

# MONTHLY WEATHER REVIEW

Editor, JAMES E. CASKEY, Jr.

Volume 78  
Number 8

AUGUST 1950

Closed October 5, 1950  
Issued November 15, 1950

## ATMOSPHERIC OZONE AT WASHINGTON, D. C.

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[Manuscript received July 7, 1950]

### ABSTRACT

Total atmospheric ozone measurements at Washington, D. C., for the period February 18, 1948, through March 9, 1950, are tabulated and described. The usual annual variation is found. The average amount of ozone observed for the 2 years is somewhat low for 39° N. latitude.

Ozone departures from normal in relation to surface low and high pressure systems are analyzed. The distribution around Lows agrees quite well with most other analyses. Around Highs the distribution is poorly defined, and is considerably different from results at other geographical areas.

Correlations of ozone with temperatures at the 500-mb. and 100-mb. levels are examined. The correlation coefficients vary regularly from month to month, and there is a suggestion that their highest absolute values occur in some spring and winter months, and that the lowest absolute values occur in summer and early autumn. Two possible explanations of this variation are discussed, namely, (1) the annual cycle of the variability of ozone and of temperature, and (2) the annual change of the winds above 18 km.

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### INTRODUCTION

Relatively few comprehensive analyses of total atmospheric ozone measurements have been made in North America, although in recent years a few studies have been made in the United States [1, 2]. Thanks to New York University for a loan of their Dobson ozone spectrophotometer (No. 19), the United States Weather Bureau has been making regular ozone measurements in Washington, D. C. (lat. 38°54' N., long. 77°3' W.) whenever practicable since February 1948. The results of these measurements will be presented here.

### METHOD OF OBSERVATION

#### THE INSTRUMENT

The Dobson spectrophotometer has already been described [3] and will be mentioned only briefly. In order to increase the sensitivity of the instrument so that measurements of the vertical distribution of ozone could be made, the electrical portions of the instrument were completely redesigned [4] by the Naval Research Laboratory, and a photomultiplier was substituted for the original photocell,<sup>1</sup>

<sup>1</sup> The following is quoted (slightly modified as shown by brackets) from a letter by Dr. Dobson dated August 15, 1950, and received after this manuscript had been submitted for publication:

"Since multipliers have become commercially available . . . , the photocell has [generally] been replaced by a multiplier. It is understood that this has also been done for most of the older instruments in Europe and that all new instruments are fitted with multipliers."

but the optical portion of the instrument was not changed.

The instrument operates fundamentally by comparing the intensities of two wavelengths of *direct* sunlight, namely  $I$ =intensity at wavelength 3110A and  $I'$ =intensity at wavelength 3300A;  $I$  is strongly decreased through absorption by ozone, while  $I'$  is only slightly affected by ozone. A dial is attached to an optical wedge which varies  $I'$  until its electrical response equals that due to  $I$ , at which time a galvanometer indicates a zero reading. The instrument is calibrated so that, except for constants which cancel out in the end, the dial reading gives  $L=\log_{10}(I/I')$ . By taking dial readings over a large range of solar altitudes, a graphical plot of  $L$  against  $\mu$  can be made, where  $\mu$  is the optical path through the ozone layer, the vertical path being taken as unity. Once an "appropriate" height of the ozone "layer" is chosen (we used 20 km.),  $\mu$  depends on the solar altitude. On days when ozone changes very little, this graph of  $L$  vs.  $\mu$  will be a straight line, and extrapolation to  $\mu=0$  gives  $L_0=\log_{10}(I_0/I'_0)$ , the value of  $L$  "outside" the earth's atmosphere. Except for instrumental changes,  $L_0$  is assumed to remain constant with time. Moreover, instrumental changes which affect  $L_0$  can be checked periodically with lamps which have been supplied with the instrument.

#### CALCULATION

Having determined  $L_0$  and  $L$ , the total amount of ozone,  $x$ , in a vertical column above the observer [5] is given by

$$x = \frac{L_0 - L}{1.17\mu} - 0.085 \quad (1)$$

The unit of  $x$  (or  $O_2$ ) is the centimeter; i. e., the height which a column of ozone of unit cross section would occupy if it were reduced to standard temperature and pressure. The 0.085 is a factor which is supposed to take account of differential scattering of energy in the two wavelengths by air molecules and by dust. Actually the instrument contains provision for examining the intensity of a third wavelength,  $I''$  at 4450A, which together with  $I'$  should yield information about the change in the dust scattering from observation to observation. But dust scattering is improperly understood at present, and several different formulae have been used from time to time to account for it. However, all the Washington data presented here have been calculated from equation (1).

Equation (1) applies only to observations made by utilizing direct sunlight. With reduced accuracy, the instrument can also be used in the daytime during overcast conditions, when the sun is not visible, but only direct solar measurements have been included in this paper. During some of our observations the sky was indeed overcast, but the sun's disk was visible and the energy received by the instrument was sufficiently intense to make a dial reading possible.

#### ACCURACY OF THE OBSERVATIONS

The accuracy of an ozone observation depends on (1) the accuracy in setting the dial (or wedge), (2) the accuracy of  $L_0$  and (3) the effect of dust and other atmospheric contaminants.

#### DIAL SETTING

Dobson [3] found that the effect of dial setting introduced a negligible error, and this appears to be true also of our observations. Our practice is to make three dial readings in the space of about one minute, the average of the three readings comprising a single observation. If we assume that the amount of ozone does not change appreciably in one minute, the greatest difference between any two of the three dial readings indicates the error due to dial setting. On the average, we found the difference to correspond to about 0.002 cm. of ozone.

#### VALUE OF $L_0$

At irregular intervals, values of  $L_0$  were determined by making series of observations during one day, and also by checking with the lamps. We found that sometimes  $L_0$  did change. In September 1948, following a period during which the instrument had been transported for use at White Sands, N. Mex.,  $L_0$  changed for an undetermined reason. Although the instrument had not been moved from Washington after June 1949, on December 6, 1949, the dial readings and hence the  $L_0$  underwent a marked permanent change, which presumably was due either to a slippage of the connection between the dial and the wedge, or to some discontinuity in the photomultiplier response. However, this change was abrupt and very obvious, so that an appropriate  $L_0$  could be determined and applied to the observations following December 6. For reference purposes, the individual values of  $L_0$  and the values which we used (generally an average of the values in any period) are given in table 1. On several

TABLE 1.—Values of  $L_0$

Date	$L_0$	$L_0$ used
1948		
Feb. 27	2.666	} Feb. 18, 1948 through July 6, 1948: 2.619
Mar. 1	2.581	
Mar. 5	2.620	
June 8	2.609	
Sept. 1	2.685	
Sept. 13	2.676	
Oct. 26	2.702	
Oct. 27	2.687	
Dec. 7	2.732	
1949		
Mar. 29	2.667	} Aug. 31, 1948 through Dec. 5, 1949: 2.680
Mar. 30	2.677	
July 27	2.680	
July 28	2.680	
Aug. 25	2.650	
Aug. 29	2.680	
Aug. 30	2.652	
Sept. 2	2.674	
Sept. 9	2.698	
Sept. 20	2.677	
Sept. 21	2.684	
1950		
Jan. 24	2.800	} Dec. 6, 1949 through Mar. 9, 1950: 2.800
Jan. 26	2.805	
Mar. 6	2.800	

occasions calibrations with the lamps offered supplementary values for aid in evaluating the finally used values of  $L_0$ . Since equation (1) also involves  $\mu$ , the maximum error due to inaccuracies in  $L_0$  is probably of the order of 0.01 cm. of ozone.

DUST

Equation (1) does not allow for changes of dial reading caused by changes in "dust" although several different ways of allowing for dust have been used in the past [6]. The dust effect is complicated and improperly understood, and it appears that the error introduced by use of equation (1) in comparison with the formula of Ramanathan and Karandikar [6] usually is less than 0.01 cm. (at least for India), although on very hazy days it may reach 0.02 cm.

In summary, it seems that a difference of 0.01 cm. or less between two observations, taken on different days or in different months, is of doubtful reality, and perhaps on occasion a difference of 0.02 cm. or even more may be caused by something other than a change in ozone.

THE OBSERVATIONS

As mentioned earlier, each observation consists of three dial readings taken in rapid succession. For each day, the observations have been averaged and these average values appear in table 2 and in figure 1. Table 2 also contains the number of observations in each day and the range of the ozone values, that is, the difference between the highest and lowest ozone values observed on the particular day.

