

N95- 11053

## Volcanic-Aerosol-Induced Changes in Stratospheric Ozone Following the Eruption of Mt. Pinatubo

W. B. Grant, E. V. Browell, J. Fishman, V. G. Brackett<sup>1</sup>, M. A. Fenn<sup>1</sup>, C. F. Butler<sup>1</sup>, D. Nganga<sup>2</sup>, A. Minga<sup>2</sup>, B. Cros<sup>2</sup>, S. D. Mayor<sup>1</sup>, G. D. Nowicki<sup>1</sup>, R. E. Veiga<sup>1</sup>, L. L. Stowe<sup>3</sup>, C. S. Long<sup>3</sup>

NASA Langley Research Center, MS 401A, Hampton, VA 23681-0001

1. SAIC, Hampton, VA, 23666; 2. Université Meuen Ngouabi, Brazzaville, The Congo; 3. NOAA NESDIS, World Weather Bldg., Washington, DC 20233

### ABSTRACT

Measurements of lower stratospheric ozone in the Tropics using electrochemical concentrations cell (ECC) sondes and the airborne UV Differential Absorption Lidar (DIAL) system after the eruption of Mt. Pinatubo are compared with The Stratospheric Aerosol and Gas Experiment II (SAGE II) and ECC sonde measurements from before the eruption to determine what changes have occurred as a result. Aerosol data from the Advanced Very High Resolution Radiometer (AVHRR) and the visible and IR wavelengths of the lidar system are used to examine the relationship between aerosols and ozone changes. Ozone decreases of 30% at altitudes between 19 and 26 km, partial column (16-28 km) decreases of about 27 D.U. and slight increases (5.4 D.U.) between 28 and 31 km are found in comparison with SAGE II climatological values.

### 1. INTRODUCTION

Reductions of mid-latitude stratospheric ozone were observed following the volcanic eruptions of Mt. Agung on March 17, 1963 [Pittock, 1966; Grams and Fiocco, 1967], Fuego in October 1974 [Jäger and Wege, 1990], El Chichón on March 28 to April 4, 1982 [Adriani et al., 1987; Angell, 1988; Newell and Selkirk, 1988; Jäger and Wege, 1990], and low values of tropical stratospheric ozone were observed following the eruption of Mt. Pinatubo on June 15 and 16, 1991 [Waters et al., 1992; Grant et al., 1992; Schoeberl et al., 1992]. Various theories have been developed to explain the observed mid-latitude changes including: (1) heterogeneous chemistry involving reactions of nitrogen species with the sulfuric acid aerosols, freeing chlorine to be more active in attacking ozone [Hofmann and Solomon, 1989; Michelangeli et al., 1989]; (2) aerosol effects on solar radiation including heating [Fiocco et al., 1978; Adriani et al., 1987; Michelangeli et al.,

1989]; and (3) transport of perturbed tropical air masses to mid-latitude regions without further changes in ozone after arrival [e.g., Pittock, 1966; Dobson, 1973].

The eruption of Mt. Pinatubo, being the largest of this century, placed 20 to 30 megatonnes of material into the stratosphere [McCormick and Veiga, 1992], and thus it provided a good opportunity to continue the study of the effect of volcanic aerosols on stratospheric ozone. To this end, historical data from SAGE II [McCormick et al., 1989], and recent data obtained using ECC sondes in the Tropics [Grant et al., 1992] and the UV DIAL system [Browell, 1989; Browell et al., 1990a] are used for the ozone change determinations. For aerosol information, data obtained using the UV DIAL system [Browell 1989; Browell et al., 1990b] and the AVHRR instrument on NOAA/11 [Stowe et al., 1992] are used.

### 2. INSTRUMENT DESCRIPTIONS

Three ozone data sets are employed for the analysis of the effects of volcanic aerosols on tropical ozone -- SAGE II data for the appropriate phase of the quasi-biennial oscillation (QBO) from November 1984 to May 1991; ECC sonde data from Brazzaville, The Congo (4°S, 15°E), from June 6, 1990, to Jan. 3, 1992, and Ascension Island (8°S, 14°W) from July 28, 1990 to Feb. 27, 1992; and UV DIAL data from several flights during the Airborne Arctic Stratospheric Expedition (AASE-II) on January 28 and 30 and February 20, 1992. For aerosols, zonally averaged AVHRR data are used for determination of the aerosol optical depths (A.O.D.), and lidar data using the visible and IR wavelengths of the UV DIAL system are used for vertical profiles of surface area loadings.

