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Ozone in the boundary layer of the equatorial Atlantic Ocean

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ABSTRACT

Shipboard (~ 7 m) ozone and carbon monoxide measurements made in the equatorial Atlantic Ocean, south of the intertropical convergence zone, between 5° N and 10° S and 10°–30° W in August and September of 1986 exhibited variability in excess of a factor of 2. Ozone mixing ratios ranged from < 25 to over 50 ppbv while carbon monoxide mixing ratios ranged from < 50 to over 120 ppbv along the western edge of the equatorial Atlantic in spite of the fact that the prevailing, surface winds were from the east to southeast at between 5 and 15 m/s. Ozone and carbon monoxide mixing ratios were generally lower and exhibited less variability in the eastern tropical Atlantic where a lighter, south to southeasterly wind regime predominated. Isobaric trajectory analyses indicated that the high ozone and carbon monoxide mixing ratios appear to be related to long-range transport off of the African continent. Time-series analysis of the ozone data indicates a diel cycle in ozone mixing ratios with a morning maximum and afternoon minimum.

1. Introduction

As we continue to develop a better understanding of mid-latitude tropospheric ozone (O_3) distributions, the processes that determine the distribution of O_3 in the tropical boundary layer, especially the marine boundary layer, remain major uncertainties. Liu et al. (1987) have modeled tropospheric ozone in the mid-latitudes of the northern hemisphere with a major conclusion of that work being that the residence time of O_3 may vary by over an order of magnitude between winter and summer with the largest variations occurring where the levels of nitric oxide (NO) plus nitrogen dioxide (NO_2), collectively referred to as NO_x, are lowest. In mid-latitudes of the northern hemisphere during winter they calculate a residence time (τ) of nearly two hundred days. This is the result of a long photochemical lifetime and a relatively low deposition velocity of O_3 to the ocean surface due to the high surface resistance of water to ozone transfer (Galbally and Roy, 1980). It appears that most tropospheric ozone production in the northern hemisphere occurs over land in both winter and summer. With residence times of days to weeks, a large fraction of the O_3 formed within the boundary layer appears to be introduced into the free troposphere and transported. Liu et al. (1987) point out that transport of mid-latitude O_3 to the tropics may be an important sink for O_3 and that, in winter, a good portion of this destruction probably has to occur within the boundary layer rather than within the free troposphere. In an earlier model, Levy et al. (1985) found that they required an additional chemical and/or photochemical loss mechanism in the tropical and subtropical Pacific in summertime and that, in general, the low deposition velocities of O_3 within the marine boundary layer necessi-

tated additional destruction within the marine boundary layer to balance their model with observations.

In addition to questions regarding its rôle in global and hemispheric ozone distributions, the tropical marine boundary layer has its own specific questions. One is what is the source of ozone in the remote, marine tropics where in situ production may not be occurring due to insufficient levels of nitric oxide to support O_3 production (Liu et al., 1983). A second is the occurrence of an intense O_3 minimum below 1.5 km over at least a portion of the equatorial Pacific during the northern hemisphere spring (Piotrowicz et al., 1986; Routhier et al., 1980). Is there a diel cycle in O_3? Stallard et al. (1975) observed a diel cycle in the tropical Atlantic with a 0000 to 0800 (local time) maximum when they had a southerly, oceanic flow. Piotrowicz et al. (1986) and Liu et al. (1983) did not observe a diel cycle in the tropical Pacific but this may be because lower O_3 mixing ratios in the Pacific, when combined with the predicted magnitude of the cycle of $\sim 10\%$ (if $\tau = 10$ days), may result in any day/night differences being similar to the analytical uncertainties. If NO is the key species controlling O_3 production in remote regions (Chameides and Walker, 1973; Crutzen, 1979; Liu et al., 1983) then any peak in O_3 associated with in situ production rather than transport should coincide with the formation of NO, the production of which occurs upon the introduction of light into the atmospheric system (Liu et al., 1983).

In August–September of 1986 we conducted a research cruise in the equatorial Atlantic Ocean to investigate whether the distribution of O_3 in that relatively confined region of the oceans (as opposed to the more open equatorial Pacific) is extensively influenced by the surrounding land masses and to investigate whether there may be a diel cycle in ozone that may be detectable in a region with higher ozone mixing ratios.

