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TROPOSPHERIC OZONE IN THE WESTERN PACIFIC RIM : ANALYSIS OF SATELLITE AND SURFACE-BASED OBSERVATIONS ALONG WITH COMPREHENSIVE 3-D MODEL SIMULATIONS

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ABSTRACT

Tropospheric ozone production and transport in mid-latitude eastern Asia is studied. Data analysis of surface-based ozone measurements in Japan and satellite-based tropospheric column measurements of the entire western Pacific Rim are combined with results from three-dimensional model simulations to investigate the diurnal, seasonal and long-term variations of ozone in this region. Surface ozone measurements from Japan show distinct seasonal variation with a spring peak and summer minimum. Satellite studies of the entire tropospheric column of ozone show high concentrations in both the spring and summer seasons. Finally, preliminary model simulation studies show good agreement with observed values.

1. INTRODUCTION

The characteristics of tropospheric ozone production and transport in the middle latitudes of the western Pacific Rim region are being studied. This region, which includes the countries of China, Japan, and Korea, is undergoing accelerated economic growth and industrial expansion resulting in, among other things, unprecedented increases in the anthropogenic emissions of ozone precursors such as nitrogen oxides and hydrocarbons. It has already been shown that this region of Asia has the highest tropospheric ozone column values anywhere on earth (Fishman et al., 1990). Thus, the effects of tropospheric ozone in this region are clearly significant and, furthermore, its impact on the global troposphere remains unquantified. In this paper results from both satellite and surface data analyses and those from preliminary three-dimensional simulations on a regional scale using a detailed tropospheric trace gas model (i.e., the STEM-II model) are presented.

2. SURFACE DATA ANALYSIS

The surface-based observations of ozone are from the Background Surface Ozone Monitoring Network of Japan which was established in the 1980's (NIES Reports, 1989). There are over ten monitoring stations dispersed throughout Japan mostly at sites such as mountain tops and remote islands. The time span of data collection varies with station ranging from six years to three months of hourly values. Over the entire sampling period, about half of the stations, most of them from the island of Honshu, show an increase of 1-5% in the monthly average of hourly measurements while the rest of the stations show no visible trend in either direction. Figure 1 shows the monthly maximum, average and minimum values and a simple linear regression of the three parameters for two different stations for approximately a three year period. Hourly values have been averaged for each month. Figure 1(a) is data collected at Mt. Tokusagamine in Yamaguchi prefecture on the southern tip of Honshu, the main island of Japan, while the measurements in Figure 1(b) are from the Amami station, located on a small island several degrees to the south of Kyushu island. The former station shows a small uniform increase in the average values as well as the maximum and minimum values. This station is at an elevation of about 500 (m) above sea level and its geographic location makes it susceptible to influences of transport of anthropogenic precursors of ozone such as nitrogen oxides and of ozone itself. The remoteness of the Amami station most likely keeps it away from much anthropogenic influence. Figure 1(b) shows a very slight decrease in all three indices but there is a gap in the data from the summer of 1986 to spring of 1987 which is not taken into account. Though the maximum hourly value of the month has decreased with time the trend is not statistically significant.

The seasonal variation for these two stations is common to all the other background stations in Japan showing a peak in the spring and a minimum in the summer. Amami reaches its peak in late March or early April while Yamaguchi has its peak in late April/ early May. The maximum in spring is slightly earlier compared to similar stations in the eastern United States and western Europe which usually have their maximums in late spring and summer (Logan, 1989). Some have attributed this relatively early spring maximum to natural peaks in the stratospheric intrusion process during this season (Ogawa and Miyata, 1984). The higher frequency of stratospheric-tropospheric exchange during this season bring higher ozone concentrations into the upper troposphere. Others claim a photochemical explanation (Liu et al., 1987). Colder temperatures during winter and early spring allow for longer transport distances and accumulation for ozone and its precursors followed by peaks in concentration due to warmer temperatures and increased photochemical activity. The summer minimum may be possibly attributed to a shift in the dominant meteorology over the region from continental northeasterly winds from China brought on by the Aleutian low-pressure system in the winter season to