The SAGE II data are used to provide climatological values for ozone profiles before the eruption of Mt. Pinatubo. Since SAGE II is a solar occultation instrument, it is unable to measure ozone in regions with

dense aerosols that occur after volcanic eruptions, such as in the tropics. Nevertheless, the data obtained with it prior to the eruption are very useful for determining ozone climatological values, standard deviations, and QBO excursions which can be used for comparison with measurements made by instruments that were not making long-term measurements. By combining profiles for each month of interest for the years with similar phases of the QBO, deviations from recent historical variations can be determined. This is especially important near the tropics where the QBO plays an important role in determining ozone values [Bojkov, 1987; Newell and Selkirk, 1988]. For 1991, a year in which the stratospheric winds above Singapore were changing to easterly near the beginning of the year [Trepte and Hitchman, 1992], the years 1986, 1988, and 1991 prior to the eruption can be used for the climatology [Schoeberl et al., 1992]. For 1992, values from 1987 and 1989 can be used, assuming that it is the second year of the easterly phase.

The type of ECC sonde used was originally developed by Komhyr [1969] and is manufactured by Science Pump Corp., Camden, NJ. The sonde is launched on a balloon which typically reaches altitudes of 31 km before the balloon bursts. Air containing ozone is pumped through a KI solution and causes the release of electrons from the iodine, from which the amount of ozone present can be determined. The ECC sonde is assumed to be insensitive to the presence of aerosols [Komhyr, NOAA, private communication], although in the presence of high aerosol concentrations this assumption may be wrong. The accuracy of the ECC sonde can be 6% in the stratosphere (17 to 32 km) and the precision 3% [Komhyr et al., 1992].

The UV DIAL system uses a pair of Nd:YAG lasers to pump dye lasers which are doubled to generate the ultraviolet wavelengths required for the ozone measurements [Browell, 1989]. For stratospheric measurements, wavelengths of 301.5 and 310.85 nm are employed, and the absorption coefficients of Molina and Molina [1986] are used in the analysis. The residual dye laser and Nd:YAG laser energies are used to measure atmospheric backscatter in the visible (603 nm) and near-IR (1.06  $\mu\text{m}$ ) spectral regions in order to determine aerosol distributions. The UV DIAL system has been shown to make measurements of stratospheric ozone with an accuracy of 10% [Browell, 1989]. This system has been used to study ozone depletion in the 1987 Airborne Antarctic Ozone Experiment (AAOE) [Browell et al., 1988], the 1989 AASE mission [Browell et al., 1990a,b], and the 1992 AASE-II field expedition. During the AASE-II mission, the NASA DC-8 was typically flown at altitudes of 33 to 37 kft, and the UV DIAL system was pointed only in the zenith.

When there is a strong aerosol gradient present in the lidar signals, additional care is required when processing the signals to determine the ozone profile. In general, a Bernoulli solution to the lidar equation is used to correct the DIAL calculation for the wavelength dependence of the aerosol scatter and extinction [Browell et al., 1985]. Without the correction, the DIAL calculation would show a derivative signal about a strong aerosol gradient, going high before the layer and low after the layer. The backscatter correction requires a knowledge of both the normalized aerosol backscattering phase function ( $P\pi$ ) and the aerosol backscatter wavelength dependence ( $\alpha$ ) at the measurement wavelengths. The lidar signals in the IR, visible, and UV regions are used to determine the wavelength dependence of the aerosol backscatter. The value of  $\alpha$  used was generally 0.7, while the value of  $P\pi$  was set to 0.028  $\text{sr}^{-1}$  [Fenn et al., 1992]. After the corrections were applied, no obvious features associated with these aerosol effects remained in the DIAL ozone profiles.

### 3. TROPICAL DATA

Since the ECC sonde is expected to provide the measurement of ozone profiles without interference from aerosols, we will examine the ECC sonde data first. A preliminary discussion of the ECC sonde measurements showing tropical ozone reductions as great as 20% and column reductions of about 5-8% was presented by Grant et al. [1992].

In this work, ECC sonde data are compared with SAGE II climatology for individual months. While not shown, before the eruption of Mt. Pinatubo, the sondes for the first 5 months of 1991 gave mean ozone values that agreed with SAGE II climatology within statistical expectations. After the eruption of Mt. Pinatubo, there were deviations of the sonde values from the SAGE II values outside the  $2\sigma$  level for August, the first month that sonde launches were made after the eruption (see Figure 1a). The ozone decrease below about 28 km was greatest for the month of October (approximately 27 D.U. or 10% of the total column) (Figure 1b) and then gradually decreased.