2. Experimental

Cruise RP-03-RE-86 of the National Oceanic and Atmospheric Administration (NOAA) Ship *Researcher* took place between 12 August and 27 September 1986 from Bridgetown, Barbados to Dakar, Senegal to Miami, Florida. The portion of the cruise from Bridgetown to Dakar (Fig. 1) was designed to investigate the atmospheric chemistry of the equatorial Atlantic boundary layer, especially as it reflected transport off of the African continent. The cruise proceeded southeast out of Barbados and crossed the region of InterTropical Convergence Zone (ITCZ) between 14 and 15 of August at approximately 8°N, 50°W after which the prevailing winds shifted from northeast to southeast. On the eastern edge of the transect, the ITCZ actually progressed over and through a station being occupied off of the coast of Liberia on 8 September. Hydrographic stations were occupied along the north/south sections at 26°W and 10°W for periods ranging from one to three days. The west to east portions of the cruise were primarily transits.

Throughout the cruise, especially when on station, the bow of the ship was kept into the wind as much as possible. With prevailing southeasterly winds (Table 1) from 4 to 8 m/s, occasionally reaching 11 m/sec along 26°W, sampling could be conducted without interruption due to potential shipboard contamination until 30 August. Sampling during the south to north section along 10°W was strictly controlled to occur only when on station into the wind or underway when the relative wind was within a sector of $\pm 90°$ of the bow with a velocity greater than 1.1 m/s. Because the true wind speed was less than 6.5 m/s for much of the 10°W section, sampling could be conducted virtually unhindered until the ITCZ was crossed on 8 September and northerly winds encountered.

Ozone measurements were made by continuously operating a Dasibi Model 1008-AH ozone analyzer at a sampling frequency of ever 10 s. The instrument was calibrated immediately before and after the cruise. The instrument was mounted just under the bow of the ship and samples collected through a Teflon tube extending out in front of the ship and about 7 m above the sea surface. The experimentally determined (Piotrowicz et al., 1986) instrumental uncertainty for this instrument is ± 1.2 ppbv. Carbon monoxide (CO) measurements were made from duplicate, discrete samples collected into 2.5 l, electropolished, stainless steel flasks on the bow of the ship. These samples were then returned to

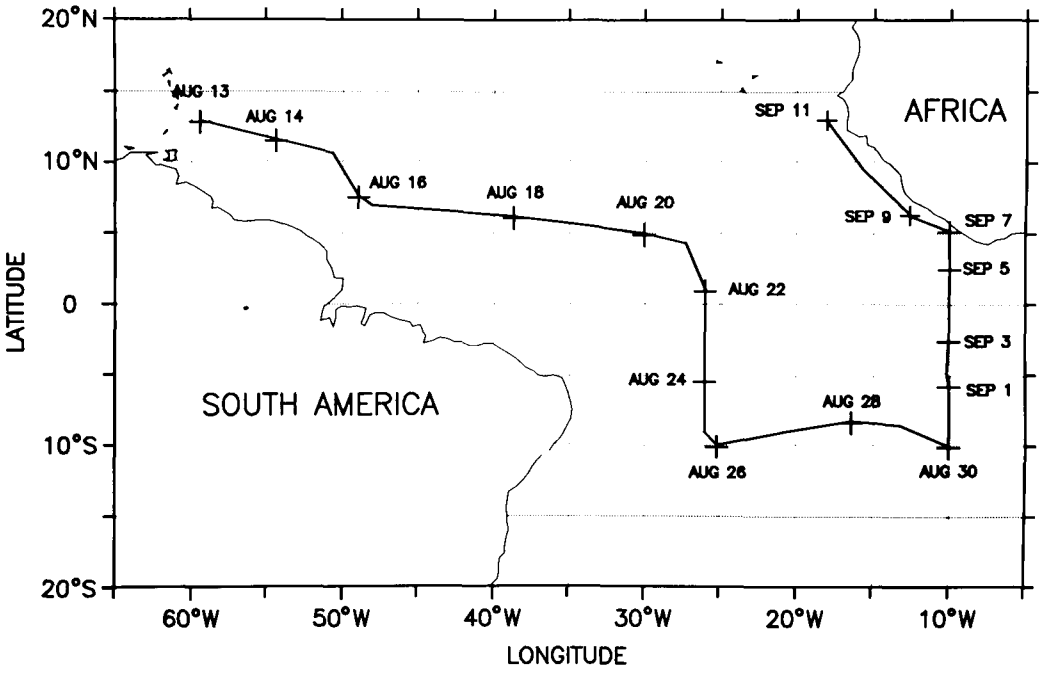


Fig. 1. Cruise track of the NOAA Ship *Researcher*, 13 August–11 September 1986.

the laboratory for analysis at a later date by gas chromatography using both a methanator (Kahlil and Rasmussen, 1984) and a reduction gas detector.

