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FORMATION AND TRANSPORT OF OZONE IN THE NORTHEAST QUADRANT OF THE UNITED STATES *

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MASTER

ABSTRACT:

The very high concentrations of ozone measured in Connecticut and elsewhere along the Northeast and Mid Atlantic Coast were investigated through analysis of data from the entire 1976 ozone season from nineteen northeastern and midwestern states, National Weather Service meteorological data, and air parcel trajectories. For several high ozone episodes, the relationship between the movement of and circulation within a high pressure system across the northeast quadrant of the U.S. and the buildup of ozone concentrations was clearly demonstrated. Air parcel trajectory analysis coupled with source density information was utilized for several well defined episodes in 1976 to show ozone measured at various locations throughout the study area is partially locally generated and partially transported from substantial distances.

KEY WORDS: ozone, photochemical oxidant, transport, trajectory

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DISCLAIMER

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INTRODUCTION

Beginning about 1973, Connecticut and other states in the northeast began to recognize what were then inexplicable photochemical oxidant phenomena. What had at the time of development of state plans for the implementation of the Clean Air Act been thought to be a relatively minor problem soon to be solved by the Federal Motor Vehicle Emission Control Program[1] was discovered to be a severe regional problem in many places in the northeastern quadrant of the country.

Since 1973, many researchers have investigated photochemical oxidant levels in the northeast and have documented air mass relationships,[2,3] high rural concentrations in many widespread areas,[4,5] and urban plumes.[6,7,8] Because it became clear that the photochemical oxidant problem along the east coast was much more than could be dealt with through individual metropolitan transportation plans, the "Moodus Ozone Conference" attended by air pollution officials from 19 states in the northeast quadrant of the U.S. was held in September, 1975. One outcome of this conference was an agreement to set up a task force to compile and share 1976 ozone data in an effort to better understand the origins of the measured ozone concentrations throughout the region.

Earlier papers have described the task force techniques and discussed three specific episodes - one in April, 1976[9] and two in August, 1976.[10] The purpose of this paper is to review the base of data acquired between April and September, 1976, demonstrate the repetitive nature of the northeast quadrant ozone episodes, and to show how the path of summertime anticyclonic systems, air parcel trajectories, and measured ground level ozone concentrations together describe the build-up of ozone concentrations in an air mass and lead to a cohesive explanation of the very high concentrations observed in the northeastern portion of the country.

METHODOLOGY

The nineteen conference attendees agreed to share 1976 ozone data and participate in an instrument calibration check. While many of the participants operate extensive networks of ozone instruments, a total of only 81 sampling locations were chosen for a uniform geographical distribution and the depicting of the regional nature of the ozone concentrations while avoiding local interferences. For example, of twelve ozone monitoring sites in Connecticut, the four most removed from city centers were selected. All of the instruments used where chemiluminescent or ultraviolet type. Two ozone calibration units were verified at the National Bureau of Standards in the spring and again near the end of the study period. Through several Regional Offices of EPA, the verified calibration was transferred to each of the participating state agencies to ensure consistency of the data.[11]

Each participant provided the maximum hourly ozone concentration for every day at all selected sites. For each of the 183 days in the study period (April 1 - September 30, 1976), a map of ozone isopleths was prepared, over which were superimposed isobars and front locations. These maps formed the primary tool of the study, for they clearly showed the relationship between air mass location and ozone concentration. Periods of time which the maps indicated were of particular interest were studied in detail using air parcel trajectories.

RESULTS AND DISCUSSION

Ozone Distribution in a High Pressure System

Researchers have shown that ozone concentrations increase within a summertime anticyclone as the system travels through the Northeastern Quadrant of the U.S.[5,9,10] National Weather Service surface maps show that between April 1 and September 30, 1976, some twenty-four individual anticyclonic systems traversed the study area. With only a few exceptions, the ozone isopleths indicate a build-up of ozone within each system as it first crossed the urban-industrial areas of the midwest and a continued spreading and a further accumulation of ozone as the system circulated and traveled eastward.

Typical Episode

Although most episodes followed this pattern, detailed analysis of the many occurrences of elevated ozone indicates each episode was somewhat unique. However, it is possible to describe some general characteristics common to most episodes and to develop a description of a "typical" episode which occurred between April 1 and September 30, 1976.

1. The time interval from the air mass's entry into western portions of the area to departure over the Atlantic varied from 4 to 15 days but averaged 7.

2. The most frequent path of the system center was southeastward from Canada into Minnesota, on into Illinois-Indiana-Ohio, then eastward into Pennsylvania or Maryland/Virginia and continuing out into the Atlantic off North Carolina.

