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Tropospheric Ozone Production, Transport, and Distribution in East Asia

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I. Introduction

The characteristics of tropospheric ozone production and transport in the middle latitudes of East Asia have been studied using surface measurements, satellite observations, and regional-scale model simulations. This western Pacific Rim region, which includes the countries of China, Japan, Taiwan, and Korea, is undergoing accelerated population growth and industrial activity resulting in marked increases in the anthropogenic emissions of ozone precursors such as nitrogen oxides and hydrocarbons. The impact of these unprecedented increases of pollutants on the regional and global environment is expected to be very big. However, investigations have only recently been initiated in this region.

The long range transport (LRT) and chemical transformations of these ozone precursors and other trace species, pollutants, and aerosols in this region is a potentially important issue for tropospheric ozone, both regionally and globally. Long range transport of gases such as ozone and aerosols in the atmosphere is mostly governed by the prevailing synoptic wind fields. In the western Pacific Rim, as is the case in many mid-latitude locations of the northern hemisphere, the dominant flow is westerly owing to the jet stream activity. Because of the rapid industrialization of China and the continued accelerated growth of many Eastern Asian countries the eastward flux into the relatively clean Pacific air of trace gases attributable to anthropogenic sources is expected to grow substantially in the next few decades. This westerly flow is already responsible for the well-known dust storm episodes in Korea and Japan caused by the LRT of dust originating in the desert areas of China and Mongolia. This dust is transported all the way to the islands in the mid-Pacific. Observational data and modeling studies show possible dynamic and/or heterogeneous chemical interactions between the dust-rich and ozone-rich air masses for this region (Zhang et al., 1993). Coincident measurements of dust and ozone at a high altitude surface site in Japan show that during dust events the ozone and dust concentrations behave similarly on the time scale of days (i.e., ozone increases as the dust concentration increases) but are anti-correlated on hourly time-scales (Kotamarthi et al., 1991). Dust, with a well characterized surface source, provides a valuable tracer which can be exploited to help quantify the source of the tropospheric ozone budget.

Most of the global surface ozone measurement stations in the Northern Hemisphere report a spring maximum in their data (Singh et al., 1978; Ogawa and Miyata, 1985). It has been argued whether the spring ozone peaks have been caused by photochemical production from anthropogenic emissions of precursors or intrusion of stratospheric air with high concentrations of ozone. For many years it was believed that meteorological conditions during the spring were more conducive to stratospheric intrusions (Danielsen, 1968). The argument for stratospheric intrusion is centered on the claim of increased frequency of stratospheric intrusion events in the spring season compared to other seasons. However, it has also been suggested that the nonlinear characteristics of ozone photochemical production from nitrogen oxides and hydrocarbons are the cause of the spring peaks due to longer lifetimes and longer transport distances under colder temperatures. Liu et al. (1987) suggests that the main cause of the spring maximum observed in most rural surface ozone monitoring sites is caused by gradual accumulation of ozone over the winter months when the lifetime of ozone is significantly longer than in summer. There is no conclusive evidence which of the hypotheses is correct but it seems likely that both would be a contributing factor in causing these spring peaks.

In this paper some sample results from surface data analyses of background ozone, satellite analyses of tropospheric column ozone, and those from a comprehensive three-dimensional simulation on a regional scale are presented.

II. Behavior of Background Surface Ozone

The background ozone monitoring network in Japan was set up in the 1980's to study the diurnal, seasonal, and long-term behavior of background ozone in the Japan islands. The ozone data are monitored at relatively clean "remote" sites in the islands ranging from 24°-44° N and 124°-143° E (NIES Reports, 1989). Most of these monitoring sites are located on top of high mountains or on remote islands. All the stations show definite seasonal variations, with the peaks usually coming in the spring (usually April or May) and smaller values in the summer and autumn. Figure 1 shows the monthly maximum, average and minimum values for one site for approximately a three year period. Hourly values have been averaged for each month. The data was collected at Mt. Tokusagamine in

Yamaguchi prefecture on the southern tip of Honshu, the main island of Japan. 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 seasonal variation for this station is common to all the other background stations in Japan showing a peak in the spring and a minimum in the summer. 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. Some have attributed this relatively early spring maximum to the aforementioned natural peaks in the stratospheric intrusion process during this season. 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 mostly southerly winds bringing moist air from the lower latitudes in the summer.

