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Periodic Behaviors

in the Observed Vertical Column Abundances

of Atmospheric Hydroxyl

NSF ATM 86-07193

NASA NAGW-989

January 23, 1989

by

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Introduction

The data base for the vertical column abundance of atmospheric hyroxyl (OH) for Fritz Peak Observatory, Colorado (40°N, 105°W), now extends from 1976 through 1988 and is composed of 8849 independent data sets, averaging about 15% uncertainty and 20 minute time resolution each. The dominant solar zenith angle (χ) dependence of the OH abundance is characterized by an empirical curve, N(88), which has been updated from N(82) [Burnett, et al, 1988] to include all valid data from 1980 through 1988. The χ-dependence of the OH abundance has been, to a first order, removed from the data base by a normalization procedure in which each data point is divided by the N(88,AM) value for the corresponding solar zenith angle. The resulting normalized OH values may then examined for other systematic effects, particularly for periodic variations.

Observations have also been made at Boca Raton, Florida (26°N, 80°W) and at Truk, Federated States of Micronesia (7°N, 152°E). These data bases are much less extensive and, as such, are less amenable to analysis for periodic behaviors. Some comparisons with the Colorado data may be made, however.

Characterization of the Fritz Peak Data Base through 1988

The Fritz Peak data base for 1980-1988 is shown in Figure 1 where the vertical OH abundances are plotted against the secant of the solar zenith angle (sec χ). The empirical χ-dependence, N(88), determined by a polynomial least-squares fit, is also shown. Earlier data, 1977-1979, have a quantitatively different χ-dependence, particularly at high sun. The differences in the two time periods are discussed in Burnett, et al [1988].

The monthly averages of the normalized OH abundances through 1988 are shown in Figure 2. The OH values gradually increase during the 1977-80 period; from 1980-1988, the average abundance for any year differs from the overall average by 3% at most. (Excluding the 4 summer months in 1982 affected by the El Chichon event - see Burnett and Burnett [1984].) The annual variation, maximizing in winter, described for the 1977-1983 data [Burnett and Burnett, 1984] persists through 1988. A summertime sub-maximum is also seen to be a regular feature of the normalized OH abundances. Its magnitude is about 10%. The unusually high summer values in 1982 may be described as a combination of this sub-maximum and the OH increase, about 9%, which occurred during the passage of the EL Chichon cloud.

Examination of this extended data base shows that the winter-time normalized OH abundance peaks are amplitude-modulated with a maximum in 1980. The amplitudes decrease to a minimum in 1985, and increase through 1988. The summer sub-maximum is similarly modulated. These modulations are in phase with solar activity as described by sunspot cycles 21-22 and shown in Figure 3.

In order to examine these periodic features in more detail, the long-term behavior of the OH abundance is removed from the normalized data base. The dashed line in Figure 2, which represents the abundance increase through late 1979 and the average PM abundance bias through 1988, is subtracted from the data. The El Chichon enhancement is also removed. The resulting set of residual normalized OH abundances is shown in Figure 4. The basic annual variation is a sinusoid of 22% amplitude maximizing in January and which is

modulated by the solid lines in Figure 3. The superimposed summer sub-

maximum, which is also modulated in phase with the current solar cycle, is a

squared sinusoid maximizing in July and extending through May and August. Its

maximum amplitude is 19% above the corresponding minimum in the annual curve.

The rms variation of the OH residuals about the composite curve shown in

Figure 4 is 50% less than the rms variation of the OH abundances about the

dashed line in Figure 2. This corresponds to a correlation coefficient

between the data and the Figure 4 curve which is significant at the 99% level.

The data in Figure 4 have also been Fourier-analyzed. The resulting

power spectrum is shown in Figure 5. The annual and semi-annual features are

prominent. A small biennial component not evident in the data base also

appears, but not at a significant level. There are no other significant

periodicities.