3. Ozone concentrations in the air mass entering the U.S. from Canada averaged 30-50 ppb.

4. As the high pressure system began to influence the study area, the areas which first began experiencing ozone levels above 80 ppb were immediately downwind of urban and industrial areas.

5. As the high moved eastward, the areas of high ozone intensified, expanded and eventually merged. After several days in the U.S., most of the areas under the influence of the high pressure system experienced levels in excess of 80 ppb.

6. On subsequent days areas of elevated ozone merged, but "urban plumes" of more elevated ozone remained detectable downwind of midwest and northeastern urban centers.

7. Fairly uniform elevated ozone concentrations over broad areas were noted by the time the east coast was on the backside of the high.

8. The highest concentrations were found downwind of urban areas in the Northeast at times when the air flow direction was such that urban plumes were superimposed on substantially elevated air mass-borne ozone concentrations on the backside of the high pressure system.

9. A cold front marked the end of the episode and provided a clear demarkation between the ozone laden air mass and a new, cleaner air mass as it traveled from west to east.

Figure 1 shows an example of an episode beginning on July 17 and ending on July 21, which follows this typical description fairly well. Table 1 lists characteristics of each of the twenty-four episodes.

Trajectories and Ozone Half-Life

While the maps of ozone isopleths clearly demonstrate the change in position with time of the highest ozone concentrations, they do not, by themselves, provide definitive answers to two questions:

1. Is the ozone, precursors, or some combination, transported, or are we merely observing the movement of meteorological conditions conducive to local generation?
2. Is the ozone primarily anthropogenic in origin, or formed from naturally-emitted precursors?

Measurements of ozone life-times in the troposphere of sufficient length to allow long-range transport help answer the first question. Calculations[10] based on Stasiuk and Coffey's data[2] and Research Triangle Institute's recent work[11] have shown ozone half-life above the nocturnal inversion layer (and away from sources of oxides of nitrogen) of 16 to 34 hours. Wolff et al[10] reported an average of 29.4 hours based on a series of aerial measurements. Half-lives of this magnitude clearly support the possibility of long-range transport.

Trajectories, showing the path of air parcels which arrived coincidentally with elevated ozone measurements help answer both of the questions. Air parcel trajectories for the entire summer were prepared. [The technique is described in Reference 13.] Figure 2 shows back trajectories for 7/20, while isopleth maps for the July 17-21 episode are shown in Figures 1a-d. Isopleth maps and trajectories for the episode of June 2-11 are shown in Figures 3 and 4. Trajectories terminate at 1300 EST, for Chicago; Youngstown, Ohio; Lexington, Kentucky; New York; Norfolk, Virginia; Marquette, Michigan. The numbers indicate the previous position of the air parcel in six-hour intervals (e.g., '1' is the position 6 hours before termination, '2' is 12 hours, etc.).

July 17 - July 21

The anticyclone which defines the episode of July 17-21 first enters the study area from the west early on July 17. Under the influence of the high pressure system, the maximum ozone concentrations at sites near high emission density areas in Ohio and Indiana slightly exceed 80 ppb on the seventeenth.

By July 18 the high is centered in southern Illinois with all of the northeast quadrant of the U.S. under its influence. Strong source areas are still discernible by their elevated ozone concentrations, but the area enclosed by the 80 ppb isopleth in Figure 1b is growing. On July 19 the center of the system is over West Virginia, with slow, clockwise circulation and conditions conducive to oxidant formation. Monitors throughout the central portion of the study area report levels above 80 ppb. In Figure 1c, the influence of local emissions can be seen in the 160 ppb area in Ohio. Figures 1d and 1e show the ozone isopleths and trajectories, respectively, for July 20. The trajectories indicate air parcel flow generally from the west-southwest. The 0100 h New York trajectory indicates that the air which was over Ohio containing very high ozone concentrations on July 19 has reached New York City and surroundings early on July 20. As can be seen in Figure 1d, New York, New Jersey, and the three southern New England states experienced very high ozone levels on July 20. The cause of these extremely high concentrations (hourly averages as high as 230 ppb) downwind of the very high east coast emission areas is superposition of urban plumes on already elevated air-mass-wide ozone levels. During a very similar episode in August, 1976, reported on elsewhere,[10] concentrations entering the New York area from the Midwest were shown to average between 100 and 125 ppb, while sources in the Pennsylvania-New York-New Jersey-Connecticut area added 80 ppb or more.

The cold front located in Minnesota on July 20 marks the "back-end" of the episode, and it sweeps rapidly across the area on July 21 pushing the polluted air completely out of the area by July 22.

June 2 - June 11

This episode is atypical in its path and speed of the system's travel. Nonetheless, it provides an excellent opportunity for insight into the process of ozone formation and transport in the study area. Apparently because of its slow, meandering travel, the air mass acquired such quantities of ozone that some of the year's highest concentrations were recorded under its influence.

