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TROPOSPHERIC OZONE AND AEROSOL VARIABILITY OBSERVED AT HIGH LATITUDES WITH AN AIRBORNE LIDAR

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ABSTRACT

Large-scale summertime (July-August) distributions of O_3 and aerosols were observed in a broad range of atmosphere conditions over the tundra, ice, and ocean regions near Alaska in 1988 and over the lowlands and boreal forests of Canada in 1990. The tropospheric O_3 budget in the high-latitude regions was found to be strongly influenced by stratospheric intrusions, and deposition at the surface was found to be the main sink for O_3 in the troposphere. Enhanced levels of O_3 were observed in plumes from fires in Alaska and Canada. This paper discusses the large-scale variability of O_3 and aerosols observed in the high-latitude regions during these field experiments.

1. INTRODUCTION

The NASA Atmospheric Boundary Layer Experiment (ABLE-3A) was conducted near Barrow and Bethel, Alaska, from July 10 to August 12, 1988, and a second field experiment (ABLE-3B) was conducted over eastern Canada from July 6 to August 16, 1990. The primary objective of these experiments was the investigation of the sources and sinks for tropospheric ozone (O_3) in the high-latitude regions of North America. During these field experiments, the NASA Langley Research Center's airborne DIfferential Absorption Lidar (DIAL) system [Browell et al., 1983; Browell, 1989] was flown on the NASA Wallops Electra aircraft to provide simultaneous remote measurements of O_3 and aerosols above and below the aircraft from near the surface to above the tropopause. An O_3 measurement accuracy of less than 10% or 3 ppbv (parts per billion by volume), whichever is larger, was obtained [Browell et al., 1983, 1985]. In addition, comparisons between airborne DIAL and in situ O_3 measurements were made throughout the field experiments to verify that the DIAL accuracy was in agreement within the above stated accuracy. Detailed characteristics of the current airborne DIAL system and the O_3 DIAL technique are given by Browell [1989].

This airborne lidar system has been used in many previous experiments to study tropospheric O_3 and aerosol distributions [see e.g., Browell et al., 1987, 1988, 1990]. This paper discusses the results of the airborne DIAL measurements of O_3 and aerosols that were made during ABLE-3A&B in a broad range of atmospheric conditions over the ice, tundra, and ocean regions of Alaska and over eastern Canada.

2. ARCTIC AND SUBARCTIC MEASUREMENTS

A total of 33 missions were flown as part of ABLE-3A, including 23 flights in the vicinity of Barrow and Bethel, Alaska, and 10 survey flights between the NASA Wallops Flight Facility at Wallops Island, Virginia, and Barrow. The tropospheric O_3 budget in the summertime high-latitude regions near Alaska was found to be strongly influenced by stratospheric intrusions. Regions of low aerosol scattering and enhanced O_3 mixing ratios in the troposphere were usually correlated with descending air from the lower stratosphere. In the vicinity of Barrow, Alaska, O_3 was generally in the range of 20-30 ppbv below 2 km; however, air with O_3 >40 ppbv and low aerosol scattering was found as low as 1 km with continuity in air mass characteristics to the tropopause level. Ozone measurements in the vicinity of Barrow showed large regions where O_3 exceeded 45 ppbv extending down to about 1 km altitude. Using the zenith DIAL O_3 data, these regions of elevated O_3 were traced back to large-scale stratospheric intrusions that generally increased the O_3 levels in the upper troposphere [Shipham et al., 1992]. Ozone mixing ratios of over 100 ppbv were found as low as 6 km in the presence of strong intrusion events. The variability in O_3 in the mid to upper troposphere affects the O_3 distribution in the lower troposphere, and due to the frequency of the observed intrusions, this process plays an important role in the tropospheric O_3 budget at high latitudes.

Several cases of continental polar air masses were examined during this experiment. The aerosol scattering associated with these air masses was very low, and the atmospheric distribution of aerosols was quite homogeneous for those air masses that had been transported over the ice for at least 3 days. The average O_3 profile (Figure 1) derived from three cases of continental polar air masses had a nearly constant gradient of 7 ppbv/km from 30 ppbv at 600 m to 55 ppbv at 4 km. This distribution reflects the influence of downward transport of O_3 from the upper troposphere at high latitudes and the destruction of O_3 near the surface.

