THE DISTRIBUTION OF BERYLLIUM-7 WITHIN HIGH-PRESSURE SYSTEMS IN THE EASTERN UNITED STATES

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Abstract. During a 1976 rural ozone field study conducted in McKee, Kentucky, and Busick, North Carolina, simultaneous measurements of ozone and beryllium-7, a tracer of stratospheric air, were obtained. The purpose of this study was to assess the importance of naturally produced stratospheric ozone on ground-level ozone concentrations observed in rural areas in the Eastern United States. It is currently accepted that the stratospheric contribution to the surface ozone is about 30-50 ppb, and that the occurrence of this is confined to the front part of high-pressure systems immediately behind a cold front. Furthermore, it is thought that the stratospheric contribution is negligible on the back side of the high-pressure system where higher levels of ozone occur. This study seriously questions both of these hypotheses because the highest levels of beryllium-7 were observed on the back side and not on the front side of high-pressure systems.

Introduction

Tropopause folding associated with large-scale eddy mixing near the jet stream during the passage of surface cold fronts has been identified as an important mechanism for transporting stratospheric ozone into the lower troposphere [*Danielsen*, 1968]. When this occurs, stratospheric air containing ozone and ^{7}Be (a natural tracer of stratospheric air) [*Husain et al.*, 1977; *Ludwick et al.*, 1976] enters the troposphere and is transported downward behind the surface cold front [*Danielsen*, 1968; *Reiter*, 1977]. Mean surface ozone concentrations attributed to these stratospheric intrusions have been estimated to vary between 15 and 50 ppb [*Reiter*, 1977; *U.S. EPA*, 1975; *Singh et al.*, 1977; *Mohnen*, 1977]. This concentration varies with the season, with the maximum concentrations occurring in the late winter and early spring and the minimum occurring in the late summer and early fall [*Singh et al.*, 1977].

Recently, it was estimated that the upper limit of stratospheric ozone at Whiteface Mountain in New York during midsummer was 37 ppb [*Husain et al.*, 1977]. Localized intrusions which produced concentrations exceeding 80 ppb have also been identified, but these occurrences seem to be infrequent [*U.S. EPA*, 1975; *Singh et al.*, 1977], especially during the summer months when photochemical production creates additional quantities of ozone in the lower troposphere.

Because intrusion of stratospheric ozone is associated with the passage of cold fronts, it has been postulated that the maximum ozone concentrations from this mechanism will occur just behind the cold front on the front side of a high-pressure system [*U.S. EPA*, 1975; *Ludwig et al.*, 1977]. Consequently, it was also concluded that stratospheric ozone does not significantly contribute to the widespread Eastern United States summertime ozone problem [*U.S. EPA*, 1975; *Ludwig et al.*, 1977], which has been shown to be associated with the back side of high-pressure systems [*Wolff et al.*, 1977].

If stratospheric ozone is not a factor on the back side of high-pressure systems, concentrations of ^{7}Be should decrease from east to west in a high-pressure system in the absence of precipitation. To test this hypothesis, daily ^{7}Be concentrations were measured for a three-month period during August through October, 1976, at two rural sites in the Eastern U.S. The sampling days were stratified according to the relative position of the sites within the high-pressure system.

The two sites were McKee, Kentucky, from August 3 to September 13, and Busick, North Carolina, from September 24 to October 24, 1976. Both sites are located in the area which, according to *Reiter* [1977], is quite prone to be influenced by stratospheric intrusions. The McKee, Kentucky, site was located in an open field in Jackson County, about 10 km south of downtown McKee on County Road 270. The nearest major city was Lexington, 85 km north. The elevation at the site was 410 meters. The Busick, North Carolina, site was located about 50 km east of Asheville, North Carolina, in the Pisgah National Forest at an elevation of 980 meters.

Methodology

Two high-volume air samplers were operated simultaneously to process a specific volume of air which ranged from 1500 to 6500 cubic meters. The samples were collected on Gelman spectro-grade, type-A glass-fiber filters. The two filters obtained during each sampling period were folded together in a 1 x 5 x 6.4-centimeter packet and treated as one sample. Sampling periods were 24 hours.