3. Spatial variability in ozone and carbon monoxide

Fig. 2 presents the hourly ozone and discrete carbon monoxide mixing ratios throughout the cruise track. The vertical lines of the time axis indicate 0000 Local Standard Time (LST) of the days indicated. The ozone data represents an average of 10 instrumental cycles while the carbon monoxide represents each discrete sample. In most cases, duplicate samples were collected during each sampling for CO. Ozone mixing ratios vary by over a factor of two throughout the cruise from a low of around 20 ppbv on 14 August and intermediate lows of around 25 ppbv on 16–18 August and 29 August to highs of slightly over 50 ppbv on 25 August and 40 ppbv on 2 September. In the course of the cruise track, very abrupt changes in O_3 Mixing Ratios (O_3 MRs), e.g., $\geqslant 30\%$, are occasionally observed. These large, rapid changes were observed to always occur in precipitation events with O_3 MRs being reduced while an event is occurring and rapidly returning to pre-event levels upon emerging into a non-precipitating region. Except for 6 September, the carbon monoxide mixing ratios exhibit similar trends to the ozone mixing ratios with relatively high CO mixing ratios occurring where O_3 is rising, or has risen, and lower CO mixing ratios where lower O_3 MRs are occurring.

There are two distinct increases in the O_3 data. The first occurs between 18 and 26 August and a smaller rise between 29 August and 5 September. During the cruise it was observed that the high O_3 between 20 and 26 August was associated with the presence of black aerosols being collected on the fifth stage and final filter of a high volume cascade impactor (S. R. Piotrowicz, unpublished data). These two periods of higher O_3 MRs also have elevated levels of CO as compared with the period from 26–30 August. Because this time of the year is associated with biomass burning in the tropics, e.g., Amazonia, the rise in O_3 and CO could be associated with

Table 1. *True wind speed and direction at 0000 LST*

Date	Direction (° True)	Speed (m/s)
13 August	053	5.1
14 August	055	5.1
15 August	066	4.6
16 August	100	5.1
17 August	068	2.6
18 August	150	8.7
19 August	140	8.7
20 August	150	7.2
21 August	155	6.7
22 August	190	4.6
23 August	120	5.1
24 August	100	7.2
25 August	113	9.8
26 August	113	10.3
27 August	125	8.2
28 August	120	9.3
29 August	126	9.3
30 August	120	9.3
31 August	095	8.7
1 September	117	7.2
2 September	125	7.2
3 September	165	4.1
4 September	105	1.5
5 September	205	5.1
6 September	215	7.7
7 September	210	7.2
8 September	230	6.2
9 September	260	6.2
10 September	225	7.7
11 September	200	2.1

long-range transport. Isobaric back trajectories were obtained for the entire cruise track from the NOAA Geophysical Monitoring for Climatic Change Program (Harris, 1982) to investigate the potential sources of the air being sampled during this period.

Fig. 3 are the ten-day, back trajectories at 700 and 850 mbars representative of the major events and inflection points in the O_3 and CO mixing ratios in Fig. 2. Even though large uncertainties can occur beyond three to four days in such trajectories, especially in remote regions, they remain useful in investigating general flow patterns. The trajectories for 16 August represent the flow in the vicinity of the ITCZ with the free trophspheric flow out of the north and, in terms of transport, possibly containing components derived from the North Atlantic trade winds while the flow at the top of the boundary layer is from the south Atlantic. The flow on 24 August represents the typical flow encountered during this high O_3 and CO event. It appears that this event is very likely associated with long-range transport off of tropical Africa. During this time of the year, extensive biomass burning is occurring in tropical Africa and O_3 associated with that biomass burning has been traced some 800 km off the coast following the prevailing winds at this latitude Cros et al. (1988).