The solid curve in figure 1 represents the average annual variation, and was determined by eye (with minor adjustments) from a plot of 10-day mean values of ozone overlapping by 5 days. The curve shows many of the

TABLE 2.—Daily ozone values

Date	Average ozone (0.001 cm.)	Number of observations	Range (0.001 cm.)	Date	Average ozone (0.001 cm.)	Number of observations	Range (0.001 cm.)
<b>1948</b>				<b>1948</b>			
Feb. 18	196	22	29	June 17	226	2	15
19	207	2	2	22	208	11	-----
26	230	3	5	24	208	7	6
27	215	9	17	25	222	4	26
Mar. 1	211	12	11	28	209	7	8
5	198	9	17	29	225	3	33
8	237	4	15	30	212	9	9
10	224	3	4	July 1	209	2	2
12	210	5	45	6	198	1	-----
15	212	2	0	Aug. 31	222	7	10
May 10	225	4	26	Sept. 1	215	25	9
11	224	2	11	2	214	3	8
12	208	4	20	3	210	3	49
13	214	1	-----	4	223	2	3
18	276	2	5	5	205	3	2
19	256	3	12	8	202	4	7
20	254	2	9	9	224	3	18
21	241	5	7	12	199	3	1
24	240	2	21	13	210	26	24
25	229	1	-----	14	214	3	4
27	231	1	-----	15	221	3	36
28	230	2	19	16	234	6	53
June 1	253	1	-----	17	226	3	5
2	267	3	20	20	209	2	6
4	238	6	8	23	198	3	16
7	222	2	0	27	200	4	10
8	224	25	16	28	192	2	10
9	246	9	9	29	190	3	1
10	244	19	19	Oct. 1	193	2	1
11	222	9	13	7	201	3	14
14	228	4	6	12	187	11	24
15	220	4	9	14	194	16	30

TABLE 2.—Daily ozone values—Continued

Date	Average ozone (0.001 cm.)	Number of observations	Range (0.001 cm.)	Date	Average ozone (0.001 cm.)	Number of observations	Range (0.001 cm.)
<b>1948</b>				<b>1949</b>			
Oct. 15	173	2	1	July 18	199	4	3
18	208	1	-----	19	211	4	9
19	213	3	4	20	207	2	4
20	195	2	1	21	209	2	1
21	202	6	9	22	206	3	18
25	176	4	11	25	216	3	3
26	176	21	7	26	223	1	-----
27	175	21	10	27	212	34	13
28	194	4	2	28	218	23	11
29	220	2	20	29	215	2	21
31	190	1	-----	Aug. 1	227	2	11
Nov. 4	183	4	9	4	200	2	13
8	177	3	2	8	236	3	10
10	153	3	1	9	217	3	10
12	154	3	5	10	220	3	20
16	167	2	0	11	224	2	15
17	173	4	6	12	222	2	2
18	175	5	9	16	205	4	6
20	153	1	-----	19	215	3	20
23	161	1	-----	22	211	5	31
26	167	2	3	23	201	1	-----
27	158	2	4	24	210	3	10
30	186	1	-----	25	218	12	22
Dec. 1	192	2	14	26	207	12	2
2	187	3	15	27	191	12	6
6	174	3	4	30	206	19	18
7	179	18	6	Sept. 1	204	5	9
9	194	1	-----	2	207	22	7
10	243	5	4	5	201	1	-----
13	188	5	15	8	214	5	13
14	185	3	9	9	211	22	13
17	182	3	5	12	197	5	3
20	234	2	1	13	192	3	3
21	185	4	7	15	197	4	12
22	194	5	3	19	200	7	6
23	178	3	3	20	212	17	13
24	177	3	6	21	200	50	11
27	221	4	3	22	206	7	13
28	200	4	9	26	208	4	9
31	216	2	6	30	189	5	23
<b>1949</b>				<b>1949</b>			
Jan. 3	185	3	10	Oct. 3	181	2	0
6	179	10	13	4	180	5	3
7	203	3	5	5	196	1	-----
10	230	1	-----	10	176	4	5
11	181	3	3	11	172	10	9
12	225	2	4	12	173	4	9
13	221	6	5	18	185	2	8
14	201	2	1	20	175	2	15
15	172	2	6	21	179	2	10
17	218	4	6	24	190	1	-----
18	213	2	10	26	196	3	13
23	226	2	7	27	195	4	24
24	208	3	10	Nov. 2	183	3	28
25	215	2	3	9	211	1	-----
Mar. 1	269	2	1	14	172	2	4
2	255	3	7	15	182	2	14
4	236	2	4	17	223	3	5
7	259	3	24	18	234	2	2
8	206	3	14	21	194	3	4
9	215	1	-----	22	209	3	2
14	231	2	3	23	188	2	2
16	274	3	18	25	215	1	-----
21	212	2	4	28	189	1	-----
23	217	3	12	30	191	3	3
29	216	20	22	Dec. 1	198	2	7
30	217	9	12	5	222	2	5
Apr. 4	236	3	11	6	231	1	-----
6	252	3	2	7	239	1	-----
7	245	2	0	8	249	1	-----
8	289	5	5	9	270	1	-----
15	220	3	13	10	236	1	-----
19	262	4	15	14	206	2	3
20	237	4	3	15	199	1	-----
21	241	3	1	17	200	2	0
25	218	4	9	20	227	1	-----
28	229	3	15	21	212	1	-----
29	243	3	3	24	214	2	9
May 4	265	4	16	29	232	2	2
5	241	2	7	30	228	2	3
9	212	4	10	<b>1950</b>			
12	245	5	28	Jan. 6	186	1	-----
13	266	3	17	9	211	1	-----
17	251	3	7	10	196	4	5
18	232	5	4	11	212	2	2
19	219	3	0	18	187	1	-----
20	211	2	1	23	212	1	-----
23	234	4	16	24	226	8	14
24	235	3	2	25	219	4	10
25	235	4	11	26	198	14	7
27	254	2	3	Feb. 3	229	14	17
31	249	4	13	8	250	7	12
June 1	275	4	21	15	203	5	-----
5	243	2	7	20	259	1	16
6	232	3	14	21	231	3	13
8	222	3	10	23	251	2	6
11	221	2	2	27	290	2	12
13	213	2	2	Mar. 1	239	1	-----
14	221	3	9	2	301	2	2
15	218	1	-----	6	232	33	6
				7	230	1	-----
				9	254	3	8

