of this contradiction is the transboundary air pollution from Asian industrial countries (Ohara et al., 2001). Pochanart et al. (2002) shows from monitoring results at remote stations and backward trajectory analysis that polluted air masses from the Asian Continent influenced the air quality in springtime in Japan. However, Pochanart et al. (2002) also shows the influence of the transboundary air pollution is relatively lower in summertime than in the other seasons. In the case of Kanto region of Japan (the area in and around metropolitan Tokyo on the Pacific coast of Japan) during summer, dominant flows come from the south or southwest, bringing clean air from the Pacific. Therefore, the influence of the transboundary air pollution is lower in summertime Kanto than in the other seasons and areas in Japan. In other words, the influence of local emissions in Kanto region becomes important. It could be possible to detect the relationships between ozone concentrations and emissions of its precursors in summertime Kanto.

In Japan, various measures to control anthropogenic emissions have been enacted to mitigate the air pollution by ozone (Wakamatsu et al., 2013). Actual measures taken between 2000 to 2005, for example, are the Road Transport Vehicles Act (amendment) for CO, NOx and hydrocarbons emissions in 2000 and The Law Concerning Special Measures for Total Emission Reduction of NOx and PM Automobiles in Specified Areas (the Vehicle NOx, PM law) in 2001. Emission regulations with respect to the stationary emission sources have been improved successively by amendment of the Air Pollution Control Law. In this period, the annual-mean daytime maximum ozone concentration for the entire country showed a tendency of increase. However,
differences in annual-mean daytime maximum concentrations between 2000 and 2005 in Tokyo metropolitan were not detected. A possible cause is inter-annual meteorological variability to ozone concentrations.

The purpose of this study is to verify the effect of emission controls between 2000 and 2005 on the ozone concentration in the Kanto region by using meteorological and chemical transport models. Meteorological conditions in 2005 are commonly used for both 2000 and 2005 simulations to make clear changes in ozone concentrations due to emission reductions between the two years. The effect of emission measures carried out from 2000 through 2005 will be evaluated by comparing the result of the simulation.

2. Method

In this study, we used the Weather Research and Forecasting (WRF) model version 3.2.1 (Skamarock et al., 2008) and the Community Multiscale Air Quality (CMAQ) model version 4.7.1 for air quality simulations. The calculation period was 25 December 2004 to 31 December 2005, with the first 7 d for spin-up. Within this period, we analyzed the four months, June to September 2005. As shown in Fig. 1, the model consists of three domains of D1, which covers a wide area of East Asia, D2, which almost includes the major four islands of Japan, and D3, which focuses on the Kanto region.

In the WRF simulation, horizontal resolutions are 64 km, 16 km, and 4 km, and the numbers of grids are 108×92, 116×124, 72×72 for D1, D2 and D3, respectively. Number of the vertical layers is 30. As input data to the WRF model, for meteorology we used final analysis data provided by the National Center for Environmental Prediction (NCEP), and for sea surface temperature we used OISSTHR (Reynolds et al., 2007) provided by National Oceanic and Atmospheric Administration (NOAA). Three-dimensional analysis nudging was applied to the west-east and north-south wind components in D1 and D2, with the nudging coefficient set to 1.0×10⁻⁴ s⁻¹ for the entire simulation period. WRF model was configured with the planetary boundary layer of Janjic (2002), the WRF single-moment 6-class microphysics scheme (Hong and Lim, 2006), Kain-Fritsch scheme (Kain, 2004) for the cumulus parameterization of D1 and D2, Noah land surface model (Chen and Dudhia, 2001), the RRTMG scheme (Iacono et al., 2008) for long and short wave radiation simulation.

In the CMAQ simulation, horizontal resolutions of D1 to D3 are the same values of WRF simulation and numbers of grids are 80×96, 108×100, 56×56 for D1, D2 and D3, respectively. The model of gas phase chemistry used in CMAQ simulation is SAPRC-99 (Carter, 2000). For initial and boundary condition of the base case CMAQ calculation, we used the default value of CMAQ for D1, and simulated results for the outer domains D2 and D3.