The seasonal behavior of the AM-PM asymmetry in OH has been reported

earlier [Burnett and Burnett, 1984; Burnett, et al, 1988]. The nature of the

asymmetry is illustrated in Figure 6 where the average values of OH abundance

for spring and fall for 1980-1988 are plotted as a function of solar zenith

angle. Figure 7 shows the covariation of the OH diurnal asymmetry with total

ozone for 1977-1988. It is seen that the amplitudes of both quantities remain

relatively constant through 1983 and then decrease to a minimum in 1984-1986

and subsequently increase in 1987-1988. This modulation is similar in both

relative amplitude and phase to the modulation of the annual cycle in the

normalized OH abundances.

Other OH Measurements

Observations have been reported from Florida for 1980-1986, but the data base has several long gaps so that analysis for periodicities is inconclusive. However, the even-year winters of 1980, 1984, and 1986 (there was no data for 1982) show excursions which decrease in amplitude at approximately the same rate as the Colorado winter amplitudes for the same period. (See Burnett, et al [1988], Figure 3b.)

The observations begun at Truk, FSM, in January of 1987 are to be continued through early 1989 [Minschwaner, et al, 1987]. While variations of the order of 50% have been observed during 1987-1988, the periodic nature of these variations, if any, is as yet unclear.

Discussion

General Remarks

The normalized abundances shown in Figure 2 are not entirely free from seasonal bias. For example, observations for sec χ near 1 (overhead sun) occur only in the summer; so the summertime normalized abundances may show a diminished departure from the annual average. Similarly, atmospheric observing conditions tend to introduce a seasonal bias at large zenith angles. Nevertheless, we find that the variations described by the curve in Figure 4 do not change significantly when a restricted data base, including only zenith angles for which all seasons are represented, is analyzed.

The calculation of OH abundances from the absorption data assumes an effective temperature of 250°K for the entire column. A small seasonal error resulting in the underestimation of the OH abundance in winter is thus

introduced. A 15°K difference between effective summer and winter temperature corresponds to a seasonal error of about 3% amplitude.

Since these are column measurements and are thus insensitive to altitude, the observed OH behaviors might be due to variations in tropospheric OH. The tropospheric contribution to the OH column should vary with seasonal differences in solar zenith angle, water vapor, etc. The effect of the expected high-sun increase in tropospheric OH in summer will be minimized by our normalization procedure. It is also not clear how the tropospheric column would contribute an annual winter maximum, since tropospheric OH is expected to maximize in the summer [G.H. Mount, personal communication, 1988].

In any case, the seasonal difference attributable to tropospheric OH must be less than the average tropospheric OH column abundance which is estimated to be about $1 \times 10^{12}/cm^2$ [WMO, 1985], or about 1-2% of the total observed OH column. Recent measurements by Platt, et al [1988], give an average mid-day, summer, mid-latitude concentration of $6 \times 10^6/cm^3$. If a constant vertical distribution is assumed, this corresponds to a tropospheric OH column of about $3 \times 10^{12}/cm^2$ or 3% of the observed summer mid-day OH column abundance.

The dominant role of solar UV flux in the photochemistry of OH is evident in the empirical dependence on sec as shown in Figure 1. Much of the diurnal and seasonal change in the OH column abundances may be characterized by this single variable. The amplitude of the annual variability in OH abundances due to changes in solar flux because of the eccentricity of the earth's orbit has been calculated and is about 2% with a winter maximum [Burnett, 1986]. This gives a combined contribution from annual solar flux and temperature variability of about 5% amplitude.

The direct effect of solar UV variability on OH abundance during a solar cycle has also been examined [Burnett & Burnett, 1982]. If changes in solar UV flux were the only factor involved, OH column abundances would be expected to increase modestly between solar minimum and solar maximum. OH abundances were observed to increase by 50% coincident with the approach to solar maximum during 1977-1980, whereas subsequent OH abundance levels have remained essentially constant through the declining phase of the past solar cycle (1980-1986) and the initial increasing phase of the current cycle (1986-1988). In contrast to the long-term OH abundance behavior, observed OH periodicities show a clear amplitude modulation which is in phase with recent solar activity (see Figures 3 and 4). Since a direct photochemical response to UV flux changes during the solar cycle does not adequately describe the observed OH behaviors, the possible role of transport must be considered.

