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Tropical Tropospheric Ozone (TTO) Maps

from Nimbus 7 and Earth-Probe TOMS

by the Modified-Residual Method.

1. Validation, Evaluation and Trends based on

Atlantic Regional Time Series

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ABSTRACT.

The well-known wave-one pattern seen in tropical total ozone [Shiotani, 1992; Ziemke et al., 1996, 1998] has been used to develop a modified-residual (MR) method for retrieving time-averaged stratospheric ozone and tropospheric ozone column amount from TOMS (Total Ozone Mapping Spectrometer) over the 14 complete calendar years of Nimbus 7 observations (1979-1992) and from TOMS on the Earth-Probe (1996-present) and ADEOS platforms (1996-1997). Nine- to sixteen-day averaged tropical tropospheric ozone (TTO) maps, validated with ozonesondes, show a seasonality expected from dynamical and chemical influences. The maps may be viewed on a homepage: <http://metosrv2.umd.edu/~tropo>. Stratospheric column ozone, which is also derived by the modified-residual method, compares well with sondes (to within 6-7 DU) and with stratospheric ozone column derived from other satellites (within 8-10 DU). Validation of the TTO time-series is presently limited to ozonesonde comparisons with Atlantic stations and sites on the adjacent continents (Ascension Island, Natal, Brazil; Brazzaville); for the sounding periods, TTO at all locations agrees with the sonde record to ± 7 DU. TTO time-series and the magnitude of the wave-one pattern show ENSO signals in the strongest El Niño periods from 1979-1998. From 12°N and 12°S, zonally averaged tropospheric ozone shows no significant trend from 1980-1990. Trends are also not significant during this period in localized regions, e.g. from just west of South America across to southern Africa. This is consistent with the ozonesonde record at Natal, Brazil (the only tropical ozone data publicly available for the 1980's), which shows a not statistically significant increase. The lack of trend in tropospheric ozone agrees with a statistical analysis based on another method for deriving TTO from TOMS, the so-called Convective-Cloud-Differential approach of Ziemke et al. [1998].

1. Introduction

The original concept of deriving tropospheric column ozone in the tropics by a residual method consists of using total column ozone from TOMS (the Total Ozone Mapping Spectrometer) and subtracting stratospheric column ozone from an independent sensor. The key assumption is that stratospheric ozone is constant with longitude so that variations in total ozone detected by TOMS are due only to variability in tropospheric column ozone.

There are three general limitations of this type of residual approach. First, there may be a mismatch in orbital and sampling characteristics between TOMS and the other sensor. This occurs with SAGE (Stratospheric Aerosol and Gases Experiment), which typically samples the 10N-10S latitude band 50-60 days per year, sometimes missing the tropics for several months in a row. Thus, the original tropospheric ozone residual technique of Fishman and coworkers [Fishman et al., 1990; 1991; update in Fishman and Brackett, 1997], which used SAGE, reported climatology based on 2-3 month-averaged tropospheric column ozone. Second, TOMS total

ozone does not detect near-surface ozone over surfaces with low albedo with 100% efficiency [Hudson et al., 1995; Kim et al., 1996; Hudson et al., this issue, 1998]. Third, stratospheric column ozone derived from the independent sensor may have considerable uncertainty in the lower stratosphere. This is a particular concern for SBUV (Solar-Back-Scatter Ultraviolet), the instrument on-board the NOAA operational satellites [Ziemke and Chandra, 1998; Hudson et al., 1998]. Lower stratospheric uncertainties also affect stratospheric ozone from the UARS (Upper Atmosphere Research Satellite) sensors, MLS (Microwave Limb Sounder) and HALOE (Halogen Occultation Experiment), which have been differenced with TOMS in a residual approach [Ziemke et al., 1998].

These factors have motivated the development of two methods that use TOMS total ozone with physical parameters other than a separate satellite measurement to distinguish stratospheric ozone from tropospheric ozone. The two methods, the convective-cloud-differential (CCD) method [Ziemke et al., 1998] and the modified-residual approach [Hudson and Thompson, 1998], are described in detail in the 20 Sept 1998 Journal of Geophysical Research.

In the CCD method, stratospheric ozone is set equal to the TOMS total ozone measurement over highly reflecting, high altitude clouds in the western Pacific Ocean. At cloud-free pixels, signified by reflectivity < 0.2 , tropospheric column ozone is obtained by subtracting the above-cloud stratospheric ozone amount from TOMS total ozone. This is referred to as the CCD tropospheric ozone column. Because there is no cloud height information in TOMS, a primary assumption in the CCD method is that the high reflectivity clouds have cloud-top at the tropopause. Monthly averaged maps between 20N and 20S are shown in Ziemke et al. [1998]; no correction is made for reduced detection efficiency near the surface.

In the modified-residual (MR) method, the well-known wave-one pattern in total ozone [Shiotani, 1992] is used to normalize total ozone over the entire tropical band to a single value in the region of the ozone minimum, near 180 longitude. Tropospheric column ozone taken from ozonesondes over and near the Atlantic maximum is subtracted from TOMS total ozone to give the stratospheric column ozone value [Figure 1]. Stratospheric column ozone subtracted from total ozone yields tropospheric ozone at all other longitudes; an efficiency correction is applied because only cloud-free (low reflectivity) TOMS data are used. Fourier analysis of the wave-one pattern sets the latitudinal limits of the tropical air mass within which the modified-residual method is assumed to be valid (Figure 4 in [Hudson and Thompson, 1998]).

In the modified-residual method regular ozonesondes in the vicinity of the ozone maximum were only available for 1991-1992, during the pre-TRACE-A (Transport and Atmospheric Chemistry near the Equator - Atlanctic) and SAFARI (Southern African Fire Atmospheric Regional Initiative)/TRACE-A intensive. A seasonal regularity observed in the ozonesonde data set (Figure 5 in Hudson and Thompson, 1998) suggests that the method can be applied in other years. Thus, in early 1997, we initiated use of

the MR method to process real-time Earth Probe (EP) and ADEOS (Advanced Earth Observing System) data. Daily, 3-day- and 9-day-averaged images appear on a homepage:
[http://metosrv2.umd.edu/~tropo/\(Real-time\)](http://metosrv2.umd.edu/~tropo/(Real-time)).

In early 1998 the modified-residual method was extended back in time to cover the remainder of the Nimbus 7 (=N7)/TOMS record (Section 2, below). For this purpose, TOMS total ozone was averaged over two 13-16-day periods per month. These images, referred to as TTO (tropical tropospheric ozone) maps, also appear on the homepage.

Validation and evaluation of the TTO maps is presently underway. This paper presents some of the first findings in this effort. Validation of the Nimbus 7 and EP/TOMS TTO products focuses on the Atlantic and adjacent continents (Section 3), where tropical sonde data are available from public archives: Natal (Brazil, 1979-1992), Ascension Island (1990-1992, 1997-1998), Brazzaville (1990-1992). Derived stratospheric ozone and TTO in the Natal region from the MR method are compared to the same parameters taken from the CCD method [Ziemke et al., 1998; Chandra et al., 1998a].

Evaluation through analysis with Atlantic and near-Atlantic regional time-series uses the N7 record (1979-1992) because the EP/TTO is only two years and is dominated by the unusually intense 1997-1998 El Niño-Southern Oscillation (ENSO) [Chandra et al., 1998a]. The following questions are addressed in Section 4:

- (1) What do seasonal cycles and interannual variability in TTO look like and how do they compare with those observed in ozonesondes?
- (2) Do TTO and derived stratospheric ozone, O_3^{str} , show signals typical of ENSO influences?
- (3) Can trends in tropospheric ozone over eastern South America and the eastern Pacific be detected for the 1980's, as reported using indirect approaches with TOMS ozone [Jiang and Yung, 1996; Kim and Newchurch, 1996]?

Preliminary answers to these questions demonstrate that the MR method gives TTO maps of sufficient accuracy for tropical climatological studies and time-series analyses. Furthermore, with 1x2 degree resolution and 2-3 maps/month, TTO maps by the modified-residual method are more highly resolved spatially and temporally than tropical tropospheric ozone obtained by other techniques.

2. Modified-Residual Method

A. Technique and Application to Observations Before and After 1991-1992

Figure 1 illustrates the major features of the modified-residual method. **Figure 2** in Hudson and Thompson [1998] describes the steps in deriving stratospheric column ozone, excess ozone, background tropospheric ozone and tropospheric column ozone (TTO). The fundamentals of the method are given here, for easy reference, and to clarify where the most critical assumptions are made:

(1) Normalization of total ozone (TOMS version 7, Level 2 data, cloud-free as defined by reflectivity < 0.15)) at or near 180° longitude, where ozone is a minimum. This permits the assumption of a single stratospheric ozone value, $O_3)_{str}$, over the latitude range in which the wave-one pattern is observed in total ozone.

(2) An underlying total ozone amount (the thick smooth curve in Figure 1) follows a wave-one pattern; this is referred to as "background total ozone," $O_3)_{total}$ in Hudson and Thompson [1998] and Kim et al. [1996].

(3) The wave-one feature also delineates a background tropospheric ozone column amount, $O_3)_{tr}^{back}$, in Figure 1. "Excess ozone," $O_3)_{excess}$, which is the amount of ozone normally ascribed to pollution [Fishman et al., 1991; Thompson et al., 1996a] is defined as follows:

$$O_3)_{excess} = O_3)_{total} - O_3)_{tr}^{back} \\ = O_3)_{tr} - O_3)_{tr}^{back},$$

where $O_3)_{total}$ is total ozone from TOMS Level 2 data.

(4) Total ozone at the minimum (at or near 180 degrees) is used to derive stratospheric ozone from:

$$O_3)_{str} = O_3)_{total}(180) - O_3)_{tr}^{back}(180),$$

where the latter parameter is designated "A" in Figure 1.

(5) The magnitude of the wave-one is obtained by Fourier analysis, with the wave optimized to fit along total ozone minima, as shown in Figure 1. The assumption that the wave lies in the troposphere is signified by flat stratospheric ozone in Figure 1.

Features (4) and (5) represent a key assumption in the modified-residual method, namely that the wave-one pattern is in the troposphere. This appears to be supported by analysis of sondes and stratospheric ozone derived from SAGE [Fishman et al., 1990; Shiotani and Hasebe, 1994], MLS and HALOE [Ziemke et al., 1996], although neither the sampling frequency of the sondes nor the accuracy of lower tropospheric profiling is definitive.

In Hudson and Thompson [1998], $O_3)_{tr}^{back}(180)$, which is used to obtain stratospheric ozone in (4), was derived from the wave pattern and tropospheric ozone data from sounding stations near the ozone maximum. The reason is, that for the period used in developing the MR method, just prior to and during the SAFARI/TRACE-A campaigns of 1992, no ozonesondes were launched near the tropical Pacific ozone minimum. However, at Natal, Ascension, and Brazzaville, there were more than 130 soundings during 1991-1992 [Diab et al., 1996; Nganga et al., 1996; Olson et al., 1996; Thompson et al., 1996a]. Values for each two-week period for $O_3)_{tr}^{back}(N, A, B)$, where N, A, B represent the locations at Natal (6S, 35W), Ascension (8S, 15W) and Brazzaville (4S, 15E), were obtained through the constraint that excess ozone plus the background tropospheric ozone at each location must equal integrated tropospheric ozone from the ozone sondes:

$$O_3)_{excess}(N, A, B) + O_3)_{tr}^{back}(N, A, B) = O_3)_{sonde}(N, A, B)$$

Averaging of $O_3)_{tr}^{back}(N, A, B)$ over the three sites was used to obtain $O_3)_{tr}^{back}(0)$ and $O_3)_{tr}^{back}(180)$ from the longitudinal

dependence of the wave. The relationship in (4) gave stratospheric ozone, $O_3)_{str}$, by subtraction from total ozone. When averaging over the three sounding sites was carried out, a seasonally varying signal for $O_3)_{tr}^{back}(0)$ could be fit with a sinusoidal function: $O_3)_{tr}^{back}(0, t)$, where "t" refers to time (Figure 5 in Hudson and Thompson, 1998). Our best estimate of $O_3)_{tr}$ at each latitude/longitude point in the 1991-1992 period, referred to "TTO", was obtained using this function and the above procedure.

Preliminary extension of the 1991-1992 background tropospheric ozone signal at the wave maximum, $O_3)_{tr}^{back}(0, t)$, to TOMS total O_3 data from 1990 gave TTO in excellent agreement with tropospheric ozone measured by balloon-borne sondes at Ascension, Natal, and Brazzaville (Figure 2). Hence, it was decided to use the MR method with the 1991-1992 $O_3)_{tr}^{back}(0, t)$ function during the EP and ADEOS periods (1996-present) and backward through the Nimbus 7 era to give two time-series of TTO.

For each 13-15-day period over which version 7 total ozone from Nimbus 7 is averaged, the procedure used is identical to that described in Hudson and Thompson [1998]. A set of normalization constants for total ozone prescribes the location of the ozone minimum and permits derivation of a single $O_3)_{str}$, subject to the magnitude and geographical boundaries of the wave-one pattern. The first set of N7/TTO maps was posted as gif images on the <http://metosrv2.umd.edu/~tropo> homepage in April 1998. A sample map, with two panels per month appears in Plate 1A. For real-time processing, the averaging procedure has been modified to compute running averages of total ozone. Daily, 3-day and 9-day averaged TTO maps appear on the "real-time" section of the homepage (Plate 1B).

B. Uncertainties in TTO from the Modified-Residual Method

Hudson and Thompson [1998] and Kim et al. [1996] presented a thorough error analysis of TTO, ascribing imprecisions to assumptions made in adopting a seasonally varying background tropospheric ozone column near the ozone maximum: $1\sigma = 4.8$ DU (Table 1 in Hudson and Thompson, 1998). This is nearly identical to the $1-\sigma$ deviation (5.3 DU) between individual ozonesondes and the 2-week averages used in referencing $O_3)_{tr}^{back}(0)$ to the sondes (Table 4 in Hudson and Thompson, 1998). Thus, to 2σ , an uncertainty of 10 DU applies. This may be as small as 15% of tropospheric column during the seasonal Atlantic tropospheric O_3 maximum or localized pollution events in other regions. It is a larger fraction near the Pacific tropospheric ozone minimum, which averages 20 DU over the course of a year.

3. Validation of Derived Stratospheric Ozone and Modified-Residual TTO (1979-1992; 1997-1998)

A. Stratospheric Ozone

Equatorial stratospheric column ozone, $O_3)_{str}$, derived from the MR method agrees, to 8 DU, with pre-Mt Pinatubo-eruption SAGE II measurements recorded between 10N and 10S in 1985-1991 (Figure

3). A similar level of agreement is found for derived stratospheric ozone and ozone integrated from 1-100 hPa from the UARS/MLS launched in September 1991. Discrepancies between the N7 derived stratospheric ozone from September 1991-December 1992 compared to MLS stratospheric O_3 are similar to those from the EP period shown in Figure 4. Deviations of 8-11 DU between $O_3)_{str}$ and observations from the other satellites are within the precision of the MR method and SAGE and MLS column ozone; for the latter two sensors, the profiles are highly uncertain below 20 km. The fact that $O_3)_{str}$ is lower than the SAGE and MLS stratospheric column values may reflect systematic differences in effective tropopause height. Integrated tropospheric ozone from the sondes at Ascension, Brazzaville and Natal includes balloon data from the surface to 100-120 hPa. This is where the ozone gradient changes sharply (see, for example, Figure 5 in Thompson et al., 1996a) and it also corresponds to the tropopause typically reported in NCEP analyses.