Five cases were studied to determine the average background O_3 profile over the tundra region of southwestern Alaska (Figure 2). Near the surface, the tundra and continental polar O_3 levels are about the same; however, the average O_3 value above 1.5 km is 10-20% less than for the continental polar cases. This is thought to be due to the influence of more frequent stratospheric intrusions at the higher latitudes associated with the continental polar air masses. Zenith O_3 profiles in the region of 56-62°N showed good agreement from 2.5-10.0 km in altitude with a

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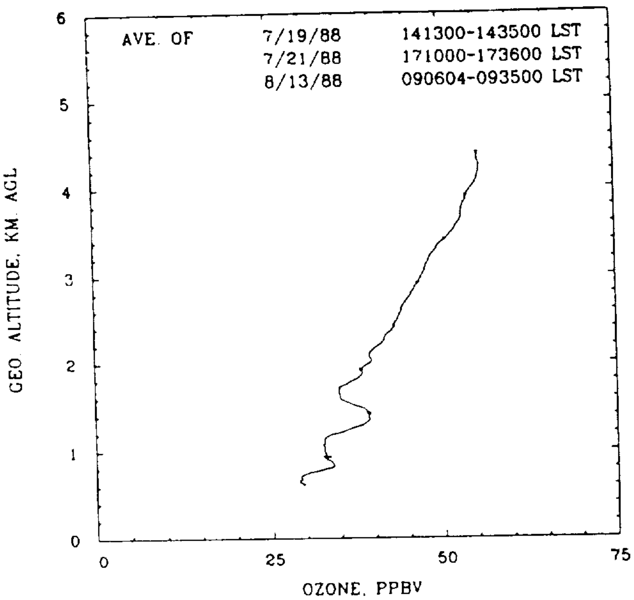

Fig. 1. Average continental polar O_3 profile derived from DIAL measurements on the dates shown.

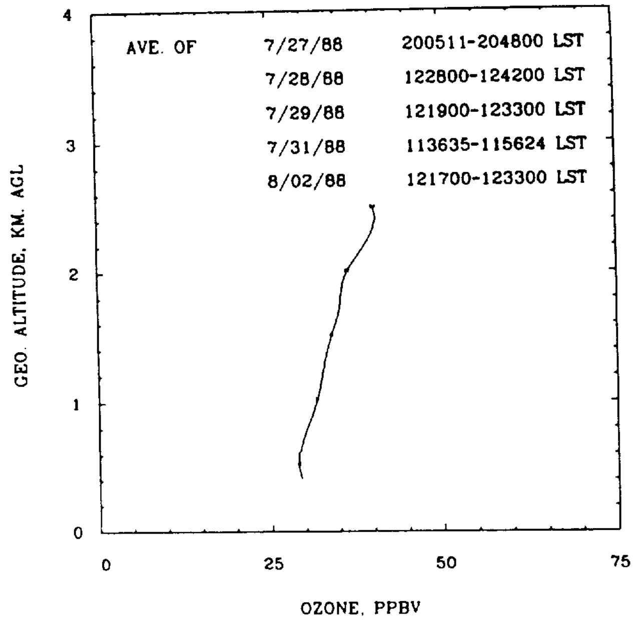

Fig. 2. Average tundra O_3 profile derived from DIAL measurements on the dates shown.

5.5 ppbv/km gradient determined below 2.5 km over the tundra.

Several cases of enhanced O_3 were observed during ABLE-3A in conjunction with enhanced aerosol scattering in layers in the free troposphere. Multiple aerosol layers were often observed coming from the many fires in Alaska. In some cases, O_3 in these layers was found to be more than 50% higher than the average background O_3 distribution over the tundra region. The products of biomass burning was found to significantly alter the chemical composition of the troposphere in the Arctic [Wofsy et al., 1992], as was shown to happen over the Amazon Basin during the dry season [Andreae et al., 1988].

An analysis of the percentage of the troposphere containing air having significantly different characteristics was conducted using the nadir and zenith DIAL data. The troposphere was found to contain air that fell into four general categories with the following properties: (1) mixed-layer air with enhanced aerosol scattering and with O_3 <30 ppbv; (2) plumes in the free troposphere with significantly enhanced relative aerosol scattering; (3) stratospheric intrusions in free troposphere with significantly enhanced O_3 levels and with low aerosol scattering; and (4) background air in the free troposphere with none of the above characteristics.