The episode begins on June 2 with an anticyclone centered near Hudson's Bay. The broad circulation around this high pressure system affects most of the study area. Ozone concentrations are low at all monitoring sites and trajectories indicate flow entering the area from Canada. On June 3 and 4 the center of the system moves slowly southward with trajectories indicating air parcel movement generally east-to-west through the center of the region. Isopleth maps (Figure 3) indicate high ozone spreading from eastern urban areas and other high source density areas westward indicating a case of transport of ozone from the east coast into the midwest. Westerly flow continues on June 5 and 6 with wind speeds decreasing and the area enclosed by the 80 ppb isopleth growing as individual source areas become indistinguishable.

The center of the high continues southward on June 7 and 8; air parcel trajectories indicate a dramatic change in flow as most of the region becomes affected by return flow around the system from a generally westerly direction. As the maps of Figure 3 indicate, although the wind direction in the center of the region has changed by 180°, the same air mass, which has acquired substantial quantities of pollutants in the previous six days, continues to affect the area. On the ninth and tenth, the center of the system progresses in a more typical fashion toward the Virginia-North Carolina area. In this position, the direction of circulation as indicated by the trajectories in Figure 4 is such that the high pollutant levels throughout the air mass are added to plumes from large eastern source areas and sites along the east coast experience extremely high levels of ozone. The high pressure system eventually progresses eastward carrying the polluted air out over the Atlantic.

Trajectories from Source Areas

The trajectories of these two episodes indicate that air parcels pass over areas of high emission density and demonstrate transport over long distances in a pattern consistant with the observed and mapped ozone concentrations. [Similar results for April 11-19 are reported in Reference 9.]

CONCLUSIONS

The data show an air mass typically carries 30-50 ppb when it enters the study area from Canada. Trajectory analysis shows that areas of high ozone precursor emission density produce elevated ozone plumes downwind. Following the progress of many anticyclones produces a repetitive picture of "merging" of plumes from high emission areas and spreading of the high ozone areas to a point where a generally uniform air-mass-wide elevated ozone concentration can be measured. The concentration is apparently a function of the forward speed, size and path of the air mass, together with the time of year and other parameters affecting oxidant formation rate. A description of the sequence of meteorological events leading up to the very high ozone concentrations frequently observed in and downwind of the metropolitan areas on the east coast has been developed. Isopleth maps of ground level ozone concentrations depict the superposition of urban plumes upon air-mass-borne ozone transported long distances. Analysis of twenty-four episodes in 1976 indicates the repeated occurrence of this phenomenon in areas under the influence of the backside of a high pressure system.

The existance of discernible plumes confirms the importance of short-range contribution to the oxidant problem, but it is the regionally-transported portion of the measured ozone which generally causes the most severe concentrations. Data for July 17-21 and several other episodes allow a rough assessment of the relative contributions from distant versue more nearby sources - fifty percent or more of the measured ozone apparently is advected into the New York-Philadelphia area on most days.

While it has been noted that each episode is unique, the repetitive aspects of the twenty-four episodes in 1976 are striking. It would appear that a cataloguing of characteristics of episodes would allow knowledgeable forecasters to follow an air mass from its inception and predict the severity of the episode.

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TABLE 1
1976 EPISODES

First Day	Last Day	Entry Point	Position on last day	Max. Conc. (ppb)	Date of Max.	Duration (Days)	Location of Max.
4/11	4/21	MN	E of NC	195	4/19	12	Morris Dam, CT
4/28	5/2	MN	E of VA	110	5/2	5	Eastford, CT
5/3	5/6	MN	E of VA	128	5/6	4	High Point, NJ
5/7	5/11	MN	E of MA	132	5/10	5	Allentown, PA
5/12	5/15	MN	E of NJ	130	5/15	4	Sandy Hook, NJ
6/2	6/16	WS	E of NC	259	6/9	15	Babylon, NY
6/16	6/19	IL	N of VT	158	6/18	4	Morris Dam, CT
6/19	6/30	IL	E of GA	255	6/24	12	Middletown, CT
7/1	7/9	MN	E of NC	230	7/9	9	Sandy Hook, NJ
7/7	7/11	MN	N OF ME	180	7/11	5	Middletown, CT
7/12	7/17	MN	E of VA	175	7/15	6	Norfolk, VA
7/17	7/21	IL	ALA	230	7/20	5	Eastford, CT
7/20	7/24	MN	E of ME	140	7/24	4	Henrico Co., VA
7/24	7/28	MN	E of VA	250	7/28	5	Cheverly, MD
7/31	8/6	MN	E of NC	220	8/5	7	Amherst, MA
8/6	8/14	MN	E of SC	265	8/12	9	Middletown, CT
8/13	8/23	MN	VA	250	8/22	11	Sandy Hook, NJ
8/23	8/29	MN	E of ME	210	8/26	7	West Newbury, MA
8/28	9/1	MN	E of VA	145	9/1	5	Morris Dam, CT
9/1	9/5	MN	E of MA	115	9/3	5	Miamisburg, OH
9/4	9/10	MN	E of VA	189	9/9	7	Fairfax Co., VA
9/10	9/15	IL	E of DEL	205	9/13	6	Sandy Hook, NJ
9/16	9/20	MN	E of GA	130	9/20	5	Middletown, CT
9/23	9/26	MN	E of ME	115	9/24	4	Norfolk, VA