Anthropogenic emissions in Japan were derived from emission inventories for vehicles (Morikawa et al., 2012) and for the other sectors (Nakatsuaka et al., 2012). Ship emissions were derived from emission inventories developed by the National Maritime Research Institute (NMRI) and by the Ocean Policy Research Foundation (OPRF) (OPRF, 2012). Emissions in East Asia except Japan were produced from the dataset described by Shimadeara et al. (2014). For the estimation of biogenic VOC, we used the Model of Emissions of Gases and Aerosols from Nature (MEGAN, Guenther et al., 2006) version 2.04.

In this study, we have selected 2000 and 2005 as target years. Table 1 shows that total amount of the anthropogenic ozone precursors (NOx and VOC) emitted in D3 in 2000 and 2005. The NOx emission is reduced by about 6% and the VOC emission is reduced by about 20% in 2005 compared to 2000.

Figure 2 shows the distributions of the anthropogenic NOx and VOC emissions and changes in emissions from 2000 to 2005. NOx emissions are concentrated in and around of the central Tokyo area. VOC emission is concentrated like NOx emissions, but there are also high emissions in inland of the Kanto region, around Kumagaya and Iesaki (points of these two points are shown in Fig. 1). For the changes in emissions in Fig. 2, reduction of emissions in the metropolitan area was particularly significant for both VOC and NOx. In addition,

![Fig. 1 Horizontal model domain using for CMAQ. Black points in D3 indicate locations of diagnostic points in Kanto region; Iesaki, Kumagaya, Oomiya and Shinjuku.](image-url)
there are areas where the VOC emission has been reduced greatly in inland. The points where emission increased are also sparsely shown. As shown in Table 1, emissions of the anthropogenic precursors of ozone in the whole Kanto region were decreased from 2000 to 2005.

In this study, we conducted three cases of simulations by changing the anthropogenic emissions in Japan (Table 2). Other computational conditions like meteorological fields, initial and boundary conditions for D3 of CMAQ simulations, and emissions from the other anthropogenic sources in Japan were common to base case simulation. This makes it possible to simply simulate effects of emission reduction, apart from meteorological variations and the transboundary pollution, on the ozone concentration in the Kanto region.

From Tables 1 and 2, [Base case O₃ Conc.]–[NOₓ2000_VOC2000 O₃ Conc.] regard as if reduce the NOₓ and VOC emission at the same time.

In this study, the high ozone concentration criterion was defined as 120 ppbv, in accordance with the standard criterion for photochemical oxidant advisories in Japan. A high concentration day was also defined as the day when daytime maximum concentration at any grid in D3 exceeds 120 ppb in the basic case. Here, in accordance with the definition of the Ministry of the Environment, daytime was defined as the period from 5:00 to 20:00 Japan Standard Time (JST, UTC+9 h; times are given as JST below). Observed ozone concentration data used in this study were preliminary figures of hourly ozone concentration also used to compare with the daytime maximum ozone concentration involved in the D3.

3. Results

3.1 Reproducibility of the calculation and diurnal variation of ozone concentration in the high concentration day

Figure 3 shows the comparison of observation and Base case daytime maximum concentration in high concentration days at four points shown in Fig. 1. From Fig. 3, there is a tendency of overestimation regarding the average of the daily maximum value. In high concentration days, seven data exceed twice or lower than half, but most data is within half and twice.

In the early morning, ozone concentrations are low over the whole region, especially around the coast of the Tokyo
Bay. At noon, ozone concentration was increased mainly by photochemical production. At this moment, pollutants from the coastal area do not reach the inland areas. However, an increase of the ozone concentration was found in the inland area. Kiriyama et al. (*Asian J. Atmos. Environ.*, under submission) showed that this morning increase in the inland area was strongly influenced by other sources than the central Tokyo and surrounding area of the Tokyo Bay with high NOx and VOC emissions and downward transport of ozone remaining aloft by development of the boundary layer. In the afternoon, winds from the sea flow in the coastal area of southwest Kanto. Also over the Tokyo Bay, the sea breeze has begun to approach towards the land. Pollutants emitted from sources along the coast cause high-concentration ozone in the inland area.