The characteristic diurnal patterns for the stations of Amami, located on a small island several degrees to the south of Kyushu island, and Yamaguchi are shown in Figures 2a&b. No appreciable diurnal cycle in the winter month of February, 1986 is evident for the Yamaguchi site while a slight dip is seen during the daytime hours in Amami. The mean value for the southern island site of Amami is slightly higher than Yamaguchi, at 49 ppb and 44 ppb, respectively, but the Amami data seems to indicate that 1986 might have been an abnormal year for this site. Figure 2b shows the diurnal patterns for a summer month, August 1985. The remote low latitude island site of Amami consistently shows a shallow daytime dip, possibly from photochemical reactions resulting in a small but net sink of ozone due to low NO_x concentrations. This may also be a result of a dilution effect due to the development of the mixing layer during daytime hours. Yamaguchi has a photochemistry-induced diurnal cycle with an afternoon peak that is about 15 ppb higher than the averaged nighttime concentrations. The Yamaguchi site is located in a mountainous terrain with an elevation of 530 m. In summer, the mesoscale transport of polluted air masses from nearby Yamaguchi City and industrial areas sometimes occurs along the coast to this measurement site in the late afternoon or early evening.

III. Tropospheric Ozone Analysis from Satellite-based Measurements

Fishman and Larsen (1987) have developed a method of estimating tropospheric ozone using satellite measurements of total ozone and stratospheric ozone. The total ozone data comes from the TOMS (Total Ozone Mapping Spectrometer) instrument aboard the polar-orbiting Nimbus-7 satellite. This instrument works on the principle of backscattered ultraviolet radiation. The stratospheric ozone data comes from the SAGE II instrument aboard the Earth Radiation Budget Satellite. The two data sets of total and stratospheric ozone have several data "points" which overlap with respect to time and space. From each of these pairs, we can calculate an implied tropospheric residual ozone value by subtracting the stratospheric ozone column from the total ozone.

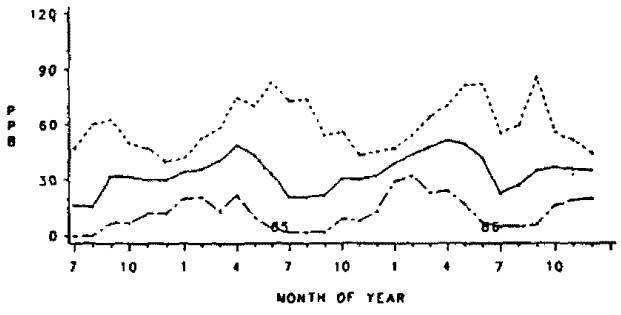

Figure 1. Maximum 1-hour ozone concentration value for the month, the monthly mean, and the minimum for the month for Yamaguchi.

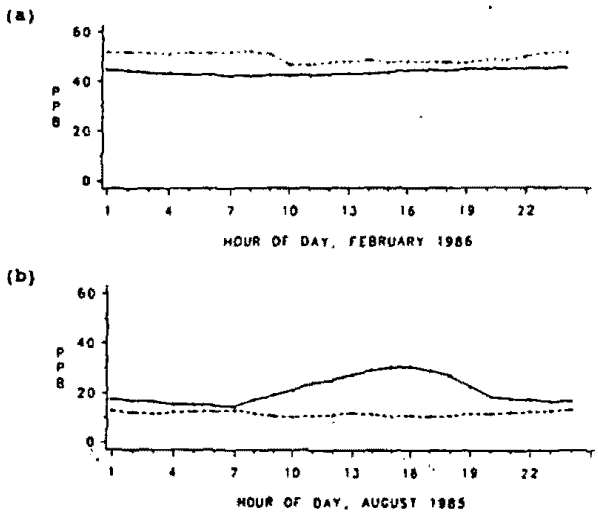

Figure 2. Dirunal variation of the measured ozone concentrations for Amami and Yamaguchi: (a) Mean values for each hour of day in February, 1986 (b) Means for August, 1985 (The dashed lines are the Amami data).

Results show a peak for the tropospheric column in East Asia in the spring and summer seasons of over 40 Dobson units (1 Dobson unit = 2.69 x 10^{16} O_3 molecules cm^{-2}). 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. Our results are in close agreement with the tropospheric ozone values on Fishman's global plots for the Pacific Rim region (Fishman et al., 1990). The key issue in tropospheric ozone in this region is the relative contributions of the stratospheric intrusion process and the photochemical production process to the tropospheric ozone budget. However, the roles of anthropogenic activity and tropopause folding in the tropospheric ozone budget of this region remains unquantified.

The tropospheric ozone distributions from the residual analysis show not only a seasonal dependency but is a function of latitude as well. The frequency distribution of the residual column with respect to latitude is given in Figure 3. The latitude band groups are 20-30°N, 30-40°N, and 40-50°N. The spatial differences are not as noticeable as the seasonal ones, but there is a slight peak for the middle latitude band of 30-40 N. This can be seen by the higher frequency of cases between 40-60 D.U. for this band. An interesting feature of this band, relative to the other two bands, is the concentration of major urban centers within this band. Almost all of Japan excluding the northern island of Hokkaido, the Korean peninsula, and most of the major cities of China lie in this area. This supports the hypothesis that photochemical production of ozone from anthropogenic precursors is responsible for the high residual values appearing in the middle latitude zone.