Transport and OH Variation

The direct transport of odd-hydrogen radicals would not be expected to give rise to the observed periodicities because of the very short chemical lifetimes of these species, on the order of minutes in the stratosphere and a few hours in the mesosphere [Shimazaki, 1985]. Long term transport-related changes in source or sink species would give rise to equivalent changes in average odd-hydrogen levels which, since 1980 at least, have not been observed. Hence, those transport mechanisms which may give rise to periodicities in either the source and sink species for odd-hydrogen or in their spatial gradients should be considered as possible factors influencing the observed OH behavior.

There are annual cycles of 1) middle atmospheric zonal winds with winter

westerlies and summer easterlies and 2) mesospheric cross-equatorial

circulation from the summer hemisphere to the winter hemisphere [Andrews, et

al, 1987]. A tropical quasi-biennial oscillation (QBO) in stratospheric

winds, primarily within 20 degrees of the equator, but with some effects at

higher latitudes, is well-established [Reed, 1964; Gray and Pyle, 1988].

A semi-annual oscillation (SAO) in zonal winds also occurs. The SAO has

maximum amplitude at the solstices at the stratopause and a maximum amplitude

at the equinoxes at the mesopause. The SAO has minimum amplitude at 40°

latitude, about 50% of its tropical value, and shows little latitudinal phase

shift [Andrews, et al, 1987].

Both the QBO and SAO have been tentatively correlated with solar

activity. Labitzke [1987] has reported a covariation of middle atmospheric

temperatures with solar activity when the temperature data base is partitioned

according to the phase of the tropical QBO. An amplitude modulation of about

50% in the semi-annual zonal winds in phase with solar activity has been

reported by Nastrom and Belmont [1980]. That study failed to detect a similar

modulation of the annual winds, however. No physical mechanism has yet been

identified for either of the above apparent correlations with solar activity.

The QBO, annual, and SAO variations in middle atmospheric transport have

well-documented effects on the total abundance and vertical distribution of

ozone, especially in the stratosphere where ozone is a reliable dynamic tracer

(see, for example, Wilcox, et al [1977] and Andrews, et al [1987]). The

effect of transport on the distribution of middle-atmospheric water vapor is

less well-known due to the difficulties involved in obtaining frequent and reliable observations. Transport effects for most minor constituents in the mesosphere are complicated by short photochemical lifetimes. The transport of odd- hydrogen free radicals should not be involved at any altitude since their chemical lifetimes are always short compared to dynamic lifetimes.

The curve shown in Figure 4 involves a sinusoidal annual cycle added to a summer sub-maximum which is represented by a squared sinusoid. This suggests that separate mechanisms, possibly at different altitudes, are responsible for the net seasonal behavior of OH. It may also be noted that the periodic behavior of OH in 1977-80 has been assumed to be independent of the high-sun enhanced abundance increase observed during that period. Both periodicities contain a modulation which is proportional to solar activity as measured by the sunspot number index. The AM-PM asymmetry (Figure 7) and the Florida abundances also show modulations which are similar in relative amplitude and in phase to those described above. These effects could imply a general modulation of atmospheric circulation, perhaps due to solar activity.

On the other hand, the data shown in Figure 4 could be represented, but not as well, by a constant 12-month period sinusoid added to a solar activity-modulated 6-month period sinusoid. This situation would be consistent with other observations which find solar activity correlated with the SAO but not with the annual circulation [Nastrom and Belmont, 1980]. The results of the Fourier-analysis shown in Figure 3 are not inconsistent with either of the above possibilities. In the absence of a firm theoretical framework, any number of possible combinations of periodic and long-term variation could be invoked which would "fit" the observed OH behavior.