FIGURE CAPTIONS

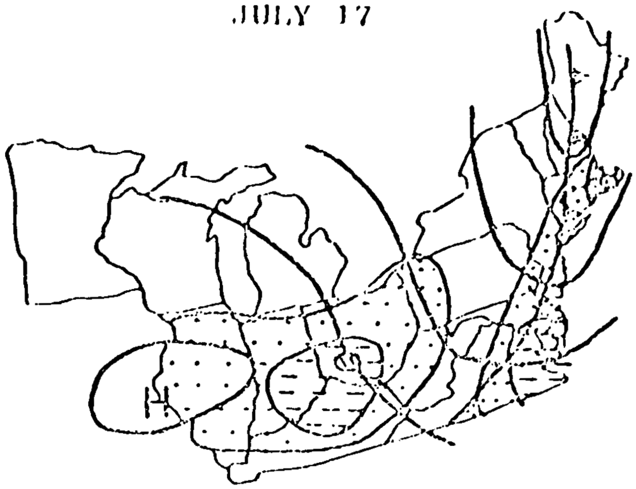
Figure 1: Ozone isopleth maps for the period July 17-20, 1976.

Figure 2: Backward air parcel trajectory terminating in New York City at 0100h on July 20. The numbers 4 and 6 indicate the approximate position of the air parcel 24 and 36 h prior to termination.

Figure 3: Ozone isopleth maps for the period June 2-12, 1976.

Figure 4: Backward air parcel trajectories terminating at 1300h in N.Y., N.Y., Norfolk, Va., Youngstown, Oh., Lexington, Ky., Chicago, Ill. and Marquette, Mich. Numbers indicate the approximate previous position: 1=6h, 2-12h, 3=18h etc.

1a
JULY 17



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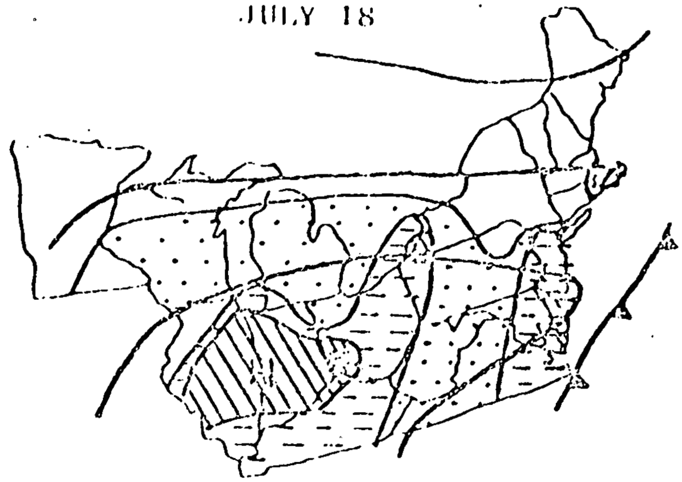
JULY 18



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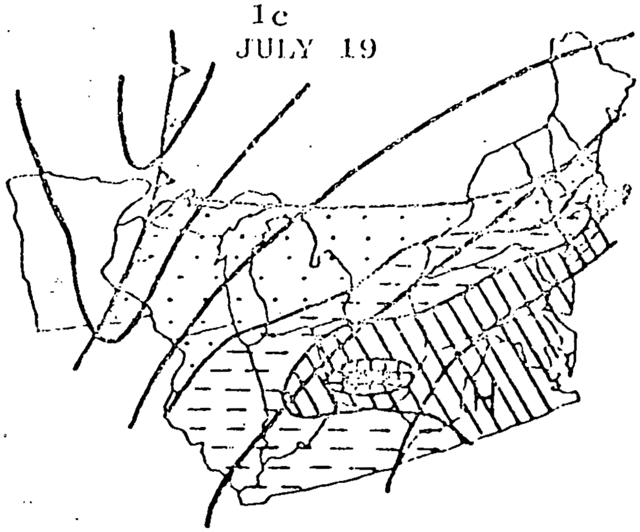