The 30 August trajectories can only be traced back five days, however, the flow here is from the

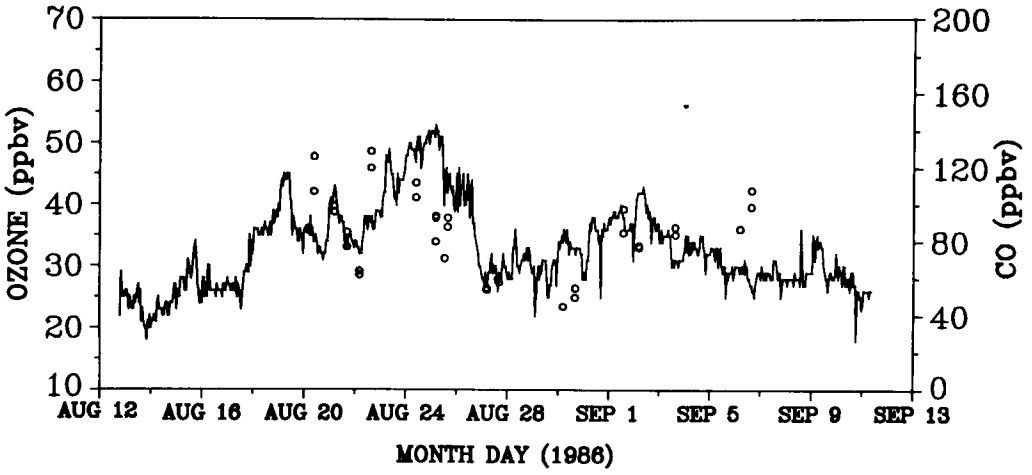
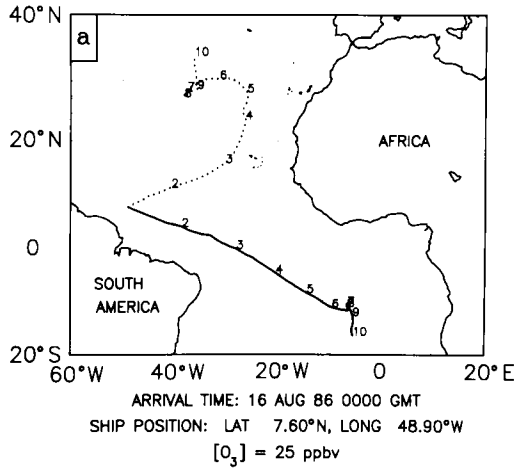
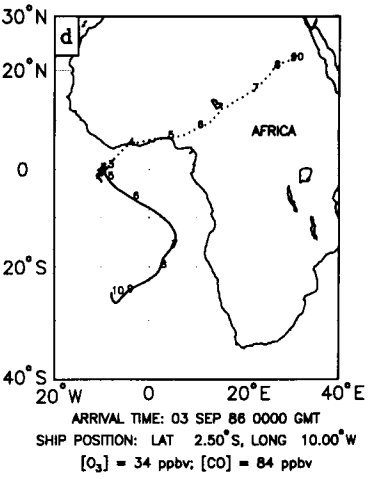
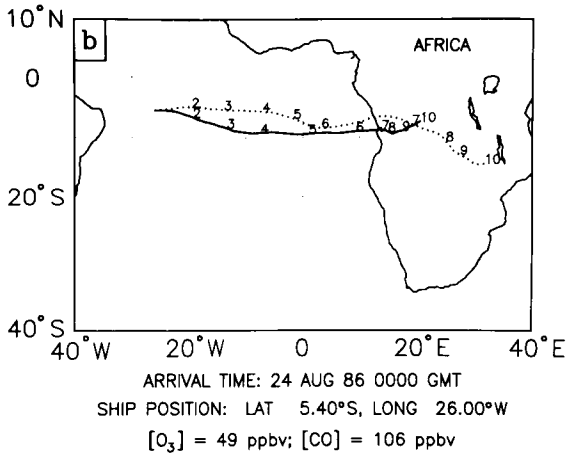
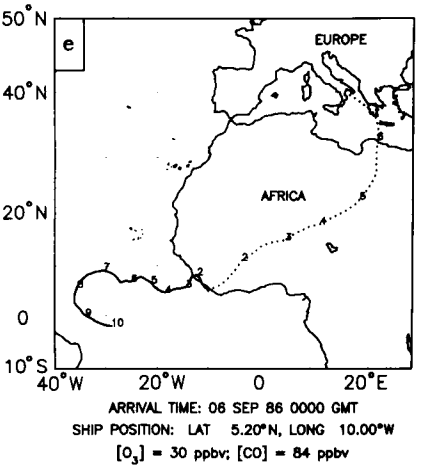