Summary

We have identified clear periodicities in the seasonal variation of the
Colorado OH data base. The winter maximum and summer sub-maximum are both
modulated in phase with the 11-year cycle in solar activity. The Colorado
AM-PM asymmetry and the Florida data base show similar modulations. An
11-year modulation of the average Colorado OH abundances is not, however,
observed. We suggest that the observed periodicities and their 11-year
modulation result from variations in the transport of the longer-lived source
and sink constituents for the odd-hydrogen free radicals. At this point
neither the altitude nor the particular transport process(es) involved can be
positively identified, nor can specific physical mechanism which produced the
modulations and their correlation with solar activity.

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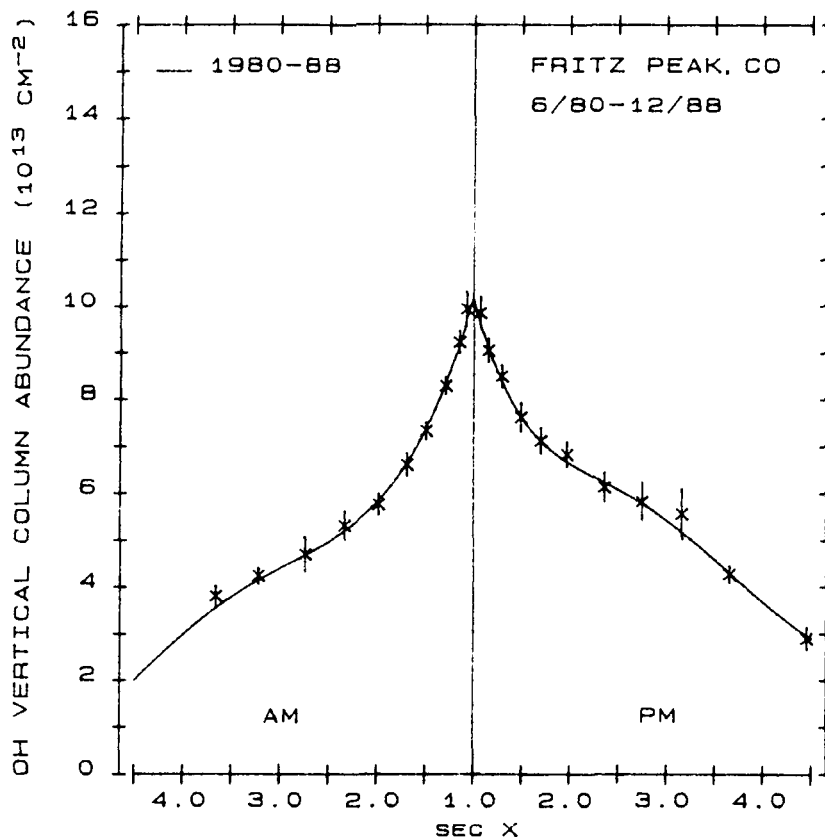
Figure 1 Average solar zenith angle dependence of the
vertical column abundance of atmospheric OH at Fritz Peak,
Colorado (40°N, 105°W) 1980-1988. The average OH vertical
column abundances, Nv, are calculated from 8849 independent
absorption measurements and are displayed as a function of
the secant of the solar zenith angle, χ. The empirical
curve shown is a non-linear least squares fit to the data
and has a standard error of fit of $0.12 \times 10^{13}/cm^2$. Error
bars represent the standard error of the mean, $\sigma/\sqrt{N}$.

