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# Trends of ozone in the troposphere

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**Abstract.** Using a set of selected surface ozone (nine stations) and ozone vertical profile measurements (from six stations), we have documented changes in tropospheric ozone at a number of locations. From two stations at high northern hemisphere (NH) latitudes there has been a significant decline in ozone amounts throughout the troposphere since the early 1980s. At midlatitudes of the NH where data are the most abundant, on the other hand, important regional differences prevail. The two stations in the eastern United States show that changes in ozone concentrations since the early 1970s have been relatively small. At the two sites in Europe, however, ozone amounts increased rapidly into the mid-1980s, but have increased less rapidly (or in some places not at all) since then. Increases at the Japanese ozonesonde station have been largest in the lower troposphere, but have slowed in the recent decade. The tropics are sparsely sampled but do not show significant changes. Small increases are suggested at southern hemisphere (SH) midlatitudes by the two surface data records. In Antarctica large declines in the ozone concentration are noted in the South Pole data, and like those at high latitudes of the NH, seem to parallel the large decreases in the stratosphere.

## Introduction

Ozone in the troposphere is acknowledged to be an important greenhouse gas with a complex chemistry [*WMO*, 1995; *Houghton, et al*, 1996]. The potential for human alteration of its distribution is also generally recognized. There is evidence from observations [*Bojkov*, 1988; *Staehelin et al.*, 1994] and modeling studies [*Levy II et al.*, 1997] that ozone in the troposphere has increased as a result of increases in human produced ozone precursor emissions. Observational evidence for this conclusion is primarily based on several European records where the methodology is traceable to current ozone measurement techniques [*Volz and Kley*, 1988; *Staehelin et al.*, 1994]. Continuous observational records of ozone in the troposphere extend back nearly 30 years for a few ozone vertical profiling sites (ozonesondes) and about 20 years for the longest in situ surface measurements. The relatively short lifetime of ozone in the troposphere (typically a few days to 2 months) means that a relatively dense network of observing sites is required to get a globally representative determination of long-term changes. Such a network does not currently exist, nor is it likely to exist in the near future. For this study we have selected observational records that are from differing geographic regions and that have used consistent measurement techniques over the period we are investigating. This means, for instance, that in Europe we consider profile data only from Hohenpeissenberg in Germany and surface data only from the Zugspitze even though there are several other relatively long records available. In Japan there are three long-term ozonesonde sites. Tsukuba was chosen because it has the most complete (although still somewhat spotty) record of the three. In Canada four ozonesonde stations also have long records. Resolute was selected because of its high latitude location to get a feel for north polar changes. In addition, the tropospheric changes at the Canadian sites have been extensively discussed [*Tarasick et al.*, 1995]. Though the focus here is primarily on records of 15 years or longer, a few shorter records (~10 years) are considered because they are the longest available in data-sparse regions.

## Surface Ozone

Nine longer-term surface ozone records are considered here (Figure 1). Three of these sites (Zugspitze, Germany; Izaña, Canary Islands; and Mauna Loa, Hawaii) are at altitudes over 2000 m and the data considered in this analysis are selected to represent free tropospheric conditions. This is done by using the nighttime (downslope flow) observations. Two other locations (Whiteface Mountain, New York, and South Pole) are at higher altitudes but are not considered to be free tropospheric sites. The other four locations (Barrow, Alaska; American Samoa; Cape Point, South Africa; and Cape Grim, Australia) are remote sites that sample marine air that comes over long ocean fetches. All of the sites are part of measurement networks maintained by the authors' laboratories. The ozone mixing ratios for each site are plotted as annual averages to emphasize the longer-term variations. The numerical trend analyses were performed on both the monthly anomalies and the annual values.