Fig. 2. Shipboard ($Z \simeq 7$ m) ozone (——) and carbon monoxide (o) mixing ratios from 12 August to 11 September 1986 in the equatorial and south Atlantic Oceans. Ozone is hourly; carbon monoxide are flask samples.









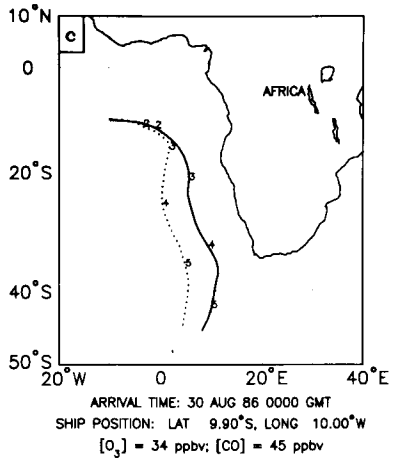


Fig. 3. Isobaric back trajectories for representative flow patterns along cruise track. —— = 850 mbars; ····· = 700 mbars.

south Atlantic where, even given uncertainties in such an analysis, the air has probably not been in recent contact with any land mass and a corresponding low is observed in both O_3 and CO mixing ratios. There is data on O_3 and CO in the boundary layer of mid-latitude, oceanic regions in the southern hemisphere over the Pacific ocean. The O_3 mixing ratios of around 30 ppbv are typical of mid-latitude, Pacific air (Routhier et al., 1980). The CO mixing ratios to 40–50 ppbv that are associated with south Atlantic flow are very similar to mid-latitude, southern hemisphere Pacific regions (Heidt et al., 1980; Fraser et al., 1986). This period is the austral winter. If the residence time of O_3 is considerably longer at mid-latitudes in the winter than in the summer in regions of low NO_x (Liu et al., 1987), then both CO and O_3 are likely to be well mixed in mid-latitudes in the winter. It is not unreasonable, therefore, that the south Atlantic and south Pacific exhibit similar levels of these species at this time of the year.

A mixed flow with free tropospheric air coming from the African continent while the surface flow remains oceanic is encountered as the cruise approaches the African coast on 3 September. The data from 6 September indicates that this is the only period when the CO and O_3 trends do not covary. The sampling location at this time is some 30 km off the coast of Liberia, south of the ITCZ, and although the O_3 mixing ratios are representative of marine air, the CO levels appear to have a continental influence. The trajectories indicate a terrestrial influence at 700 and 850 mbars. Even though there appears to be transport of terrestrial CO, it does not appear that the O_3 is being formed over the continent and transported to this site which is in contrast with the 26°W section where transport of terrestrially produced O_3 appears to be occurring.

4. Temporal variability in ozone

Even though the major variability in ozone in this region is associated with whether or not transport from terrestrial environments is occurring, the variabiliry in Fig. 2 appears to have some high frequency components. A harmonic analysis (Panofsky and Brier, 1963) was performed on the hourly O_3 time series in Fig. 2 for the data collected south of the ITCZ, 0800 (LST) on 15 August to 1900 (LST) on 8 September. The width of the observational period contained therein is 590 h and contains 586 measurements of O_3 mixing ratios with winds from the proper direction relative to the bow of the ship. The periodogram analysis indicated that even with the high O_3 transport events dominating the O_3 variability ($\sim 86\%$ of the system variability) there was a small, but significant, contribution to the variability due to some higher frequency processes. These occurred at intervals of 24.4 and 42 h with each contributing approximately 1.5% to the total variability. Other harmonics contributed $<0.5\%$, and generally $<0.1\%$ each to the total variation. To evaluate the magnitude and shape of the $\sim$24-h harmonic, an average O_3 mixing ratio deviation was computed for each hour. This is the average of the hourly deviations (LST) from the daily means throughout the cruise. Computing the daily mean and the deviations of each hour within that day minimizes the low frequency contributions to O_3 variability and assumed that the distribution around the daily mean within any particular day is normal such that an average with an error estimate can be calculated. The average and standard errors of the hourly O_3 deviations from the daily mean for the cruise track south of the ITCZ are presented in Fig. 4.