FRITZ PEAK, COLORADO
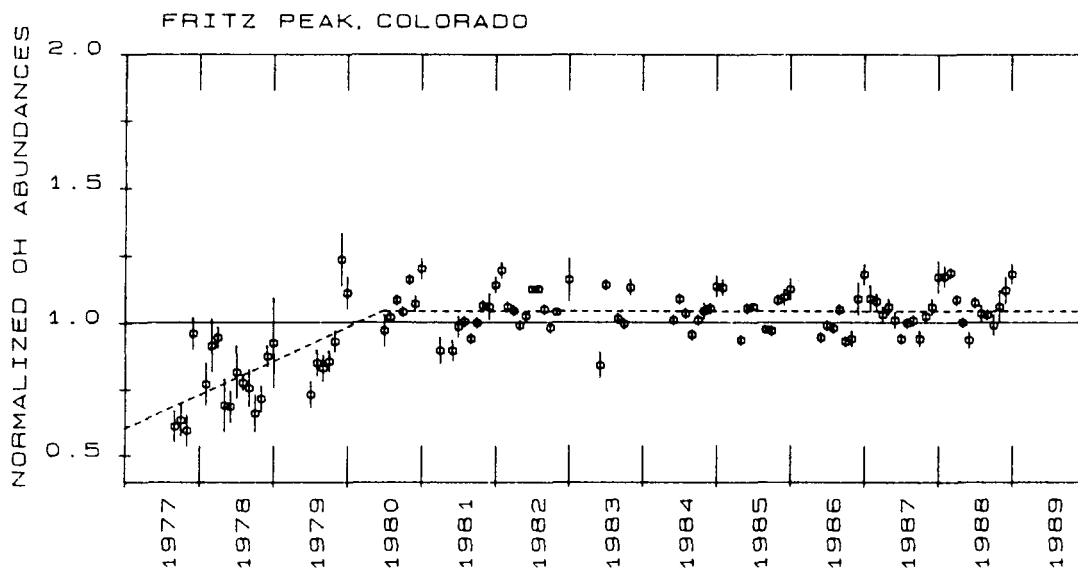
Figure 2   Monthly averages of normalized OH abundances for the
period 1977-1988.   OH abundances have been normalized by dividing
by the N(88,AM) empirical curve from Figure 1.   The dashed line
shows the approximately linear abundance increase observed from
1977 through 1979 and the subsequent constant average OH abundance.
The horizontal portion of the dotted line is off-set from zero due
to the persistant OH abundance bias toward PM values.   The rms
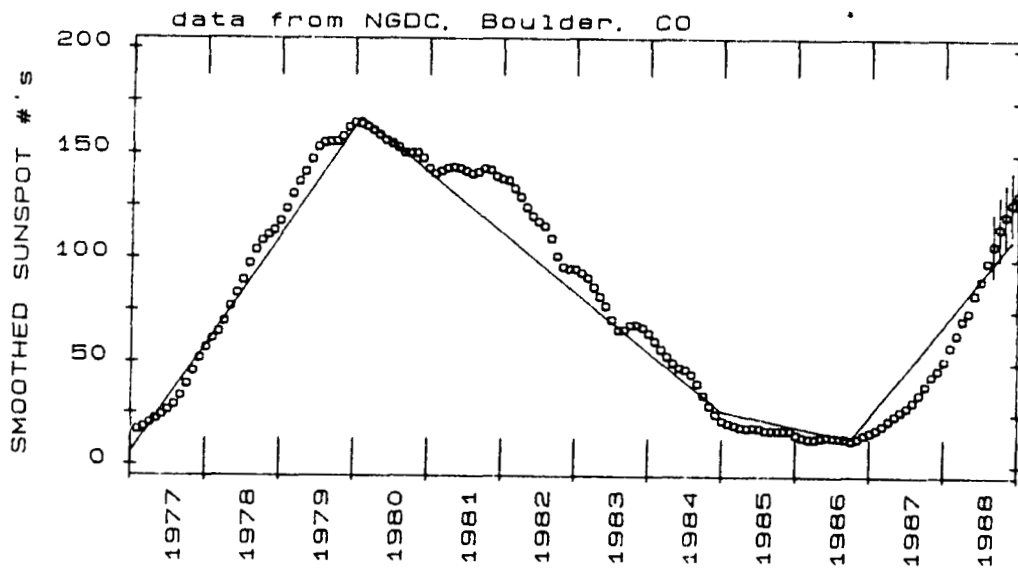deviation of normalized OH abundance about the dashed line is 0.085.

