AIRBORNE LIDAR MEASUREMENTS OF OZONE AND AEROSOL DISTRIBUTIONS OVER NORTH AMERICA AND THE WESTERN ATLANTIC OCEAN DURING THE INTEX-NA FIELD EXPERIMENT

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ABSTRACT

The NASA Langley airborne Differential Absorption Lidar (DIAL) made extensive measurements of tropospheric ozone (O₃) and aerosol distributions during the Intercontinental Chemical Transport Experiment - North America (INTEX-NA) conducted from July 1 to August 14, 2004. The airborne DIAL system was operated simultaneously in nadir and zenith modes from the NASA DC-8 aircraft for complete atmospheric cross sections from near the surface to above the tropopause along the flight track. Largescale distributions of O₃ and aerosols were obtained over North America (NA) and the western Atlantic Ocean on seventeen long-range flights of the DC-8. Air masses with enhanced O₃ and aerosols in the lower troposphere were observed in conjunction with the outflow of pollution from the United States (US) over the western Atlantic. Biomass burning plumes from distant Alaskan fires were frequently observed in the mid troposphere over NA as were air masses with mixtures of stratospheric air and polluted tropospheric air. The amount of O₃ from the stratosphere was found to be about 20% at low latitudes (<30°N) and more than 40% at high latitudes (> 40° N). The first average latitudinal and longitudinal distributions of O₃, aerosols, and potential vorticity (PV) were determined from the INTEX-NA data, and the relative occurrence of different air mass types was examined in different latitude and longitude regions. These results are discussed with respect to sources, sinks, and transport of air masses observed during this field experiment.

1. INTRODUCTION

NASA Langley Research Center's airborne DIAL system participated in the INTEX-NA field experiment to investigate the large-scale variations of O_3 and aerosols associated with sources of pollution over NA

and the transport of this pollution over the western Atlantic Ocean. The airborne DIAL system was operated on the DC-8 aircraft which was based in three locations for this field experiment: NASA Dryden Flight Research Center (DFRC), Edwards Air Force Base (AFB), California; Mid-America Airport, St. Louis, Missouri; and Pease Airport, Portsmouth, New Hampshire. Remote profile measurements of O_3 and aerosol backscatter and depolarization were made from near the surface to above the tropopause along the flight track of the DC-8. In situ measurements of O₃, aerosols, and a wide range of trace gases were made onboard the DC-8 and correlated with the remote DIAL measurements. Meteorological analyses of PV distributions along the flight track were used to indicate the fraction of observed O_3 that could be attributed to stratosphere-troposphere exchange. Fiveday backward trajectories were also used to indicate the possible origin of observed air masses.

The airborne DIAL system used ultraviolet (UV) DIAL wavelengths of 288 and 300 nm for the measurement of O_3 profiles and multiple-wavelength aerosol backscatter profile measurements at 576/599 (nadir/zenith) and 1064 nm. Aerosol depolarization profiles were also made simultaneously below and above the aircraft at 576 and 599 nm, respectively. This airborne DIAL system and the techniques used for these measurements have been described in detail in many previous publications [see e.g., 1-3].

Seventeen long-range flights of the airborne DIAL system were accomplished during the INTEX-NA field experiment for a total of more than 130 hours of data. There were two data flights from Edwards AFB over the eastern Pacific Ocean to examine the types of air masses coming into the US from the west. There were two separate deployments to St. Louis with a total of five data flights, and there were ten data flights from Portsmouth. The locations of the various flight tracks

are shown in Fig. 1 along with the different longitude regions that were used for grouping the flight results. Examples of the airborne DIAL O_3 and aerosol measurements made during INTEX-NA are described in Sections 2 and 3. The data obtained during INTEX-NA were analyzed to provide a comprehensive understanding as to the large-scale distribution of O_3 and aerosols over NA and the relative contribution of various processes in determining the tropospheric O_3 and aerosol budgets, and examples of these results are discussed in Sections 3.

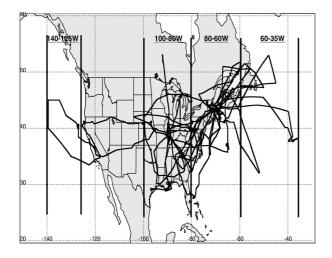


Fig. 1 INTEX-NA flight tracks.

2. OZONE INTERCOMPARISONS

In situ ozone measurements (chemiluminescent technique) made on board the DC-8 during aircraft ascents and descents were compared with DIAL O_3 profiles in the nadir and zenith direction on as many flights as possible during INTEX-NA. In addition, ozonesonde profiles were sometimes used to compare with DIAL O_3 profiles, and this allowed for O_3 comparisons at altitudes above the ceiling of the DC-8. Since the airborne DIAL O_3 measurements have been previous validated in many other field experiments, only two examples of these comparisons are shown below from INTEX-NA confirming that the airborne DIAL system continued to make excellent O_3 profile measurements during this field experiment.