If stratospheric ozone is set equal to TOMS total ozone less the integrated tropospheric ozone column from sondes, a comparison can be made between derived stratospheric ozone, $O_3)_{str}$, and observations at Natal (1979-1992; Figure 5) and Ascension (Figure 6, 1997-1998). Agreement of stratospheric ozone with the sondes is better (mean deviation = 6-7 DU) than with SAGE and MLS; deviations are more evenly distributed between positive and negative values.

The CCD method also derives stratospheric column ozone appropriate for the tropics. A comparison of $O_3)_{str}$ for the equator, based on the MR and CCD methods, appears in Figure 7. Agreement between the two techniques is within 4 DU, which is remarkably good, considering how different are the assumptions used in deriving the stratospheric ozone column. A signature of the QBO (Quasi-Biennial Oscillation) appears in both records.

The difference between stratospheric ozone derived from the MR and CCD methods (diamonds for MR stratospheric ozone - CCD stratospheric ozone in Figure 7) appears to have a seasonality: positive in December-February, negative in the middle of the year. Both techniques have assumptions which could contribute to the pattern. The high clouds used in the CCD method are assumed to be always at the tropopause. More variable cloud heights, subject to changes in large-scale circulation or migration of the ITCZ (Intertropical Convergence Zone) could be signified by the variation with respect to the MR value for $O_3)_{str}$. The MR method in turn, is based on a seasonal variation in the $O_3)_{tr}^{back}$ parameter near the Atlantic ozone maximum that usually transmits to a seasonal variation in $O_3)_{tr}^{back}$ near 180° longitude ("A" in Figure 1), where the stratospheric ozone column is derived. The parameter $O_3)_{tr}^{back}(180)$ maximizes near the middle of the year (Figure 5c in Hudson and Thompson, 1998), which would give lower stratospheric column ozone from a given total ozone value. Unfortunately there is no long-term tropical Pacific ozonesonde record with which to interpret the seasonal differences in MR-derived and CCD-derived stratospheric ozone. The Samoan tropospheric ozone record is ambiguous about ozone seasonality. [Komhyr et al., 1989; Oltmans et al., 1998]. However, the wave-one boundaries (Figure 4 in Hudson and Thompson, 1998) show that

Samoa is too far south to be used reliably for comparisons with equatorial ozone in any case (Figure 10 and discussion below).

B. Tropospheric Ozone

Validation of tropical tropospheric column ozone during the pre-1991 Nimbus 7 period and during ADEOS and EP is complicated by a geographically and temporally uneven ozonesonde data base. For the 1980's, only Natal ozonesonde data are readily available [Kirchhoff et al., 1991, 1996; Logan, 1994]. For comparison with the sondes, TTO from the MR method is averaged over the 9 pixels from 5-7°S and 32-38°W. The mean agreement with the TTO obtained at Natal, (mean deviation = 6.6 DU, Figure 8A) is excellent, considering that the sonde record of 1-3 sondes/month (except during intensive campaigns [Kirchhoff et al., 1996]) may not capture accurate half-month averages. The same level of agreement with Natal sondes was found with the CCD method (Figure 8B, adapted from Figure 7b in Ziemke et al., 1998). As in the derivation of stratospheric ozone, agreement between the two methods during the N7 period is strong.

Figure 2A shows excellent agreement between twice monthly averaged TTO from Nimbus 7/TOMS (1990-1992) and integrated tropospheric ozone from sondes launched at Ascension Island before and during the TRACE-A field experiment. Ascension sonde launches, at two-per-week frequency, were resumed in mid-1997. Comparing the sonde data with 9-day-averaged TTO from late July 1997 through June 1998 (Figure 9) shows slightly better agreement than for the N7/TTO validation. This may be due to a shorter averaging period for the sondes: 5.1 DU mean deviation for the EP/TTO record.

Comparison between TTO and ozonesonde data from American Samoa (14°S, 171°W; Figure 10, Komhyr et al., 1989) is performed by extending the MR method beyond strictly tropical air, as indicated by Fourier analysis of the wave-one. Accordingly, agreement between TTO and Samoan ozonesondes during 1986-1990 is not as good as at Natal or Ascension; the mean deviation is 11 DU.

4. Evaluation of Nimbus 7 TTO through Regional Time-Series Analysis

In this section, the quality of TTO and other parameters derived in the MR method is evaluated by using time-series to address questions about climatology and atmospheric processes that affect tropical ozone. Most of the time-series are based on N7 data because the EP/ADEOS data records are two years or less. Furthermore, the dominance of the 1997-1998 ENSO in the EP tropospheric ozone record [Chandra et al., 1998a] probably renders the EP time-series atypical. Nimbus 7 TTO data during the period in which total ozone was suppressed by the Mt Pinatubo eruption (mid-1991 through June 1992) are also used sparingly.

A. Seasonal Cycles and Interannual Variability at Ozonesonde Sampling Sites

Time-series of TTO data over given regions are used to address the question:

☛ What do seasonal cycles and interannual variability in TTO look like and how do they compare with variations observed in ozonesondes?

Figures 2A and 8 show comparisons between ozonesonde data in the Atlantic region and TTO for the 9 pixels surrounding Ascension, Brazzaville and Natal. The distinctive feature at all three sites is a regular seasonal variation, with tropospheric ozone peaking 20-25 DU higher in the latter half of the year than in the second quarter when ozone is a minimum.

Figures 11A-C shows the variability of TTO at the Natal, Ascension and Brazzaville locations, along with larger regions between the eastern Pacific just west of South America across the Atlantic to southern Africa (Figures 11D-G). The seasonality of TTO in the regions shown in Figure 11 is determined by fitting a linear regression model [Hollandsworth et al., 1995] to the 14-year twice-per-month time-series. The model, which has been used extensively for analysis of trends in total ozone [Stolarski et al., 1996], includes the assumption of a seasonal cycle and linear trend (Section 4B). The regression model can be modified to take into account solar cycle effects, ENSO signals and the QBO; this has not been done in the current analysis. The reader is referred to Ziemke et al. [1998] and Thompson et al. [1996b], respectively, for discussion of QBO effects on derived stratospheric ozone and total ozone in the Nimbus 7 records.

Figure 12 shows the regression model best-fit for seasonality in the TTO (solid line) and ozonesonde data at each site (----). For Natal (Figure 12A), there are 155 of a maximum possible 336 data points based on twice-per-month averaging of the sondes over 14 years; see the density of * in Figure 8A. For Ascension and Brazzaville, there are only 39 and 42 data points compared to 72 twice-monthly intervals in the 1990-1992 record. Discuss...(to be added) - stress agreement of MR-TTO with sondes and coherence of three locations.//

Time-series over the Nimbus 7 period are also used to explore the question:

☛ Do the wave amplitude and TTO derived from the MR method show signals expected from ENSO influences?

The amplitude of the wave-one feature (Figure 1) from 1979-1992 (Figure 13; open circles, in DU) is a measure of the contrast between the Atlantic background tropospheric ozone maximum, $O_3)_{tr}^{back}(0)$, and the Pacific minimum, $O_3)_{tr}^{back}(180)$. The maximum wave amplitude in the March-May period corresponds to greater convective activity over the Pacific Ocean, which brings ozone-poor air from surface up through the free troposphere, reducing the tropospheric ozone column amount. In this respect, the wave may be thought of as signifying an eastern Pacific ozone deficit relative to the Atlantic. In the August-October period the maximum wave amplitude reflects "excess" ozone over the Atlantic, which is attributed to advection of photochemically produced ozone from the adjacent continents, followed by subsidence [Chatfield et al., 1996; Krishnamurti et al., 1996; Thompson et al., 1996a].

<u>Region</u>	<u>TTO mean, 1979-1992</u>	<u>TTO mean, 8/96-6/98</u>
0-12S, zonal mean	26.2 DU	31.8 DU
E. So. Am. (0-12S, 40-70W)	31.4 DU	37.2 DU
So. Africa (0-12S, 0-30E)	25.4 DU	29.5 DU
Natal region (4-8S, 32-38W)	34.2 DU	39.7 DU

Note that comparison of EP/TOMS with the ground-based Dobson network of instruments for measuring total ozone has shown a possible higher bias of EP/TOMS relative to the N7 record (G. Labow, R. McPeters and R. Stolarski, personal communication, 1998). However, the MR method should be independent of this difference (Figure 1). Ultimately, determining the relative strength of the 1982-1983 and 1997-1998 ENSO events on tropospheric ozone will require a longer time-series with EP/TOMS. It would also be desirable to fill in the 1993-1994 period with a TTO record derived from METEOR/TOMS; this has not yet been done due to the different orbital characteristics of METEOR/TOMS compared to N7, EP and ADEOS.

B. TTO Trends during the Nimbus 7/TOMS Period

In contrast to residual methods [Fishman et al., 1991, 1996; Ziemke et al., 1998] which are based on multiple sensors, with mismatches in sampling period, footprint, calibration and operational lifetime, tropospheric ozone determined from the TOMS-only MR method is well-suited for analysis of trends. Because of the brevity of the EP/TOMS TTO and its strong ENSO signature, only TTO data from the N7 record are used in trends analysis here.

We note that from mid-1991 through 1992, record low total ozone, due to the eruption of Mt Pinatubo, introduces a strong bias in the latter part of the N7 record. For this reason, and because the first year of Nimbus 7 operation may have had some startup sampling inconsistencies, only TTO from 1980 through 1990 are used for analysis of trends. The linear regression model [Hollandsworth et al., 1995] used for determination of seasonality (Figure 12) is the basis for determination of linear and seasonal trends with the N7 TTO record. Model results for Natal appear in Figure 14. The model fit (----) to the TTO time-series (solid line in Figure 14A) is excellent. The ozonesonde data with model fit appear in Figure 14B. The straight line represents the linear trend, with the deseasonalized mean TTO and sonde integrated tropospheric ozone values in Table 1, as the starting point. In Figures 14C and D the seasonal trends for the TTO and sonde data sets appear, in DU/year, with the dotted line indicating 2σ .) There is no significant linear trend at Natal, according to the TTO record: -0.10 ± 0.20 DU/yr; the Natal mean TTO over 1980-1990 is 36.3 ± 0.11 DU. The sonde data, which are quite sparse (see also Figure 8A), give a mean of 2 DU less than TTO (Table 1) and a trend that is insignificant to the 2σ level: 0.24 ± 0.26 DU/yr. Both the TTO and sonde analyses show that, although the annual trend is negligible, this could be due to opposing factors operating at different times of year. Namely, a small but possibly

significant increase in tropospheric ozone in the early part of the year is followed by a negative trend.

Table 1. Regional TTO (DU) and Trends (DU/yr) in Tropical Tropospheric Ozone from MR method, 1980-1990

	Mean column TTO (DU, +/- 2 σ)	Trend (DU/yr) (+/- 2 σ)	Figure No.
Natal, sondes 6S, 35W	33.4 (2.8)	+0.33 (0.42)	14B, D
Natal, TTO 5-7S, 32-38W	36.3 (1.1)	-0.08 (1.6)	14A,C
Ascension Is. TTO 7-9S, 12-18W	36.6 (1.2)	0.010 (0.20)	15A
0-12°N, zonal mn	27.1 (.82)	-.077 (.10)	n/a
0-12°S, zonal mn	27.4 (.82)	-.075 (.10)	n/a
S. America, 0-12°S, 40-70W	34.6 (1.2)	-0.10 (.18)	15B
E. Pacific 0-12°S, 80-110W	25.5 (.90)	-0.41 (1.4)	15D
S. Africa, 0-12°S, 0-30E	37.1(.47)	-.0222(1.2)	n/a
S, Atlantic, 0-12°S, 0-40W	37.8 (.45)	0.71 (1.4)	15C

The linear regression analysis of TTO for other regions shows that seasonality, interannual variability and trends at Natal are representative of tropospheric ozone over the entire south Atlantic Basin. Figure 15 shows TTO corresponding to Ascension Island, eastern South America, the South Atlantic, and the Eastern Pacific, with the model fit for each case and the linear trend. As for Natal, there are no significant trends on an annual basis, but small seasonal trends are marginally significant in the first part of the year (Figure 16). The South Atlantic region encompasses both Natal and Ascension Island, whereas South America should typify most of northern Brazil. Uniformity in seasonality and insignificant or borderline significant seasonal trends appear in all regions from the eastern Pacific across the south Atlantic Basin. For completeness, analysis of the 14-year TTO record in the zonally averaged band from 0-12S and 0-12N is also given in Table 1. No significant trend is apparent throughout the tropical band.

The regression analysis suggests that tropospheric column ozone in regions well-known for seasonal burning - Brazil, and south central Africa, for example - did not change significantly during the 1980's. Unfortunately, although a number of satellite sensors for detection of fires were operational in the 1980's, no single fire count product is available to compare with the N7 TTO record. Although smoke as a proxy for fires is difficult to correlate consistently with tropospheric ozone due to different characteristics of low-altitude detection of ozone and aerosols, the TOMS absorbing aerosol product (detecting smoke and dust) is available for the entire N7 period (see, the TOMS homepage: <http://toms.gsfc.nasa.gov> to view monthly-averaged absorbing

aerosol maps). In general, no strong trend in absorbing aerosol appeared during 1979-1992 [Herman et al., 1997].

The fact that any significant trend in TTO occurs during the first part of the year, which is out-of-phase with the southern hemisphere savanna burning season, would be consistent with little or no aerosol (fire) trend. However, an increase in background tropospheric ozone could explain the tendency toward an increase in the early part of the year. Global methane and CO increases occurred throughout the 1980's (Ref), even though the increase in tropical CO appears to have leveled off or even reversed in the 1990's [Novelli et al., 1998]. Such increases alone would have produced a small ozone increase, which might be more noticeable when tropospheric ozone is near its annual minimum.

The finding of no significant trends in tropical tropospheric ozone agrees with Chandra et al. [1998b], who have examined the 1979-1992 tropical tropospheric ozone time-series obtained by the CCD technique. How do our results compare to trends in lower tropospheric ozone (surface to 6 km) inferred from TOMS by a terrain-differencing method [Jiang and Yung, 1996; Kim and Newchurch, 1996; 1998]? These groups have analyzed several smaller regions from 1979-1992:

<u>Name</u>	<u>Trend</u>	<u>Ref.</u>
West of New Guinea	+0.06DU/yr	Kim and Newchurch, 1998
East of New Guinea	no trend	Kim and Newchurch, 1998
E. Pacific (2-5N)	"slightly positive"	Kim and Newchurch, 1996
E. Pacific (0-12S)	no trend	Kim and Newchurch, 1996
E. Pacific (12-23S)	+0.14DU/yr	Jiang and Yung, 1996
East of Andes (0-12S)	+1%/yr	Kim and Newchurch, 1996

Our inference of no trend for the eastern Pacific between the equator and 12S concurs with Kim and Newchurch [1996]. However, the TTO maps show no increases east of the Andes during the 1980's (eastern South America, Figure 15b), whereas Kim and Newchurch [1998] find a 1%/yr increase. There are several explanations for this. Kim and Newchurch [1996, 1998] use only a few pixels of TOMS data for their time series. They also used the gridded TOMS product, which is subject to artifacts due to clouds and low-altitude ozone detection efficiency [Hudson et al., 1995; J. R. Ziemke, P. K. Bhartia and R. D. Hudson, unpublished results, 1998].

