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Profiling of Ozone in the Free Troposphere by the Lidar Technique

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Measurements of ozone in the troposphere have become increasingly important during the past decade with reports of ozone increases on global scales that not only threaten the vitality of the plant and the animal kingdom, but can also, since ozone is a greenhouse gas, contribute to global warming. The lidar application that is described here is specifically designed to accommodate the needs of the scientific community to accurately measure tropospheric ozone over a period of many years as one indicator of global change, and is intended as a prototype for a network of tropospheric ozone lidars.

Key Words: Ozone, Troposphere, Differential absorption lidar

The vertical distributions of ozone concentrations in our atmosphere have been studied for many decades. In recent years, substantial decreases in stratospheric ozone and increases in tropospheric ozone have been attributed to man. Not only are the tropospheric increases insufficient to offset the large decreases in the stratosphere, these ozone increases in the lower part of the atmosphere may contribute to global warming, can provide a significant human health hazard in urban areas, and appear to adversely affect the vitality of plants and animals. These and other factors have contributed to an escalation of interest in accurate measurement of ozone distributions.

Nature provides a challenge for scientists who wish to extract an anthropogenic signature of ozone change from the measurements. Ozone is naturally produced and destroyed in our atmosphere, resulting in natural ozone variations as a function of altitude, latitude, and time that are not easily separated from changes due to man. Whether we are studying ozone changes in the stratosphere, where concentrations are high, or in the troposphere, where its natural abundance is much less, we need accurate measurements at a variety of latitudes made over periods of many years and at a frequency sufficient to statistically identify increasing anthropogenic components. Requirements of accuracy, long-term consistency, and frequency of the measurements

strongly influence the final configuration of ozone lidar systems.

The measurement of ozone in the free troposphere by the differential absorption lidar technique (DIAL) has been practiced for more than a decade. The instruments are usually ground-based although aircraft applications have also been quite successful¹⁾. The particular configuration we describe is located at Fritz Peak Observatory (40°N, 105°W, 2.7 km ASL), near Boulder, Colorado, USA, and has been making routine ozone observations since September 1992. It provides profiles from 4 km ASL (above mean sea level) up to the tropopause (about 12 to 14 km ASL) day or night in from 1 to 4 minutes with a 1 km altitude resolution and better than 10% absolute accuracy. A second version will be installed at the NDSC site at Mauna Loa, Hawaii.

DIAL designs suitable for tropospheric ozone profiling are significantly different from their stratospheric counterparts²⁾, the most obvious difference results from the ultraviolet absorption cross section of ozone³⁾ (Fig. 1) and the vertical distribution of ozone in the atmosphere (Fig. 2). In general, the DIAL method requires two wavelengths with differing ozone absorption cross sections. Since ozone concentrations are much less in the troposphere than in the stratosphere (Fig. 2), wavelengths with a larger absorption cross section are selected

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Fig. 1 Ozone absorption cross sections at 25 °C from 245 nm to 340 nm from Reference 3.

Fig. 2 Typical mid-latitude ozone profile. A vertical resolution of 5 meters is provided by a dual-beam uv-absorption photometer that started measurements at approximately 1 km altitude on board a balloon launched near Palestine, Texas on October 10, 1983. The increase in ozone at approximately 15 km altitude ASL marks the transition from the troposphere to the stratosphere.

for the troposphere to optimize signal to noise. The wavelengths chosen for the stratospheric lidars are usually longer than 305 nm, while wavelengths as short as 285 nm are useful in the free troposphere. Although it is not discussed here, the DIAL technique can be optimized for measurement of ozone very near the ground (within the boundary layer) by

using still shorter wavelengths that are near the ozone maximum absorption cross section at 254 nm (Fig. 1). A judicious choice of wavelengths not only provides the sensitivity required to measure accurately the ozone concentrations present in the atmosphere, but in the free troposphere it can allow for rapid profiling 24 hours a day.

Wavelengths shorter than 285 nm are attenuated sufficiently in the atmosphere to limit the useful altitude range of ground-based measurement to the lower troposphere. Since our objective is to measure ozone throughout the free troposphere, this sets the lower limit for our measurement wavelengths. The upper limit on wavelength is determined by our requirement for daytime measurements. There is sufficient solar radiation at wavelengths longer than about 293 nm to significantly increase the background sky signal during daylight hours and again limit the maximum altitude range of the measurement. During daylight measurements we insert a solar blind 283-292 nm band-pass filter in front of the photomultiplier detector to virtually eliminate the solar photons with wavelengths longer than 293 nm, and we restrict our laser wavelengths to be between 286 and 292 nm. Night-time profiles can be made without the filter, allowing longer wavelengths to be used and providing measurements into the lower stratosphere.