In Figure 1 the results of the linear regression calculation of the monthly anomalies are plotted with the annual averages. A cursory examination of the results in Figure 1 reveals that there are sites with long-term increases (of various magnitudes) and one location with a significant decrease (South Pole). The largest trend is at the alpine station at Zugspitze [*Scheel et al.*, 1995], where an overall linear increase in the ozone mixing ratio of about 1.5% per year has occurred. The bulk of this increase took place in the 1970s and early 1980s. Because of this, a single linear trend fit to the data at Zugspitze does not best describe what is happening. If the data are divided into two equal periods the linear increase during the first half is nearly five times as large as during the latter half (3.06±1.01% per year and 0.63±0.42% per year, respectively). During the earlier period ozone concentrations increased

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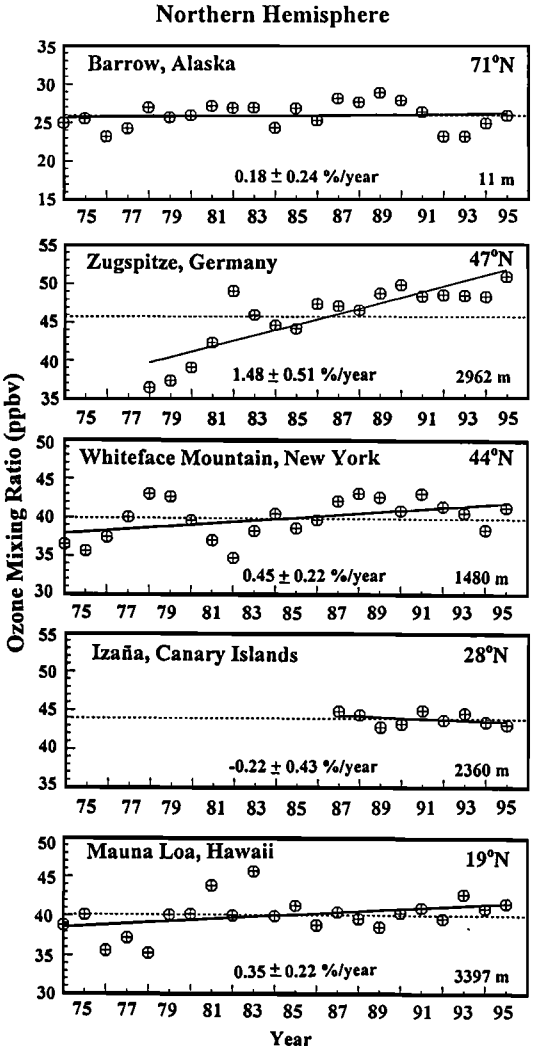


**Figure 1.** Annual average ozone mixing ratios (ppbv) for surface ozone measuring sites. The dashed line is the long term average. The solid line is the linear least squares fit to the monthly anomalies. The linear trend and 95% confidence in percent per year is given with the plot for each location.

significantly in all seasons (largest during summer and spring), while during the latter period increases were significant only in the summer months.

At Whiteface Mountain the annual ozone values (Figure 1) show an increase just under 0.5% per year. This relatively small increase is much reduced during the second half of the record. The Mauna Loa increase, though much smaller than at Zugspitze or even Whiteface Mountain, also seems to have taken place primarily in the 1970s and early 1980s. The small increase at Cape Point may reflect calibration uncertainties early in the record [*Brunke and Scheel*, 1997], however, a small overall increase that is statistically significant is also present at Cape Grim [*Galbally et al.*, 1996]. At the sites with the largest changes (Zugspitze and South Pole) computations of the trend on both the monthly anomalies and annual means yield significant results. At Whiteface Mountain, Mauna Loa, Cape Point, and Cape Grim the trends from annual means are not significant.

## Changes in the Free Troposphere

Although the primary source of information on ozone changes in the free troposphere is ozonesonde measurements, there are a number of shortcomings in this record. At some sites there has been a change of sonde type. The frequency of soundings is also a problem in some cases. A comprehensive analysis of the ozonesonde data base for trend purposes was carried out by *Logan* [1994]. A detailed analysis of the Canadian record was done by *Tarasick et al.* [1995], of the Japanese stations by *Akimoto et al.* [1994], and of Hohenpeissenberg by *Claude and Kohler* [1997]. We have selected three stations which have used consistent instrumentation and are located on three continents of the NH. Each of these sites has operated for 25 years or more. The Brewer-Mast data prior to 1980 at Resolute were not included as a single time series with the ECC data since no adequate procedure seems to be available at present for producing a homogeneous time series in the troposphere [*Logan*, 1994: *Tarasick et al.*, 1995, 1997]. At Tsukuba, Japan, and Wallops Island, Virginia, there are periods of sparse or missing data, while at Hohenpeissenberg, Germany, a very comprehensive data set is available. The data were taken from the World Ozone and Ultraviolet Radiation Data Centre (WOUDC), Toronto, Canada, data base with several corrections applied for known procedural changes. At Hohenpeissenberg and Tsukuba an adjustment was made for the shift in the total ozone scale in 1992. At Tsukuba prior to 1990, there are very few summer observations. Trends were computed separately excluding all summer observations and including them where available. The differences were small. A portion of the Wallops Island data (1970-1985) in the WOUDC archive was normalized to the total column, while the data after 1985 are not. For the purposes of this study the entire data set was renormalized to the total ozone amount reprocessed to the 1992 (Bass-Paur) scale. These three sites are located in regions of the eastern United States., western Europe, and eastern Asia where one might expect the largest anthropogenic influence on tropospheric ozone because of precursor emissions associated with high levels of population and industrialization.