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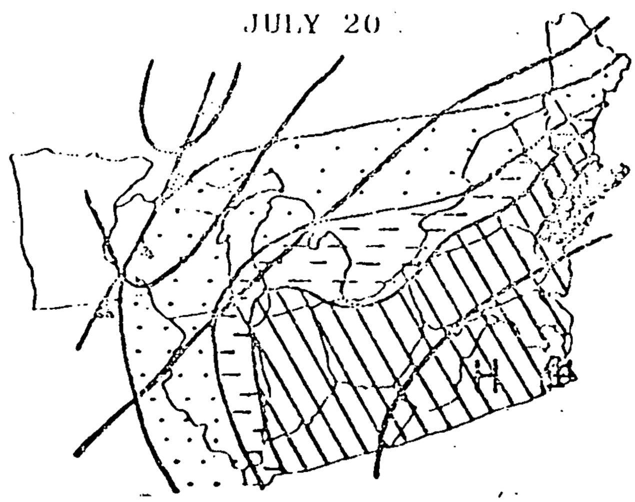

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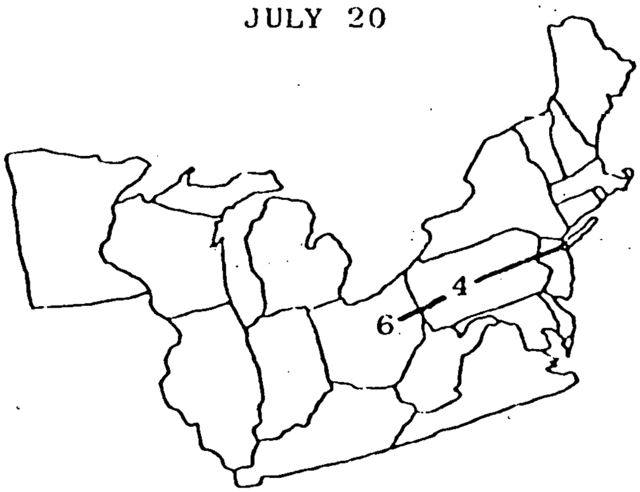
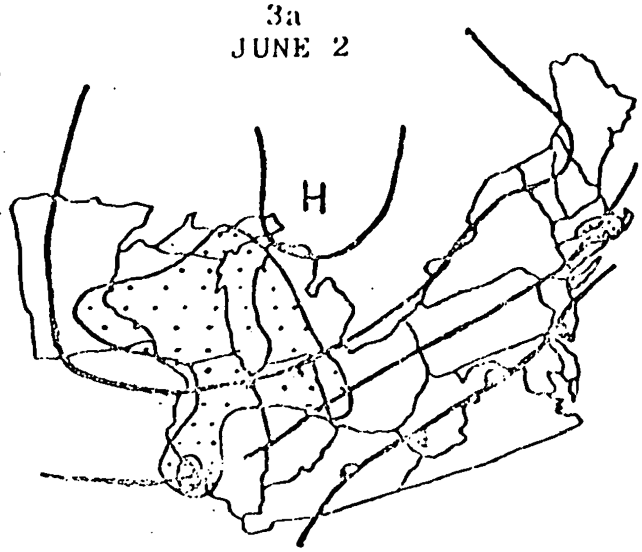


KEY FOR OZONE MAPS

- 60 ppb
- 60-79 ppb
- 80-99 ppb
- 100-149 ppb
- 150-199 ppb
- 200 ppb

3a
JUNE 2



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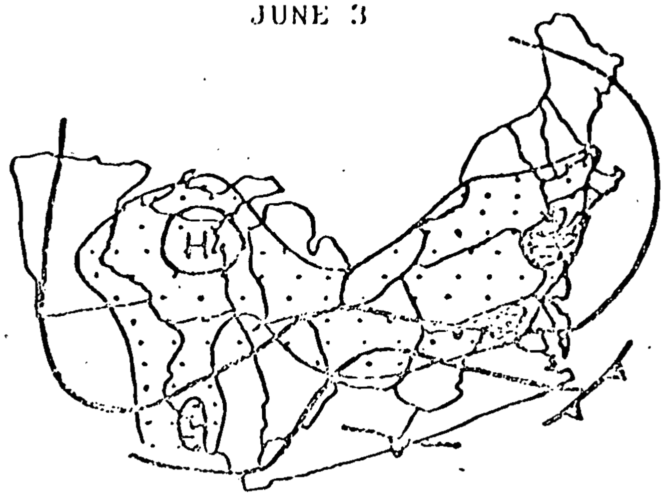