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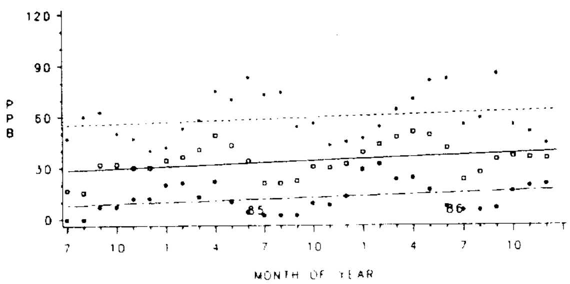
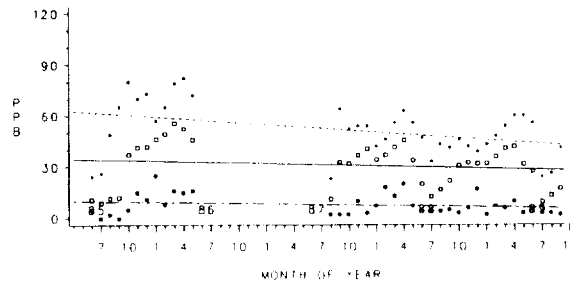


(a)



(b)

Figure 1. (a) Monthly maximum, average and minimum surface ozone values for Yamaguchi Station. Elevation : 530 (m) (b) Monthly maximum, average and minimum surface ozone values for Amami Station. Elevation : 300 (m)

mostly southerly winds bringing moist air from the lower latitudes in the summer (Whelpdale and Moody, 1990).

Figure 2(a) shows a plot of daily average ozone and temperature at Happo in the heart of Japan for a period of five months in 1987. The drop-off of the ozone concentration in early July is very clearly shown while the surface temperature continues to rise as the summer develops. Happo is located at an elevation of about 1800 (m), essentially in the free troposphere, and the simultaneous measurements of ozone, dust, temperature, pressure and relative humidity is providing valuable insights into deciphering the sources of ozone, whether it be local sources, long range transport from urban centers, in situ photochemical production, or downward transport from the stratosphere and the upper troposphere. At this stage of the analysis, positive correlations between ozone and dust concentration peaks in the spring season, along with the corresponding meteorological conditions, seem to show that a combination of a passing cold front followed by general subsidence under a stagnant high-pressure system over Japan result in downward transport of ozone-rich, dust-rich mid-troposphere air and increases in photochemical activity causing higher production of ozone.

Similar plots for Amami and Yamaguchi (for years 1988 and 1986, respectively) are shown in Figure 2(b). As discussed above, Amami reaches its spring peak sooner and the summer minimum also arrives earlier, the latter most likely due to its southerly location making it more susceptible to relatively early influences from clean lower latitude air. The overall values of ozone concentration are lower for Yamaguchi relative to Happo, and Amami lower still. Site elevation and proximity to sources are the primary reasons behind these differences.

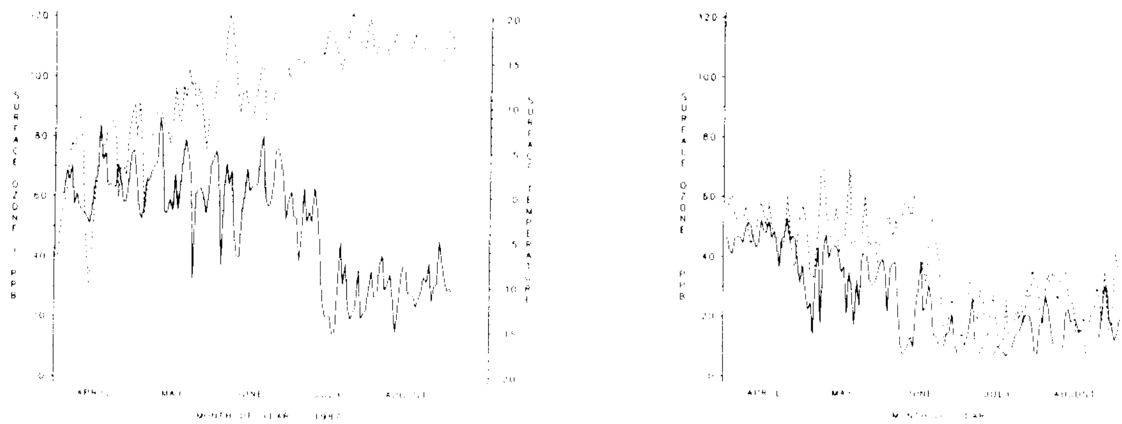
3. SATELLITE ANALYSIS OF TROPOSPHERIC COLUMN

