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VERTICAL CIRCULATION OF AIR POLLUTANTS AND OZONE DISTRIBUTION OVER LOS ANGELES

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Abstract:
In order to investigate emission source contributions and air pollutants interactions within the planetary boundary layer (PBL), air quality simulations were conducted and focused on a high-ozone episode from 14 to 19 July 2005 over Southern California. Emission inputs were obtained from the U.S. EPA 2005 National Emissions Inventory, while meteorological fields were predicted by the WRF model, and atmospheric chemistry simulations were computed using the CMAQ model version 4.7.1. The modeling domain consisted of 229x169 horizontal grids with a resolution of 4 km and 41 vertical layers to resolve atmospheric circulations as well as pollutant transformation and transport processes. A zero-out method for seven emission sources was applied for sensitivity runs. The simulations revealed formation of high ozone concentrations near the top of PBL over the Los Angeles area. The results also indicate low surface ozone concentrations due to NO titration at the center of Los Angeles compared to surrounding areas. An increase in ozone concentrations was simulated at the altitudes where the VOC/NOx ratio increased and NO concentrations approached zero. A high–concentration portion near the PBL top and above the PBL top was confirmed. Sea and land breezes appear as the main mechanisms in generating this distribution. Ozone concentration in consideration with aging was discussed using HNO_3 and H_2O_2 distributions. Biogenic emissions contributed to a domain-wide ozone formation, however, they also contributed to high ozone concentrations near the PBL top over Los Angeles. In contrast, vehicle emissions contributed to the ozone formation far from the urban area in the lower free troposphere. The study also suggests that, in order to understand air pollution mechanism in an urban area, the interaction of a natural origin gas with an anthropogenic emission should be considered in detail.

Key words: *CMAQ, ozone, atmospheric pollution, chemical mechanisms, photochemical reactions, vertical pollutant distribution, emission source contribution*

INTRODUCTION

Los Angeles (LA), CA is located in complex terrain surrounded by mountains with ridges of more than 2000m. Anthropogenic air pollutants from urban area are generally transported by sea breeze and contribute to a high concentrations of ozone on the mountain slopes in the summer. It is called “a mountain chimney effect”, and some observational studies were carried out (e.g., Langford et. al. 2010). To understand air pollutant evolution in complex terrain, it is important to consider ozone formation through atmospheric transport and photochemical reactions of pollutants. Analysis of a vertical cross-section as a function of the location and height can be analysed air pollutants separately local or long-distance transport. The behaviour of high-concentration ozone air mass and their emission source contributions seen over the LA basin was investigated using episodic simulation of the Community Multiscale Air Quality (CMAQ) Modeling System (Byun and Schere, 2006).

METHOD

Emission inputs were obtained from the U.S. EPA 2005 National Emissions Inventory processed by the EPA SMOKE model. Meteorological fields were predicted by the WRF model, and atmospheric chemistry simulations were computed using the CMAQ model version 4.7.1. To accurately resolve atmospheric circulations as well as pollutant transformation and transport processes, the modeling domain

consisted of 229x169 horizontal grids with a resolution of 4 km and 41 vertical layers. A five-day period starting from 14 July 2005 was simulated focused on a high ozone episode in the LA basin. The evaluation of the simulated ozone concentration was carried out using data from 43 EPA monitoring stations in the Southern California. The model results agree fairly well with measurements based on the recommended EPA measures.
A zero-out method for seven emission sources was applied to understand the contribution of ozone formation. The emission included on-road gasoline, on-road diesel, off-road gasoline, off-road diesel, residential, industrial, and biogenic sources.

Vertical wind velocity was estimated from the convergence and divergence of horizontal wind velocity in CMAQ ready wind data in consideration of hydrostatic balance. In order to obtain vertical cross sectional distribution which is easy to understand intuitively, including ozone concentration, layer's values of the σP coordinates was converted to the Cartesian coordinates.

RESULTS

Figure 1 shows one example of ozone distribution and wind field at 18:00 Pacific Standard Time (PST) on 17 July, 2005. This figure is a latitude cross-section including LA, the Pacific Ocean is to the left-hand side, and horizontal extension is 404km. The lower layer was expanded like grid spacing of CMAQ, so that the details of the ozone and wind field distributions can be clearly seen. High concentration air mass of ozone of more than 75ppb is shown in white. This area include Riverside, San Bernardino Mt. (SB-Mt), and the Pacific Ocean. Over LA, the ozone concentrations are lower than over the surrounding area by the effect of NO titration. Counter-clockwise air flow was simulated in the west side of SB-Mt, and some circulations were seen in the east side of SB-Mt.