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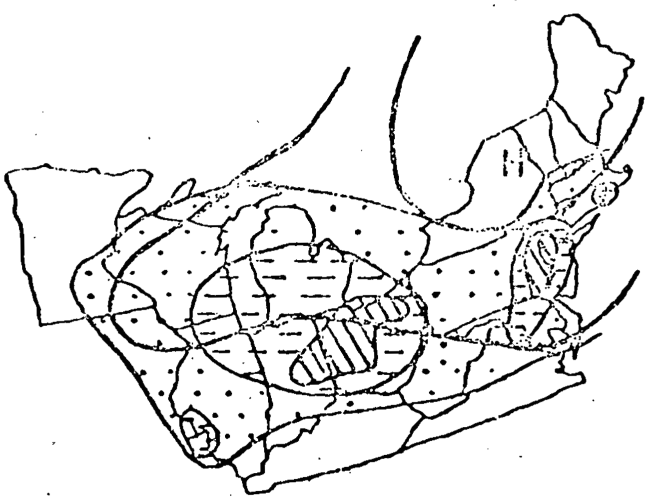

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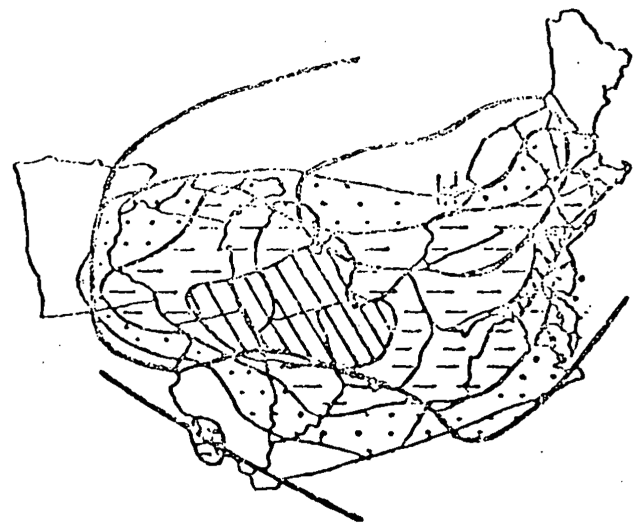

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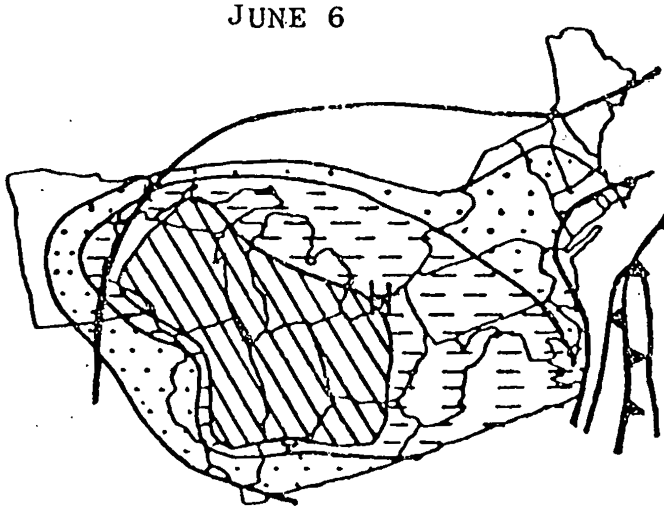

KEY FOR OZONE MAPS

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- 80-99 ppb
- 100-149 ppb
- 150-199 ppb
- 200 ppb

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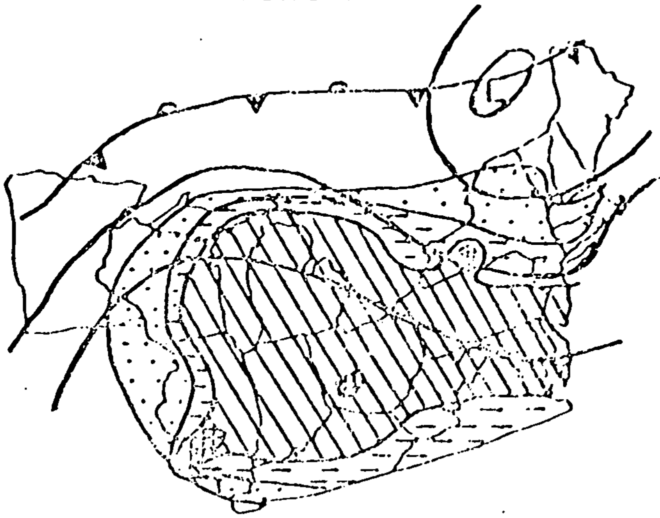