IV. 3-Dimensional Simulation of Tropospheric Ozone Production and Transport

The mechanisms of transport, chemical transformation and removal of ozone and other trace gas species have been studied using a detailed three-dimensional tropospheric trace gas model (i.e., the STEM-II model). A springtime episode in May of 1987 is used to investigate the characteristics of the photochemical oxidant cycle and its effect on the tropospheric column ozone budget and the effects of pollutant long-range transport on the distribution of ozone in this region. The STEM-II (Carmichael et al., 1986) is a comprehensive three-dimensional, transport/chemistry/removal model. In order to fully capture the potential contribution of the effects of photochemical production and LRT of oxidants, including ozone, and its precursors on the air quality of the region and the advection out into the Pacific Ocean, a significantly large domain covering about 63 degrees in longitude from 99-162°E and

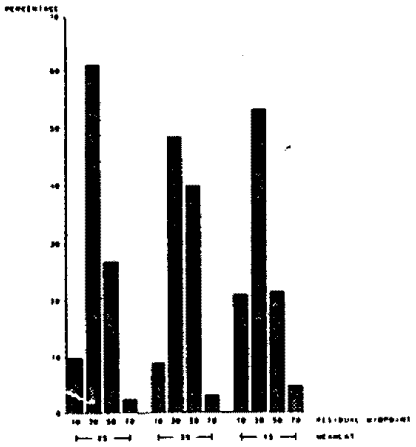

Figure 3. Frequency distribution of the derived tropospheric residual for East Asia grouped by latitude (denoted by the mean latitude of a 20° latitude band). The residuals are represented by the mean value of a 20 D.U. interval range.

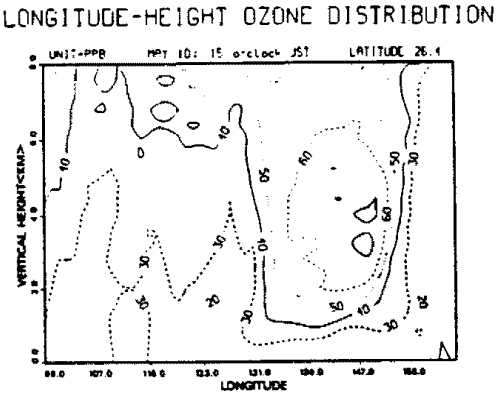

Figure 4. Longitude-height cross-section of predicted ozone distribution at 3 P.M. JST for the 10th of May, 1987 along 26.4° N.

about 36 degrees in latitude from 19.626-55.514°N was selected for this simulation. The domain includes all of Japan, North and South Korea, Taiwan, most of China, and parts of Russia and Mongolia. The vertical domain is taken to be from the surface to 8 km with the increment being 400 m for a total of 21 vertical points. The simulated period is May 6-13, 1987.

A longitude-height cross-section is shown in Figure 4 for the latitude "line" which had high column ozone over the Pacific Ocean. The latitude is 26.4° N and the 3 P.M. cross-section for one day of the simulation is shown to observe the buildup of this high columnar value. A strong downflow emerges on the 9th of May and pockets of air containing 60 ppb of ozone are formed. This air mass continues to move downward on the 10th while also building its ozone concentration. The shape of the contours signify that the increases are through downward and horizontal convergence. Values of 60 ppb reach the 2 km level. Finally, on the 11th, the ozone mixing ratio reaches values of over 90 ppb at a height of just over 2 km. It is evident that very high mixing ratios of ozone are capable of reaching the lower atmosphere. A combination of consistent downward flow and a strong horizontal convergence due to a deep low pressure trough results in almost urban-like ozone mixing ratios close to 100 ppb only a couple kilometers above sea level in the middle of the Pacific. It should be noted that if the same combination of meteorological factors occurred over a urban/suburban area or even over a rural area local or regional plumes containing high volumes of ozone and its precursors may merge with the downflowing air from the upper troposphere. If the conditions were favorable for efficient photochemical production, such as under a subsiding high pressure system which usually trails the low pressure troughs, this may result in a so-called ozone episode of over 150~200 ppb of ozone near the surface.

In addition, photochemical activity involving NO_y transported by continental plume is believed to be one of the causes of the high pocket of 60 ppb ozone seen in Figure 4. These high concentrations of NO_y at such broad marine altitudes should contribute heavily to the large mixing ratios of ozone and the high total column values. Thus, photochemical production, together with the long range transport of ozone and its precursors, and in situ production during the LRT process all combine to produce this very high ozone air mass in the middle of the Pacific.

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