The lasers we use are continuously variable over the wavelength range described above. Each wavelength is produced by an independent laser system, consisting of a Nd:YAG laser that is doubled to pump a dye laser. The resulting visible wavelength emitted from the rhodamine dye is again doubled to produce the required ultraviolet wavelengths. The two lasers are fired synchronously at 20 Hz, and their output wavelengths are independently variable. By separating the two laser outputs by 400 μ s we can use a single photomultiplier detector with the broad-band bandpass filter described above. This allows the wavelengths to be changed even from profile to profile to search for interfering molecular species and to make consistency checks. By retaining flexibility in wavelengths, errors in the calculated values of ozone due to differential Mie scattering can also be decreased. A wavemeter is used to monitor the wavelengths.

As with all lidar systems, time resolved detection of backscatter from the propagating laser pulses provides the signal. In our case, two independent telescopes and gated photomultipliers are used, one for the near range and the other for the more distant altitudes. This is necessary to cover the large dynamic range of useful signal, more than six orders

of magnitude. The signals are hardware averaged by 12-bit analog to digital converters and recorded by a devoted data acquisition computer. Data from an ozone profile can be acquired in as little as one minute. During data acquisition, a preliminary analysis of the data from the previous profile is made on a separate data analysis computer. This gives real-time information to the operator.

The lidar system we have described provides vertical profiles with an absolute accuracy that has been estimated to be better than 10%. The estimated statistical uncertainty in the detected signal is altitude dependent and apparently less than 3%, the uncertainty in the ozone cross section is known to be about 2% including temperature dependence, molecular interferences can be shown to contribute no more than 1%, and hardware errors such as laser-telescope alignment, photomultiplier signal-induced bias, and digitizer non-linearities contribute no more than 5%. A simple sum of these error estimates results in the 10% overall absolute uncertainty which we consider to be a conservative estimate under clear sky conditions. Uncertainties in the wavelength dependence of Mie scattering have not been included in this analysis, however the effect in clear air is relatively small in the free troposphere.

Null profiles (profiles using two identical wave-

Fig. 3 Typical "null profile" using 288 nm wavelength taken with DIAL system at Fritz Peak Observatory on March 2, 1994 under daylight conditions. A corresponding log ratio profile taken a few minutes later using 288 and 291 nm is proportional to the ozone concentration. The abscissa is the usual DIAL calculation for a 1 km range element of the natural log ratio of measured signal intensities obtained for each wavelength pair.

lengths) are used routinely to check laser-telescope alignment and other hardware related errors. These checks have verified our error estimate for these components to be no more than 5%. A typical zero profile is shown in Fig. 3 along with an ozone

Fig. 4 Profile of ozone concentration obtained from the same data represented in Fig. 3. The horizontal bars represent the deviations of the null profile in Fig. 3 from zero and are an indicator of hardware and statistical errors. The profile begins at approximately 1 km above ground level and extends to almost 15 km ASL.

Fig. 5 Composite of more than 170 independent ozone profiles taken from 9 AM until 10 PM local time September 15, 1993. The altitude range on the vertical scale is from 4 to 15 km ASL. Colors indicate mixing ratios of ozone with units of parts per billion by volume (ppbv) and are obtained from the measured ozone concentrations and a standard climatology of air densities.

profile (Fig. 4) taken a few minutes later. We have also verified the overall error of no more than 10% for the 4 to 7 km ASL range by direct comparison with highly accurate in-situ measurements of ozone made in-situ from aircraft flying above our lidar during operation.

Tropospheric ozone is highly variable on very short time scales, so frequent sampling is necessary to provide a statistically accurate representation of ozone distributions. The lidar technique can provide many profiles in rapid succession, however it requires a cloud-free period while taking data, thus somewhat biasing the data set. These biases cannot be completely avoided, however our rapid profiling capability allows for measurements during periods when clouds may be present but not overhead. An example of the rapid profiling capability during clear sky conditions is seen in Fig. 5 a composite of nearly 170 daylight profiles taken over a 12 hour period.

We now have more than two years of ozone observations using our lidar at Fritz Peak Observatory near Rollinsville, Colorado. The expected seasonal changes in ozone are apparent, reflecting the annual rise and fall of the tropopause and the ozone increase during spring. We have also made a few intensive studies with 12 to 24 hour periods of continuous measurement such as the one shown in

Fig. 5. The changes that we have observed on these short time scales appear to be dynamical rather than chemical in origin.

The ability to independently change wavelengths allows for many new techniques to check for consistency and assure the best accuracy in the reduced data. We have briefly mentioned the null profiles, where both lasers are tuned to the same wavelength and a log-ratio profile is calculated. Deviations from a zero profile reflect errors that might otherwise go undetected. Another capability of this DIAL system that we have not discussed is provided by varying the wavelengths from profile to profile to identify the presence of other molecular species with absorption cross sections that could produce an ozone-like signature in our DIAL measurements and introduce errors in the ozone retrieval. The ability to change wavelengths and to make measurements of ozone during all hours of the day make this lidar configuration a very flexible instrument suitable for many atmospheric applications.

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