Figure 3   Smoothed sunspot numbers for the 1977-1988 period.   The
circles represent solar activity as defined by the sunspot number
index [NGDC, 1988].   Solar maximum of cycle 21 occurred in December
1979 and the onset of solar cycle 22 occurred in September 1986.
The solid lines are proportional to the lines used to modulate the
amplitude of the 12-month cycle observed in the normalized OH data
base.
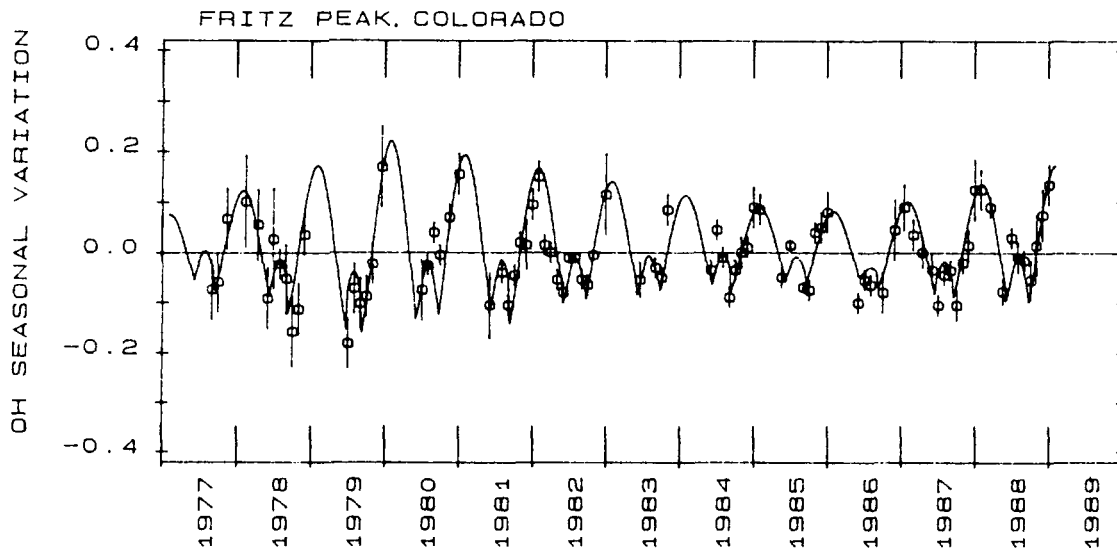
FRITZ PEAK, COLORADO