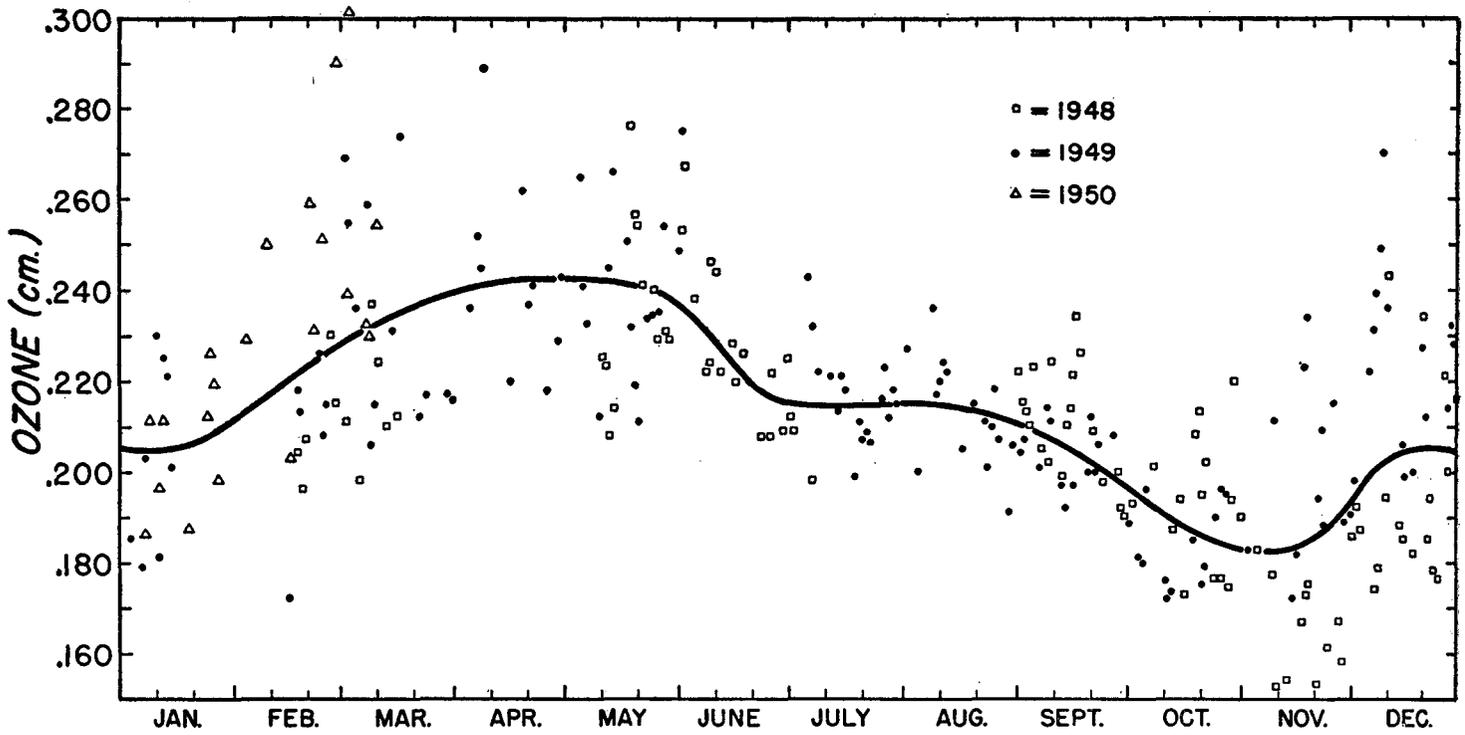


FIGURE 1.—Daily average ozone observations (symbols) and "normal" annual ozone distribution (curve).

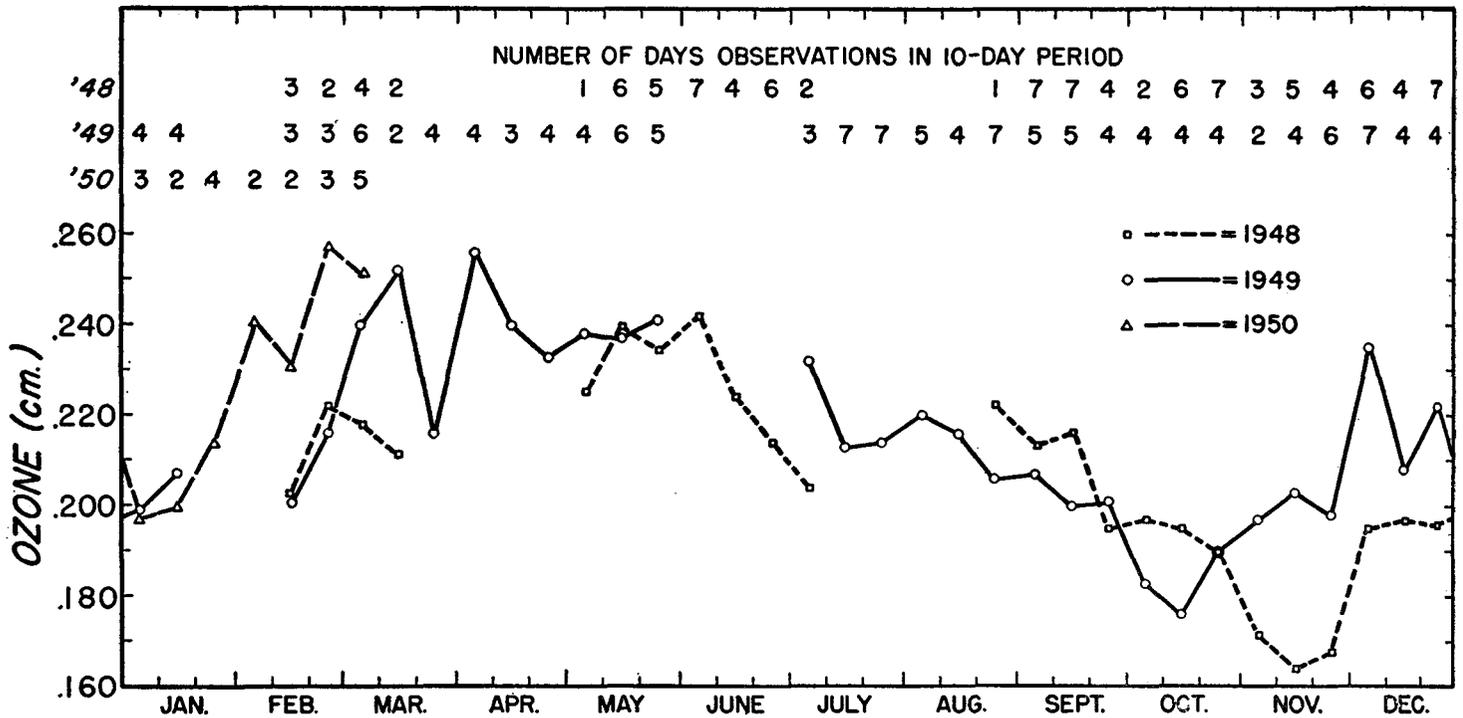


FIGURE 2.—Ten-day mean values of ozone and number of days of observations.

usual features. The annual trend is quite evident, showing the maximum in the spring and the minimum in the fall. The plateau in August is similar to that found by many observers [7], but the plateau in December-January is apparently not generally observed. It remains to be seen of course whether anything except the over-all annual trend will remain after more data have been obtained. By comparison with data from Craig's [7] graphs, it is found that the curve is about 0.02 cm. below the average for about latitude  $40^{\circ}$  N. in all seasons, but is very close to a curve of the minimum values for that latitude. The curve differs markedly from Stair's [2] values, being about 0.04 cm. lower in January, and about 0.035 cm. higher in August.

As usual, the scatter seems to be greatest in spring, although November 1949 also showed considerable scatter. We shall return to this point later.

Figure 2 shows the individual 10-day means (not overlapping), together with the number of days in each 10-day period on which observations were made. In general, the different years show some marked differences, and some interesting similarities. The two Mays have about the same average value in spite of the great daily variability (fig. 1) but the two Novembers are rather different. In 1950, the data start out with the same value as for January 1949, but in February 1950, the mean value is considerably higher than in 1949.

## DISTRIBUTION OF OZONE AROUND SURFACE PRESSURE SYSTEMS

### LOW PRESSURE SYSTEMS

Dobson, Harrison, and Lawrence [8] (D. H. L.) showed that, on the average, ozone tended to be distributed in a definite manner in relation to surface low and high pressure systems. For cyclones, Meetham [9] extended their findings, and later Dobson, Brewer, and Cwilong [10] (D. B. C.) amplified the earlier results by separating Lows into (a) "young depressions" and (b) "old occluded depressions." In the occlusions, which they say had only cold air associated with them, the highest ozone was found *in*, and *in advance of*, the low center; only *above* normal ozone values appeared in occlusions. In the young depressions, with open warm sector, the highest ozone appeared to the west of the low center, and the lowest values appeared over the surface warm front.

Tønsberg and Olsen [5], observing in Tromsø, Norway, also related their ozone data to surface low pressure systems, and by contrast to the (D. B. C.) results found *no* above normal values for a thousand miles in advance of the surface position of the occluded front; in advance of the occlusion, and in a few cases even behind it, only below normal values were observed. For the case of the open wave cyclone, their results agree fairly well with

those of (D. B. C.). As a matter of fact, in the Tromsø results there is little difference between the occluded and open wave cyclone ozone distributions.

Using the technique of Meetham, and of Tønsberg and Olsen, the Washington ozone data were analyzed as follows. On each day during which one or more ozone observations had been made, namely, on the dates in table 2, the 1800 GMT North American surface weather map was examined. If a well-developed low center was present, and if Washington lay inside the area encircled by a cyclonic isobar, Washington was considered to be under the influence of the cyclone. On some occasions Washington lay between a deep Low and a well-developed High; the nearly straight isobar through Washington did not immediately enclose either pressure system, but rambled around several systems. In these cases, Washington was considered to be under the influence of both the cyclone and the anticyclone. Two intersecting perpendicular lines were drawn on a sheet of translucent paper, as shown by the north-south and east-west lines in figure 3. The translucent paper was then placed on the weather map with the intersection over the low center, and the perpendicular lines were placed tangent to the latitude line and along the longitude line through the low center. The position of Washington was then marked on the translucent paper; and, considering the curve of figure 1 to be the normal, the departure from the normal ozone for that day was written near the position of Washington. Occasionally, the Low was of questionable dominance, or it was not clear that Washington was under the influence of the Low; these cases have been marked with a question mark (?) in figure 3. Very questionable cases were discarded.