5. Summary

A time-series of maps based on TOMS ozone data, two per month with a 1° latitude by 2° longitude grid, have been produced for tropical tropospheric column ozone between 20N and 20S. Periods include the 1979-1992 Nimbus 7 record and ADEOS and EP-TOMS, the latter from 1996 to the present. The technique used, the modified-residual method, is based on high density, cloud-free TOMS ozone in combination with ozonesonde data to separate stratospheric and tropospheric ozone. In this validation and evaluation study, the focus is on the tropical

Atlantic and near-Atlantic stations. The major findings are:

(1) Comparison with sonde data shows excellent agreement within the uncertainty of the MR method ($1\sigma = 5$ DU) at Natal, Brazil (1979-1992), Brazzaville (1990-1992) and Ascension Island (1990-1992; 1997-1998).

(2) Analysis of time-series from the 1979-92 TTO data set show seasonality consistent with the ozonesondes; highest ozone occurs in the second half of the year.

(3) The wave-one pattern in equatorial tropospheric ozone is a persistent feature, with seasonally varying amplitude and correlation with markers of dynamical variability, eg SST, prominent during ENSO events. Regional signatures of the 1982 ENSO appeared as localized TTO maxima in eastern South America and as minima in the eastern Pacific and southern Africa.

(4) From 110°W to 30°E , TTO shows insignificant trends from 0 - 12°S during 1980-1990, which agrees with the findings of Kim and Newchurch [1996] for the eastern Pacific but not for eastern South America. The Natal ozonesondes, the only operational site in the tropics throughout the 1980's, shows a positive, but barely significant, trend in the first half of the year.

Comparisons between the MR method and the CCD method [Ziemke et al., 1998], which is another TOMS-only approach to retrieval of tropospheric column ozone, show excellent agreement between derived stratospheric ozone and tropospheric column ozone, with the latter performed using the Natal data set as a standard. The assumptions of the two methods, as well as the selection of TOMS high-density (Level 2) data, allow extraction of stratospheric ozone from total ozone by very different approaches.

Validation, evaluation and intercomparison of the MR TTO data is continuing, with expansion to regions beyond the Atlantic as more tropical ozone data become available. An appealing feature of the MR method is the time-averaging (2-3 maps/month) and spatial resolution, which should render them useful for process studies and field campaigns, as well as for climatological investigations of the type presented here. Readers are encouraged to use the maps on the homepage and to correspond with us about correlative ozone data.

Acknowledgments. Thanks to R. M. Todaro for producing the Nimbus 7 1979-1990 TTO maps and to H. Guo for the EP- and ADEOS TTO maps and validation. A. D. Frolov has helped with sonde and SAGE II analysis. Ascension Island sondes since 1997 are courtesy of F. J. Schmidlin at the GSFC/Wallops Flight Facility. Real-time TOMS processing for EP- and ADEOS-TOMS is possible thanks to the Goddard Ozone Processing Team. We appreciate discussions with J. R. Herman, P. K. Bhartia and O. Torres on TOMS absorbing aerosol data and comments on the manuscript from S. Chandra, J. R. Ziemke, R. D. McPeters and J. F. Gleason. Many thanks to S. M. Hollandsworth, T. L. Kucsera, M. G. Seybold (SM&A at NASA/Goddard) and A. V. Cresce for assistance with the trends program. Support for this analysis, and for some of the ozonesondes, is provided by the NASA ACMAP (Atmospheric Chemistry Modeling and Analysis Program). Additional support comes from the NASA Tropospheric Chemistry Program and an EOS Interdisciplinary Science study.

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Captions.
17 Oct 98

Fig. 1 Schematic of the modified-residual (MR) method for deriving tropical tropospheric ozone and stratospheric ozone, given total ozone from TOMS, $O_3)_{total}$, a wave-one pattern (with amplitude λ) and excess ozone, $O_3)_{excess}$. The basis of distinguishing stratospheric and tropospheric ozone is a 2-year climatology of ozonesondes at three sites near the wave maximum (0° longitude). These imply a fixed, seasonally varying $O_3)_{tr}^{back}(0)$ which is assumed to apply over the duration of Nimbus 7 (1979-1992), Earth-Probe (Aug. 1996-present) and ADEOS (Sept. 1996 - April 1997) TOMS and from which A and $O_3)_{str}$ are derived. A corresponds to $O_3)_{tr}^{back}(180)$, which refers to the location of the total ozone minimum, at or near 180° longitude.

Fig. 2 Comparison of tropical tropospheric ozone (TTO) derived from the modified-residual method for 1990-1992 at (A) Ascension Island ($8^\circ S, 15^\circ W$); (B) Brazzaville, Congo ($4^\circ S, 15^\circ E$) (C) Natal, Brazil ($6^\circ S, 35^\circ W$). Line denotes twice-per-month averaged TTO from Nimbus 7/TOMS and * symbol is integrated ozone from the sounding, surface to 100 hPa. The precision of the MR method is ± 5 DU (Dobson Units, $1 \text{ DU} = 2.69 \times 10^{16} \text{ cm}^{-2}$), owing to averaging tropospheric ozone over 1-2-week periods.

Plate 1. (A) Typical Nimbus 7 period map as it appears on the TTO website. URL = <http://metosrv2.umd.edu/~tropo>. Each month's record consists of two maps. The first is based on averaging Days 1-15 Level 2, low-reflectivity TOMS total ozone; the second image is based averaging TOMS from Day 16 to the end of the month. Processing with the MR method is carried out from 20N to 30S but values are shown only within the range of the wave-one pattern. (B) same as (A) except for an ADEOS map during the southern hemisphere 1996 burning season. Real-time maps show $10^\circ S-10^\circ N$, for easy reference during field campaigns.

Fig. 3 Comparison of derived stratospheric ozone, $O_3)_{strat}$, with SAGE II ozone, averaged over $10^\circ S-10^\circ N$, and recorded from 1985-1991. From June 1991-1993, stratospheric aerosols from the Mt Pinatubo volcanic eruption made reliable SAGE ozone retrievals impractical. Nimbus 7/TOMS derived stratospheric ozone record is compared with the UARS/MLS after September 1991 (Figure 6 in Hudson and Thompson, 1998).

Fig. 4 Comparison of derived stratospheric ozone, $O_3)_{str}$, with UARS MLS stratospheric column ozone, averaged over $10^\circ S-10^\circ N$, 1996-1998. Integration of ozone is from 1-100 hPa; below 46 hPa, MLS ozone precision is 50%.

Fig. 5 Comparison of derived stratospheric ozone, $O_3)_{str}$, solid line, from the modified-residual method for 1979-1992 at Natal, Brazil ($6^\circ S, 35^\circ W$). Natal is the only tropical sounding station with regular sondes since 1978 [Kirchhoff et al., 1991; 1996]. * denotes stratospheric ozone computed by subtracting twice-per-month averaged tropospheric ozone from Natal sondes from TOMS total ozone).

Fig. 6 Same as Fig. 5 except that derived stratospheric ozone, $O_3)_{str}$, is obtained from the twice-weekly ozonesonde launches at Ascension Island during mid-1997-mid-1998.

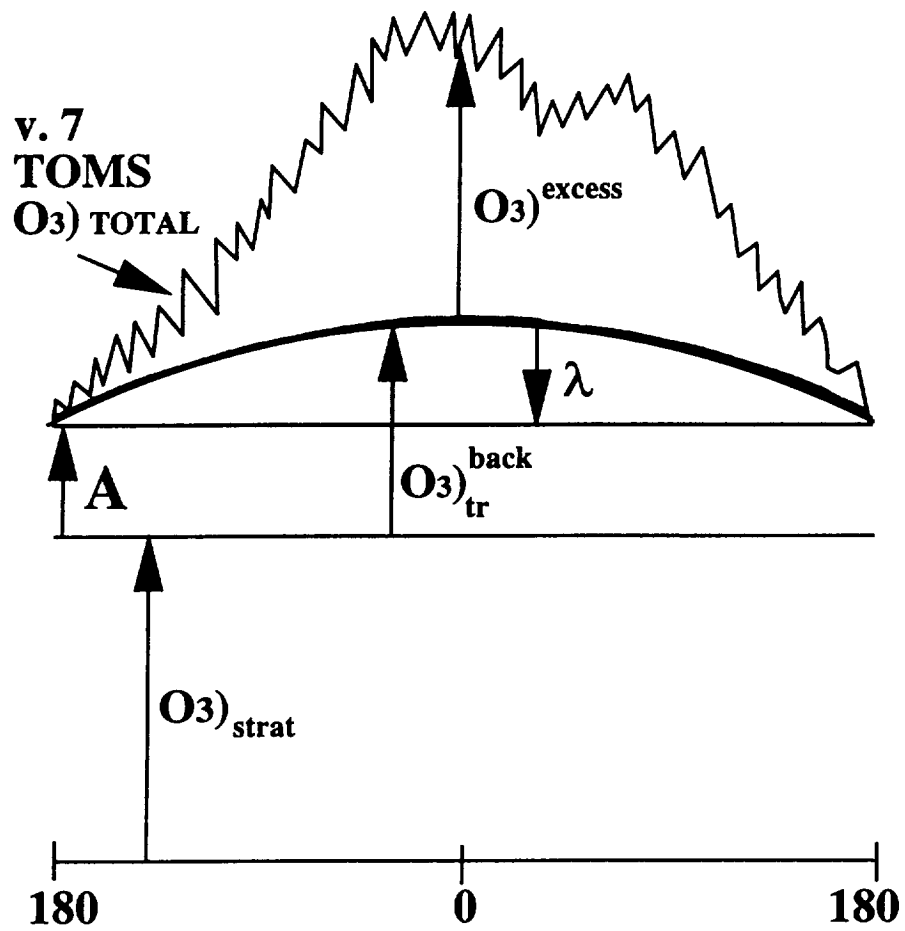
- Fig. 7 Comparison of derived stratospheric ozone from the modified-residual method and from the CCD method. CCD method gives monthly values. For comparison a monthly value from MR method has been obtained by averaging two values for O_3^{str} each month.
- Fig. 8 (A) Comparison of integrated tropospheric ozone from ozonesondes with TTO from the modified-residual method for 1979-1990 at Natal, Brazil (6°S, 35°W). Natal is the only tropical sounding station with regular sondes since 1978 [Kirchhoff et al., 1991; 1996]. Line denotes twice-per-month averaged TTO from 4-6S and 32-38W; * symbol is integrated ozone from the sounding, surface to 100 hPa. Deviation of TTO from sonde value appears at bottom of figure. (B) Same for monthly averaged ozonesondes and derived TTO from CCD method; in the latter a single 5x5° pixel is used.
- Fig. 9 Comparison of TTO from Earth-Probe/TOMS for late July 1997-May 1998 at Ascension (8°S, 15°W). Ascension soundings were re-activated in 1997 after a hiatus of nearly 5 years; launch frequency in 1997-1998 is twice per week. Line denotes twice-weekly averaged TTO and * symbol is integrated ozone from the sounding, surface to 100 hPa. Deviation of TTO from sonde is \diamond .
- Fig. 10 Same as Figs. 8 and 9 except that comparison is between integrated tropospheric ozone from Samoan sondes (14°S, 171°W) and N7/TTO obtained by extending processing to 14S, which is usually south of the wave-one pattern denoting tropical air masses.
- Fig. 11 TTO derived from the MR method over the Nimbus 7 period at three ozone sounding sites, for which latitude-longitude given in caption for Figure 2 (A = Natal; B = Ascension; C = Brazzaville). Latitude is 0-12°S for the other regions, with longitudes as follows: D = Eastern Pacific, 80-100°W; E = eastern So. America, 40-70°W; F = south Atlantic, 0-40°W; southern Africa, 0-30°E. Deseasonalized mean, determined by linear-regression model, appears in each frame.
- Fig. 12 Model determined seasonality for Natal (A), Ascension (B) and Brazzaville (C) where solid line signifies analysis of 14-year TTO corresponding to each site and dashed line refers to seasonality based on integrated tropospheric ozone from the sonde record: 1979-1992 at Natal, 1990-1992 for Ascension and Brazzaville.
- Fig. 13 Amplitude of tropospheric wave, in Dobson Units (DU, outer scale, open circles), over 14-year Nimbus 7/TOMS record [cf Ziemke et al., 1998]. Pattern of minima twice per year (typically in May-June and December-January) represents N-S transition in Intertropical Convergence Zone, ITCZ. Stable ITCZ in either hemisphere is associated with convective transport causing dilution of the tropospheric ozone column over the Pacific relative to the Atlantic [Piotrowicz et al., 1991]. Effect of the 1982-83 and 1987 ENSO events on wave amplitude is illustrated by positive correlation of wave amplitude with the sea-surface temperature (filled circles, ΔSST , deg K) anomaly. Positive anomaly signifies more convection over the Pacific, a greater Atlantic-Pacific ozone contrast, ie larger wave amplitude.

Fig. 14 (A) Time-series of TTO (solid line) at Natal (6°S, 35°W), with best-fit (----) from a linear regression model with seasonal cycle and trends assumed over 1980-1990. Linear, deseasonalized trend is indicated with straight line. (B) Seasonal cycle from the model fit (%/yr), with the 2σ significance indicated by the dotted line. C, D. Same except that the Natal ozonesonde record, with sondes averaged to twice-per-month frequency, are the basis for the model analysis.

Fig. 15 (A) Ascension Island TTO for the whole calendar years of the Nimbus 7 period, 1980-1990. As in Fig. 14, model best-fit (dashed line) and deseasonalized trend (straight line) are given. B, C, D. Same as A for eastern South America (0-12S, 40-70W); southern Atlantic (0-12S, 0-30E); east southern Pacific (0-12S, 80-110°W). Southern African TTO (0-12S, 0-30E), not shown, is similar to the south Atlantic.

Fig. 16 Seasonal cycle from the model fit (%/yr), with the 2σ significance indicated by the dotted line. Same analyses as Fig. 15. A = Ascension; B = eastern South America; C = south Atlantic; D = eastern south Pacific.