Figure 4   Modulated seasonal variations of normalized OH column
abundances at Fritz Peak, CO.   The circles represent residuals
from the normalized OH abundances when the long-term average OH
behavior (the dashed line shown in Figure 2) is subtracted.   The
curve represents the sum of a) an annual sinusoid of amplitude
0.22, maximizing in January and amplitude-modulated proportional
to solar activity (solid line in Figure 3); and b) a modulated,
squared sinusoid extending from May through August, maximizing
in July, and having a maximum amplitude of 0.19 above the
corresponding July minimum in the annual curve.   The rms deviation
of the OH residuals from the curve shown is 0.041.   The minimum
statistically significant rms deviation of a monthly mean for a
data base of 8850 observations with an average experimental
uncertainty per observation of 15% is 0.016.
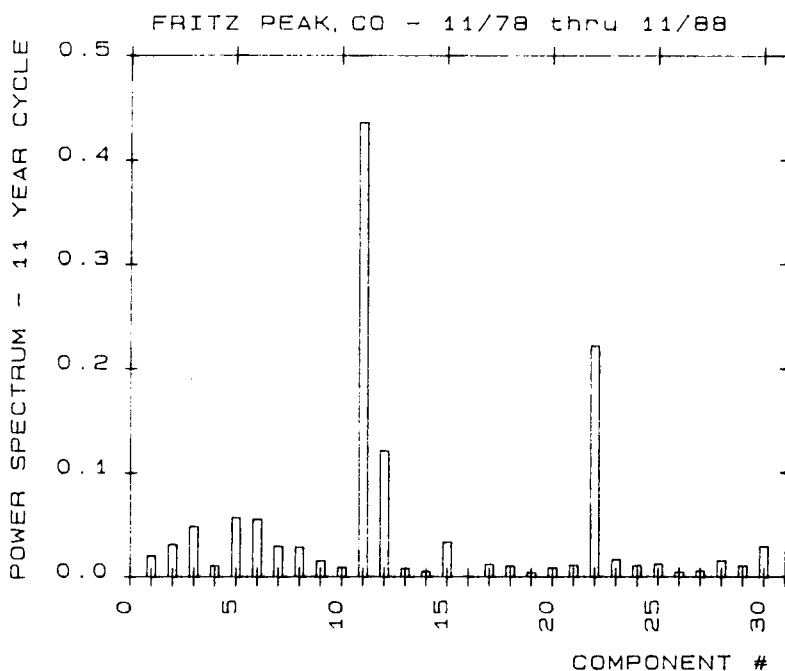
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FRITZ PEAK, CO - 11/78 thru 11/88

Figure 5 Discrete power spectrum for the normalized
OH abundances for 1977-1988. The power spectrum from a
full Fourier analysis of the normalized OH abundances is
shown. Components 11, 12, and 22 are significant.
Components 11 and 12 have a frequency corresponding to
an annual cycle in the data base. The phase of these
components is consistent with the observed January maximum.
Component 22 is consistent with either a true semi-annual
cycle, an annual cycle out of phase with the primary annual
periodicity, or an annually-occurring sub-maximum. Its
phase shows a July maximum. Components 5 and 6 correspond
to a biennial cycle and are of marginal significance; such
a periodicity is not apparent in the Colorado data base.

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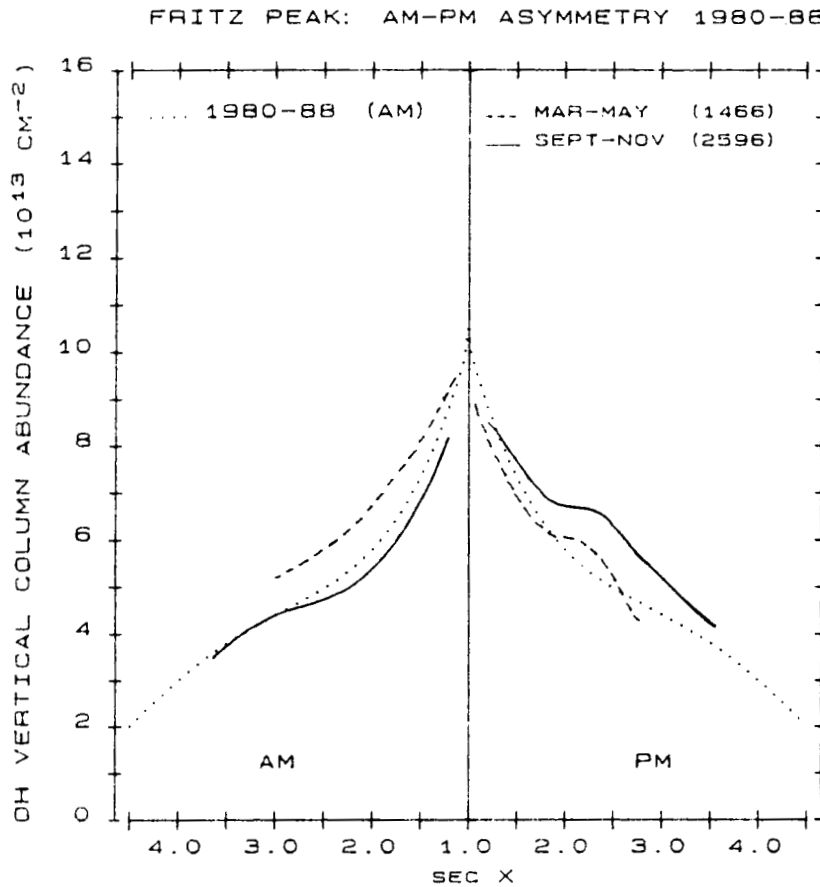OF POOR QUALITY