The results are shown in figure 3 where, for emphasis, all departures of less than 0.01 cm. in absolute value were deleted because the sign of smaller departures is in some doubt. It should be noted that these data do not represent the ozone distribution around a single cyclone, nor do the isolines represent average values of ozone departure; the isolines have been included to facilitate visual examination of the magnitudes. But, insofar as cyclones are similar to each other, the "average" pattern of ozone distribution may be found by this technique. The similarity to the pattern of Tønsberg and Olsen, to that of (D. H. L.) and, for open wave cyclones, to that of (D. B. C.) is quite striking. Although there is no single center of maximum ozone departure, for this group of relatively large departures there is a broad region to the west of the cyclone where, without exception, the ozone was above its normal value; to the east, only below normal values are found. As was the case for the Tromsø data, negative values were associated with occlusions (in at least two cases) as well as with open wave cyclones.

As for the distance from the cyclone center, Meetham [9] had found the maximum ozone departures in a region

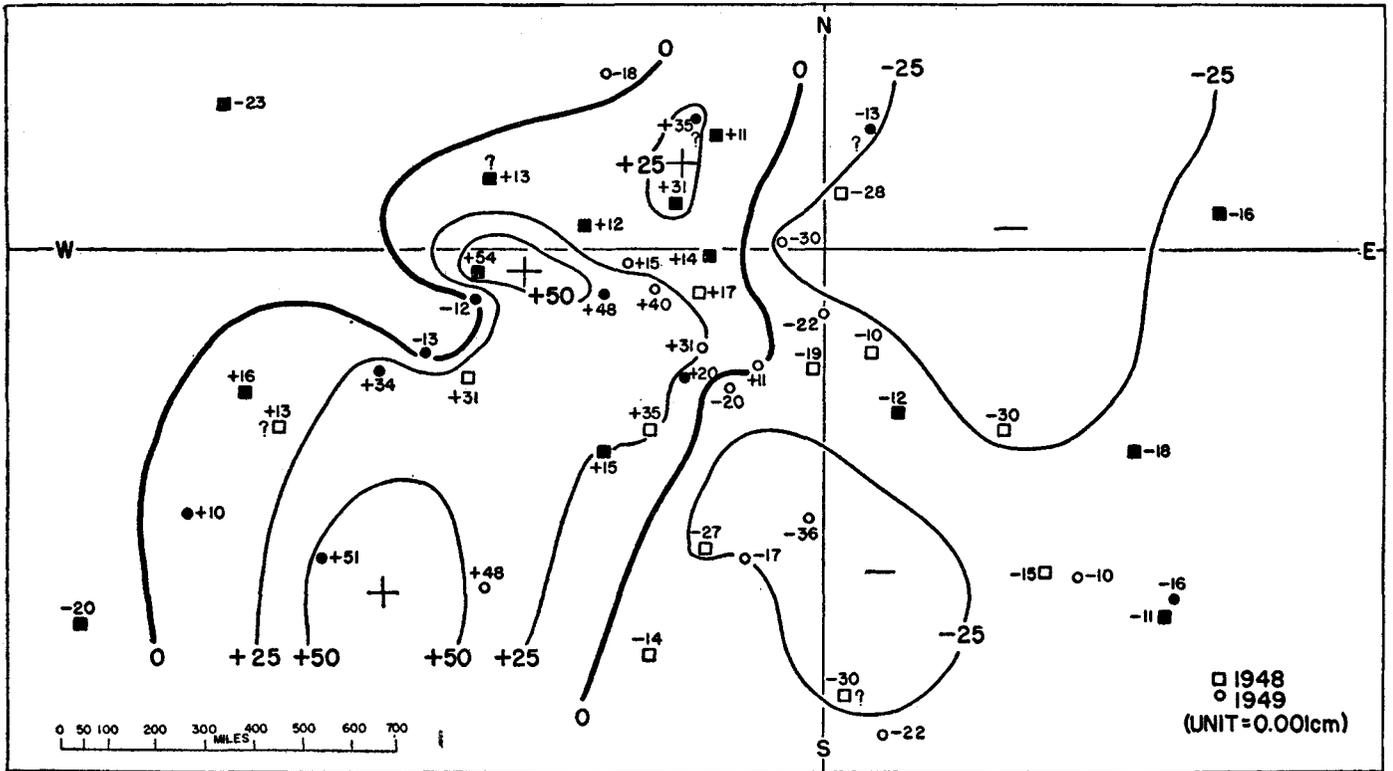


FIGURE 3.—Ozone departures from normal distributed relative to surface low pressure systems; departures with absolute values smaller than 0.01 cm. are excluded. Solid symbols represent cases which appear also in figure 4.

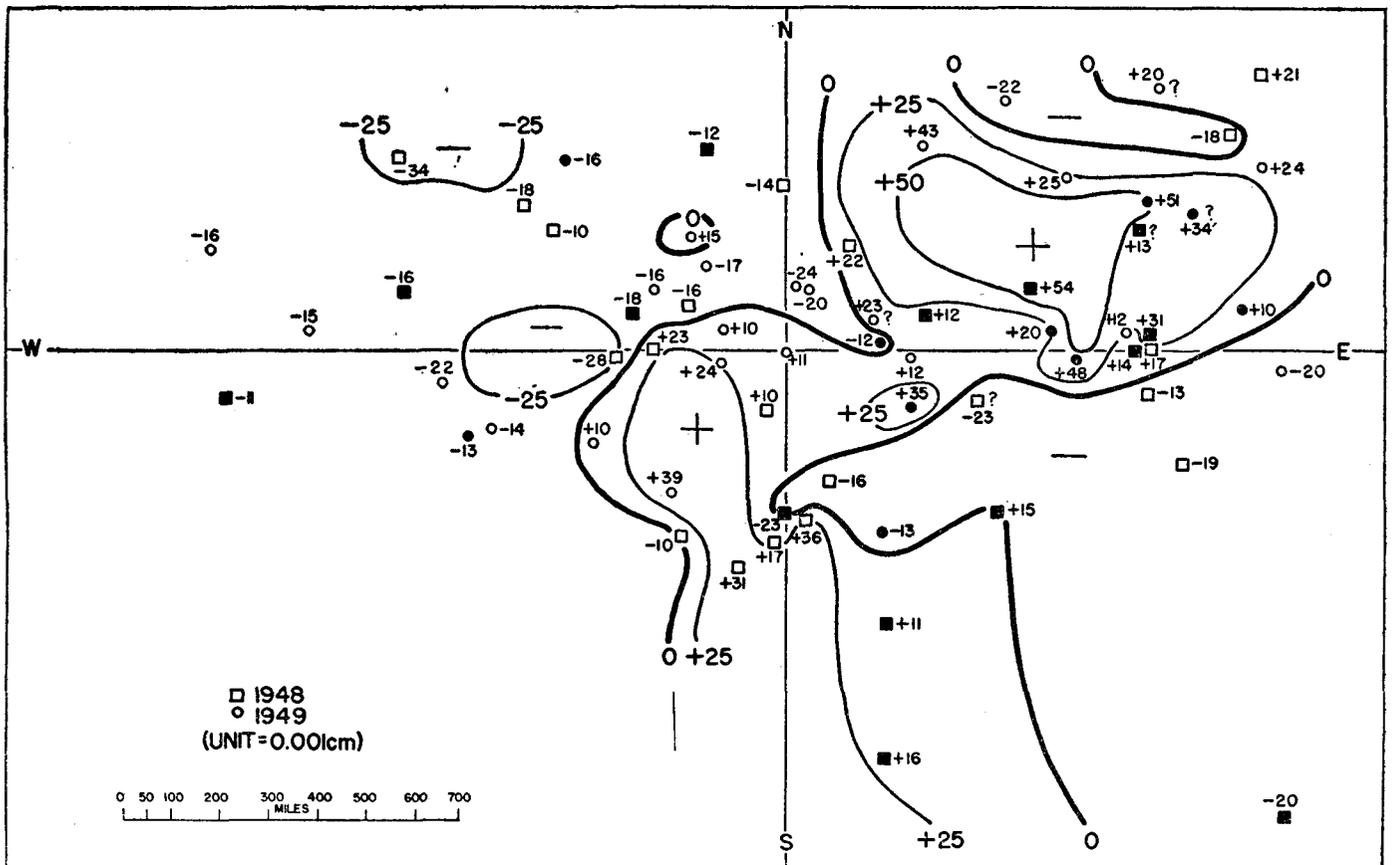


FIGURE 4.—Ozone departures from normal distributed relative to surface high pressure systems; departures with absolute values smaller than 0.01 cm. are excluded. Solid symbols represent cases which appear also in figure 3.