Modified – Residual Method



[Hudson & Thompson, 1998]

Fig. 1

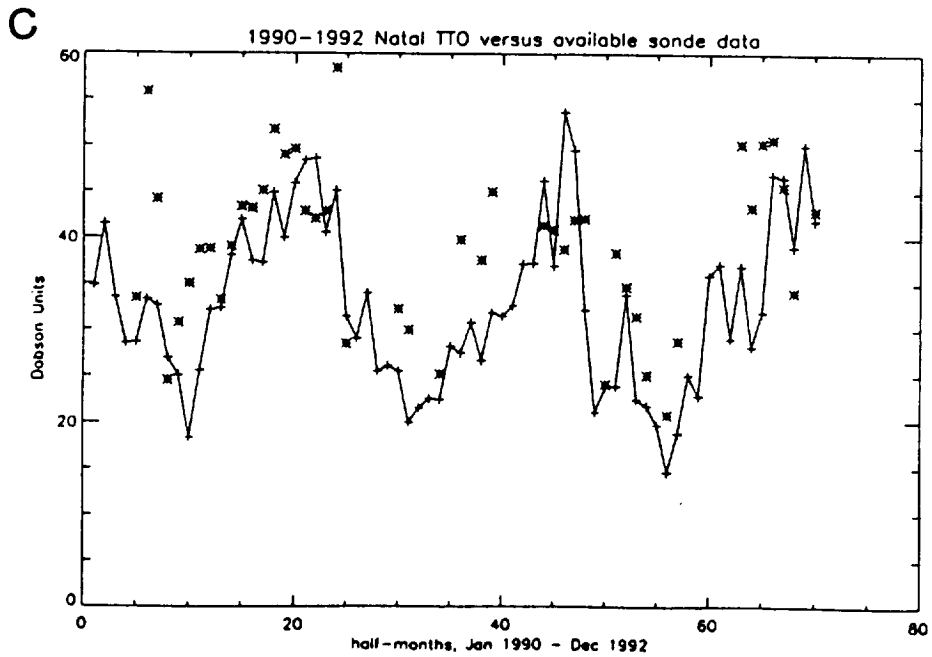
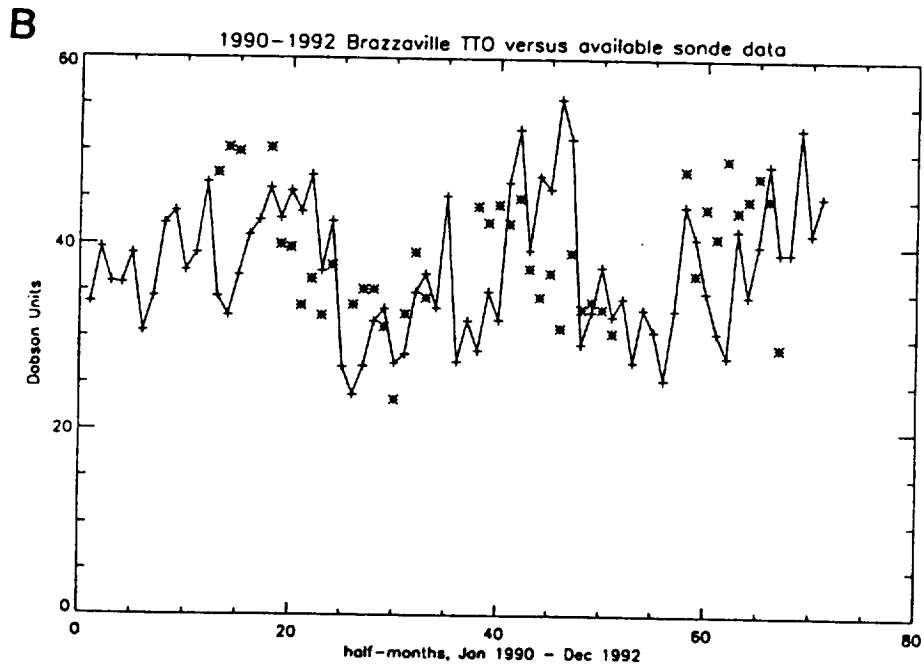
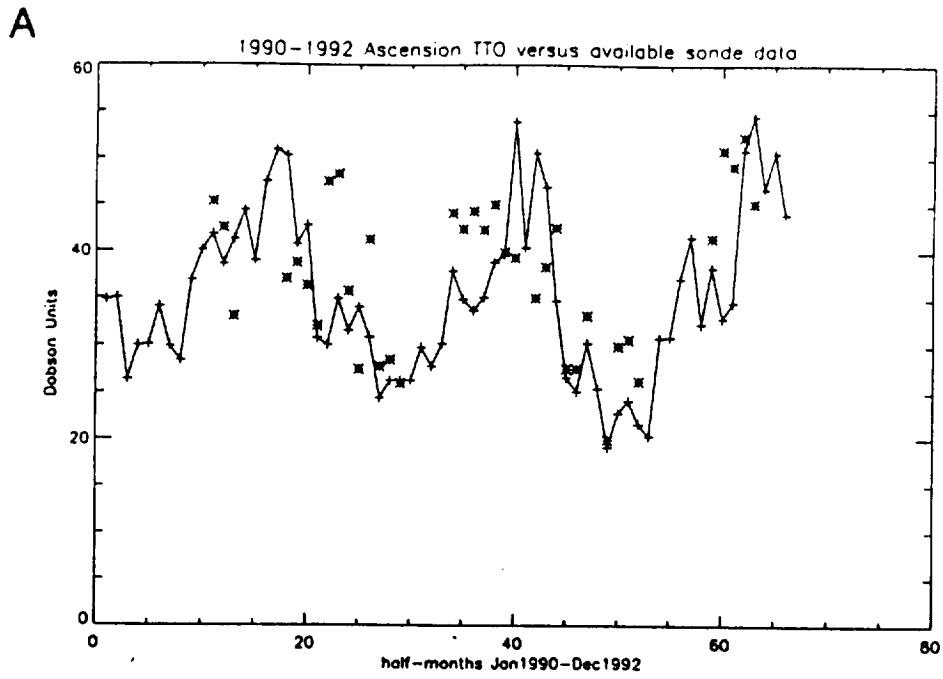


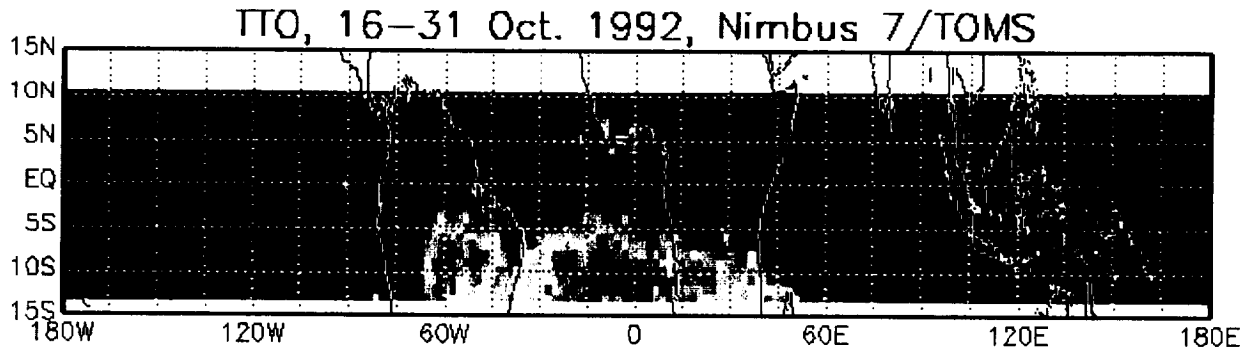
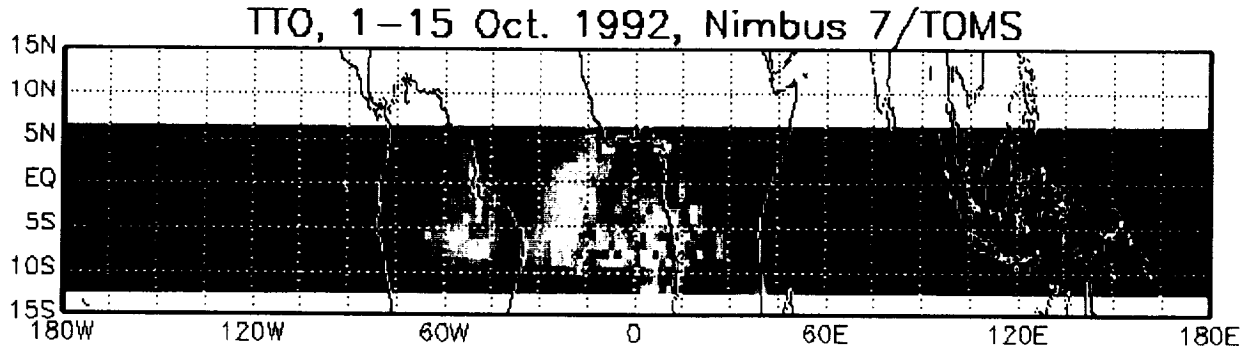
Fig. 2

1979-92 Tropical Tropospheric Ozone(TTO), Nimbus 7/TOMS

Year 1992

Month October

Go



no data 0 10 20 30 40 50 60 70 DU UMD/GSFC

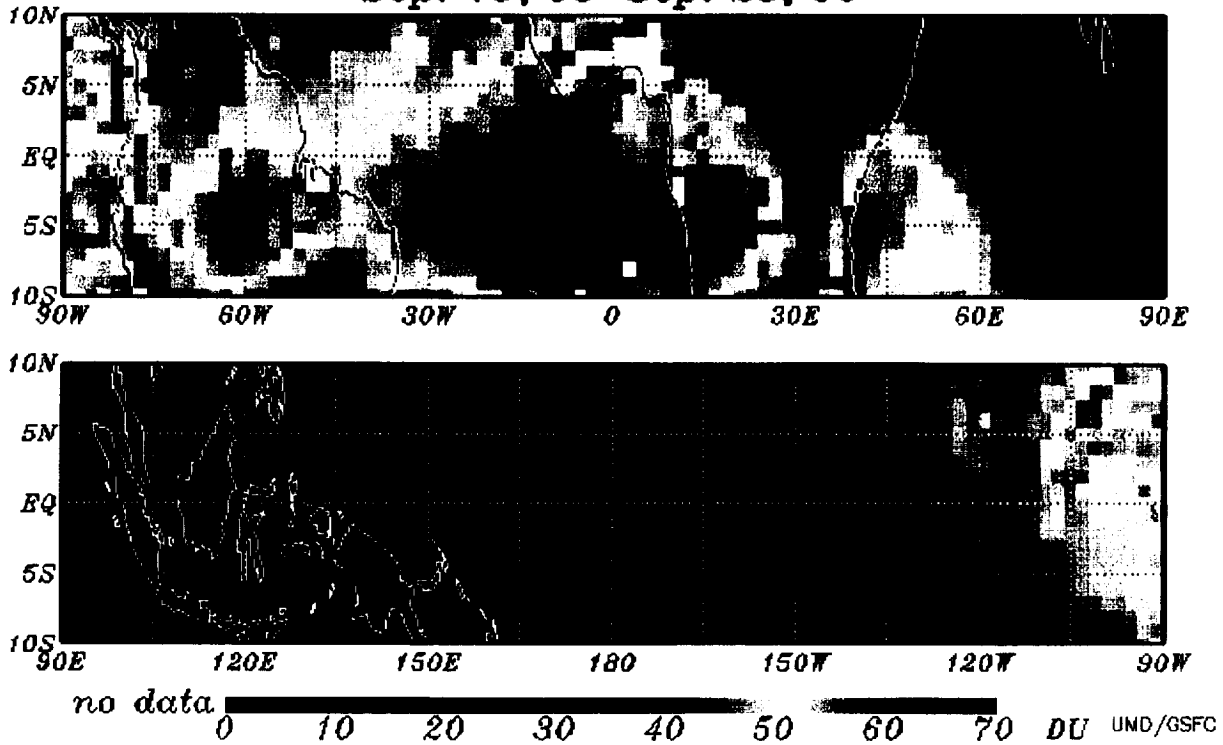
Plate 1A

Tropical Tropospheric Ozone from ADEOS/TOMS

Daily based 3 days average 9 days average
Year 1996 Month Sep Day 19

Go  

Tropical Tropospheric Ozone, ADEOS/TOMS Sep. 15, 96-Sep. 23, 96



Click "Real-time" on the menu to see TTO (Earth Probe/TOMS).

Plate 1B

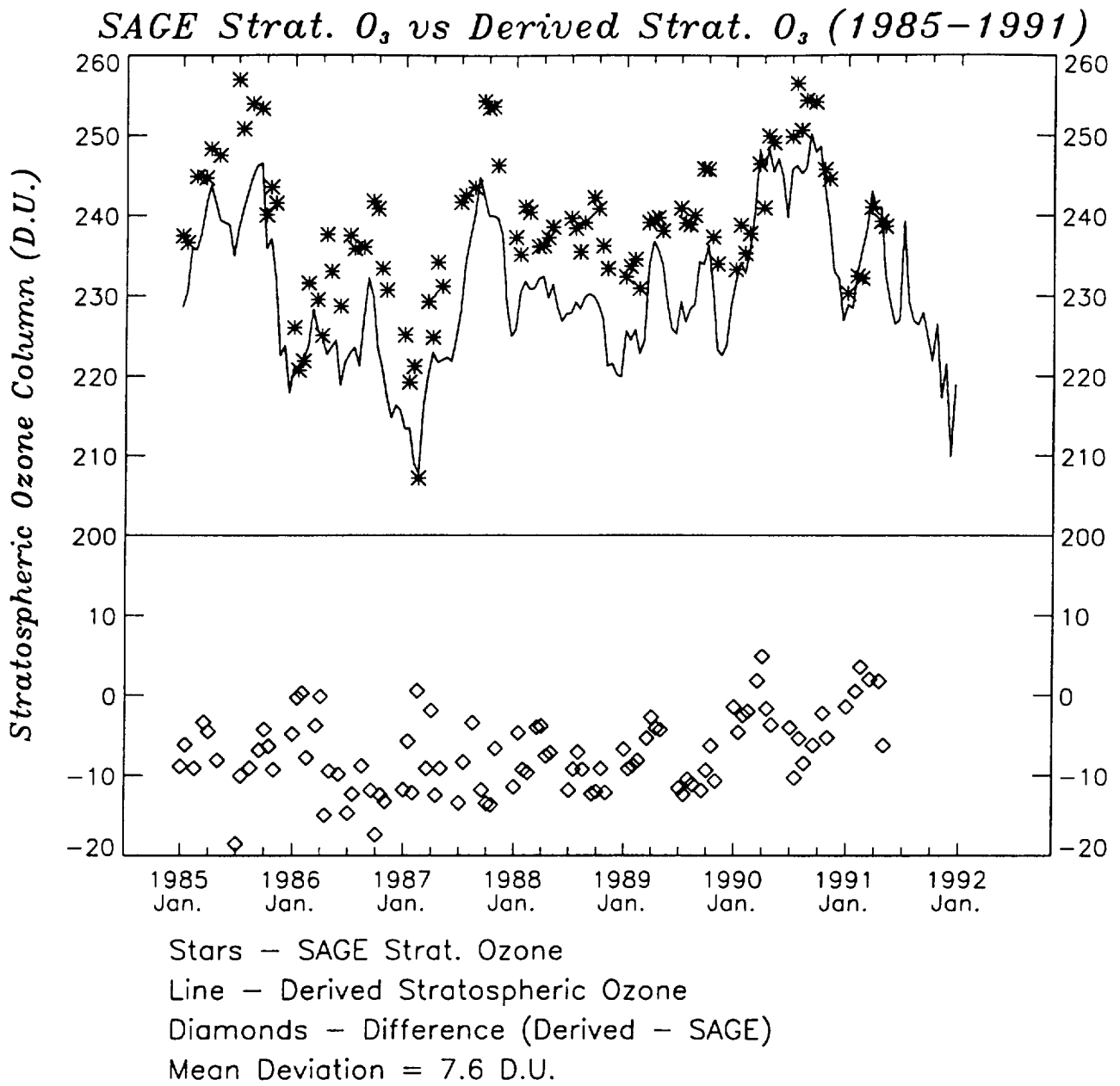
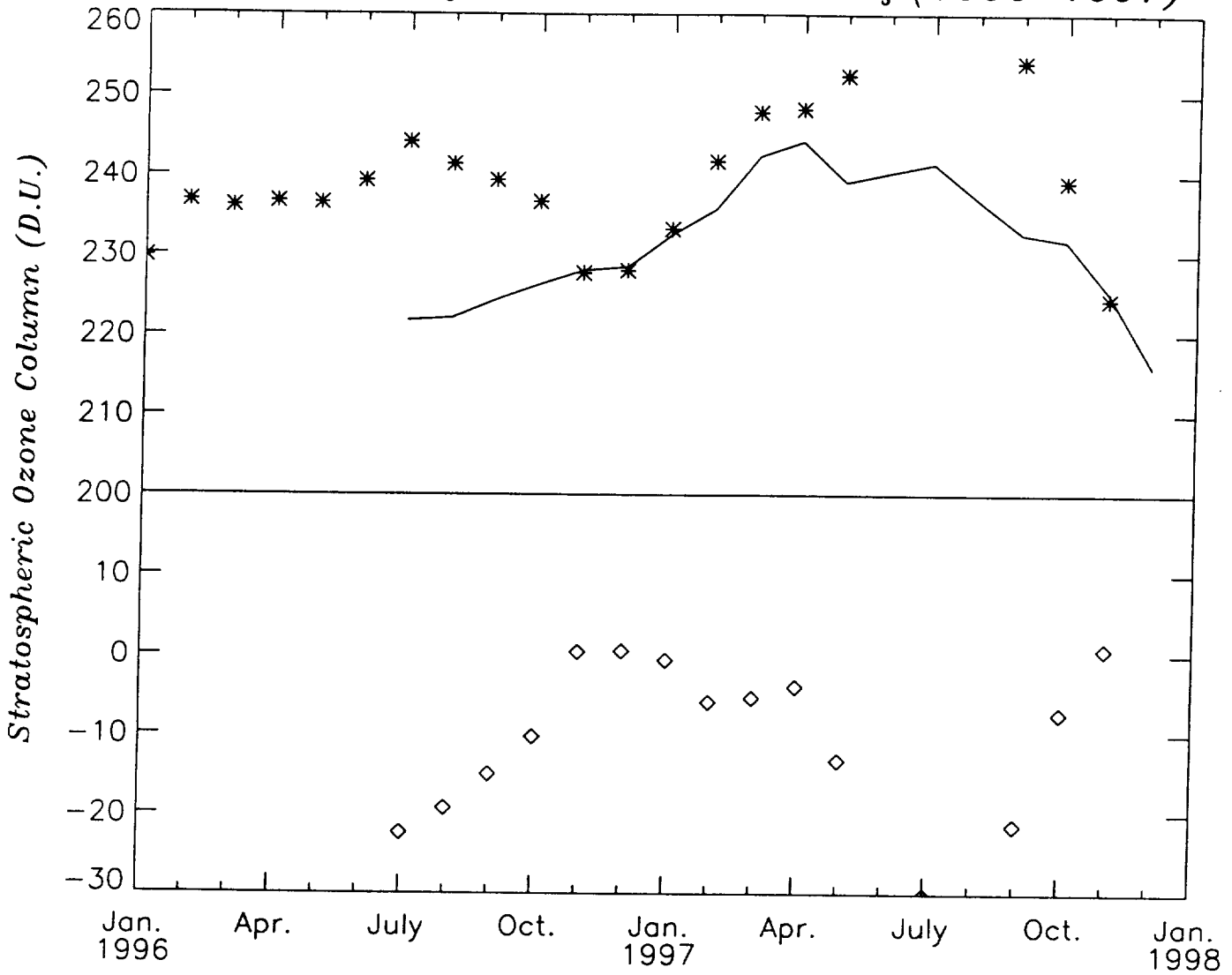