For the three longer ozonesonde records, data are shown (Figure 2) for a lower layer of 850-700 hPa (~1.5-3 km) and an upper tropospheric layer of 500-300 hPa (~6-9 km). For the shorter period since 1980 (chosen to coincide with the beginning of ECC

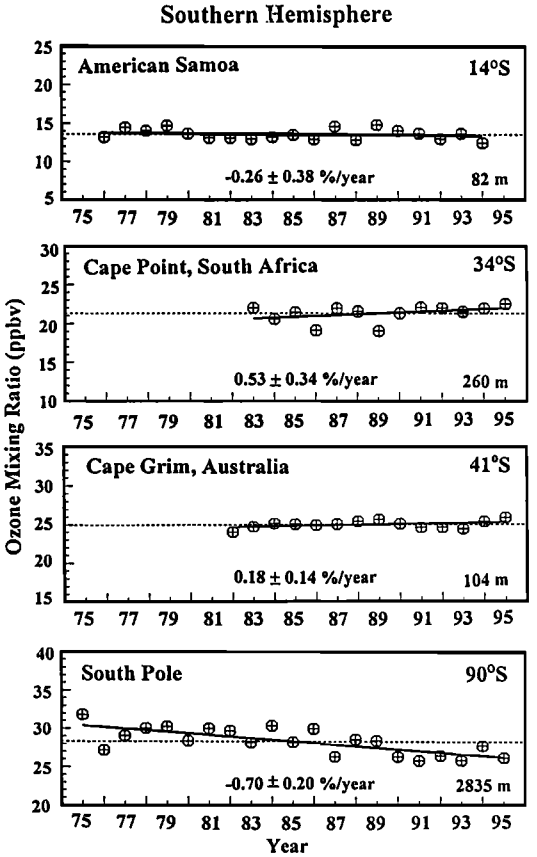


**Figure 1.** (continued)

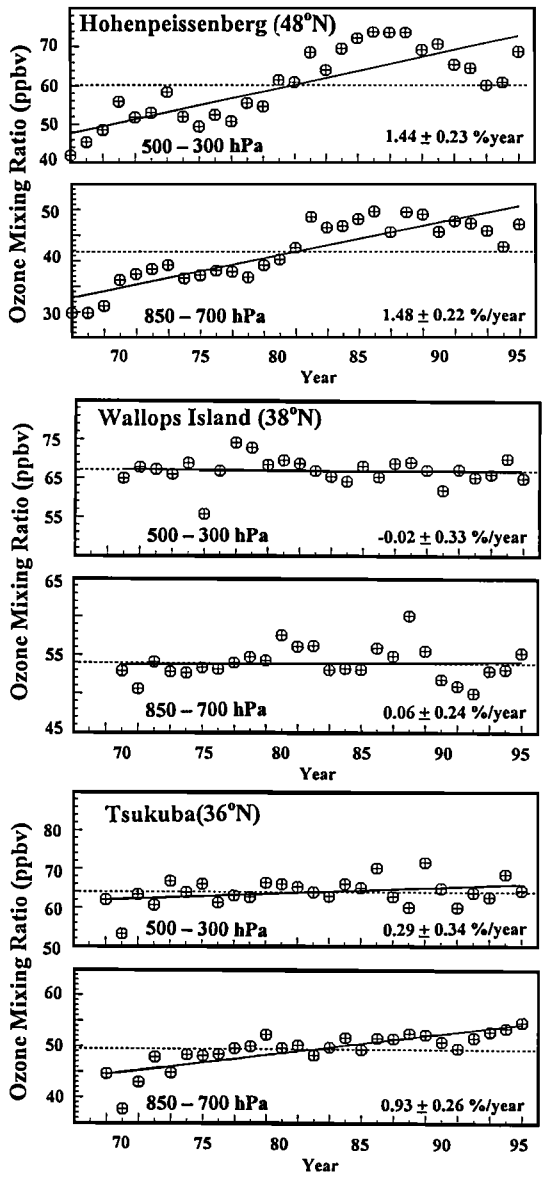


**Figure 2.** Annual average ozone mixing ratios and trends from ozonesonde data for two layers in the troposphere (850-700 hPa and 500-300 hPa) at Hohenpeissenberg, Tsukuba, and Wallops Island.