The 24.4-h harmonic appears to represent a diel cycle in O_3 mixing ratios. The maximum positive deviation occurs in the early morning with a sharp decrease in the late morning to an afternoon minimum (predicted in Chameides and Stedman, 1977) which remains somewhat stable throughout the night. During the cruise, sunrise generally occurred between 0530 and 0600 LST with first light some 30 to 45 min prior to that. The peaks in Fig. 4 cannot be considered to be exact hourly positions of O_3 maxima and minima because no attempt has been made to take cloudiness, declination and relative position within the time zone into account in deriving Fig. 4. The general trends of a morning maximum and afternoon minimum, we believe, can be considered representative of a diel cycle of O_3. Stallard et al. (1975) in a cruise in the equatorial and south Atlantic during the Austral summer, and much closer to the coastline, observed a similar pattern with an afternoon minimum ex-

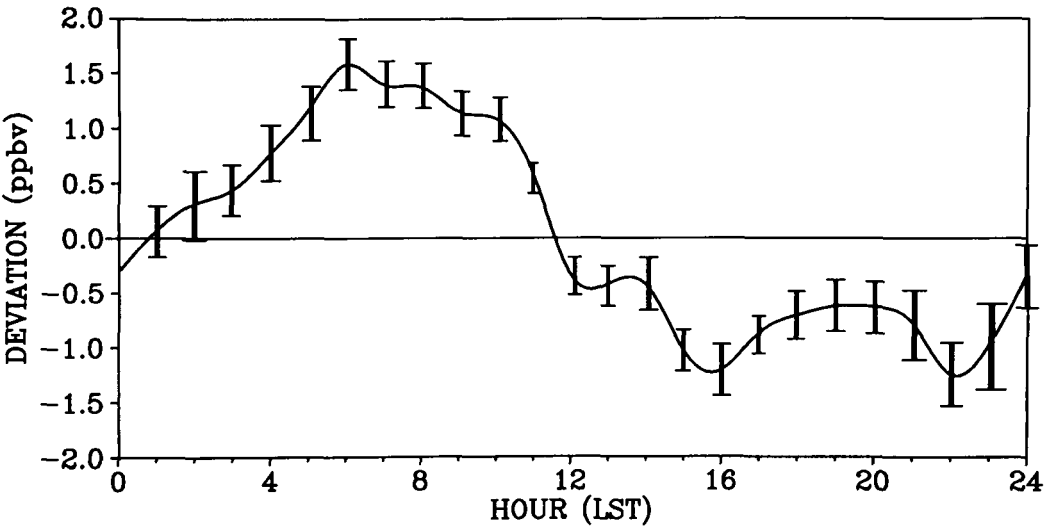


Fig. 4. Deviation of the hourly mean from the daily mean (34.5 pppbv) ozone mixing ratio, south of the intertropical convergence zone, 0800 (LST) on 15 August–1900 (LST) on 8 September 1986.

tending into the evening (1600–2000) and a mixed set of observations of increases and decreases between 2000 and 2400. They found a 0400–0800 (LST) maximum but their highest frequency of observations of maxima occurred from 0000 to 0400. The data in Fig. 4 is not inconsistent with Stallard et al. (1975) given that the deviations are positive between 0000 and 0400 and tending towards zero in the 2000–2400 period. Stallard et al.'s (1975) higher frequency of O_3 maxima occurring between 0000 and 0400 (LST) is the major difference between the two data sets. Stallard et al. (1975) present a very comprehensive data set and an examination of their data suggests that the reason may be location. Their 0000 to 0400 maxima appear to occur most frequently at higher O_3 mixing ratios which coincide with the northern portion of their cruise track, portions of which were north of the ITCZ at that time, and other segments running 50–100 km off of the west African coast as they paralleled the coast on a track from Dakar to Walvis Bay. This is in the range of a land breeze effect, thus, terrestrially produced ozone may account for some of their 0000–0400 occurrences of O_3 maxima. The data of Stallard et al. (1975) also show a very sharp decrease in O_3 mixing ratios around the period of the solar zenith as occurs in Fig. 4.