Fig. 3

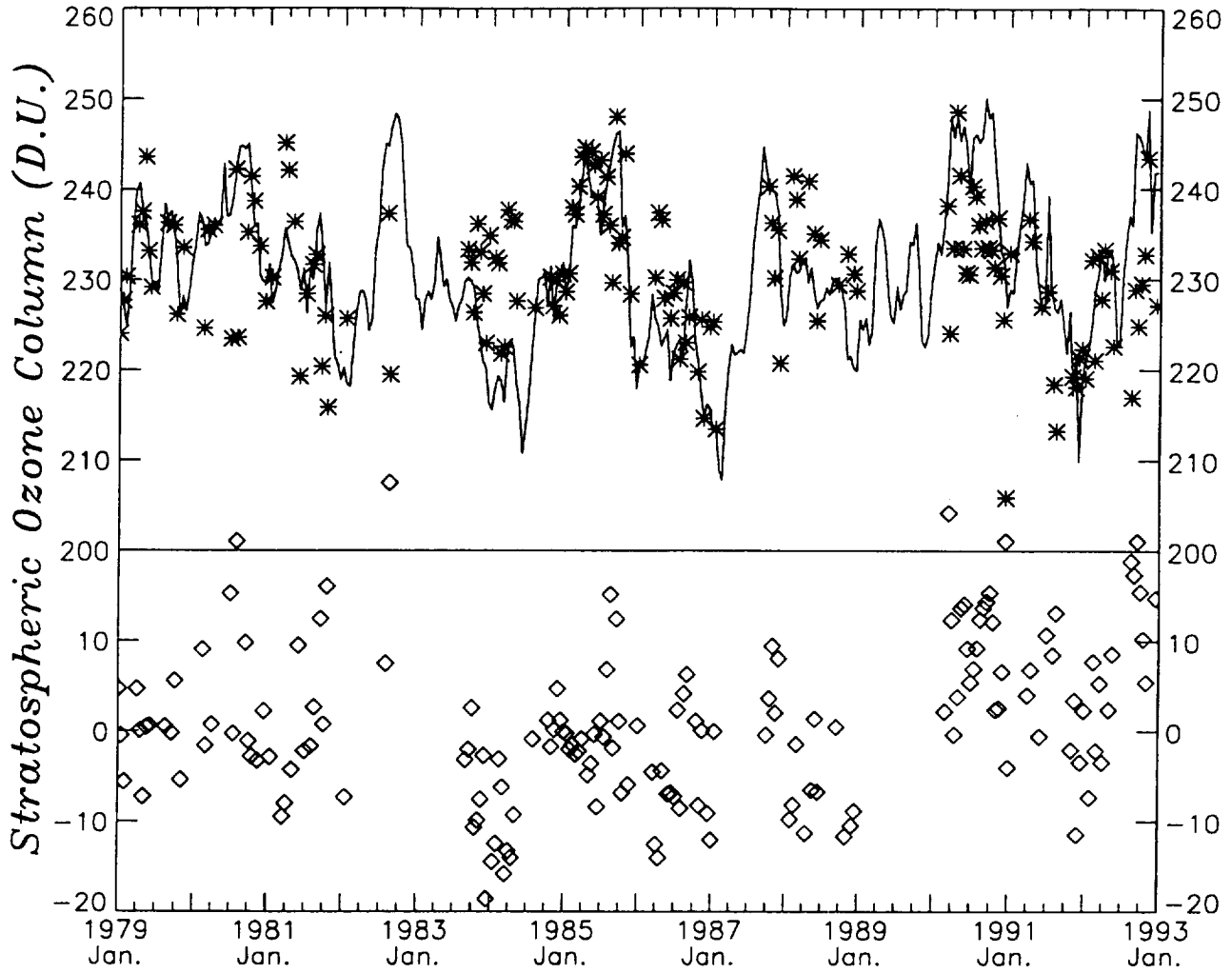
MLS Strat. O₃ vs Derived Strat. O₃ (1996-1997)



Stars - MLS Strat. Ozone
Line - Derived Stratospheric Ozone
Diamonds - Difference (Derived - MLS)
Mean Deviation = 10.5 D.U.

Fig. 4

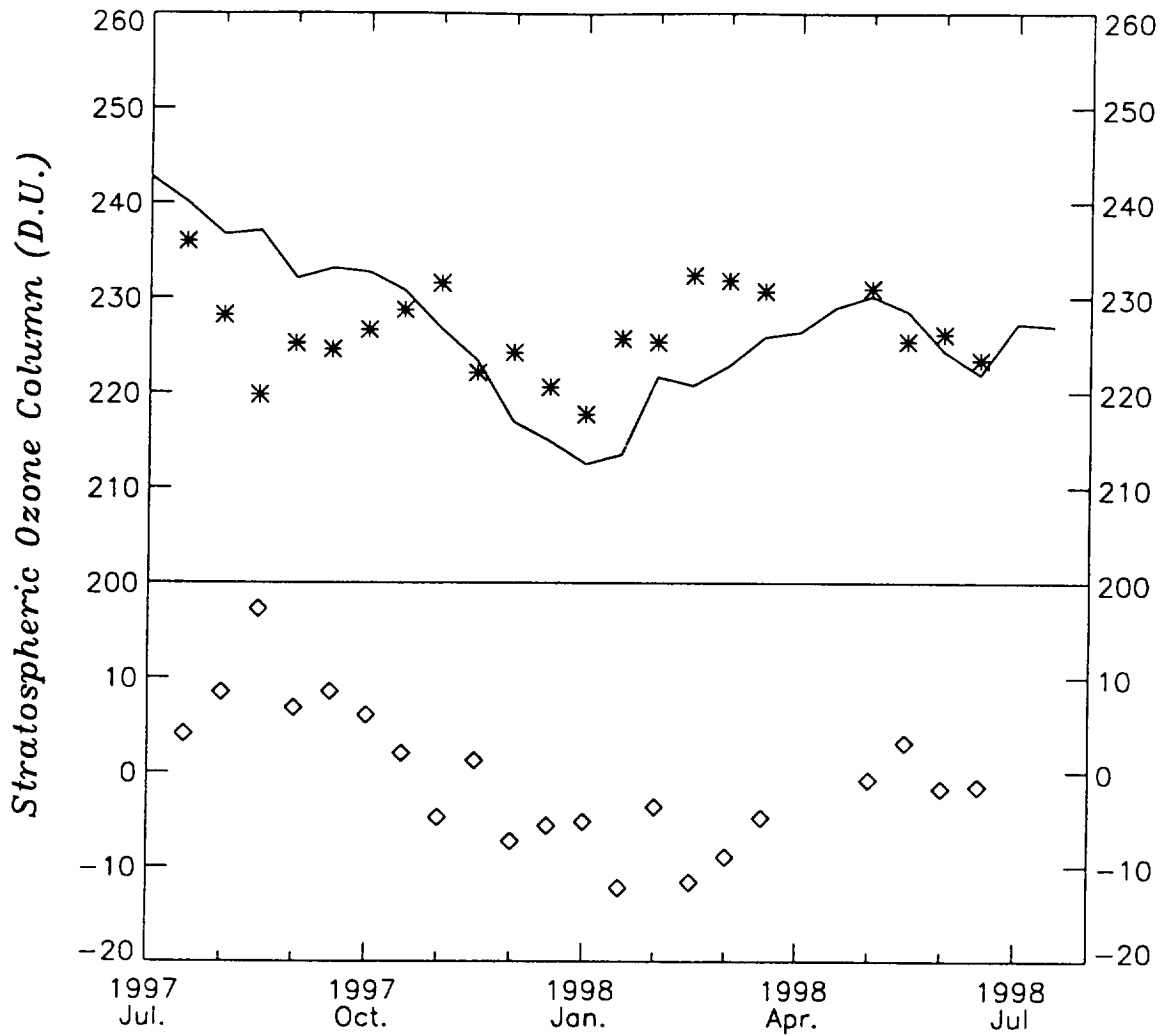
*(TOMS-Tropospheric O₃ from Natal Ozonesondes)
vs Derived Strat. O₃, 1979-1992*



Line - Derived Strat. O₃
Stars - (TOMS-Tropospheric O₃ from Natal Ozonesondes)
Diamonds - Difference (Derived - Natal)
Mean Deviation = 6.7 D.U.

Fig. 5

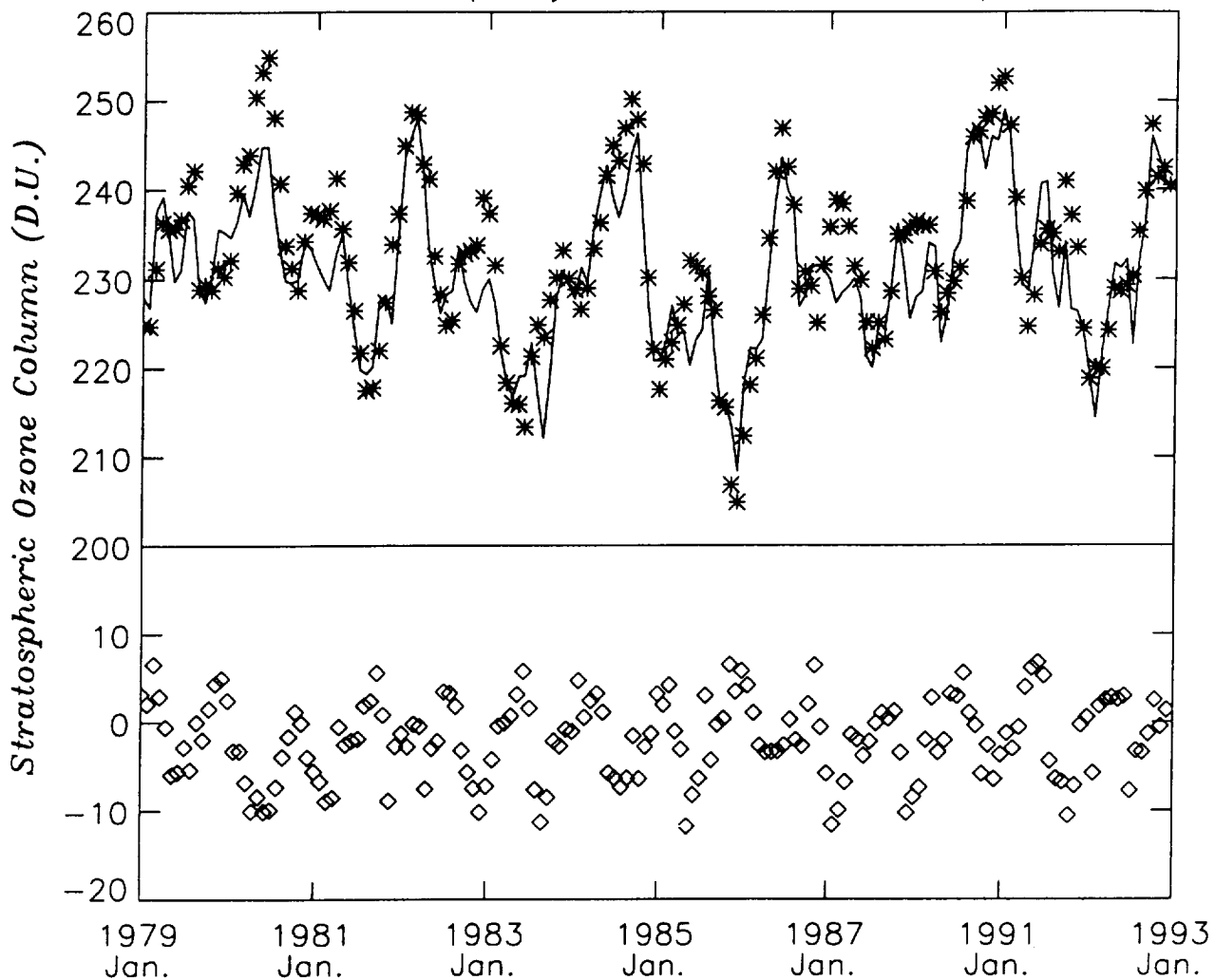
*(TOMS-Tropospheric O₃ from Ascension Ozonesondes)
vs Derived Strat. O₃, July 1997-July 1998)*



Line - Derived Stratospheric ozone
 Stars - (TOMS-Tropospheric O₃ from Ascension Ozonesondes)
 Diamonds - Difference (Derived - Ascension)
 Mean Deviation = 6.0 D.U.

Fig. 6

Derived Strat. O₃ (MR) vs CCD Strat. O₃ (1979-1992)



Stars - CCD Strat. Ozone
Line - Derived Stratospheric Ozone
Diamonds - Difference (Derived - CCD)
Mean Deviation = 3.9 D.U.