An unperturbed background O_3 profile was defined as having a gradient of 5.2 ppbv km^{-1} from 29 ppbv at 500 m to 68 ppbv at 8 km. Table 1 presents the results of an analysis of the DIAL data from ABLE-3A to determine the percentage of the troposphere containing the above types of air in particular altitude and latitude regions. The troposphere was divided into 2-km intervals from the surface to 8 km, and the data were grouped into two latitude intervals: a subarctic region (57-65°N) and an arctic region (>65°N).

Mixed-layer air was found to be contained almost entirely within the 0-2 km altitude range, and its vertical extent was a factor of 3 larger in the tundra region of southwestern Alaska than at higher latitudes (>65°N) with extensive regions of ice. Plumes were most often observed below 4 km where they accounted for almost 10% of the atmosphere. Plumes were sometimes found at the higher altitudes but with very reduced extent. The measured tropospheric extent containing plumes at

Table 1. Percentage of Troposphere in ABLE-3A Containing Different Air Types

	Altitude								Tropospheric Coverage	
	0-2 km		2-4 km		4-6 km		6-8 km		0-8 km	
N Lat. Range:	57-65	≥65	57-65	≥65	57-65	≥65	57-65	≥65	57-65	≥65
Air Type										
Mixed Layer	17	6	0	0	0	0	0	0	4	1
Plumes	9	10	10	11	1	3	2	2	5	7
Strat. Int.	12	18	30	37	51	57	57	42	38	38
Backgnd (Free Trop)	62	66	60	52	48	40	41	56	53	54

high-latitudes (>57°N) during ABLE-3A was 6% with a maximum of 11% for plumes in the 2-4 km range. While the percentage for plumes may be relatively small, they are a concentrated source of gases and aerosols that once mixed with the ambient air can significantly alter the chemical composition of the lower troposphere. The influence of stratospheric intrusions on the O_3 distribution in the troposphere increased with altitude over the entire high-latitude region investigated (>57°N) from an average of 16% in the 0-2 km range to over 53% above 4 km. At the higher latitudes (>65°N), the tropospheric extent of the intrusions was 18% below 2 km. The measured tropospheric extent containing stratospheric intrusions at high-latitudes (>57°N) was 38% with a maximum extent of 53% in the 4-6 km range. The background air was defined as tropospheric air that did not fall into any of the above categories, and it accounted for 44-64% of the troposphere (0-8 km). When the extent of the types of air is compared in the two latitude regions (57-65°N;>65°N), there is no apparent trend in any of the categories, except for the mixed layer extent, which was discussed earlier.

3. MEASUREMENTS OVER EASTERN CANADA

During ABLE-3B, 22 flights were made in eastern Canada from bases in North Bay and Goose Bay, Canada. In addition to the types of air identified for ABLE-3A, two additional types of air were observed during ABLE-3B: (1) convective outflows in the free troposphere with significantly enhanced aerosol scattering and with O_3 lower than the background level at that altitude; and (2) low O_3 air with O_3 more than 20% below the background value at that altitude and with low aerosol scattering. Background O_3 profiles over Canada are shown in Figure 3 for the lower troposphere and in Figure 4 for the upper troposphere. An analysis similar to that conducted for ABLE-3A was done for the data collected during ABLE-3B. Table 2 presents the results of an analysis to determine the percentage of the troposphere containing different types of air during ABLE-3B. An additional category was added to the table to take into account the cases where the tropopause was below 12 km.

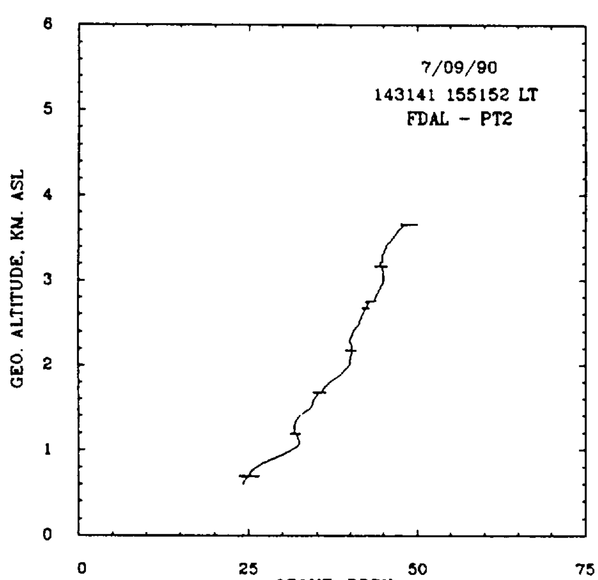

Fig. 3. Average background O_3 profile for lower troposphere observed over James Bay lowlands on July 9, 1990.