To study the trends and regional distribution of the entire tropospheric column in the western Pacific Rim, satellite analyses have been performed utilizing the TOMS instrument on board the Nimbus-7 satellite which measures the total ozone column using back scattered UV-radiation and the SAGE-II, charting the vertical distribution of ozone in the stratosphere and above by the solar occultation method, on the Earth Radiation Budget Satellite (Fishman and Larsen, 1987). Data from the years of 1985-87 for the region covering 20-50° N and 100-160° E have been analyzed by subtracting the integrated SAGE-II measurements above the tropopause from the total column data provided by TOMS. It should be pointed out that though the concept is very simple, there are still some uncertainties in the actual implementation of this methodology such as the accuracy of the TOMS instrument itself (especially in the treatment of clouds), inherent problems of the analysis such as the time difference of the TOMS and SAGE-II measurements (local noon versus local sunrise/sunset), and application to the relatively unstable midlatitude circulations, particularly with front and jet stream activity.

Results show a peak for the tropospheric column in East Asia in the spring and summer seasons of over 40 Dobson units as is seen in Figure 3 (averaged for all three years). An accurate interpretation of the data is hampered by the missing months of June and August, for which the number of data points were too few to be considered significant. However, it is clear that the tropospheric column values for the summer season are much more prominent when compared to the very low values obtained from surface measurements during this season in Japan. The regional distribution of the tropospheric column ozone shows good correlation with the prevailing seasonal wind patterns for the middle to upper troposphere. For example, the seasonal peak locations are downwind of heavy emission areas. There is also some evidence that the production and transport of tropospheric ozone in the upper half and lower half of the covered region of 20-50° N latitude are predominantly governed by differently weighted combinations of influencing factors, both meteorological and photochemical. The relatively small number of overlapping "points" between the two satellite instruments is still a substantial problem but with continued data collection expanding the data base this methodology should prove invaluable in studying the tropospheric column ozone behavior in the upcoming years.

4. PRELIMINARY 3-DIMENSIONAL SIMULATION

The STEM-II model (Carmichael et al., 1986) is



(a) (b)

Figure 2. (a) Daily averages of surface ozone and temperature at Happo station, elevation 1800 (m), during five months of 1987. The units for ozone is ppb and that for temperature is °C. Ozone is the solid line and temperature is the dashed line.
(b) Daily averages of surface ozone for Amami (for 1988; solid line) and Yamaguchi (for 1986; dashed line)

being utilized to conduct three-dimensional simulations of tropospheric ozone transport, both horizontal and vertical, and photochemistry in the study region of the western Pacific Rim. The STEM-II is a comprehensive three-dimensional, transport/chemistry /removal model. The current version of the model uses the photochemical mechanism of Lurmann et al. (1986), which includes 112 chemical reactions and 53 chemical species. This mechanism is best suited for analysis of trace gas cycles under conditions where NO_x levels are > 0.1 ppb. In order to extend the mechanism to lower NO_x conditions and to explicitly include the role of isoprene chemistry this mechanism has been modified as discussed in Jacob and Wofsy (1988). The complete mechanism includes 70 species and 200 reactions. In addition to gas phase photochemistry, the model treats cloud chemistry, and cloud micro-physics. A dust transport and chemistry module has been added to look more closely at the problem of "yellow sand" transport that is quite prevalent in East Asia. This phenomenon is thought to significantly influence the atmospheric chemistry and trace gas budgets of this region. Preliminary results show good agreement between simulated and observed data.

A springtime episode in May of 1987 is used to investigate the characteristics of tropospheric ozone production and transport during this season, including diurnal cycles. Figures 4 (a) and (b) show the results of a preliminary simulation. Both figures display ozone concentrations at surface level in ppb for the fourth day of simulation, one at 8 AM and the other at 8 PM local time. The first plot shows low values of surface ozone on the continent while the pocket of high ozone lies in south central Honshu with peak values of 80-100 ppb. This region of high ozone moves to north central Japan after a 12-hour period under the influence of a south-southeasterly wind and also grows in size. Also prominent is the 50-80 ppb region on the northern Chinese coastal areas at this time. The peak values in the early afternoon reaches values of over 100 ppb for the simulation results. Finally, slightly higher level