Fig. 7

1979-1992 Natal TTO versus available Sondes

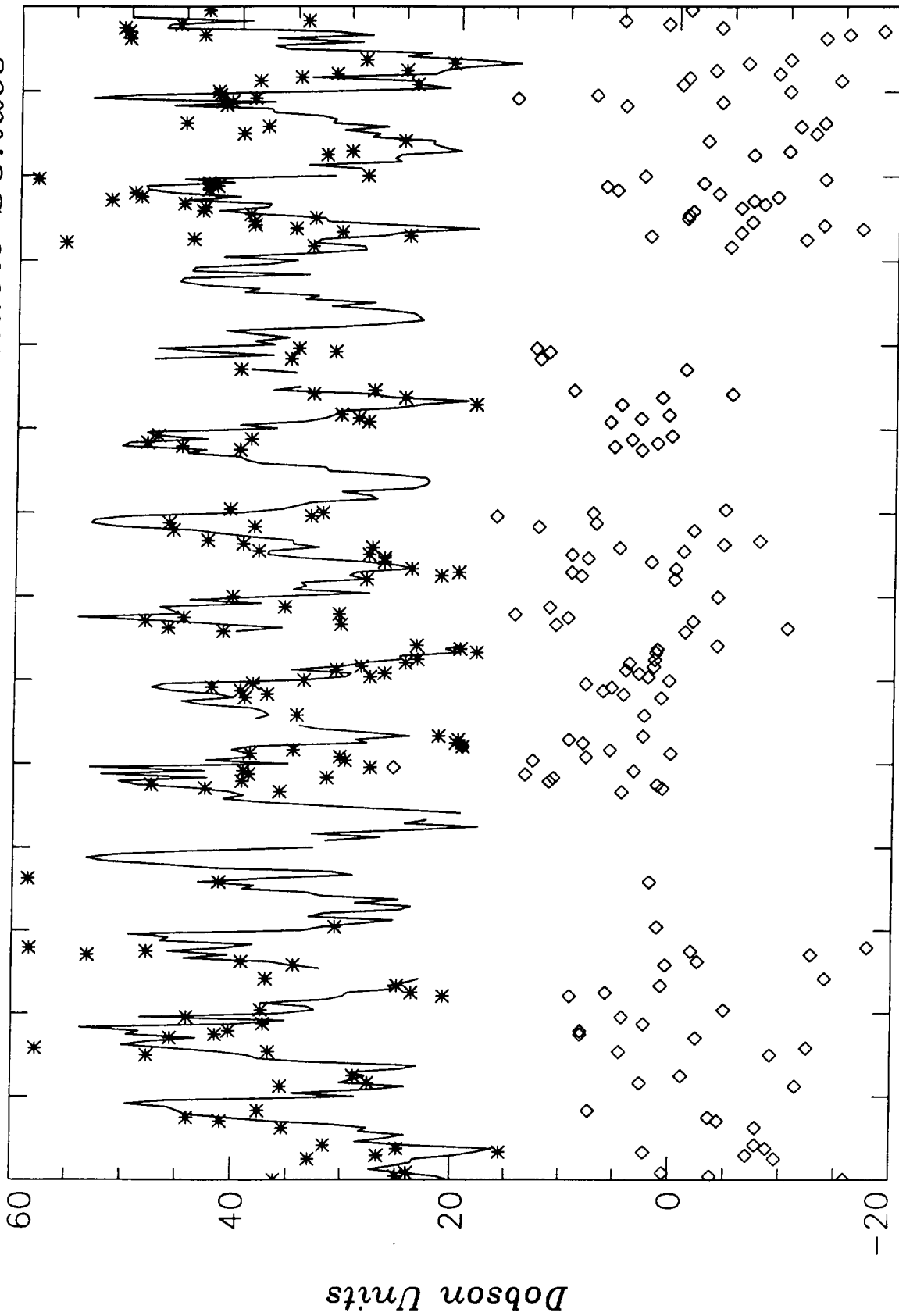


Fig. 8A
Year, January 1979 - December 1992
Mean Deviation = 6.6 D.U.

1979-1992 Natal CCD versus available Sondes

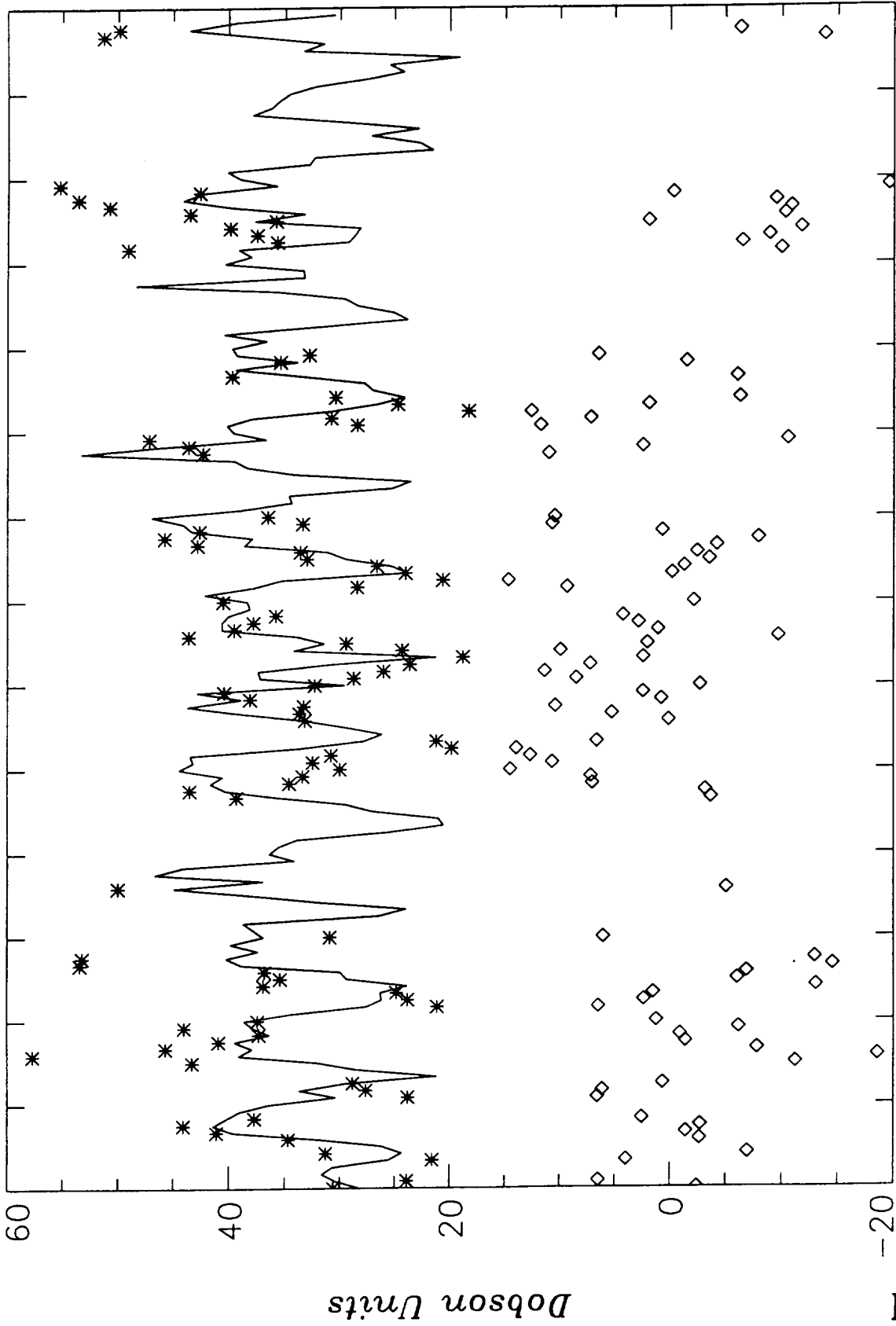
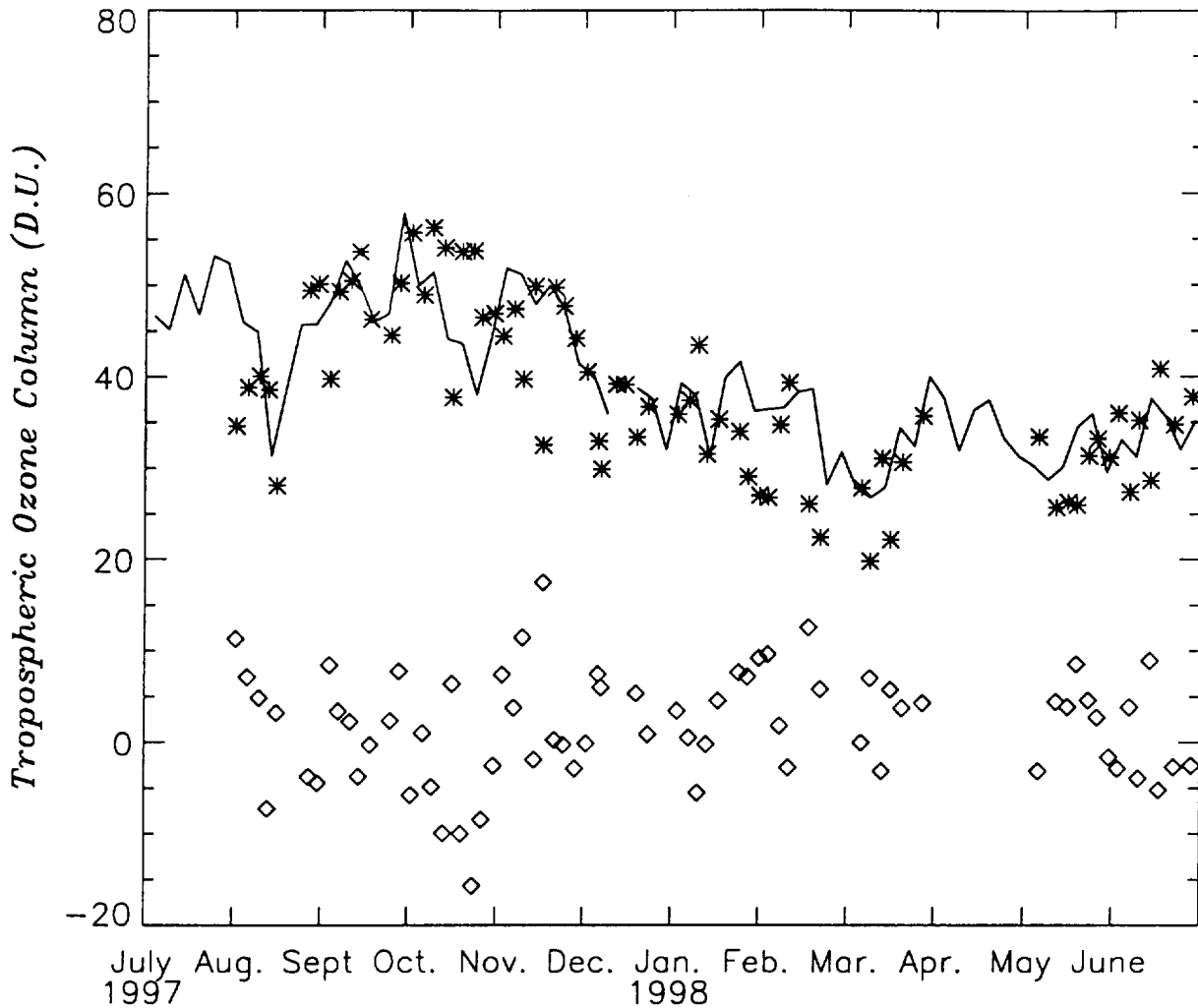


Fig. 8B

Year, January 1979 - December 1992

Mean Deviation = 6.6 D.U.

Tropospheric O₃ from Ascension Island Ozonesonde vs TTO (MR Method), July 1997–June 1998



Line – Tropical Tropospheric Ozone(TTO) from MR Method
Stars – Tropospheric Ozone from Ascension Ozonesondes
Diamonds – Difference (TTO–Ascension)
Mean Deviation = 5.1 D.U.