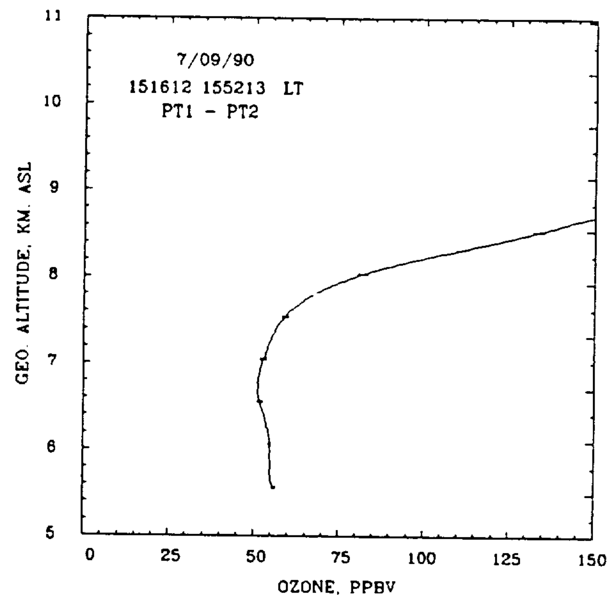

Fig. 4. Average background O_3 profile for upper troposphere observed over James Bay lowlands on July 9, 1990.

The mixed-layer air over the lowlands and boreal forests of Canada was found to cover over 25% of the 0-2 km altitude range. This is more than twice the area occupied by the mixed-layer air over the tundra in ABLE-3A. Plumes were most often observed below 4 km where they accounted for almost 25% of the atmosphere. The amount of plumes observed during ABLE-3B over Canada was more than 2.5 times the amount observed over the tundra regions of Alaska during 1988. Of the plumes observed over Canada, over 60% had elevated O_3 (>20% above the background O_3 level). While these plumes were old enough to exhibit substantial photochemical O_3 production, a significant number of young plumes from near-by fires were observed that did not have enhanced O_3.

Stratospheric intrusions were observed on 68% of the missions flown during ABLE-3B, where they were found to significantly influence the O_3 distribution in the middle and upper troposphere. Over 40% of the atmosphere between 4-8 km had enhanced O_3 and low aerosol scattering associated with these intrusions. Even in the lower troposphere there was a substantial occurrence of intrusions. In the 45-55°N latitude range, the area of the troposphere that contained intrusions was about 20% lower than the area containing intrusions at the higher latitudes in ABLE-3A. A combination of the lower tropopause, the influence of the polar jet, and the large-scale subsidence in the Arctic is thought to be the reason for the difference in the observations. Stratospheric intrusions represented 33% of the entire troposphere over Canada during ABLE-3B. This is only slightly lower than the 38% found during ABLE-3A in the Arctic and Subarctic.

Low O_3 air was observed on several occasions in the upper troposphere, and in total, it covered almost 20% of the atmosphere above 6 km. These air masses had their origins in the tropics according to their back trajectory analyses. An additional source of low O_3 to the free troposphere is associated with the vertical transport of boundary layer air resulting from cloud activity. The amount of air associated with convective outflows is generally less than 5%; however, this is probably an underestimate due to the limitation of having to make the lidar measurements outside of optically thick clouds.

Table 2. Percentage of Troposphere in ABLE-3B Containing Different Air Types

Air Type	Altitude Range 0-2 km	2-4 km	4-6 km	6-8 km	8-10 km	10-12 km	Tropospheric Coverage 0-12 km
Mixed Layer	36	8	0	0	0	0	7
Background (Free Trop.)	17	40	43	40	34	23	33
Plumes	26	24	2	2	2	1	10
Stratospheric Intrusion	8	24	45	41	31	51	33
Low Ozone Air	4	1	9	13	26	19	12
Convective Outflow	9	3	1	4	5	3	4
Low Tropopause	0	0	0	0	2	3	1

The background air accounted for 33% of the troposphere (0-12 km). This compares to 33% for stratospheric intrusions, which was the largest component of the troposphere. The low O_3 air, plumes, and boundary layer air all had comparable amounts of tropospheric coverage; however, each has distinctly different chemistry and influence of the troposphere.