A more detailed look at the individual sonde profiles indicates that the minimum in ozone at 23.5 km occurred October  $13 \pm 3$  days. In addition, an enhancement of ozone is apparent above about 28 km in all cases, being at or above the  $2\sigma$  level for 5 of the 7 months, and at the  $1\sigma$  level for 2 months.

Let us turn now to the UV DIAL ozone data in the Tropics. The tropical ozone was measured on three separate flights during AASE II -- on January 28, 1991, on a flight from NASA Ames at Moffett Field, California (38°N, 122°W) to Tahiti (18°S, 150°W); on January

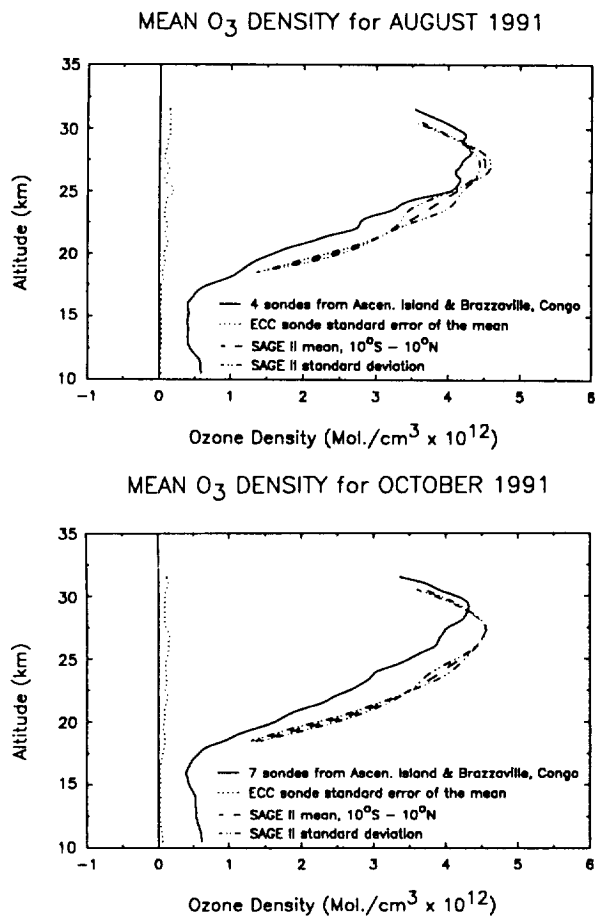


Figure 1. Monthly ECC sonde averages from Brazzaville and Ascension Island for August 1991 (a) and October 1991 (b) compared with SAGE II ozone climatology (1986, 1988).

30 on the return trip to NASA Ames from Tahiti; and February 20-21 on a flight from Bangor, Maine (45°N, 69°W) over Puerto Rico to 15°N and back. Ozone measurements were recorded to magnetic tape between 10° and 15°S on the way to Tahiti and from 6°N to near landing on the return flight.

The data can be plotted as ozone number density versus latitude for various altitudes. In this manner, the values from each of the three instruments can be plotted together, as shown in Figure 2 for 19.5 and 23.5 km. The graphs show that the ozone decrease was greatest in the 6° - 16°N latitude region, falling off rapidly from 16° - 20°N latitude as the aerosol density decreased by a factor of 10, and falling off south of 10°S. The three sets of data are mutually consistent, with the ECC sonde and UV DIAL values being quite similar, even though they were taken over 100° apart in longitude and up to a month apart in time. The larger decrease of ozone in the Northern Hemisphere is again consistent with an increased aerosol loading at that time as depicted by the AVHRR.