Fig. 9

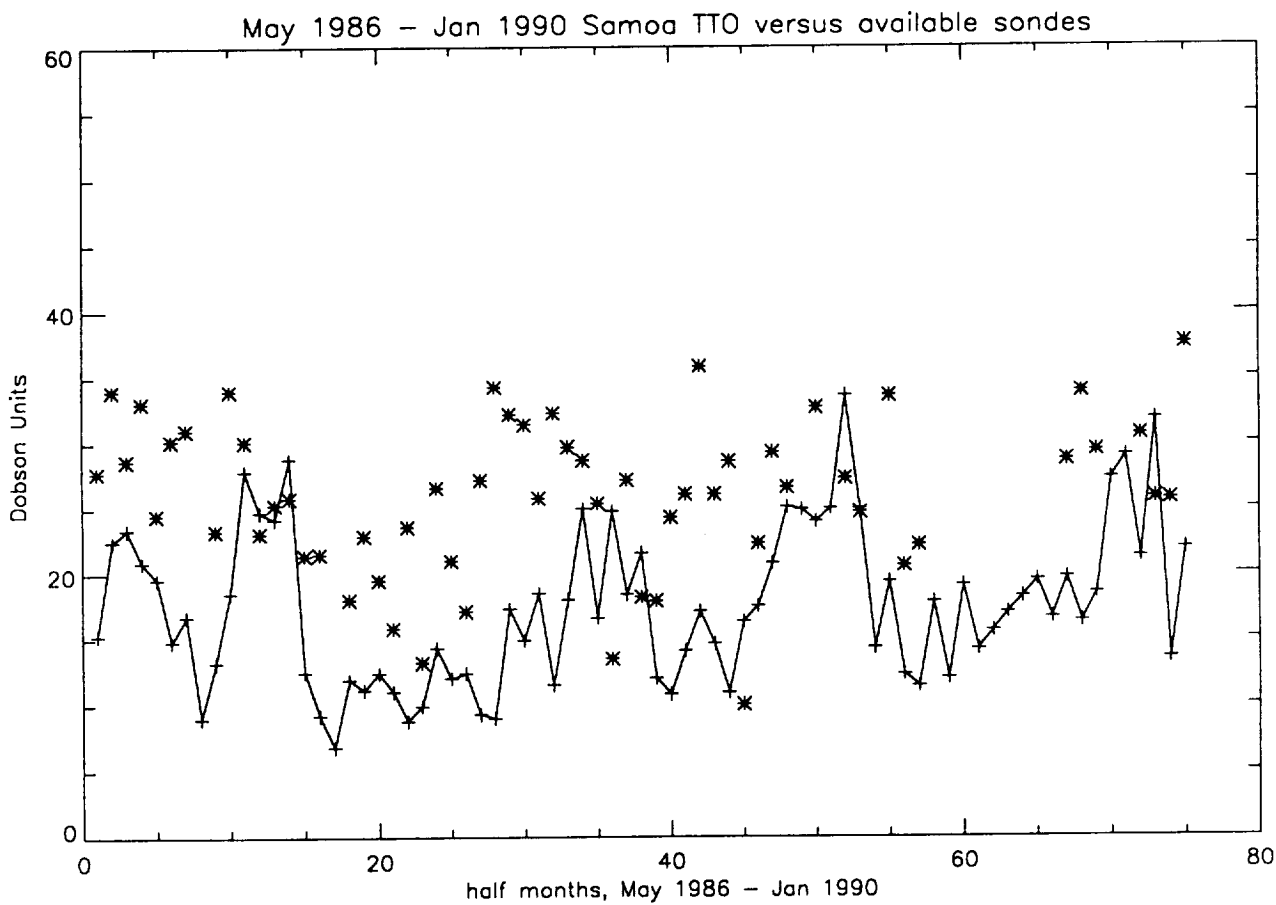


Fig. 10

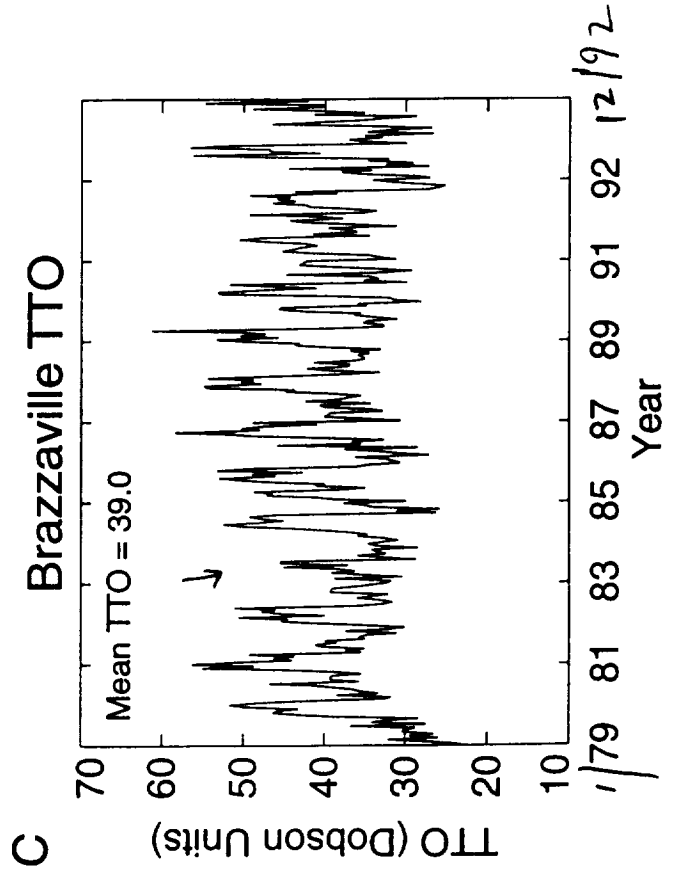
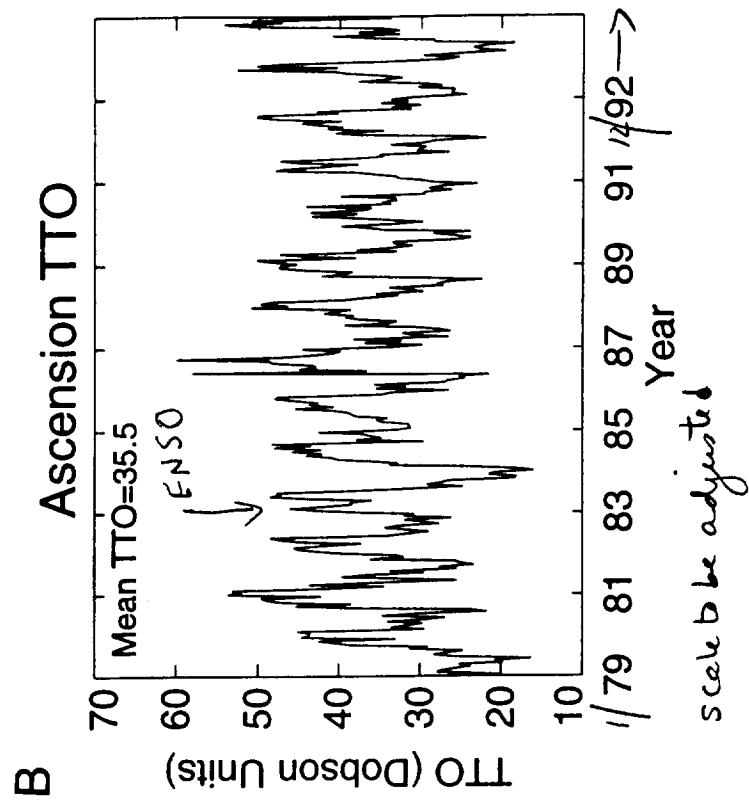
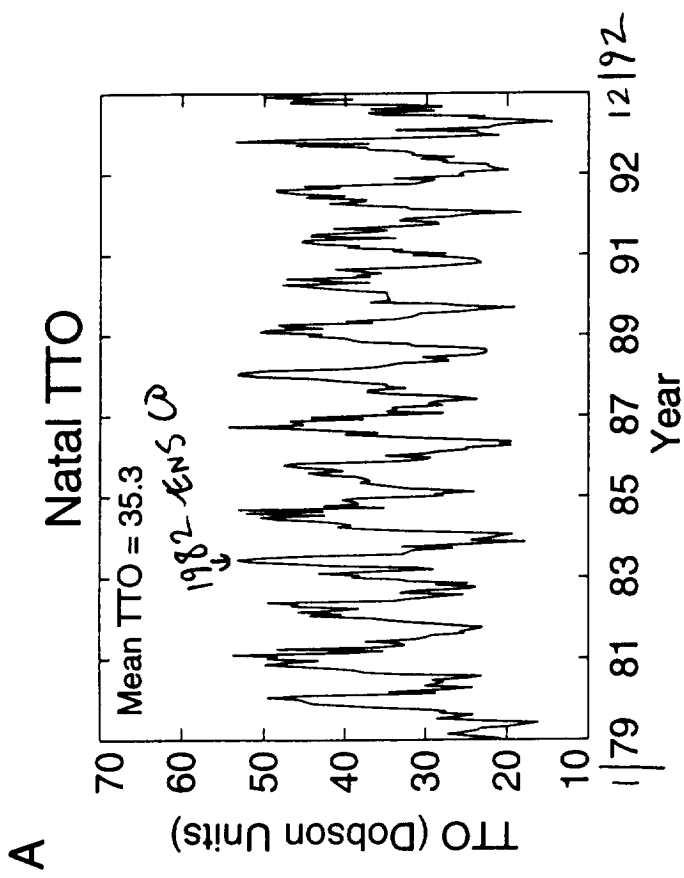


Fig. 11

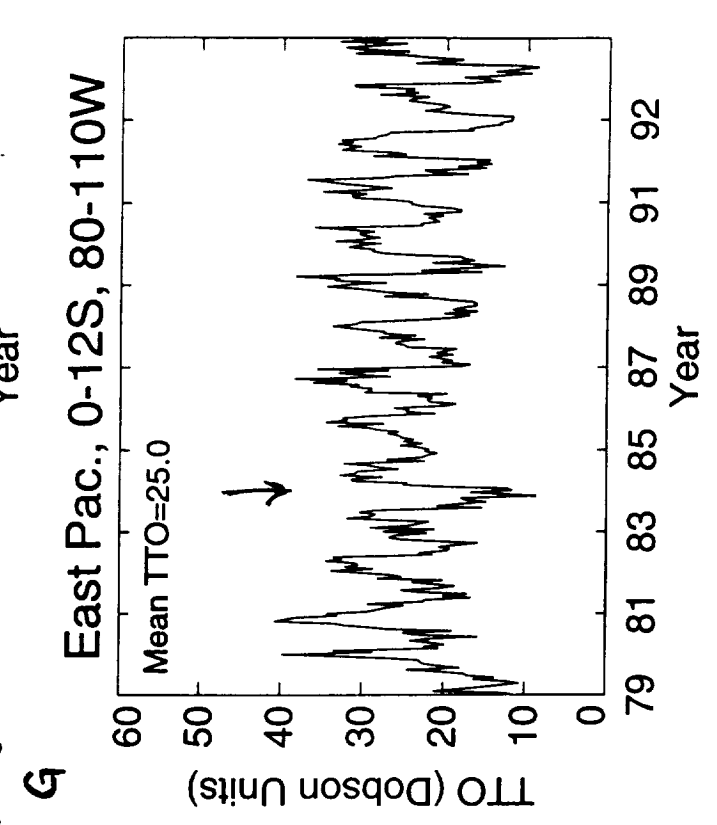
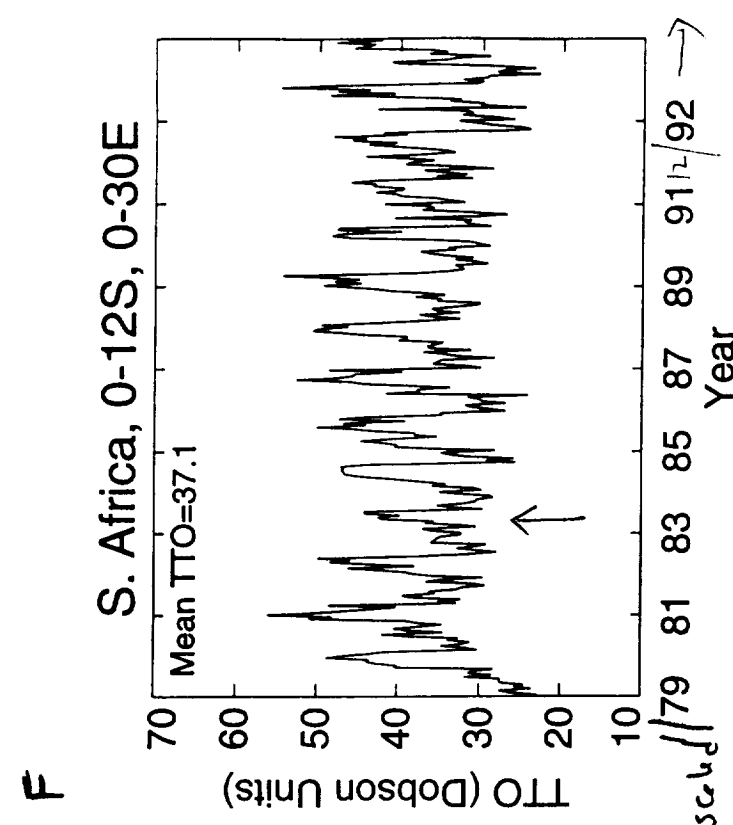
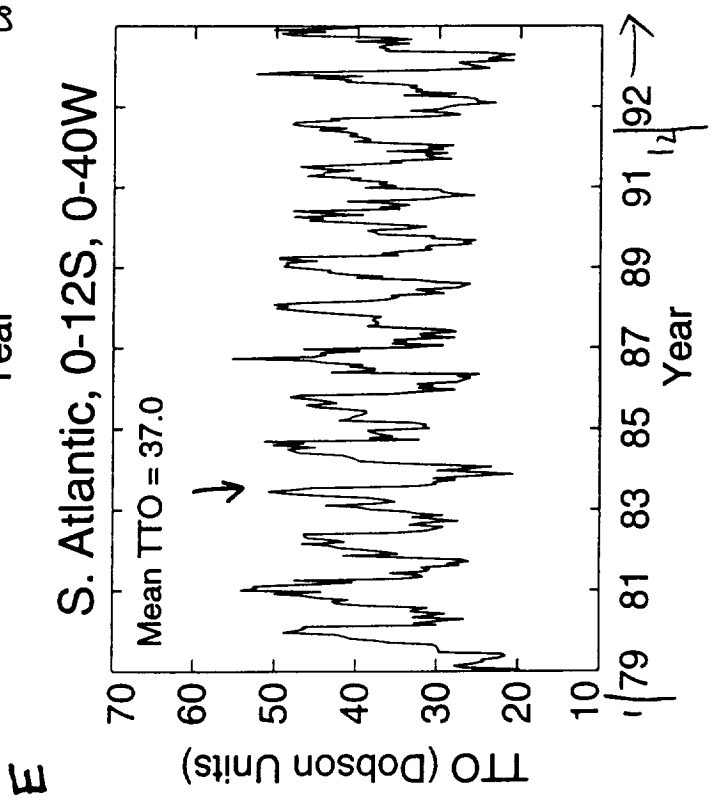
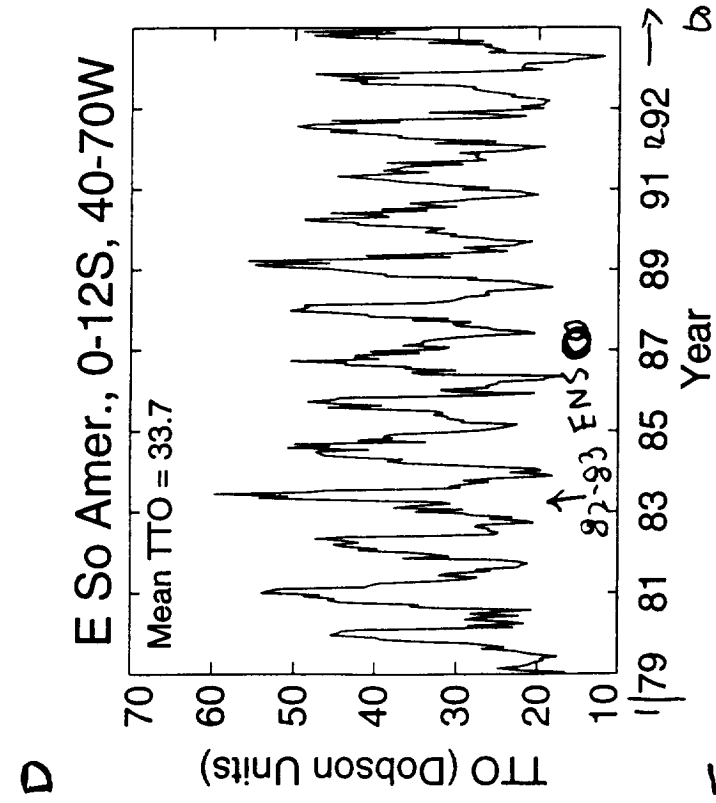


Fig. 11, cont'd.