The ^{7}Be activity was determined with a gamma-ray spectrometer which consisted of a high-resolution germanium (GeLi) detector coupled to a multi-channel analyzer. The absolute ^{7}Be concentration on the filter samples was determined through the use of counting standards which took into account sample-detector geometry, detection efficiency at

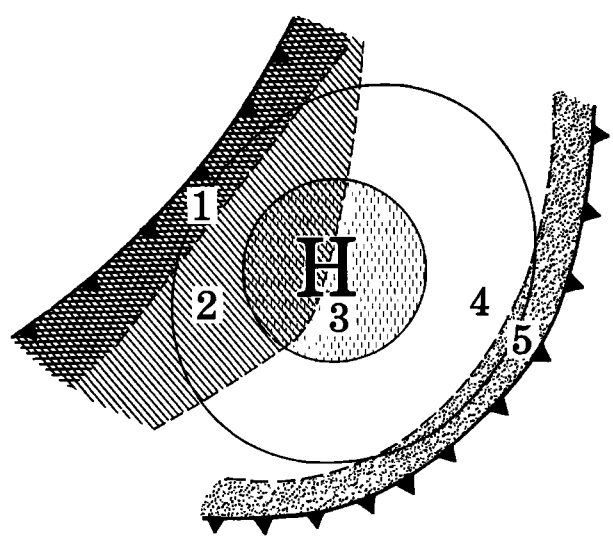

Figure 1. Classification of different areas within a high pressure system.

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Table 1. Distribution of Days According to Position in High-Pressure System.

	Classification Type[a]					
Site	1	2	3	4	5	6
McKee	2	14	4	10	3	3
Busick	4	13	5	9	3	0
Total	6	27	9	19	6	3

[a] For explanation of types 1 through 5 see Figure 1. Type 6 did not fall into any category.

various gamma energies, and the radionuclide decay scheme.

Continuous ozone measurements were made with a McMillan 1100-2 chemiluminescent analyzer calibrated by the NBKI method.

The relative position of the site in the high-pressure system was determined each day by examining surface weather maps and local meteorological parameters measured at the sites. Because precipitation would tend to scavenge the ^{7}Be aerosols, only days with no precipitation were considered.

Results

During the sampling periods, seven cold fronts and the subsequent high-pressure systems passed through McKee and seven passed through Busick. The total number of non-precipitation days on which valid data was collected was 27 at McKee and 20 at Busick.

The location of the site in the high-pressure system was classified into five overlapping regions, illustrated in Figure 1. Those days which did not fall into any of the five regions are listed as unclassified. Table 1 gives the distribution of the days during this study. Since some days fall into more than one category, the total in Table 1 exceeds 47. Only the highest 50% of the ^{7}Be concentrations are considered in Table 2, which illustrates the high proportion of ^{7}Be values on the back side of the high-pressure system. All six of the cases immediately behind the cold front in the absence of precipitation were in the lower 50%. Table 3 categorizes the days in the higher 50% of the maximum hourly ozone concentrations with respect to position within the high-pressure system. The distribution of ozone is very similar to that of the ^{7}Be, with the greatest frequency of the high ozone days occurring on the back side of a high-pressure system.

In terms of absolute concentrations, a significant difference also occurs with respect to position in the high-pressure system. For example, at Busick the average ^{7}Be values on the back side of the high-pressure system was 372 disintegrations per minute (dpm) per 1000 standard cubic meters of air while on the front side it was 261 dpm/KSCM. This represents a 43% difference. At McKee, the mean ^{7}Be concentration was 162 dpm/KSCM on the front side compared to 273 dpm/KSCM on the back side of the high-pressure system. This corresponds to a 69% difference.

The difference between the mean ^{7}Be value at Busick (326 dpm/KSCM) and McKee (228 dpm/KSCM) is not completely understood, but it may have been an instrumental artifact. Different pairs of high-volume samplers were employed at the two sites. For the pair used in McKee, the maximum flow was about 68 m3/hr while at Busick the flow was 134 m^3/hr. At McKee, however, there was evidence of the exhaust gas being recirculated because carbon (presumably from the brushes) was noted on the filter. No carbon was visible on the Busick filters. Consequently, the authors feel that the absolute ^{7}Be concentration from McKee is uniformly too low. The relative difference between front side and back side ^{7}Be values at McKee, however, should be unaffected by this artifact.