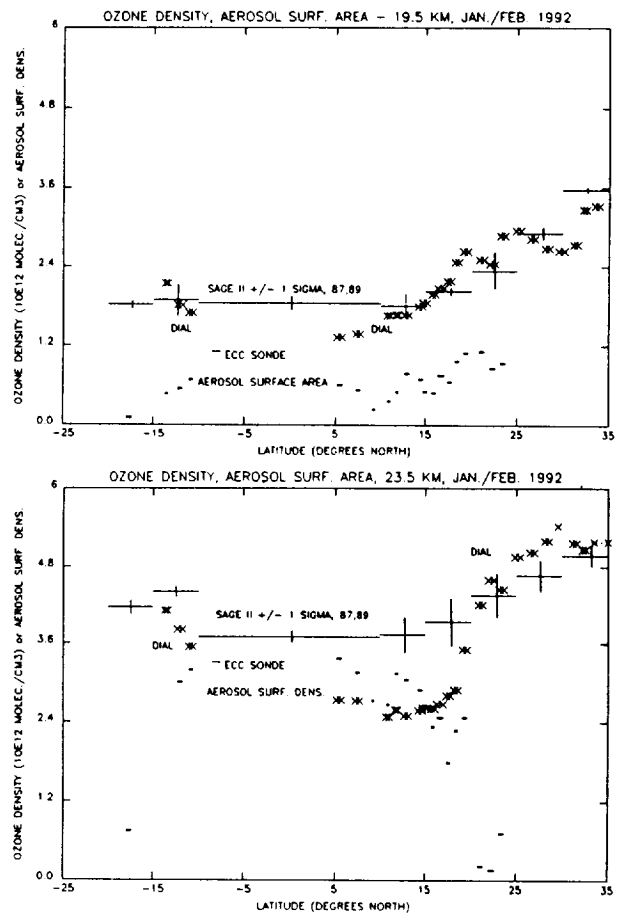


Figure 2. Graphs of tropical ozone from three instruments at altitudes of 19.5 and 23.5 km, averaged over 1 km. The SAGE II data are climatological data from 1987 and 1989. The UV DIAL are from the end of January 1992, while the ECC sonde data are from January/February 1992. The aerosol units are  $10^7 \mu\text{m}^2\text{m}^{-3}$ .

#### 4. SUMMARY AND CONCLUSIONS

Data showing large decreases in tropical ozone due to the presence of volcanic aerosol has been presented which is beyond the  $2\text{-}\sigma$  value from climatological ozone records. There also appears to be evidence for increases of ozone in the 28- to 31-km region that are about at the  $2\text{-}\sigma$  values for SAGE II data. The mechanisms responsible for the ozone changes were not well known at the time of the Symposium, in part because not enough data were available on the abundance of chemical species present in the tropical lower stratosphere in the presence of Mt. Pinatubo aerosols. It will be interesting to see what can be learned by other stratospheric ozone measuring instruments [Grant, 1989].

## 5. ACKNOWLEDGMENTS

The authors thank L. Froidevaux, Jet Propulsion Laboratory, for his encouragement in investigating the relationship between the ECC sonde data and the UV DIAL determination of Pinatubo aerosol distribution. The work was funded in part by NASA's Upper Atmospheric Research Program.