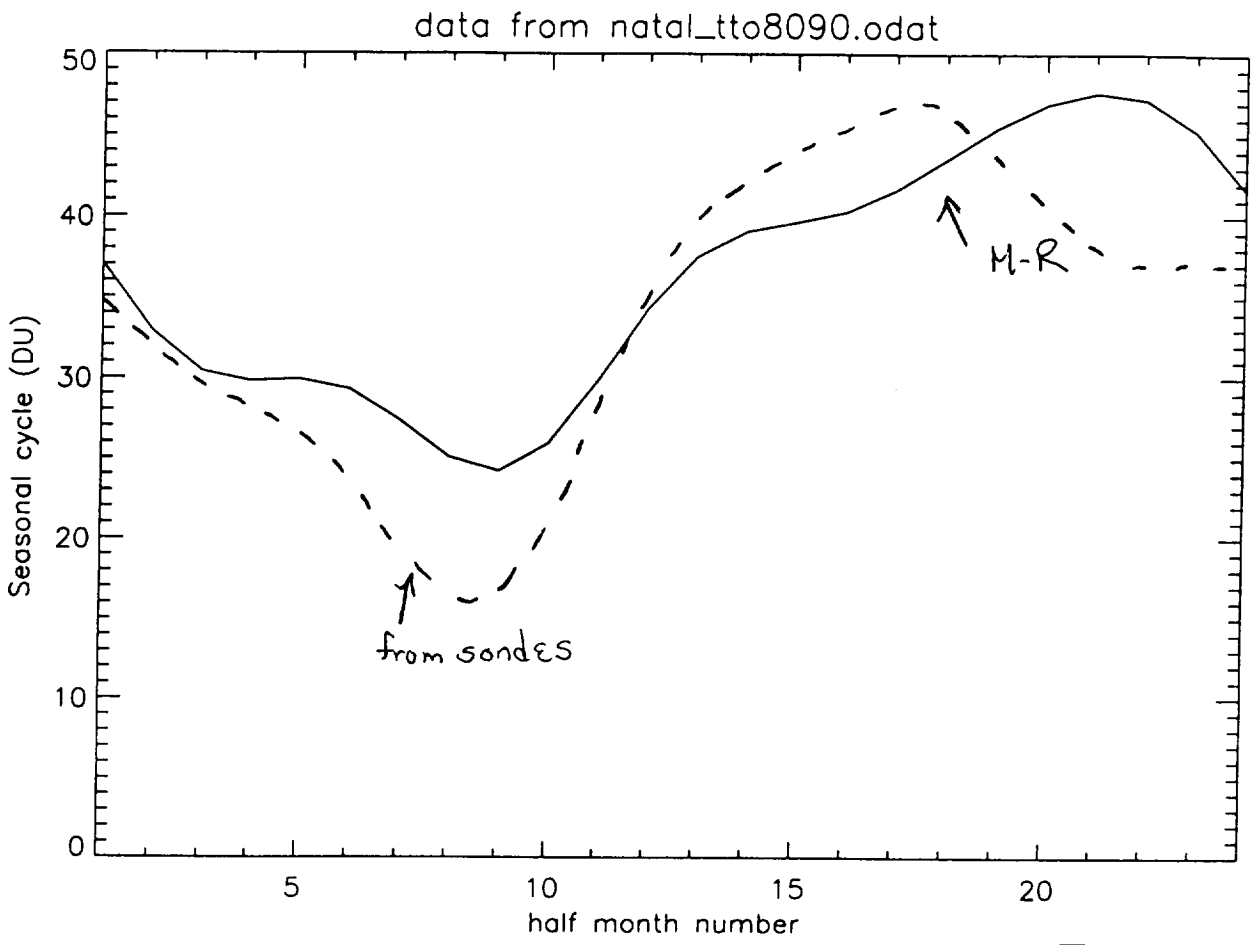


Fig. 12A

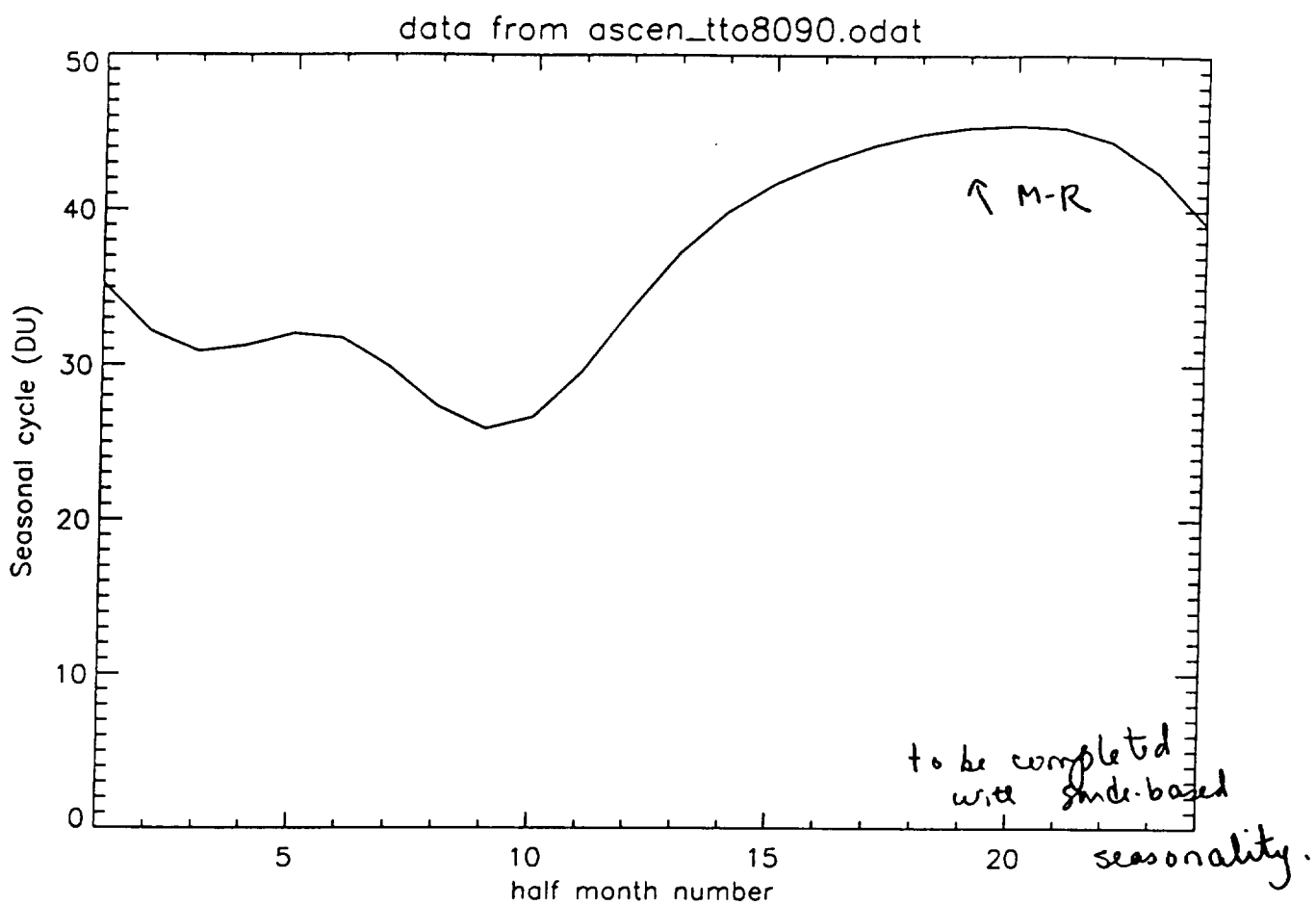


Fig. 12B

FIG 12C - H₂O & sondes
 Annual cycle at
 Brazzaville.

Monthly Ozone Wave Amplitude and Monthly Tropical SST Normalized Anomalies for 1979-1992

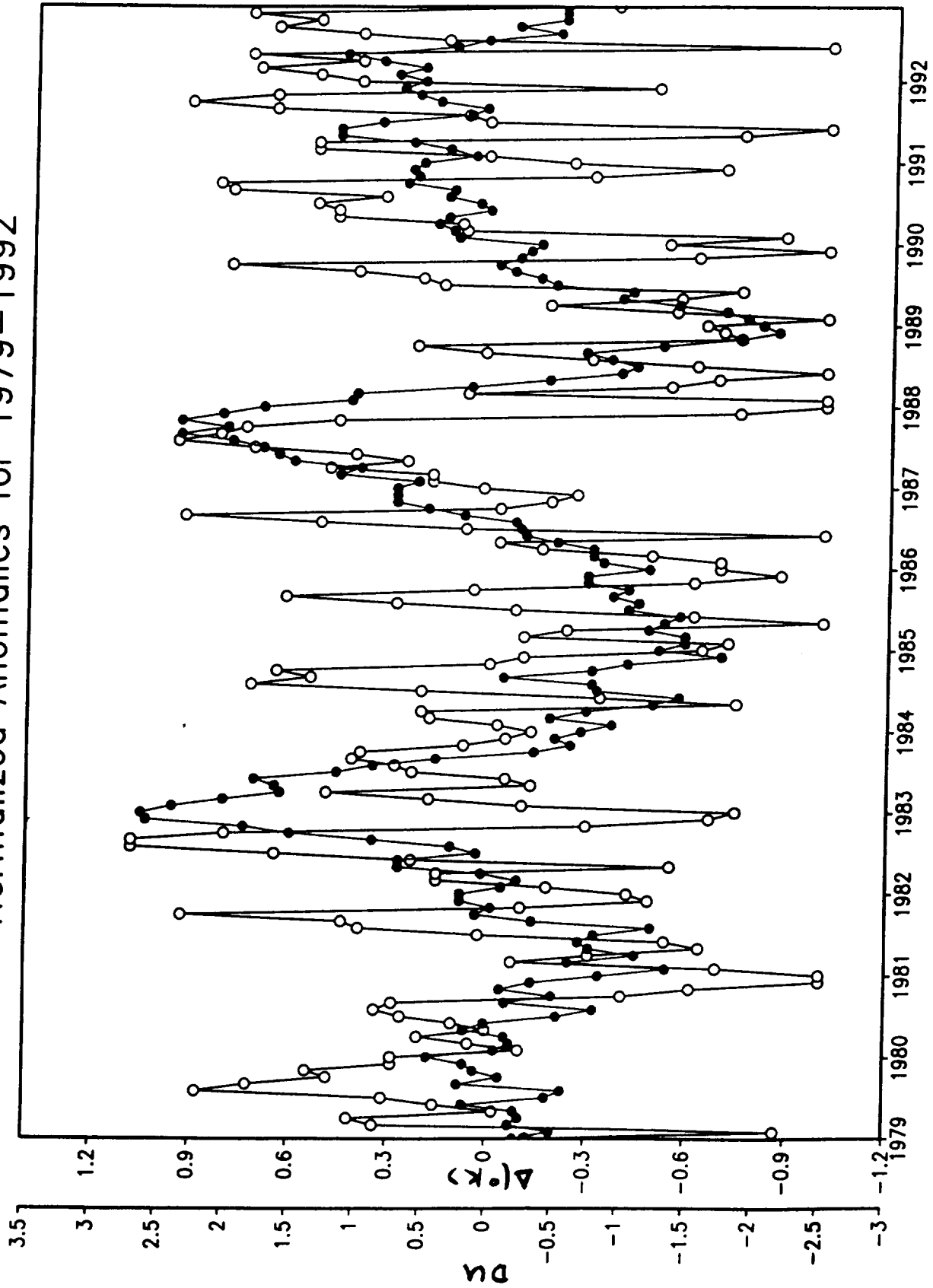


Fig. 13

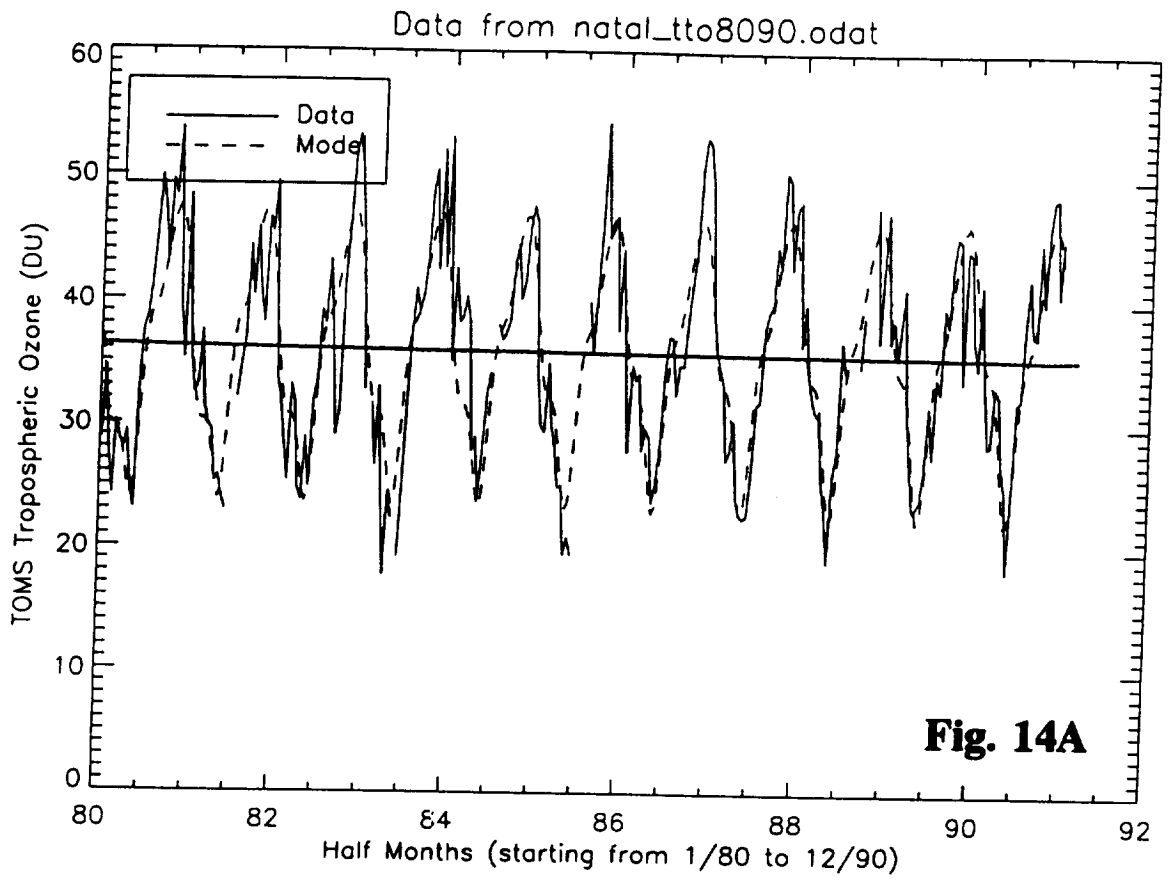


Fig. 14A

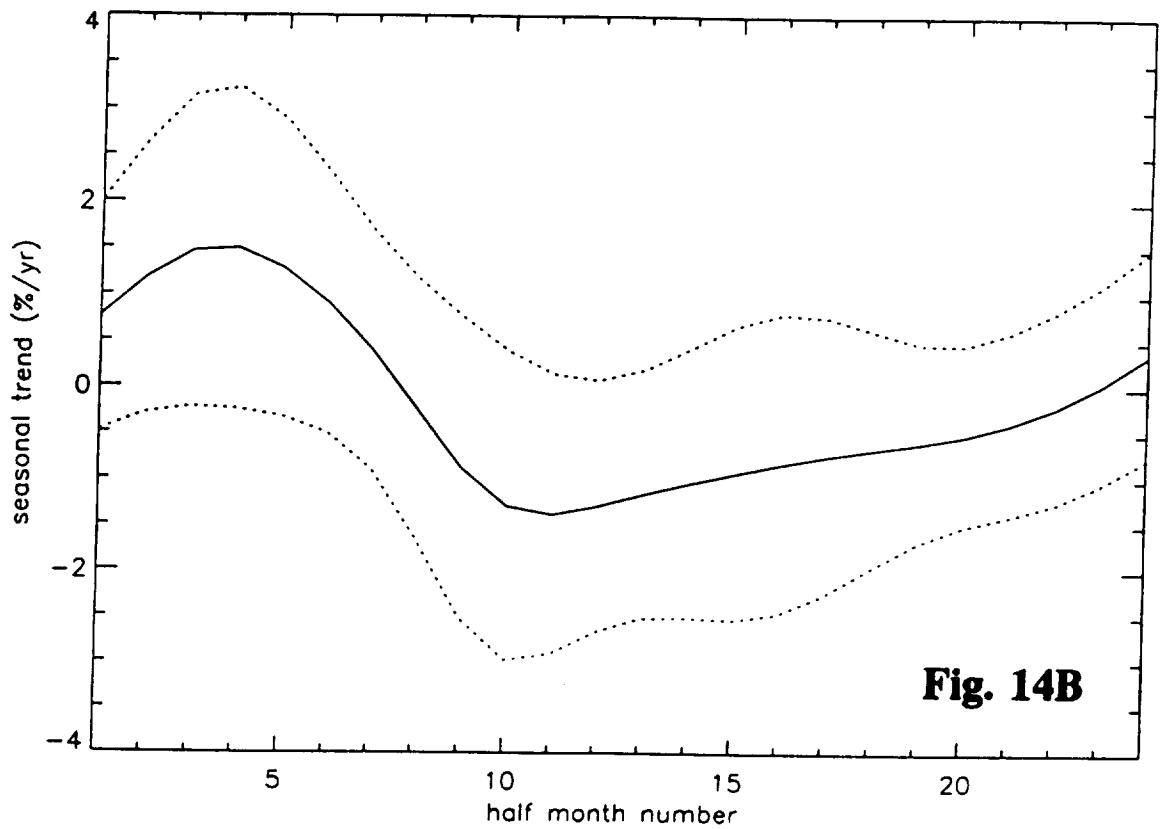


Fig. 14B