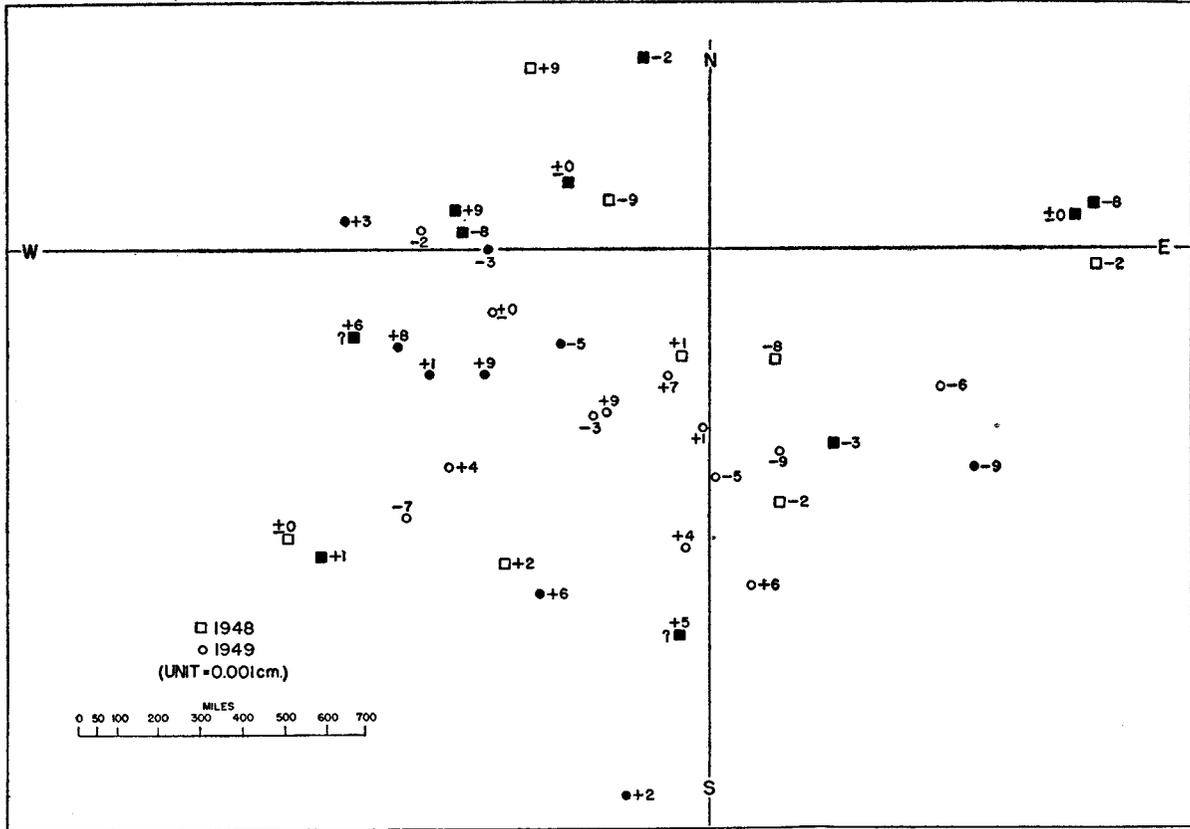


FIGURE 5.—Ozone departures from normal with absolute values less than 0.01 cm. distributed relative to surface *low* pressure systems. Solid symbols represent cases which appear also in figure 6.

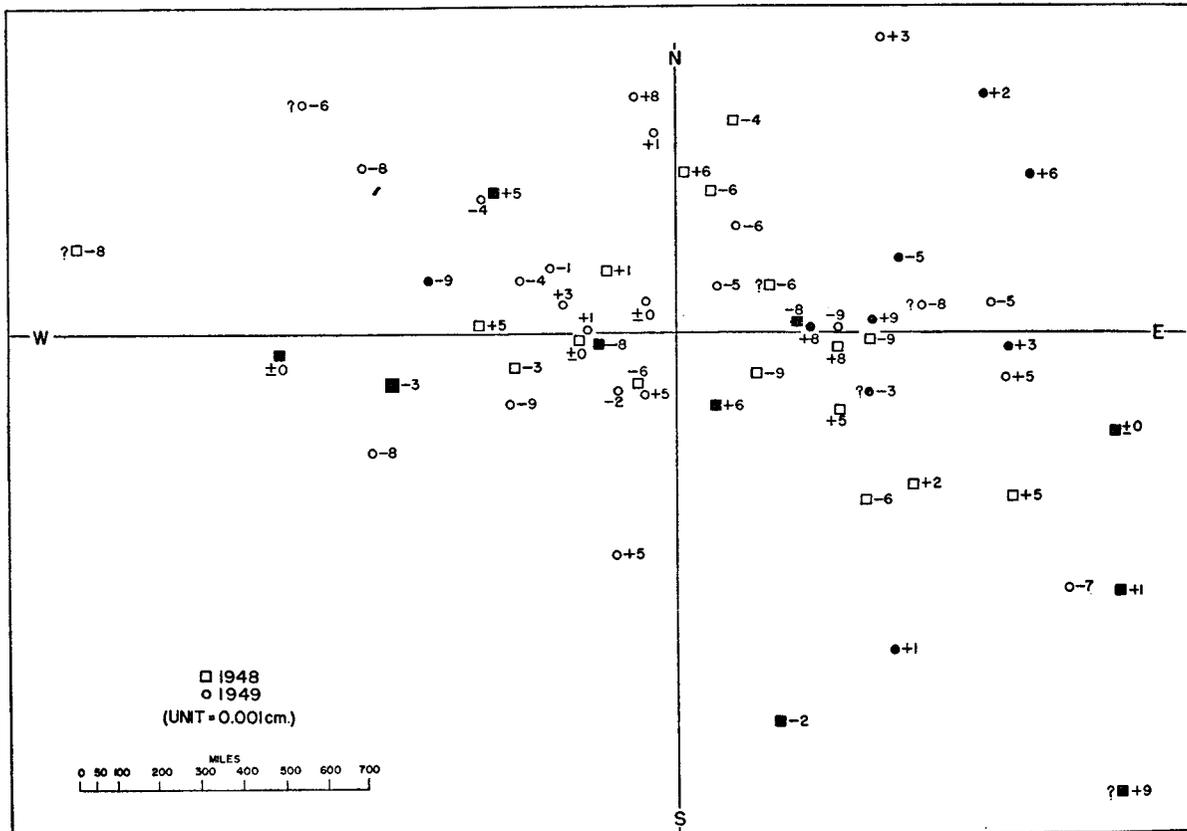


FIGURE 6.—Ozone departures from normal with absolute values less than 0.01 cm. distributed relative to surface *high* pressure systems. Solid symbols represent cases which appear also in figure 5.

0 to 400 miles from the low center; in Washington the maximum ozone departures occurred from 400 to 1,000 miles from the center.<sup>2</sup>

#### HIGH PRESSURE SYSTEMS

In the case for Highs, earlier results in Europe [8] indicated that the greatest negative ozone departures occurred in the center and somewhat to the west of the high pressure center; on the average, negative departures also were present to the east of the High, although sometimes high ozone values were observed just to the east of the high center during conditions with strong north winds.

Near Shanghai, China, Lejay [11] found that the maximum ozone values generally occurred in the northerly flow just in advance of the high pressure center; minimum ozone values were found near and in advance of low pressure troughs. The greatest contrast between Lejay's results and those of (D. H. L.) was for the ozone values in advance of and near high pressure centers. It was suggested by (D. H. L.) that variations from their own average picture were to be found in the thermal structure of the high pressure systems.