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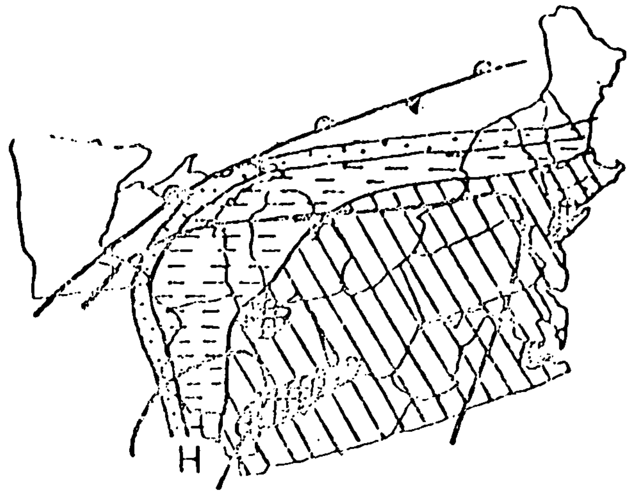

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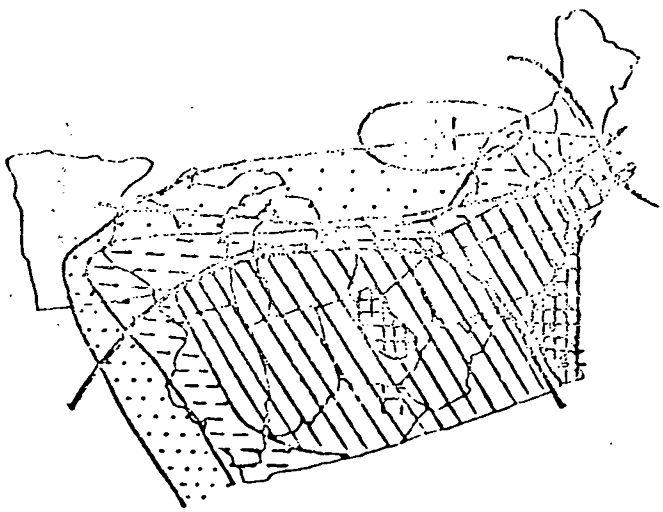

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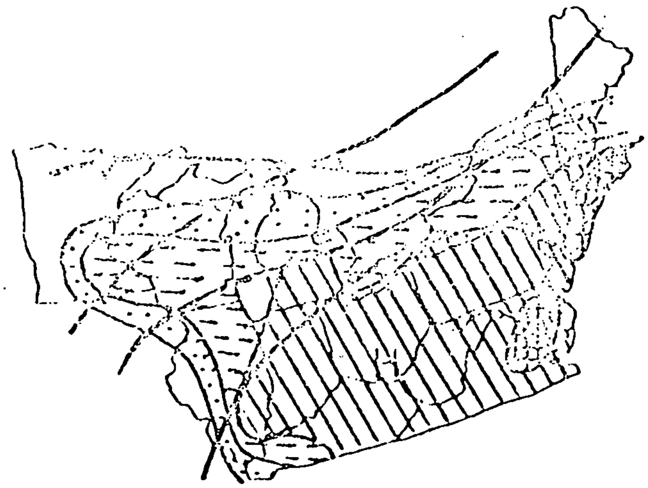

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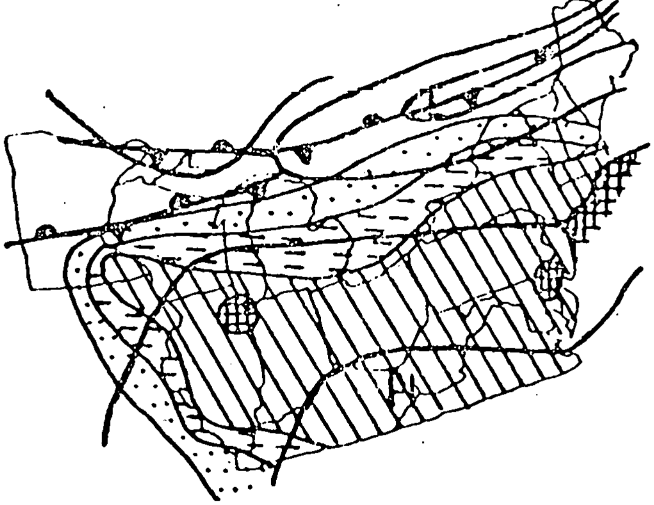