Discussion

As expected, the ozone exhibited a definite tendency towards higher concentrations on the back side of the high-pressure system. Unexpectedly, the ^{7}Be also followed the same pattern which suggests that there is more stratospheric air on the back side of the high-pressure system than on the front side. In addition, it suggests that the ozone concentration due to stratospheric intrusion is higher on the back side than on the front side which is the area of the high-pressure system just behind the cold front. These results conflict with the previously cited conclusions of EPA [U. S. EPA, 1975] and *Ludwig et al.* [1977] and suggest that the contribution of stratospheric ozone during episodes of high ozone in the Eastern U.S. may be higher than previously thought. This is supported by the results of *Ferman and Monson* [1978] who reported a 0.60 correlation coefficient between 24-hour average values of O_3 and ^{7}Be at the McKee site.

There is a plausible explanation which accounts for the occurrence of higher ^{7}Be on the back side of high-pressure systems and it is consistent with previously reported ambient observations. The intrusion occurs behind the surface cold front and the stratospheric air is transported downward but does not penetrate the boundary layer. It then is advected to the back side of the high via the clockwise circulation. Since the back side of the high-pressure systems during the

Table 2. Meteorological Factors Associated with the Highest 50 percent of the Beryllium-7 Values

	McKee		Busick		Total		% of Days in
Classification Type	No.	%[a]	No.	%[a]	No.	%[a]	Highest 50%
1. Warm air Mass before cold front	2	14	3	30	5	21	83
2. Back side of high-pressure system	10	71	8	80	18	75	67
3. Center of high-pressure system	2	14	3	30	5	21	56
4. Front side of high-pressure system	3	21	2	20	5	21	26
5. Immediately behind cold front	0	0	0	0	0	0	0
6. Unclassified	1	7	0	0	1	4	33
Total Actual Days in Upper 50%	14		10		24		
Total Category Days in Upper 50%	18		16		34		

[a] percent in the highest 50%

Table 3. Meteorological Factors Associated with the Highest 50 percent of the O_3 Concentrations

Classification Type	McKee No.	McKee %[a]	Busick No.	Busick %[a]	Total No.	Total %[a]	% of Days in Highest 50%
1. Warm air Mass before cold front	1	7	3	30	4	17	67
2. Back side of high-pressure system	9	64	9	90	18	75	67
3. Center of high-pressure system	3	21	3	30	6	25	67
4. Front side of high-pressure system	3	21	1	10	4	17	44
5. Immediately behind cold front	0	0	0	0	0	0	0
6. Unclassified	2	14	0	0	2	8	67
Total Actual Days in Upper 50%	14		10		24		
Total Category Days in Upper 50%	18		16		34		

[a] percent in the highest 50%

warmer part of the year is characterized by higher temperatures which produce convective activity, the stratospheric air is mixed to the surface there. This would account for the higher ^7Be.

Evidence for the existence of elevated layers of stratospheric ozone in the lower troposphere above the boundary layer in high-pressure systems has been presented by *Hathorn and Walker* [1977], *Sticksel* [1977], and *Johnson et al.* [1979].

Summary

The results from a 1976 study in rural Kentucky and North Carolina indicate that the highest surface concentrations of ^7Be are generally found on the back side of a high-pressure system. This result is surprising because it was previously assumed that intrusion of stratospheric air to the ground was limited to a small area on the front side of the high-pressure system behind the cold front.

In terms of ozone and photochemical oxidant-control strategies, these results could have significant implications. EPA has concluded that intrusion of stratospheric ozone is significant (30 to 50 ppb) only in this small area behind the cold front on the front side of the high-pressure system and that the higher ozone levels found on the back side of the high-pressure system are a result of photochemistry involving anthropogenic precursor emissions. This study suggests that the stratospheric ozone contribution on the back side of the high-pressure system is higher than on the front side.

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