In view of the discrepancy between the European and Asiatic results, it is of interest to see what the average condition is in the Washington area. Using the same technique as in the case for low pressure systems, we found the results shown in figure 4. Here, as in figure 3, the blackened circles or squares denote days when Washington was situated between a well-developed High and a similar low pressure system. It will be noted from figure 4 that as a rule (see figs. 5 and 6 for exceptions) when Washington was situated in the northerly flow east or northeast of a high pressure center and simultaneously west of a pronounced low center, the ozone departures were positive; actually, the highest departures of all occurred under these circumstances. Similarly, when Washington was in the southerly flow west or northwest of the surface High, and east of a Low, the ozone departure was negative. In general the plus departures which occur to the east of the high center correspond to Lejay's findings. The negative values to the west of the high center seem to correspond to those of (D. H. L.).

On the other hand, the area of plus departures to the southwest and the negative departures to the southeast of the Highs do not agree well with either of the previous investigations. Here, also, the thermal structure of the Highs may account for the variations. Perhaps also the prevalence of easterly winds (to be discussed later) at levels above 18–20 km. in summer may account in part for these departures.

Of course, figures 3 and 4 do not apply to all ozone observations, for on many occasions the ozone departures from normal were so small (absolute values smaller than 0.01 cm.) that, due to possible inaccuracies, even the

sign was really not certain. The cases when ozone departures were less than 0.01 cm. in absolute magnitude are shown in figure 5 (for low pressure systems) and in figure 6 (for Highs). For Lows the same general pattern as that of figure 3 is found; for Highs the pattern is less definite, and even suggests a reversal from figure 4. However, in both figures 5 and 6, as already stated, the sign of the departures is in doubt, although the good agreement between figures 3 and 5 supports the reality of the pattern of ozone distribution around Lows.

#### CORRELATION COEFFICIENTS

We turn next to a consideration of the relation between total atmospheric ozone amount and the temperature at specific levels in the upper atmosphere. Meetham's [9] well-known correlation coefficients showed that at Oxford, England, and separately at Arosa, Switzerland, a high correlation existed between ozone and temperature and between ozone and potential temperature at 12, 15, and 18 km. Meetham treated all his data (37 days in England, 30 days in Switzerland) as a unit considering departure from the normal in both variables in order to eliminate the effect of annual variations.

To look for a similar effect at Washington, we have examined the Washington ozone data for correlation with temperature at the 500-mb. level and with temperature at the 100-mb. level.

#### 500-MB. LEVEL

At the 500-mb. level we had considerably more data than Meetham had, and therefore decided to investigate the correlation month by month. From the radiosonde data for 1000 EST (approximately) the temperatures for the 500-mb. level,  $T_5$ , were tabulated. If in any one month both ozone and  $T_5$  had been measured on at least 10 separate days, a correlation coefficient,  $r_5$ , was computed between average daily ozone and  $T_5$ . Neither the ozone nor the temperature data were adjusted to their normals because the "annual" trend of either parameter within a single month appeared, after inspection, to be unimportant. The resulting coefficients are shown in figure 7, where each square or circle represents the  $r_5$  for a particular calendar month, except that the square for March 1948 represents data for the period February 15 to March 15, 1948. The solid lines connect points for consecutive months during which 10 or more pairs of data were available; the dashed lines pass through intervals when  $r_5$  was not computed because fewer than 10 sets of observations were available per month. The 1948 values of  $r_5$  have been repeated on the 1949 portion of the graph to show the difference between the two years.

Two facts seem noteworthy; first,  $r_5$  seems to vary fairly regularly from month to month; and second,  $r_5$  sometimes has very high negative values; e. g.,  $-0.96$  in March 1949 and  $-0.90$  in May 1948. On the other hand, the value of  $r_5$  is sometimes near zero; e. g.,  $+0.08$

<sup>2</sup> Culnan [1, p. 21] states, ". . . It appears that the large departures from normal ozone occur further to the rear of pressure systems at New York than in Western Europe."

in February–March 1948, +0.24 in July 1949, and -0.27 in August 1949. The 2 years had rather similar values during the autumn months, but seemed to be quite different in the other seasons as though the pattern were displaced about 2 to 3 months. Unfortunately, in those seasons,  $r_5$  was available for both years only in May.

100-MB. LEVEL

The 500-mb. level represents conditions in the middle troposphere. To investigate conditions in the stratosphere, correlation coefficients,  $r_1$ , of ozone with temperature at 100 mb.,  $T_1$ , were computed as in the case for the 500-mb. level, where again 10 sets of observations per month were chosen as the minimum required number. The values of  $r_1$  are shown in figure 8, together with the  $r_5$  data of figure 7 for comparison.

Here, of course, we have fewer suitable months because the radiosonde data are not as frequently obtained at 100 mb. as at 500 mb. Each square represents a computed value for  $r_1$ ; when consecutive months of  $r_1$  were lacking, adjacent points were connected with a dotted line. As might be expected from the usual inverse correlation between  $T_5$  and  $T_1$ , the left-hand scale of ordinates (500-mb. level) has the opposite sign from the right-hand scale (100-mb. level).

Although there are some marked differences between the two curves, the main features<sup>3</sup> were maintained at both the 500-mb. and 100-mb. levels.

DISCUSSION OF THE RESULTS

FIGURE 7

Figure 7 shows that in certain months, such as March 1949 for example,  $r_5$  has a high negative value. This means that during those months whenever  $T_5$  is low (cold troposphere), ozone is almost always high; when  $T_5$  is high, ozone is low. It's as though the troposphere and ozonosphere (much of which lies in the stratosphere) were geared in some way; a motion which changes the tropospheric temperature is accompanied by a corresponding (but not necessarily identical) motion which changes the total ozone by the correct amount. On the other hand, in the months when  $r_5$  is low,  $T_5$  is largely independent of ozone. Apparently, motions in the troposphere are independent of ozonospheric motions; the troposphere and portions of the stratosphere seem to be "ungeared".

FIGURE 8

By and large, figure 8 shows that  $r_5$  and  $r_1$  follow each other closely, but with opposite sign. In months when  $r_1$  is high, as in March 1949, whenever  $T_1$  is high (warm stratosphere), ozone is almost always high; when  $T_1$  is low, ozone is low. And from this, together with the discussion

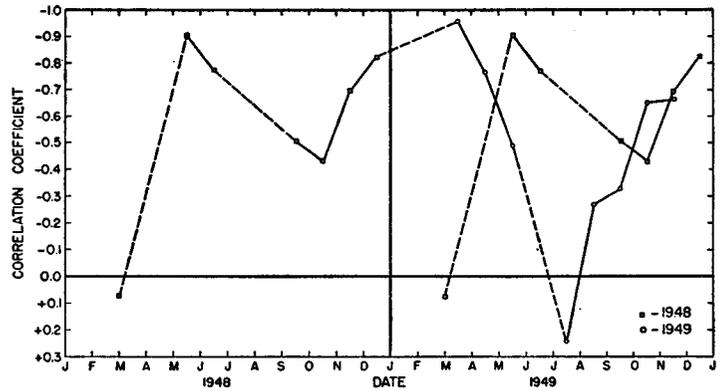


FIGURE 7.—Annual march of correlation coefficients of total ozone with temperature at 500 mb.; 1948 data are repeated on right for comparison.

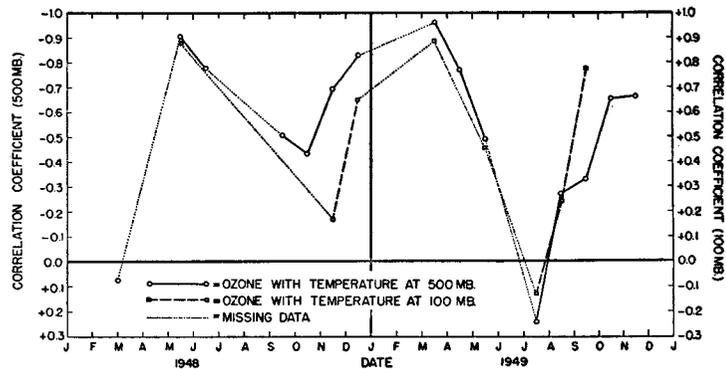


FIGURE 8.—Annual march of correlation coefficients of total ozone with temperature at 100 mb. (right-hand ordinate); 500-mb. coefficients (fig. 7) are included for comparison (left-hand ordinate).

of figure 7, the well known result is obtained that when  $T_5$  is low,  $T_1$  is generally high, and vice versa.