observations in Canada and Boulder), a midtropospheric layer (700-500 hPa) is depicted in Figure 3 at three additional sites (Resolute, Canada; Boulder, Colorado; and Hilo, Hawaii). At Hohenpeissenberg the 850-700 hPa layer shows several of the characteristics seen at the Zugspitze site as noted by *Logan* [1994]. In particular, the increase in the late 1970s and early 1980s and the smaller increase since, are seen at these nearby locations, which enhances our confidence in the result. The Hohenpeissenberg data also show that ozone was increasing throughout the 1970s and that the increase took place throughout the entire vertical depth of the troposphere. At Tsukuba the lower layer also shows large increases into the early 1980s but a slowing of the increase after that. In the midtroposphere after 1980 there is no significant trend. The upper troposphere shows no significant change over the entire record. Wallops Island shows little long-term change throughout the troposphere. At Boulder the data are quite sparse prior to 1985, but the record seems to be consistent with the other three midlatitude ozonesonde sites in showing relatively small changes since 1980. Hilo, Hawaii, in the subtropical Pacific also shows no significant change over the period of available data beginning in 1983. This is consistent with the Mauna Loa record (Figure 1), which also shows little change after the early 1980s. For the sonde data the trend estimates from annual means are significant at the same locations and levels where significant trends were found in the monthly anomalies except at Boulder.

## High Latitude Changes

At Resolute there is a significant decline in ozone mixing ratio at all levels of the troposphere that is quite similar to that shown in Figure 3 for the midtroposphere. In addition this decline is seen at almost all levels at three other ozonesonde sites in the Canadian network [*Tarasick et al.*, 1995], which verifies that this is not a local phenomenon. These longer-term declines in the Canadian arctic troposphere parallel the change that is seen in the lower stratosphere over the same period of time. The South Pole surface ozone (Figure 1) decline extends back to at least the early 1980s as well. Ozonesonde flights were begun at South Pole in 1986 well after the beginning of the earlier decreases. They do, however, encompass the period of lower amounts in the 1990s. The effect of the reduced lower stratospheric ozone concentrations following the eruption of Mt. Pinatubo, primarily shows up as a several-year decrease in the tropospheric amounts. The much more frequently measured tropospheric ozone from the surface observations (Figure 1) shows a somewhat larger decline during this period (1986-