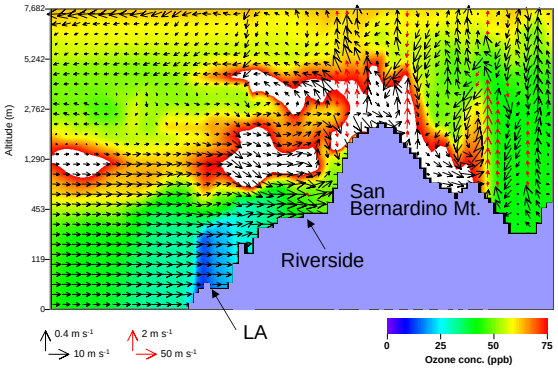

Figure 1. A latitude cross-section of ozone distribution and wind fields at 18:00 July 17, 2005

Figure 2 shows the vertical distribution of air pollutants at the same time shown in Figure 1. High NOx concentrations are distributed near the surface of the urban area between LA and Riverside as shown in Figure2a. NOx was transported to the east by sea breeze, and at the place where marine ozone merges NO from LA, ozone concentrations are reduced by the oxidation reaction. VOC distributions shown in Figure 2b were much wider than the NOx distribution. The highest concentration was observed over the west slope of SB-Mt. Up-draft flow transported VOC above the planetary boundary layer (PBL). High concentration area was simulated at the PBL top over the Pacific Ocean was caused by the VOC transported from the northwest. Figure 2c shows HNO_3 concentration coming from the NOx oxidation and fairly abundant in fresh air masses. High concentrations of HNO_3 can be seen downstream of areas with high NOx concentrations. Comparably high concentrations of HNO_3 was seen above PBL, which was caused by the vertical circulation such like VOC. In contrast, H_2O_2 shown in Figure 2d is usually seen in aged air masses. High concentrations of H_2O_2 at the PBL top over the Pacific Ocean with the same distribution as VOC, was made from aged air pollutants that were transported from the northwest.

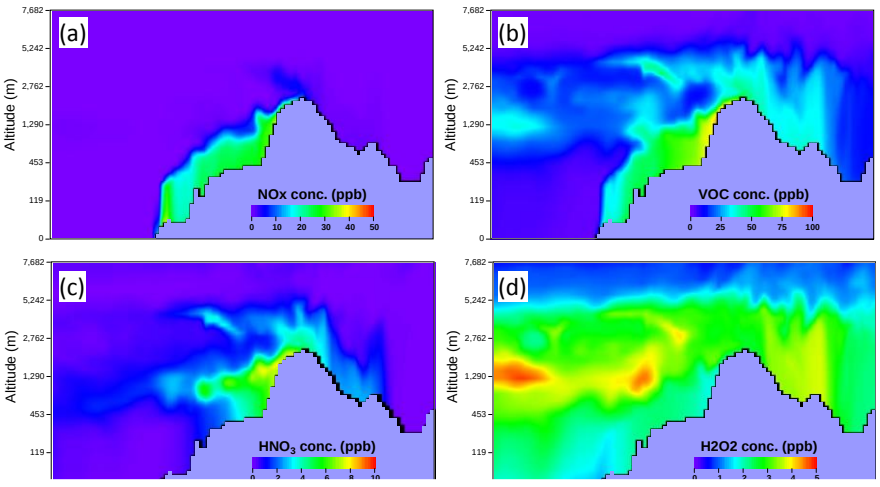

Figure 2. A latitude cross-section of air pollutants distribution at 18:00 July 17, 2005: (a) NOx, (b) VOC, (c) HNO_3, (d) H_2O_2