When  $r_5$  is low, as for example in July 1949,  $r_1$  is also low. But a study showed that  $T_5$  was fairly well correlated with  $T_1$  ( $r = -0.64$ ) in July 1949, so that up to 100 mb. all layers of the atmosphere did appear to act in unison. It will be necessary to look to atmospheric motions at higher levels to explain the low values of  $r_1$  and  $r_5$ .

In February–March 1948, the atmosphere even up to 100 mb. was apparently not acting in its usual manner. Unfortunately, there were not enough days on which both  $T_1$  and ozone had been measured, so that  $r_1$  was not included in figure 8. But considering all days in the interval February 15–March 15, 1948, on which both  $T_1$  and  $T_5$  had been measured, regardless of whether ozone had been measured or not, the correlation coefficient between  $T_1$  and  $T_5$  was low (-0.34), indicating that even the very low stratosphere was not acting in correspondence with the troposphere. Wulf and Obloy [12] have suggested that, especially in the vicinity of low pressure troughs, a warm arctic stratosphere may overlie a warm tropical troposphere. In such cases, of course, the usual relation between  $T_1$  and  $T_5$  would not be found, and perhaps such a process was operating in February–March 1948.

<sup>3</sup> Culnan [1], using 16 km. temperatures, found  $r = +0.55$  for December–May, and  $r = +0.28$  for June–November.

It should be pointed out that although the changes in  $T_5$  and  $T_1$  depend on motions at their respective levels, the ozone changes depend on motions throughout the ozonosphere. This point will be discussed more fully in a later section.

### VARIATION OF $r_1$ AND $r_5$

Many plausible reasons can be advanced for the behavior of  $r_1$  and  $r_5$ , and two of these reasons will be discussed here.

#### VARIABILITY OF $O_3$ AND $T$

It is well known that many meteorological variables, such as temperature, vary less in summer than they do in other seasons, and to an extent this is also true of ozone (fig. 1). One might suggest therefore that  $r$  might be low in the summer months because random errors of measurement might obscure any relation that exists when the variables vary over narrow limits. To see how fully the variations of ozone and  $T_5$  could account for the variations of  $r_5$ , the standard deviations,  $\sigma_{O_3}$  and  $\sigma_{T_5}$ , of ozone and  $T_5$ , respectively, were computed and plotted in figure 9. Each point was labelled with its corresponding month and value of  $r_5$ . Then isolines of  $r_5$  were drawn to delineate the general distribution of  $r_5$ .

In a general way, figure 9 shows that the very highest values of  $\sigma_{O_3}$  are indeed associated with the highest absolute values of  $r_5$ , and, with a few exceptions (notably February–March 1948), the lowest values of  $\sigma_{O_3}$  and/or  $\sigma_{T_5}$  are related to low absolute values of  $r_5$ . But within this general framework there are several noteworthy discrepancies. For example, although both Mays had rather similar values for  $\sigma$ , their values of  $r_5$  differed considerably ( $-0.90$  in 1948, and  $-0.49$  in 1949); on the contrary, although both Novembers had quite similar values for  $r_5$ , their values for  $\sigma$  differed appreciably. Despite the paucity of data at 100 mb., by and large, similar results are found when  $\sigma_{T_1}$  and  $r_1$  are utilized. At 100 mb., as perhaps in figure 9, the results are somewhat independent of  $\sigma_T$ , but  $r_1$  varies regularly with  $\sigma_{O_3}$  except that for September 1949,  $r_1$  did not fit the general pattern at all. So, while the over-all annual variations of  $\sigma_{O_3}$  and  $\sigma_{T_5}$  may account for the variation of  $r_5$  from month to month, the whole variation of  $r_5$  can perhaps not be attributed to the variations in  $\sigma$ , or by implication, to our inability to measure changes in ozone when the total range of ozone changes is relatively small.

#### ANNUAL VARIATION OF WINDS ALOFT

Aside from any seasonal change in the standard deviations, there is an observed annual variation in the high-level winds which suggests advection as a physical reason for the major variation of  $r_5$  and  $r_1$ .

The role of advection in the local variation of tempera-

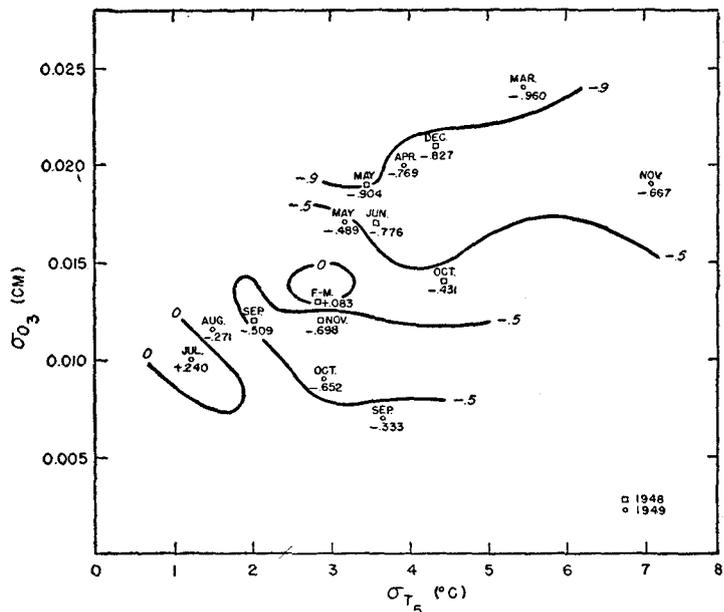


FIGURE 9.—Relation between  $r_5$  and standard deviations ( $\sigma$ ) of ozone and  $T_5$ . Numbers entered near the points and the isolines represent values of  $r_5$ .

ture and of ozone is shown in equations (2) and (3). The local variation of temperature,  $\frac{\partial T}{\partial t}$ , at the 500-mb. level, neglecting non-adiabatic effects, depends only on motions at that level and may be expressed [13] by

$$\left[ \frac{\partial T}{\partial t} \right]_5 = -\nabla_h T \cdot V_h \Big|_5 - V_z (\Gamma - \gamma) \Big|_5 \quad (2)$$

where  $-\nabla_h T$  is the horizontal temperature gradient and  $V_h$  is the horizontal wind vector,  $V_z$  is the vertical wind velocity,  $\Gamma$  is the dry adiabatic lapse rate and  $\gamma$  is the actual lapse rate. A similar equation would obviously apply for  $T_1$ .

The variation of total ozone is made up of changes occurring at all levels. This is shown in figure 10 b, c. Figure 10b [14] shows the distribution of ozone vertically above Arosa, Switzerland (lat.  $47^\circ$  N.) for a "typical" low ozone day (0.200 cm.) and for a "typical" high ozone day (0.300 cm.). Similarly, figure 10c [15] shows the vertical ozone distribution for typical low (0.155 cm.) and high (0.217 cm.) ozone days at Delhi, India ( $28.5^\circ$  N.). Presumably the corresponding ozone distribution at Washington ( $39^\circ$  N.) would be similar to either the Arosa or the Delhi distribution or to both. Note that by no means does all of the change in ozone from a low ozone day to a high ozone day take place below 18 km.; a substantial portion of the change occurs above 18 km.

The time variation of total ozone, neglecting local production or destruction of ozone, except that due to vertical motions aloft,<sup>4</sup> is

<sup>4</sup> Nicolet [16] suggests that convergence or divergence associated with vertical motion may change the amount of ozone. This effect has been neglected here.

$$\frac{\partial O_3}{\partial t} = \int_0^{50 \text{ km}} -\nabla_h(O_3)_z \cdot V_h dz - \int_{30 \text{ km}}^{50 \text{ km}} f[(O_3)_z, V_z, z] dz \quad (3)$$

where  $-\nabla_h(O_3)_z$  is the horizontal gradient of ozone at a particular height,  $z$ ; and  $f[(O_3)_z, V_z, z]$  is some unknown function of  $(O_3)_z$  and  $V_z$ , and  $z$ . Here  $(O_3)_z$  is the density of ozone (cm./km.) at the height  $z$ . The second term on the right is integrated from 30 km. to 50 km. for reasons which will be mentioned later.

ADVECTION

$|r_1|$  and  $|r_5|$  high.—In the months when  $r_1$  and  $r_5$  have high absolute values,  $|r_1|$  and  $|r_5|$ , then  $T_5$ ,  $T_1$  and  $O_3$  vary together in a manner to be expected qualitatively from advection alone.