Realizing that a great deal of the variability in O_3 and CO mixing ratios during this cruise is due to episodic events, a periodogram and hourly deviations of O_3 from the daily mean were computed for the portion of the cruise from 27 August to 8 September. This period primarily represents marine air. There is a small period around 3 September where the upper level flow might have a terrestrial component but the lower level flows in this period are marine. The purpose was to confirm whether the 24- and 42-h harmonics remained, whether the diel variability in the 24 h harmonic remained and whether the 24-h cycle was similar in shape. This period encompassed 306 h, a relatively short time series, and any confidence in conclusions based on it have considerably greater uncertainty than conclusions based on the 590-h time series. The only difference was that no harmonic was observed at ~42 h containing more variation than would be expected based on its multiple of the principal harmonic. Therefore, it appears that harmonic at ~42 h suggested in the 590-h time series is related to the major transport events and may not be related to in situ processes?

Oltmans (1981) has observed a similar diel variation in O_3 at a remote site within the marine boundary layer, the Samoa Geophysical Monitoring for Climatic Change (GMCC) site. An evaluation of the data from Samoa through 1986 (K. J. Hanson, unpublished data) continues to exhibit this pattern. A satisfactory explanation for this pattern of a morning maximum and afternoon minimum in O_3 MRs in the marine boundary layer is not readily available. The afternoon minimum is most likely the result of photochemical destruction of O_3 (Chameides and Stedman, 1977; Liu et al., 1983).

Ozone production requires the presence of sunlight. The analysis in Fig. 4, the data of Stallard et al. (1975) and the data of Oltmans (1981) all exhibit maxima in surface O_3 before sunrise and/or before photochemical production would play a significant rôle in determining the O_3 distribution near the surface. Dynamic processes may be able to explain this pattern for island sites, including Samoa, but that becomes tenuous for pure, marine sites like a ship. A positive nighttime deviation from the daily mean does not imply O_3 production but rather a redistribution of O_3 within the mixed layer such as might occur due to subsidence during the night. An early morning increase, however, is more difficult to explain. The period around sunrise is generally when the minimum in the diel variation in eddy diffusivity occurs (Geiger, 1965a). Mixing will occur as the boundary layer warms due to convection resulting in a maximum in eddy diffusivity in the late morning (Geiger, 1965a). Dynamic processes, therefore, do not appear to be able to account for an early morning maximum in the diel variability of O_3. In the free atmosphere immediately above the marine boundary layer, O_3 variability exhibits a relationship with water, both liquid (Lenschow et al., 1988) and vapor (Piotrowicz et al., 1986) phases. The period around sunrise and shortly thereafter is when rapid changes in temperature near the sea surface (Geiger, 1965b; Roll, 1965) and the relative moisture content of the atmosphere in tropical and subtropical regions (Roll, 1965) occur. Changes in the temperature/moisture relationships of the marine boundary layer are much smaller than the changes observed above the boundary layer by Lenschow et al. (1988) and Piotrowicz et al. (1986) but perhaps the same processes operating above the boundary layer are operating within the boundary layer. A satisfactory explanation for the morning maximum in a diel cycle of O_3 in tropical marine regions observed here and in other, published data is not attainable from this data. Further work on O_3 in the tropics will include more detailed investigations of the processes determining the distribution of vapor and liquid water within the tropical marine boundary layer and any relationship they may have with the distribution of O_3 in this region.

5. Conclusions

The variability in the mean O_3 mixing ratio in the boundary layer of the equatorial Atlantic Ocean during the late, northern hemisphere summer appears to be dominated by the extent of long-range transport to the region from the surrounding land masses. Once out of the immediate source region, photochemical, and possibly atmospheric dynamic processes, involving O_3 and ozone precursors determine the short term variability in O_3. There is a diel cycle in O_3 mixing ratios in the equatorial Atlantic, south of the ITCZ, with a maximum in the morning, a rapid decrease around local apparent noon to a late afternoon and nighttime minimum. The afternoon minimum can be explained by photochemical processes, however, an explanation for the morning maximum observed here and in other tropical marine regions is not attainable at this time.

6. Acknowledgments

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