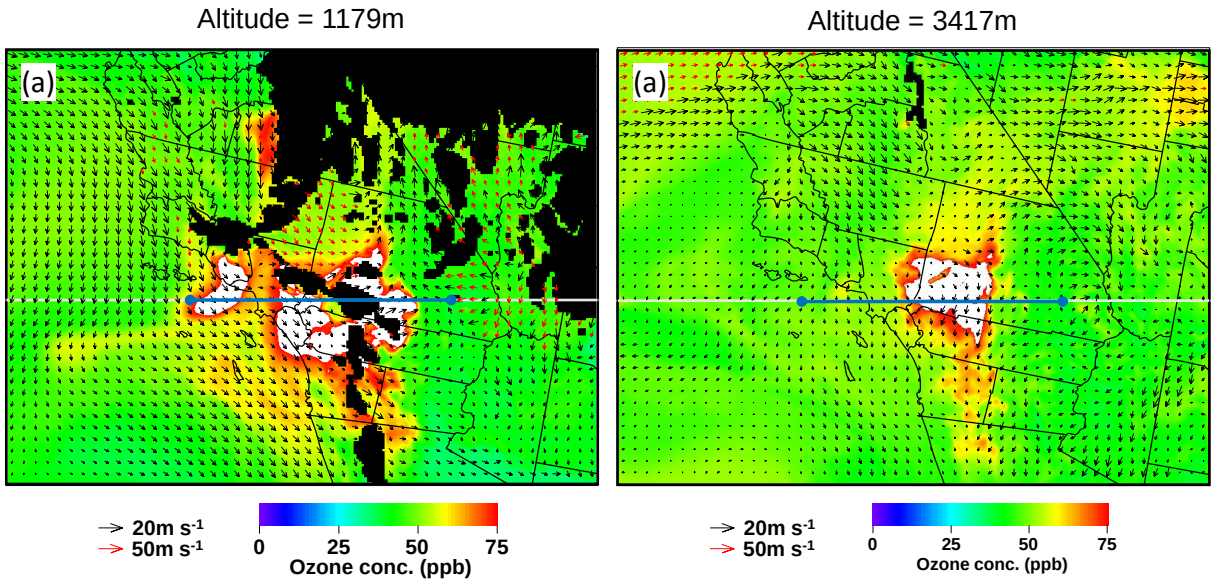

Figure 3. Horizontal distributions of ozone concentration and windfields at 18:00 July 17, 2005. (a) ozone distribution at the 1,179m height (top of the PBL) and (b) at the 3,417m. Black area indicates locations of mountains at the each height. The blue bar in the middle shows the area corresponding to Figure 1 and Figure 2.

As mentioned above, the high ozone concentration simulated at the PBL top over the Pacific Ocean (Figure 1) is most likely generated from aged air mass that was transported from the northwest. Furthermore, the high concentrations over SB-Mt. as well as the high concentrations above the PBL were possibly generated from fresh air pollutants from the urban area between LA and Riverside. In order to understand history of these two high ozone air masses, horizontal ozone distributions with constant two levels of altitude created (Figure 3). From the temporal variation of the hot spot (white area of ozone concentrations of more than 75ppb) one can see history of the pollutant transport. In Figure 3a, the hot spot over the Pacific Ocean moved from near Santa Barbara to LA by the northwest wind. A significant