3. OZONE AND AEROSOL OBSERVATIONS

Profiles of O_3 ; aerosol scattering ratios at 588 nm (average of 576 and 599 nm) and 1064 nm; and aerosol depolarization at 588 nm were obtained below and above the NASA DC-8 on long-range flights during INTEX-NA. Fig. 3 presents an example of the types of airborne DIAL data obtained on a typical flight during this campaign. The data presented in Fig. 3 are for the entire flight on 22 July 2006, which was flown from

Portsmouth, New Hampshire. The O_3 data presented in the figure includes the nadir and zenith DIAL O_3 profiles and interpolated O_3 values across the aircraft altitude using the in situ O_3 data as a hard constraint for the interpolation. On this mission the DC-8 flew over the NOAA WP-3B aircraft, the J-31 aircraft, and the NOAA Ron Brown ship, and it flew under the TERRA satellite. We were conducting coordinated studies for atmospheric chemistry, radiation science, and satellite validation objectives . The location of these encounters are indicated in the figure.

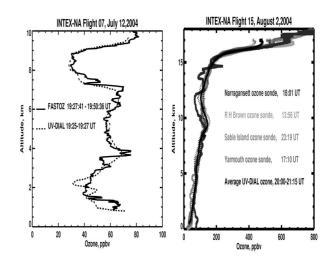


Fig. 2 In situ O₃ comparisons with DIAL O₃ profiles.

The observed O_3 distribution shows high values near the surface (<2 km) associated with the outflow of pollution from the US. At low altitudes on the northern end of the flight track, the low O₃ (<40 ppbv) was associated with the outflow of clean continental air. Throughout the free troposphere, the levels of O_3 were elevated (>75 ppbv) in many locations. Long-range transport of pollution and some stratospheretroposphere exchange (STE) was responsible for the enhanced O₃. Evidence of a stratospheric intrusion can be seen with high O_3 (>125 ppbv) descending down to about 7 km near the start and end of the flight. The aerosol scattering distribution shows high values in the lower troposphere associated with the continental pollution with some enhanced aerosol scattering in the free troposphere associated with long-range transport of pollution from Alaska. The aerosol depolarization distribution helps to delineate the source of some of the pollution since the aerosols from urban pollution tends to be dominated by sulfate aerosols which are small and spherical with no depolarization, while the aerosols in biomass burning plumes tend be large, non-spherical with some (6-8%) depolarization. Cirrus clouds can be readily detected by their very high scattering ratios and high (30-50%) depolarizations. The depolarization distribution on this flight showed the extensive presence of biomass burning plumes with enhanced aerosol loading and enhanced photochemical O_3 production. Backward trajectories indicated that these plumes had come from fires in Alaska or western Canada. Examples of O_3 and aerosol distributions observed in other regions investigated during INTEX-NA will be discussed in this paper.

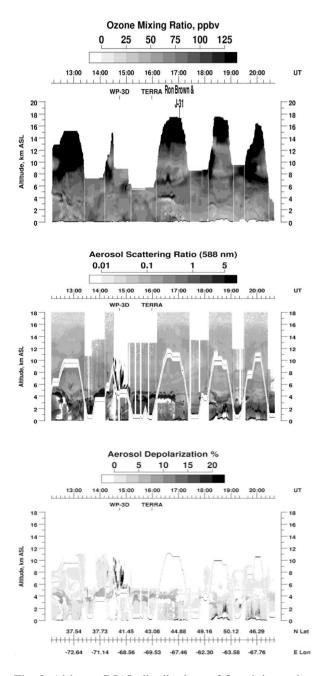


Fig. 3 Airborne DIAL distributions of O_3 mixing ratios (top), aerosol scattering ratios (middle), and aerosol depolarizations (bottom) obtained on 22 July 2004 (Flight 11 - River of Pollution).

The O₃ and aerosol data from each flight were binned

by latitude and altitude and then averaged in different longitude regions to create an average latitudinal distribution which better represented the average conditions observed during INTEX-NA. An example of these results is shown in Fig. 4. The average latitudinal distribution of O₃ measured in the 60-80°W longitude region (see Fig. 1) is shown at the top of Fig. 4. For this calculation only O_3 below the tropopause was included in the average O₃ distribution. The average low-altitude out flow of pollution in the mid latitude region (35-45°N) can be readily seen in the figure with the cleaner continental outflow at the high latitudes and the low O_3 over the western Atlantic at low latitudes. The average O₃ generally increased with altitude over the entire latitude range, and O₃ exceeded 75 ppbv above ~7 km nearly everywhere. To investigate the source of the O_3 in the mid to upper troposphere, the relationship between O₃ and PV was established in the lower stratosphere using the remote DIAL O₃ measurements and the calculation of PV from a National Weather Service (NWS) meteorological analysis. The O₃/PV ratio was found to decrease slightly with time in the lower stratosphere from about 45 ppbv/PVU on day 182 to 31 ppbv/PVU on day 226.