## REFERENCES

- Adriani, A., G. Fiocco, G. P. Gobbi, and F. Congeduti, Correlated behavior of the aerosol and ozone contents of the stratosphere after the El Chichón eruption, *J. Geophys. Res.* **92**, 8365-8372, 1987.
- Angell, J. K., An update through 1985 of the variations in global total ozone and north temperate layer-mean ozone, *J. Appl. Meteorol.* **27**, 91-97, 1988.
- Bojkov, R. D., The 1983 and 1985 anomalies in ozone distribution in perspective, *Mon. Weath. Rev.* **115**, 2187-2201, 1987.
- Browell, E. V., Differential absorption lidar sensing of ozone, *Proc. IEEE* **77**, 419-432, 1989.
- Browell, E. V., S. Ismail S. T. Shipley, Ultraviolet measurements of O<sub>3</sub> profiles in regions of spatially inhomogeneous aerosols, *Appl. Opt.* **24**, 2827-2836, 1985.
- Browell, E. V., L. R. Poole, M. P. McCormick, S. Ismail, C. F. Butler, S. A. Kooi, M. M. Szedlmayer, R. L. Jones, A. Krueger, and A. F. Tuck, Large-scale variations in ozone and polar stratospheric clouds measured with airborne lidar during formation of the 1987 Ozone Hole over Antarctica, Polar Ozone Workshop, NASA Ref. Pub. 10014, pp. 61-64, 1988.
- Browell, E. V., C. F. Butler, S. Ismail, M. A. Fenn, A. F. Carter, A. F. Tuck, O. B. Toon, M. H. Proffitt, M. Loewenstein, M. R. Schoeberl, I. Isaksen, and G. Braathen, Airborne lidar observations in the wintertime arctic stratosphere: Ozone, *Geophys. Res. Lett.* **17**, 325-328, 1990a.
- Browell, E. V., C. F. Butler, S. Ismail, P. A. Robinette, A. F. Carter, N. S. Higdon, O. B. Toon, M. R. Schoeberl, and A. F. Tuck, Airborne lidar observations in the wintertime arctic stratosphere: Polar stratospheric clouds, *Geophys. Res. Lett.* **17**, 385-388, 1990b.
- Dobson, G. M. B., The laminated structure of the ozone in the atmosphere, *Q. J. R. Meteorol. Soc.* **99**, 599-607, 1973.
- Fenn, M. A., S. Ismail, E. V. Browell and C. F. Butler, Correction of DIAL ozone measurements in the presence of stratospheric Pinatubo aerosols, Sixteenth International Laser Radar Conf., MIT, July 20-24, 1992, NASA Conf. Pub. 3158, pp 117-120.
- Fiocco, G., A. Mugnai, and W. Forlizzi, Effects of radiation scattered by aerosols on the photodissociation of ozone, *J. Atmos. Terr. Phys.* **40**, 949-961, 1978.
- Grams, G., G. Fiocco, Stratospheric aerosol layer during 1964 and 1965, *J. Geophys. Res.* **72**, 3523-3542, 1967.
- Grant, W. B., ed., *Ozone Measuring Instruments for the Stratosphere*, Vol. 1 of Collected Works in Optics, 438 pp, Opt. Soc. Am., Washington, D.C., 1989.
- Grant, W. B., J. Fishman, E. V. Browell, V. Brackett, D. Nganga, A. Minga, B. Cros, R. E. Veiga, C. F. Butler, M. A. Fenn, and G. D. Nowicki, Observations of reduced ozone concentrations in the tropical stratosphere after the eruption of Mt. Pinatubo, *Geophys. Res. Lett.* **19**, 1109-1112, 1992.
- Hofmann, D. J., and S. Solomon, Ozone destruction through heterogeneous chemistry following the eruption of El Chichon, *J. Geophys. Res.* **94**, 5029-5041, 1989.
- Jäger, H., and D. J. Hofmann, Midlatitude lidar backscatter to mass, area, and extinction conversion model based on in situ aerosol measurements from 1980 to 1987, *Appl. Opt.* **30**, 127-138, 1991.
- Jäger, H., and K. Wege, Stratospheric ozone depletion at northern midlatitudes after major volcanic eruptions, *J. Atmos. Chem.* **10**, 273-287, 1990.
- Komhyr, W. D., Electrochemical concentration cells for gas analysis, *Ann. Geophys.* **25**, 203-210, 1969.
- Komhyr, W. D., J. A. Lathrop, D. P. Opperman, R. A. Barnes, and G. B. Brothers, ECC ozonesonde performance evaluation during STOIC 1989, submitted to *J. Geophys. Res.*, 1992.
- McCormick, M. P., and R. E. Veiga, SAGE II measurements of early Pinatubo aerosols, *Geophys. Res. Lett.* **19**, 155-158, 1992.
- McCormick, M. P., J. M. Zawodny, R. E. Veiga, J. C. Larsen, and P.-H. Wang, An overview of SAGE I and SAGE II ozone measurements, *Planet. Space Sci.* **37**, 1567-1586, 1989.
- Michelangeli, D. V., M. Allen, Y. L. Yung, El Chichón volcanic aerosols: Impact of radiative, thermal, and chemical perturbations, *J. Geophys. Res.* **94**, 18,429-18,443, 1989.
- Molina, L. T., and M. J. Molina, Absolute absorption cross sections of ozone in the 185- to 350-nm wavelength range, *J. Geophys. Res.* **91**, 14,501-14,508, 1986.
- Newell, R. E., and H. B. Selkirk, Recent large fluctuations in total ozone, *Q. J. R. Meteorol. Soc.* **114**, 595-617, 1988.
- Pittock, A. B., A thin stable layer of anomalous ozone and dust content, *J. Atmos. Sci.* **23**, 538-542, 1966. 1992.
- Schoeberl, M. R., P. K. Bhartia, and E. Hilsenrath, Tropical ozone loss following the eruption of Mt. Pinatubo, submitted to *Geophys. Res. Lett.*, 1992.
- Stowe, L. L., R. M. Carey, and P. P. Pellegrino, Monitoring the Mt. Pinatubo aerosol layer with NOAA/11 AVHRR data, *Geophys. Res. Lett.* **19**, 159-162, 1992.
- Trepte, C. R., and M. H. Hitchman, Tropical stratospheric circulation deduced from satellite aerosol data, *Nature* **355**, 626-628, 1992.
- Waters, J. W., et al., The UARS microwave limb sounder experiment, paper presented at The Eighth Conference on the Middle Atmosphere, Atlanta, Ga., spons. by the Am. Meteorol. Soc., January 5-10, 1992.