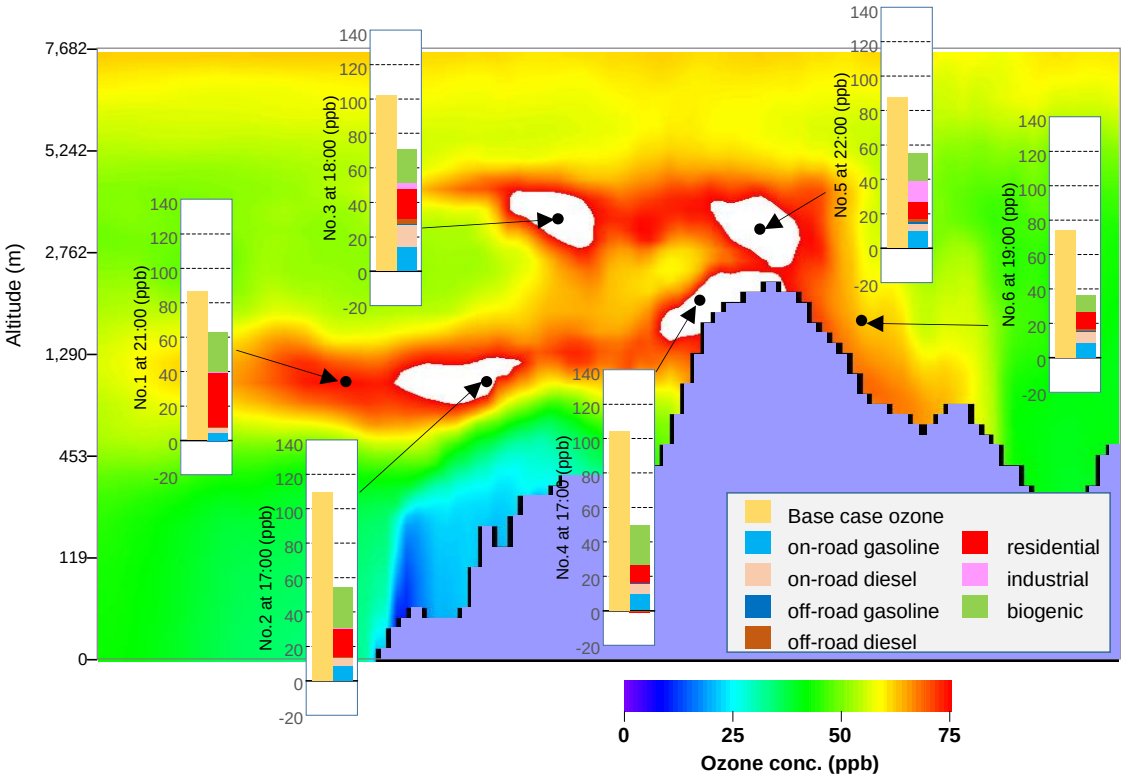

Figure 4. Time averaged hot spot distributions (high ozone concentrations of more than 75ppb) from 13:00 to 23:00 on July 17 and emission source contributions at typical points which showed the maximum concentration.

transport is not shown in the hot spot in Figure 3b, but the rotation in the same place can be seen. Angevine et. al. (2013) studied the pollutants aging by using the Lagrangian simulation and showed aging about 20 hours of automotive-emitted CO from the northwest.

Figure 4 shows the averaged hot spots distribution simulated from 13:00 to 23:00 on July 17, as well as emission source contributions at typical points where the maximum ozone concentration was simulated. Source contributions for ozone concentrations were obtained by the zero-out method for seven emission sources. Location 1 in Figure 4 was the point where high concentration of air pollutants were transported from the northwest. The residential emission contribution showed highest impact of 36.5%, and the second highest impact was by the biogenic sources of 26.1%. On-road gasoline and on-road diesel emission contributions were 4.8% and 4.1%, each. Point 2 showed a location of the highest ozone concentration of 110ppb. This is the nearest hot spot to an urban area between LA and Riverside. However, the vehicle emission contribution is smaller than the effect of the biogenic emissions (the highest contribution of 21.6%). The on-road gasoline emissions which were the highest contributor among vehicle sources contributed 7.9%, and the total vehicle contribution was 12.4%. The second important contributor were residential emissions (15.1%). Vehicle emissions showed the highest contribution at point 3 which is far from the urban area between LA and Riverside in comparison to points 2 and 4. The on-road gasoline contribution was 13.9% and the total vehicle emission contribution was 29.6%. Biogenic and residential emission contributions were 19.0% and 17.5%, respectively. They are unexpected results that the highest contributor was vehicle emission at the point away from the urban area, and biogenic contribution became small in reverse. The magnitude order of the small contributions of on-road gasoline emissions was location points 2,4,6,5, and 3, except for point 1. This suggests that air pollutants from vehicle exhausts in urban area began to be oxidized and to contribute to ozone formation gradually during the atmospheric transport. On the other hand, it is suggested that the biogenic emissions are oxidized during the transport to urban areas and become active species to react in ozone formation by interaction with anthropogenic emissions from the urban area.

SUMMARY

A five-day simulation starting from 14 July 2005 was conducted by using CMAQ air quality model focused on an observed high ozone concentration episode. In order to understand three-dimension behaviour of ozone formation and the relationship to other air pollutants, detailed numerical and graphical analysis was carried out. The main conclusions are as follows.

1. By using CMAQ ready horizontal wind data, vertical wind velocity was estimated. In a latitude cross-section including Los Angeles (LA), counter clockwise air flow was revealed in the west side of San Bernardino Mt. (SB-Mt), and some circulation patterns were seen on the east side of SB-Mt.
2. High concentrations of ozone (hot spots) with concentrations of more than 75ppb were simulated over Riverside, SB-Mt, and the Pacific Ocean near the top of planetary boundary layer (PBL), and above the PBL over Riverside.
3. A hot spot located near the urban area between LA and Riverside with high HNO_3 concentrations suggests ozone formation by fresh air pollutants. A hot spot over the Pacific Ocean with high H_2O_2 suggests ozone formation by aged air pollutants transported from the northwest. A hot spot above PBL was suggested to be formed by the air pollutants from the urban area via the vertical circulations above SB-Mt.
4. The on-road vehicle emission contribution to ozone formation appears to be small (4.8%) near the urban area but large (13.9%) above the PBL outside of the urban areas. In contrast, the biogenic contribution was large (21.6%) near the urban area and small (19.0%) at remote places above the PBL. Our results suggest that it is important to understand the transport and aging time of the anthropogenic air pollutants and biogenic emissions in order to propose efficient control strategies in reducing emission sources relevant to ozone formation and evolution especially for conditions of high-concentration episodes.

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