3.2 Changes of ozone daytime maximum from 2000 to 2005

Figure 5 shows the variation of monthly averaged daytime maximum ozone concentration in each emission reduction case in Isesaki, Kumagaya, Omiya and Shinjuku. From Fig. 5, in NOx and VOC reduction case, ozone concentration always showed a decrease in two inland points, Isesaki and Kumagaya. At Omiya, ozone concentration increased slightly in June but decreased in subsequent three months. On the other hand, ozone concentration had increased during the analysis period at Shinjuku. Therefore, it is suggested that the daytime maximum ozone concentration will be decrease by the effect of reduction of NOx and VOC emissions in the Kanto region, excluding the change of inflow from the boundary and meteorological factors. In the NOx reduction case, it is shown that a tendency to decrease in July and August and state of slightly increasing was seen in June and in September at Isesaki. Ozone concentrations in other three stations have increased in NOx reduction case, and tend to increase the variation toward the south. There was a difference of increment between Kumagaya and Shinjuku up to about 3 ppb. On the other hand, daytime maximum concentration was decreased in the VOC reduction case in these four points.

Figure 6 shows the distribution of changes in the ozone concentration from the base case for (a) the variation averaged in overall analysis period, (b) in July and (c) in high concentration days in July. In the NOx and VOC emission reduction case, the ozone concentration decreased in the Kanto region except for central Tokyo (around Shinjuku). The area where ozone concentrations increased corresponds well to the area with very high NOx emissions along the coastal area of Tokyo Bay. Therefore, the area where ozone concentrations increased was strongly affected by NOx emission reduction. From the distribution of the NOx reduction case, increases in ozone concentrations can be seen in a wide range of the Kanto region. In particular, the
tendency of the increment is greater in the coastal area of the Tokyo Bay than the central part of the Kanto region, and the effect of the NO\textsubscript{x} emission reduction is remarkable on the high concentration days. On the other hand, ozone concentrations tend to decrease over most of the Kanto region in VOC reduction case. Moreover, ozone concentrations decreased significantly around Omiya on the high concentration days.

4. Discussion

4.1 Changes in particular points of each month

In the three points where ozone concentration was decreased in the NO\textsubscript{x} and VOC simultaneously reduced case which shown in section 3.2, there is a large difference in the north–south concentration variation in the NO\textsubscript{x} reduction case. But there was no significant change in the point-to-point as NO\textsubscript{x} reduction case variation of ozone concentration in VOC reduction case. Therefore, it is considered to have the effect of decreasing the ozone concentration uniformly in month average. Large difference occurs in the amount of decrease of NO\textsubscript{x} emissions in emissions when compared to inland area, Kumagaya and Isesaki and central Tokyo, around the Shinjuku. But there is no great difference in VOC emissions as NO\textsubscript{x} emission indicated when compared with the surrounding central Tokyo and inland, around Isesaki and Kumagaya as shown in Fig. 2. Therefore, it is considered that show a uniform concentration variation in north and south VOC reduction case, but it has become a cause there is a difference between north and south in NO\textsubscript{x} reduction case.

It has been shown from a number of studies (e.g. Jacob, 1999) that the ozone concentration responds non-linearly to the change of VOC emissions and NO\textsubscript{x} emissions, and depending on the combination of NO\textsubscript{x} emissions and VOC emissions, ozone production can take two states, NO\textsubscript{x} sensitive (or NO\textsubscript{x} limited) and VOC sensitive (or VOC limited). Moreover, Sillman et al. (1998) shows that there is a state in which the ozone concentration decreases even by reducing both the NO\textsubscript{x} and VOC emission (Mixed sens). From Fig. 5, Isesaki is the Mixed sense because the ozone is decreasing in both the NO\textsubscript{x} reduction case and VOC reduction case in July and August. Moreover, VOC sensitive predominated at Isesaki in June and September because ozone concentrations increased about 0.2 ppb and 0.02 ppb in June and September, respectively in NO\textsubscript{x} reduction case but decreased in these two month in VOC reduction case. It is presumed for three points other than Isesaki that these points are often in region of the VOC sensitive when ozone concentration is increase in NO\textsubscript{x} sensitive and decreased in VOC sensitive.