FRITZ PEAK: AM-PM ASYMMETRY 1980-88

Figure 6   Average spring and fall asymmetry in OH column
abundances for 1980-1988. The empirical curve N(88,AM)
is shown on both the morning and afternoon side of the
graph in order to provide a symmetric reference. The
average spring curve (March through May) is obtained from
1466 data sets and is represented by the dashed lines.
The average fall curve (September through November) is
obtained from 2596 data sets and is represented by the
solid lines. The curves are oppositely asymmetric with
respect to morning and afternoon. The earlier, 1977-1979,
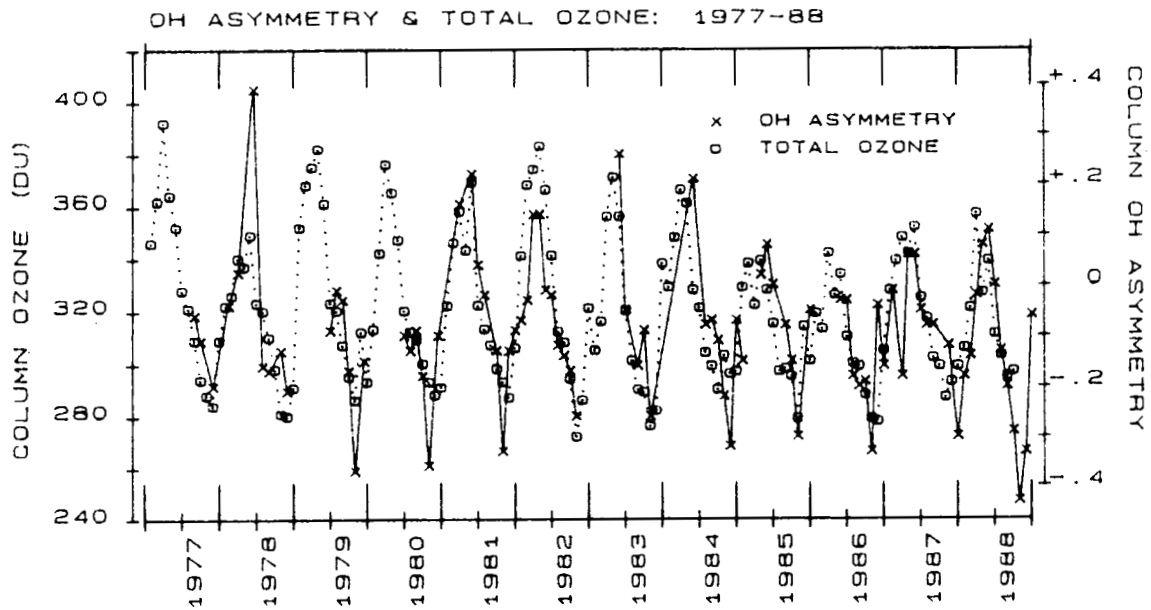OH data show a similar asymmetry.

Figure 7   Seasonally varying OH asymmetry and total ozone abundances for Colorado, 1977-1988. The crosses represent the monthly mean ozone column abundances for Boulder, CO [GMCC data courtesy of R. Grass]. The circles represent the monthly values of the OH asymmetry (see Burnett, et al, 1988 for details). In addition to the annual covariance of the two data sets, each shows an amplitude modulation of approximately 50% beginning in 1983, minimizing in 1986, and increasing through 1988.