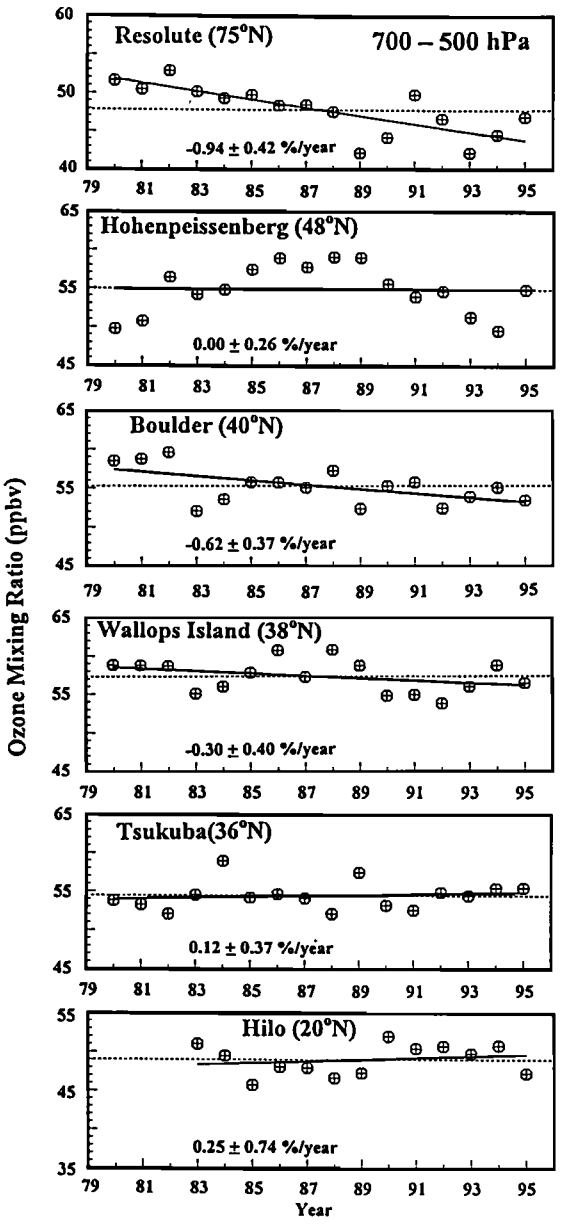


**Figure 3.** Midtropospheric (700-500 hPa) annual average ozone mixing ratios and trends for six ozonesonde sites for the record beginning in the 1980s.

1995), as well as the several-year period of low values following the Mt. Pinatubo eruption, and may better capture the trend over this relatively short period. It seems plausible that at least at these high latitude sites, the longer term decline in the troposphere is linked to the declining ozone in the stratosphere. Every year-to-year change in the stratosphere at an individual station is not, however, reflected in a corresponding change in the troposphere below. The tropospheric changes are not likely a result of processes taking place on such a local scale.

## Discussion

If, as the data from several key regions in the midlatitudes of the NH suggest, there has been a slowing or cessation of the ozone increase in areas where it had increased significantly in previous decades, what does this imply about our understanding of tropospheric ozone changes? In some regions pollution control measures have likely curbed the increase in nitrogen oxide emissions slowing the ozone increase as well [*Logan*, 1994]. In Asia, however, nitrogen oxide emissions should have continued to grow significantly in the recent period. The locations that should be most influenced by Asian emissions (Japan and perhaps Hawaii) show little growth in ozone concentration during the 1980-1995 period compared to the period prior to 1980. At altitudes above 700 hPa over Tsukuba there is no increase (Figure 3), and below this level the increase is less than half (0.44±0.43% per year) of that seen for the 1969-1995 period (Figure 2). At present it is difficult to tell whether the observed trends are at odds with expectations that ozone should be increasing downwind of the rapidly industrializing Asian continent and particularly China. Several years of continuing year-round measurements at the Japanese sonde sites should be quite helpful in assessing this trend.

Recent analysis of data from a global network (P. Novelli, et al., Distributions and recent trends of carbon monoxide in the lower troposphere, *J. Geophys. Res.*, submitted, 1997) shows that CO concentrations have declined on a global basis in recent years. This includes Asian and midlatitude Pacific Ocean locations. It is plausible that lower CO levels, if they are a result of reduced emissions, may be reflected in tropospheric ozone amounts.

In the SH midlatitudes small increases at both Cape Point and Cape Grim suggest a possible influence of increasing levels of African biomass burning [*Fishman et al.*, 1991]. Since the largest seasonal changes at these sites are during the austral winter or spring, such an influence is plausible. At both Cape Point and Cape Grim the two latest years are relatively high, but several more years of data are needed to see if this reflects a stronger signal from increasing biomass burning. In the remote western tropical Pacific little change is indicated from the surface record at Samoa.

## Conclusions

Evidence from both surface and ozonesonde measurements shows that while ozone increases were large in the troposphere over Europe through the early 1980s, that in the period since, increases have been smaller or, in some case, they have nearly ceased. Even with somewhat higher ozone amounts seen in 1996 than in recent years, this slowdown is a distinctive characteristic of the records investigated in this study. At higher midlatitudes of North America, represented by two Canadian stations, tropospheric ozone has declined since 1980. In the eastern United States the surface station at Whiteface Mountain shows an overall increase with almost no change after the mid-1980s, consistent with the pattern seen in other NH midlatitude sites. At Wallops Island little change is noted in the troposphere throughout the 25-year measurement record. In Japan during 1969-1995 ozone also increased in the lower troposphere but at a slower rate than over Europe [*Akimoto et al.*, 1994]. In the free troposphere since 1980, we found no significant change.

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