Whereas, in middle latitudes, below 18 km. both in the stratosphere and troposphere the wind direction is westerly in all seasons; above 18 km. there seems to be a well defined seasonal reversal of the observed wind direction in the ozonosphere (see for example [17, 18, 19, 20]; see also [12]). The wind above 18 km. is westerly from autumn through early spring; but in spring, the wind becomes easterly and in the summer, above 18–20 km. it remains easterly to the top of the observations which on occasion reach to 37 km. [19].

In the westerly current, that is, in winter at all elevations and in summer below 18 km., the direction of the north and south component of the air is usually (but not always) maintained at all heights from at least the 500-mb. level and above. Now, the first terms on the right of equations (2) and (3) express the advective change in  $T$  and  $O_3$  respectively; and these terms indicate that the change in  $T$  is due to advection at only one level in the atmosphere, but the change in  $O_3$  is caused by the advection summed over all levels from the ground to about 50 km.

Let us consider March 1949. If we apply the north wind condition shown schematically in figure 10a, we should expect that the entire column of air up to "great" heights would be brought toward Washington from the north by the winds, bringing with it its temperatures and ozone content (fig. 10b,c) at all heights. If further, we accept Meetham's results, that to the north of Washington ozone and stratosphere temperatures are already highly correlated, then obviously  $|r_1|$  and  $|r_5|$  will be high as far as transport from the north is concerned during March 1949.

A similar argument would of course have to hold for advection from the south. It should be mentioned however that Karandikar [21] found no "definite correlation" at Delhi between ozone and temperature at 6 km. when all data were considered, but [15] during the season of western disturbances, "air of northerly origin in the upper part of the troposphere is accompanied by increase in ozone amount and advent of southerly air by a decrease." During the winter months, at least, one should

expect  $|r_5|$  to be high also at latitudes 29° N. If so, then the high value of  $|r_5|$  at Washington and probably of  $|r_1|$  can readily be explained by the "solid" advection which is observed.

$|r_1|$  and  $|r_5|$  low.—Let us now apply the advection of figure 10a to July 1949. Since the advection is observed to be a "solid" current up to about 18–20 km. [17, 19, etc.], the previous argument will account for the observed relation between  $T_1$  and  $T_5$ . But above 18 km. the observed winds are all easterly, and we have seen (figs. 3 and 4) that on any one day the east-west distribution of ozone is far from constant. Therefore below 18 km., the winds bring in high ozone from the north, corresponding say to high  $T_1$ . But above 18 km. where part of the ozone change occurs (fig. 10b, c), the ozone contribution may be either high or low and does not depend on  $T_1$ . Therefore more than one value of total ozone is to be expected for identical values of  $T_1$ . Obviously, then  $|r_1|$  and also  $|r_5|$  will be low.

From this discussion, it seems reasonable to expect on the basis of advection that even if ozone and temperature could be measured precisely enough, in certain months high values of  $|r|$  are to be expected, while in other months, particularly during mid-summer,  $|r|$  should be low.

VERTICAL MOTION

The second terms on the right of equations (2) and (3) express the temperature change and ozone change, respectively, due to vertical motion and require some discussion. There is general agreement that in the troposphere cold air advection, for example, is accompanied by sinking motion (fig. 10a); therefore advection is the more important factor in producing local cooling there since sinking produces warming. But for the stratosphere such general agreement is lacking. One school [7] believes that warm air advection is accompanied by rising motion as

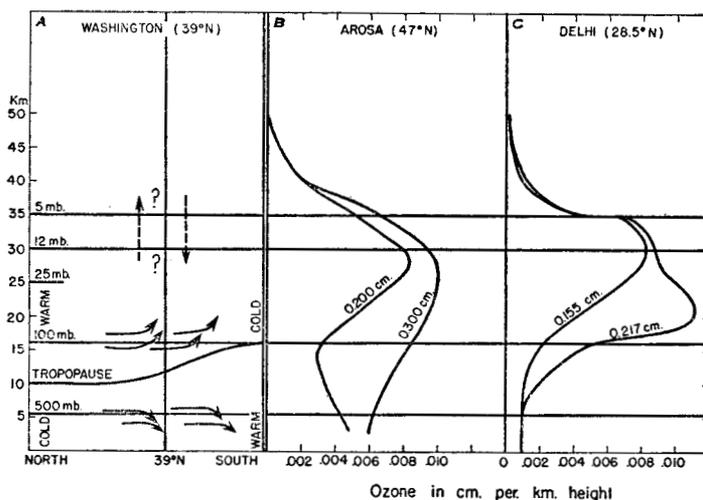


FIGURE 10.—(a) Schematic advection and vertical motion for north wind at Washington. (b) Vertical distribution of ozone at Arosa. (c) Vertical distribution of ozone at Delhi.

it is in the troposphere. Another school [16] believes that warm air advection in the stratosphere (stratospheric north winds at Washington) is accompanied by sinking motion. If on some occasion rising motion is present under such circumstances, then again advection is the predominant factor in changing  $T_1$ . But if sinking motion occurs, then either advection or vertical motion may be the more important factor.

Although the vertical motion near the 100-mb. level will affect  $T_1$ , ozone is changed only by vertical motions above about 25–30 km.; i. e., above the region of maximum ozone/km. (fig. 10). If ozone is lifted from the region below this critical height into the region above, some ozone is destroyed during the rapid return to photochemical equilibrium at the greater height [22]. Hence the total ozone in a vertical column is decreased. Likewise, if ozone is brought down from above about 30 km., the total ozone is increased. Vertical motions confined to the region below 25–30 km., by themselves do not change total ozone. In other words, vertical motion above 25 km. tends to change  $O_3$  in the same sense that it changes temperature. But there is no a priori reason for supposing that the vertical motion near 100 mb. is the same as the vertical motion above 25–30 km. (above about 25 mb.).

In view of the uncertainty regarding the sign of  $V_z$  at the various heights in the ozonosphere, it is difficult to see what its contribution is to  $r_1$  and  $r_5$ .

#### FURTHER DISCUSSION

Of course, the consideration of advection alone cannot explain all the features of figures 7 and 8. Why, for example, was  $r_5$  high in May of 1948 and relatively low in May of 1949? Ordinarily direct balloon measurements show that easterly winds above 18 km. are already observed in May. But the "normal" 19-km. pressure chart for May [23] indicates northwesterly flow over Washington at that level. Perhaps then the answer lies in the fact that in May 1948 easterly winds were not present, except perhaps at heights much greater than 18 km., while in May 1949, easterly winds were already present in much of the ozonosphere.

Other questions arise also. Why was  $r_1$  considerably higher than  $r_5$  in September 1949? Why was  $r_1$  lower than  $r_5$  in November 1948? We are of course assuming that these values of  $r_1$  are significantly different from the corresponding  $r_5$  values. If they are, explanations may have to be sought in the effects of unknown vertical motions, especially if vertical motions have different effects on  $O_3$  and on  $T_1$ . Advection can often account qualitatively for the observed facts. But, as stated earlier, the vertical motions which are surely present may not be the same throughout the ozonosphere and perhaps on occasion affect temperature changes differently from ozone changes. On these occasions, the unknown vertical motions will be reflected in  $r$ .

Also we have spoken of the air as moving "more or less"

from one direction. While this is true, the speed of the winds is often quite variable with height [19]. For example, on a particular day, the wind may be practically calm near the 100-mb. level, but have velocities of more than 50 m. p. h. at heights below and above that level. Obviously the advective effect on temperature and on ozone will be quite different on such days. Such departures from the advection of an entire column of air from a single source region will also tend to reduce the value of  $r$ , if the argument given above is valid.

#### CONCLUSION

We have analyzed the total daily ozone measurements at Washington, D. C., have related the data to surface pressure systems, and have pointed out the similarities to and the differences from previous investigations. We have also correlated ozone with temperatures in the troposphere and stratosphere and have suggested that the existence or nonexistence of a "solid" air current is related to the high or low correlation coefficients, respectively.

It will be of interest now to examine the actual trajectories of the air at upper levels for the cases shown in figures 3–6, and to see to what extent the ozone departures can be explained by a simple "solid" advection hypothesis. For those cases in which this advection hypothesis fails, we should examine either the vertical velocity or the possibility of easterly winds above 18 km. These motions may in turn suggest mechanisms for surface pressure changes.

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