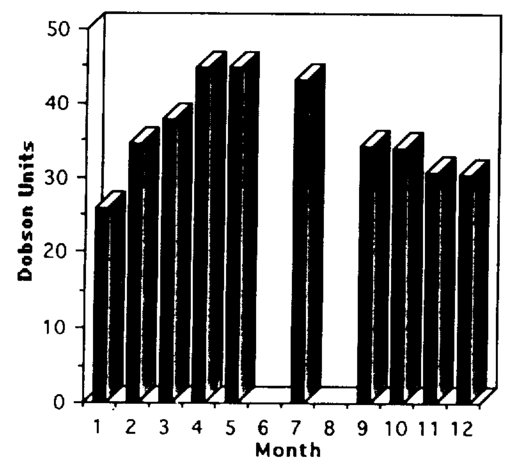
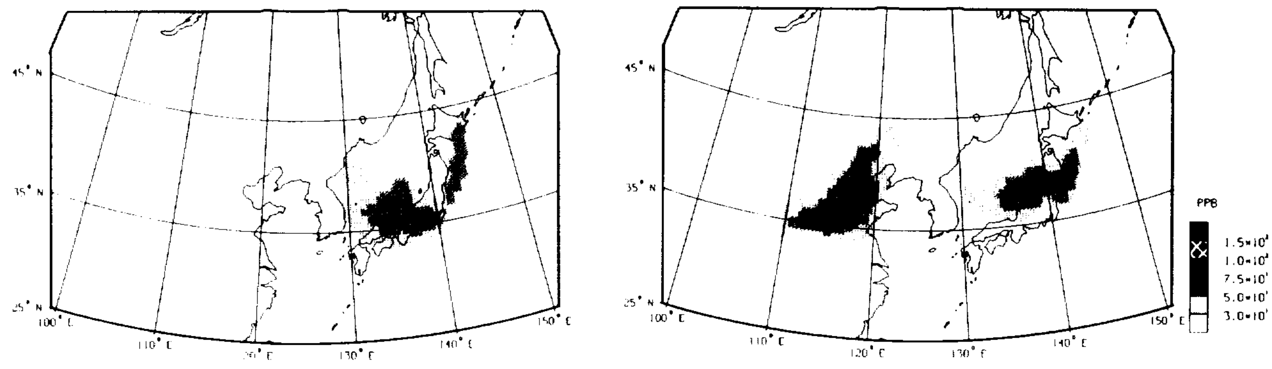


Figure 3. Monthly averages of tropospheric ozone residual column values in Dobson units (2.69×10^{16} molecules/cm^2). Averaged over the three years of 1985-87. June and August missing due to insufficient data. Overall region covered is 20-50°N and 100-160°E.

ozone concentrations reach 60-70 ppb at elevations of 1000 (m) above Japan while above the Chinese continent values of 50 ppb can be seen.

5. CONCLUDING REMARKS

The production and transport characteristics of tropospheric ozone in East Asia is being comprehensively studied utilizing three different methods. Each methodology gives a different perspective of the phenomenon and together they are adding to our insight of the tropospheric ozone production, transport and destruction pathways. The



(a) (b)

Figure 4. (a) Simulated results of surface ozone on 8 AM of May 11, 1987 (Day 4 of simulation). (b) Simulated results of surface ozone on 8 PM of May 11, 1987 (Day 4 of simulation).

continuous monitoring of background surface ozone in Japan is proving quite valuable and though the time span covered is too short to state conclusively, a uniform trend in either direction is not seen. Both downward transport of ozone-rich upper atmosphere air and accumulation and production of ozone and its precursors over the winter are possible explanations for the consistent spring peak in ozone concentration. However, the summer minimum is most likely brought about by dynamic influences of the southerlies dominating this season. Tropospheric column values show high values for the summer season along with the spring. The discrepancy in the summer may be caused by differences in dynamic regimes. Finally, preliminary simulations show that the model does a relatively good job in following the movement of pockets of elevated ozone concentrations. A more detailed and comprehensive simulation is currently under way. Also, a more rigorous statistical study of the satellite data is currently being conducted and results of an in-depth look into the surface ozone data is presented elsewhere (Sunwoo et al., 1992).

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