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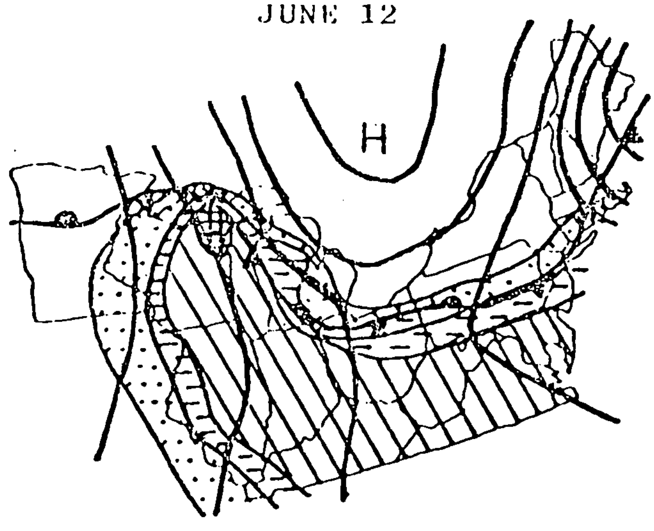

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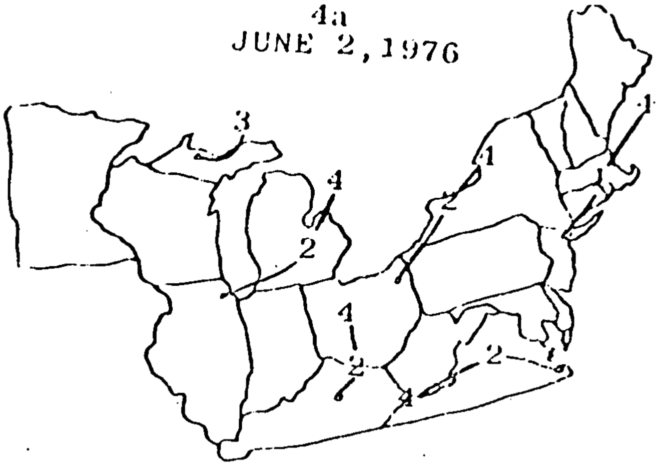

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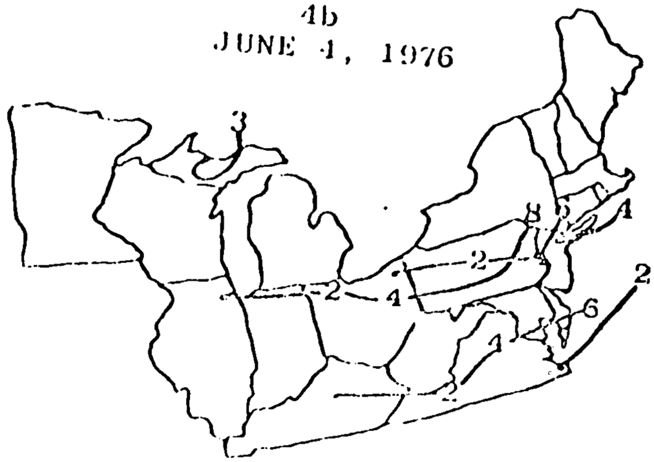

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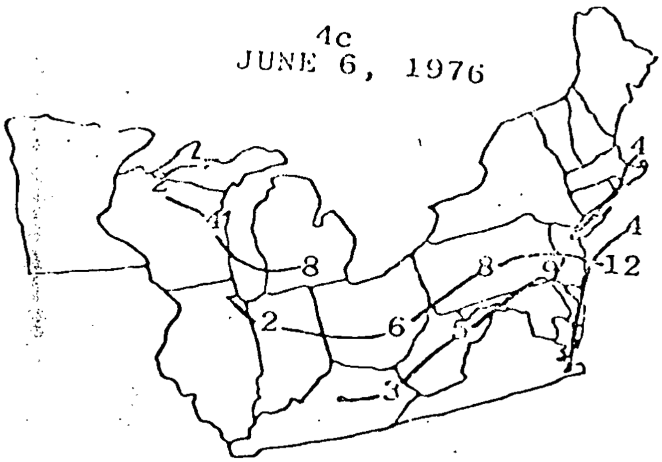

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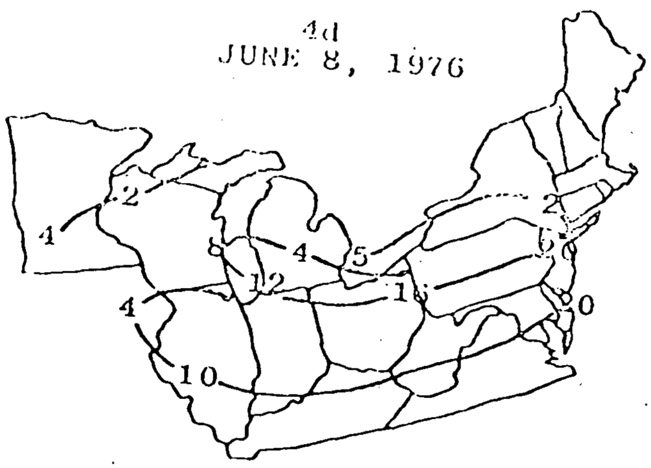

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