By taking the PV distributions from each flight and scaling it based on the O₃/PV ratio at that time, an estimate was obtained for the average amount of O₃ transported into the troposphere from the stratosphere in the same way as the average latitudinal O_3 distribution was determined. This technique has been applied in many previous campaigns to assess the recent affects of STE on the composition of the troposphere [see e.g., 3-4]. This average amount of stratospheric-derived O₃ was then subtracted from the observed amount of O_3 in the troposphere to provide the amount of O_3 associated with a tropospheric source. This is termed the "tropospheric average O₃," and its distribution is shown in the middle of Fig. 4. In addition, the ratio of the stratospheric-derived O₃ to the total average O3 is the "stratospheric fraction of average O₃," and its distribution is shown at the bottom of Fig. 4. From these figures it can be seen that in the upper troposphere (>10 km) at latitudes north of about 37° N, more than 50% of the O₃ is from the stratosphere, and on average in this latitude region, more than 40% of the O3 observed across the troposphere is from the stratosphere. South of about $36^{\circ}N$, only about 20% of the O₃ is from the stratosphere. The O_3 with a source in the troposphere can be seen to be distributed across all latitudes and altitudes with a particular enhancement in the mid troposphere due to long-range transport of air from distant locations such as Asia, and there is a particularly large amount of photochemically produced O_3 in the mid to upper troposphere at low latitudes which is also an area where long-range transport and photochemistry is enhanced. A similar analysis was performed in different longitude regions across NA, and these results are also discussed in this paper.

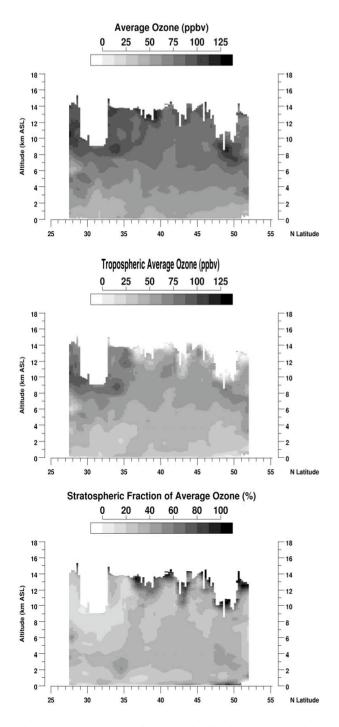


Fig. 4 Average latitudinal O_3 distribution observed in 60-80°W longitude region during INTEX-NA (top); average O_3 contributed only from tropospheric photochemical processes (middle); and fraction of O_3 attributed to transport from the stratosphere (bottom).

4. CONCLUSIONS

Enhanced O₃ concentrations and aerosol loading in the lower troposphere were found to be associated with US pollution and advection of those air masses over the western Atlantic Ocean. Long-range transport of fire plumes from Alaska was frequently observed with depolarizing aerosols over eastern North America. Average latitudinal O₃ and aerosol distributions were determined and related to large-scale atmospheric processes. Frequent mixtures of stratospheric air and polluted tropospheric air masses were observed in the middle and upper troposphere with over 40% of the O_3 observed north of 37°N was from the stratosphere. In general the composition of the troposphere was found to be a complex mixture of local and distant pollution sources and transport of stratospheric air into the This paper discusses the types and troposphere. characteristics of air masses encountered during INTEX-NA, including frequency and location (latitude and altitude) of observations, and their related atmospheric processes.

5. ACKNOWLEDGEMENTS

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6. REFERENCES

1. Browell, E. V., S. Ismail, and W. B. Grant, Differential Absorption Lidar (DIAL) measurements from air and space, *Applied Physics-B*, 67, 399-410, 1998.

2. Browell, E. V.: Global Tropospheric Ozone Investigations. Conference Digest, 19th International Laser Radar Conference, Annapolis, MD, July 6-10, 1998, NASA/CP-1998-207671/PT1, pp. 257-260.

3. Browell, E. V., et al., Large-scale ozone and aerosol distributions, air mass characteristics, and ozone fluxes over the western Pacific Ocean in late winter/early spring, *J. Geophys. Res.*, 180(D20), 8805, doi:10.1029/2002JD003290, 2003.

4. Browell, E. V., et al., Large-scale air mass characteristics observed over the remote tropical Pacific Ocean during March-April 1999: Results from PEM Tropics B Field Experiment, *J. Geophys. Res.*, *106*, 32,481-32,501, 2001.