4.2 Changes over the Kanto region of Japan.

In the coastal area with high emissions, it is expected to be in the state of VOC sensitive for the daytime maximum ozone concentration, because ozone concentrations decreased in the VOC reduction case and increased in the NO\textsubscript{x} reduction case, as shown in section 4.1. Meanwhile, when an attention is paid to around Isesaki and Kumagaya and northward, ozone concentrations are increasing over the area up to approximate latitude of Isesaki in an average of the entire analysis period and one month in July. Also, the point where the ozone concentration decreases in surroundings of Kumagaya had appeared in the average of the high concentration day. In addition, ozone concentrations decreased in the north of Isesaki. On the other hand, in the VOC reduction case, ozone concentrations decreased area had spread over the inland of the Kanto region. In the inland of the Kanto region, decrease in the precursors transported from the coastal high emission area by the sea breeze is a factor to decrease the ozone concentration. Ozone daytime maximum concentration increased over a large area in the NO\textsubscript{x} reduction case, but the increase was limited in a portion of central Tokyo in the NO\textsubscript{x} and VOC reduction case. Therefore, it can be considered in the comparison between 2000 and 2005 that the NO\textsubscript{x} reduction affects for the daytime maximum ozone concentration in the limited area of central Tokyo to increase daytime maximum ozone. In consideration of the ozone sensitivity regime similar to section 4.1, it is expected that the coastal area is in the state of VOC sensitive, and the reduction of VOC emission is effective for the decrease in ozone concentrations. Reduction of NO\textsubscript{x} and VOC emissions affects decreases in the daytime maximum ozone concentration in the inland. It is considered that the inland area is in the state of Mixed sens or VOC sensitive.

4.3 Ozone sensitivity to NO\textsubscript{x} and VOC in the high concentration day

In the previous sections, we indicated that the relationship between the variation of ozone concentration and each emission reduction cases. There are noted features and effect of each emission reduction cases which was shown from the tendency in four selected points and distribution over the Kanto region of the difference of daytime ozone maximum concentration. In this section, we try to grasp the feature by using the ozone sensitivity regime, which is used by Sillman and He (2002). Locations have been classified according to the following definitions, according to Sillman and He (2002).

(1) VOC sensitive regime: Ozone concentrations in the NO\textsubscript{x}2005_VOC2000 case are higher than those in both the
base case and the NO\textsubscript{2000}_VOC\textsubscript{2005} cases at a specific hour by at least 3 ppb.

(2) NO\textsubscript{x} sensitive regime: Ozone concentrations in the NO\textsubscript{2000}_VOC\textsubscript{2005} case are higher than those in both the base case and the NO\textsubscript{2005}_VOC\textsubscript{2000} cases at a specific hour by at least 3 ppb.

(3) Mixed sensitive regime: The NO\textsubscript{2000}_VOC\textsubscript{2005} and NO\textsubscript{2005}_VOC\textsubscript{2000} cases have ozone concentrations within 3 ppb differences from each other, and both have ozone concentrations higher than those in the base case by at least 3 ppb.

(4) Titration: Ozone concentrations in the base case is higher than those in the NO\textsubscript{2000}_VOC\textsubscript{2005} case by at least 3 ppb, and ozone concentrations in the NO\textsubscript{2005}_VOC\textsubscript{2000} case are not higher by 3 ppb or less than the base case.

(5) Insensitive: Other cases.

Sillman and He (2002) set NO\textsubscript{x} and VOC reduction rates as 35\% and 25\%, respectively. However, in this study, NO\textsubscript{x} reduction rate is about 6\% and VOC reduction rate is about 20\% as shown in Table 1. Therefore, differences in ozone concentrations between each case are expected to be small. So, we defined the threshold value as 3 ppb. Also, we determined the regime by focusing on a high concentration day, although Sillman and He (2002) used the determination for a change of ozone concentration at each time.

Figure 7 shows that an example of the distribution of regime on 28 July, one of the high concentration ozone days. The titration regime appears in the Tokyo Bay coastal area at 7:00. This area corresponds to an increasing area of ozone concentrations, and the titration effect by NO\textsubscript{x} seems to affect the increase in ozone concentrations at this time. The titration regime partially exists surrounding central Tokyo at 12:00, but largely indicate VOC sensitive. Therefore, it is likely that decrease of the ozone concentration is due to the reduction of VOC emissions. In addition, the mixed sens or NO\textsubscript{x} sensitive regimes, where ozone concentrations decrease, appear in the inland at 17:00, when sea breeze has entered.