4. CONCLUSIONS

The tropospheric O_3 budget in the Arctic was found to be strongly influenced by stratospheric intrusions. Regions of low aerosol scattering and enhanced O_3 mixing ratios were usually correlated with descending air from the upper troposphere or lower stratosphere. In continental polar air masses, the aerosol scattering was also low, and the average O_3 profile had a nearly constant gradient of 7 ppbv/km from about 30 ppbv at 600 m to 55 ppbv at 4 km. This distribution reflects the influence of downward transport of O_3 from the upper troposphere at high latitudes and the destruction of O_3 near the surface. Over the tundra of southwestern Alaska, the average O_3 level observed in the mixed layer was in the 25-35 ppbv range, and the average O_3 profile above the mixed layer was 10-20% lower than for the continental polar air masses, possibly due to fewer intrusions at lower latitudes. Several cases of enhanced O_3 were observed during ABLE-3A in conjunction with enhanced aerosol scattering in layers in the free troposphere. These layers were coming from regions where biomass burning was occurring. .

The background O_3 level observed in the mixed layer over Canada during ABLE-3B was in the 20-30 ppbv range indicating that the wetlands and boreal forest regions of Canada provided a stronger surface sink for O_3 than the tundra regions of Alaska. Except for cases of biomass burning plumes, aerosols were generally negatively correlated with O_3, which indicates that their primary source was at the surface. As with ABLE-3A, numerous cases of stratospheric intrusions were observed with evidence that their effect extended well down into the lower troposphere. Deep convective cloud activity and associated outflows were found to be the primary mechanism for vertically transporting O_3-poor air from near the surface to the upper troposphere. Air containing abnormally low O_3 was observed on a number of missions, and some of these air masses were thought to have originated in the tropics. Plumes from near-by fires had enhanced aerosol loading without any significant increase in O_3 levels. Aged plumes had only slightly enhanced aerosol scattering; however, the O_3 was enhanced to greater than 50 ppbv due to photochemical O_3 production.

5. ACKNOWLEDGMENTS

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REFERENCES

Andreae, M. O., E. V. Browell, M. Garstang, G. L. Gregory, R. C. Harriss, G. F. Hill, D. J. Jacob, M. C. Pereira, G. W. Sachse, A. W. Setzer, P. L. Silva Dias, R. W. Talbot, A. L. Torres, and S. C. Wofsy, Biomass-burning emissions and associated haze layers over Amazonia, J. Geophys. Res., 93, 1509-1527, 1988.

Browell, E. V., Differential absorption lidar sensing of ozone, Proc. IEEE, 77, 419-432, 1989.

Browell, E. V., A. F. Carter, S. T. Shipley, R. J. Allen, C. F. Butler, M. N. Mayo, J. H. Siviter, Jr., and W. M. Hall, NASA multipurpose airborne DIAL system and measurements of ozone and aerosol profiles, Appl. Opt., 22, 522-534, 1983.

Browell, E. V. , S. Ismail, and S. T. Shipley, Ultraviolet DIAL measurements of O_3 profiles in regions of spatially inhomogeneous aerosols, Appl. Opt., 24, 2827-2836, 1985.

Browell, E. V., E. F. Danielsen, S. Ismail, G. L. Gregory, and S. M. Beck, Tropopause fold structure determined from airborne lidar and in situ measurements, J. Geophys. Res., 92, 2112-2120, 1987.

Browell, E. V., G. L. Gregory, R. C. Harriss, and V. W. J. H. Kirchhoff, Tropospheric ozone and aerosol distributions across the Amazon Basin, J. Geophys. Res., 93, 1431-1451, 1988.

Browell, E. V., G. L. Gregory, R. C. Harriss, and V. W. J. H. Kirchhoff, Ozone and aerosol distributions over the Amazon Basin during the wet season, J. Geophys. Res., 95, 16,887-16,901, 1990.

Shipham, M. C., A. S. Bachmeier, D. R. Cahoon, Jr., and E. V. Browell, Meteorological overview of the ABLE-3A flight series, J. Geophys. Res., 97, in press, 1992.

Wofsy, S. C., et al., Influence of natural fires and anthropogenic emissions on atmospheric chemistry in remote areas, J. Geophys. Res., 97, in press, 1992.