Figure 7 also shows the distribution of the regime for daytime maximum ozone. From Fig. 7, the VOC sensitive regime distributed surrounding of central Tokyo. At the same time, the Mixed or NO\textsubscript{x} sensitive case sparsely exists in inland. Inoue et al. (2010) showed that the VOC sensitive regime distributed only in a part of the central Tokyo, and the regime changes from Mixed to NO\textsubscript{x} sensitive toward inland. In addition, the NO\textsubscript{x} sensitive regime is distributed in most of the Kanto area, as described by Inoue et al. (2010). Even in the present study, the VOC sensitive regime distributed in the surrounding the central Tokyo and the NO\textsubscript{x} sensitive regime distributed in the inland of Kanto. However, changes in the regime in relatively short ranges, which was highlighted by Inoue et al. (2010), have not seen in this study. Also, the NO\textsubscript{x} sensitive regime spindles compared to Inoue et al. (2010).

Figure 8 shows that numbers of occurrences of each regime for the daily maximum concentration in the four sites shown in Fig. 1.

Figure 8 indicate data in the top ten days when numbers of the appearance of grids over the 120 ppb in the base case. These ten days are 21, 25, 26 June, 18, 21, 29, 31 July, 3 August, 2 and 3 September. According to Fig. 8, Shinjuku and Omiya often take the VOC sensitive regime, but Isesaki and Kumagaya mainly take the mixed sens or NO\textsubscript{x} sensitive
regime than the other regimes. Features of effects which NO\textsubscript{x} and VOC give for high concentration ozone in each area of Kanto region had appeared.

5. Conclusion

In this study, we examined effects of the anthropogenic NO\textsubscript{x} and VOC emission reductions on ozone daytime maximum concentrations with use of WRF and CMAQ. The NO\textsubscript{x} and VOC emission reductions between 2000 and 2005 resulted in increase in ozone daytime maximum in and surrounding area of central Tokyo. But in other area, emission reduction works effectively to decrease ozone daytime maximum. Daytime maximum concentration was increased in a wide area of the plains in the NO\textsubscript{x} reduction case. On the other hand, ozone concentration decreased in the entire Kanto region due to reduction in VOC emission. Therefore, reduction of VOC emissions effectively works for decreases in ozone concentrations. In addition, ozone concentrations were lowered by either reduction of VOC and NO\textsubscript{x} emissions in Kumagaya and the north. It is considered that the ozone production decreased with amount of ozone precursors transported from central Tokyo by sea breezes and emissions from inland sources. According to the distribution of ozone sensitivity regime on the high concentration days, ozone concentration was increased by titration effect of NO\textsubscript{x} in the surroundings of central Tokyo of the early morning. But in the afternoon, VOC sensitive regime covered surrounding of central Tokyo and NO\textsubscript{x} sensitive or Mixed sensitive has appeared in inland. Occurrences of each regime on the highest ten days of the high concentration days shows that Mixed sens and NO\textsubscript{x} sensitive regimes accounted for half of the number of occurrences in Isesaki, and VOC sensitive and NO\textsubscript{x} sensitive regimes accounted for the other half. On the other hand, VOC sensitive regime is the majority in large cities such as Shinjuku and Omiya, and therefore VOC emission reductions are effective on decrease in ozone concentration in coastal, large city area. The results of present study show the effectiveness of the reduction of precursors to the decrease of ozone concentration in each area of the Kanto region of Japan.

References


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関東地方におけるNOₓおよびVOC排出削減のオゾン濃度に与える影響

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近年、日本国内において、オゾンの前駆物質濃度は減少する傾向にあるがオゾン濃度は長期的なトレンドでは増加している。越境汚染の影響が比較的少ない夏季の関東地方では国内の発生源対策が重要と考えられる。本研究ではWRFおよびCMAQを用いて、2000年から2005年にかけての排出量減少を反映した排出量データを用いた大気質シミュレーションを行い、排出量削減がオゾン濃度に与えた影響を検証した。その結果、2005年5月のNOₓおよびVOCの排出量削減により、オゾン日中最高濃度は東京都内の一部を除いた関東全域で減少を示していた。また、NOₓのみ削減した場合、オゾン日中最高濃度は東京都中心に増加するが、内陸部では減少することが示された。VOCのみの削減では関東の全域で濃度減少が示され、VOC削減の有効性が示唆された。さらに、高濃度に対する指標解析の結果、内陸部ではNOₓ sensitiveあるいはmixed sensの状態を取る事が多く、一方の都心周辺ではVOC sensitiveの状態が優勢であった。この結果よりそれぞれの地域でのオゾン濃度の低下に対する前駆物質の削減の有効性が示された。