

Correlative stratospheric ozone measurements with the airborne UV DIAL system during TOTE/VOTE

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Abstract. The airborne UV differential absorption lidar (DIAL) system participated in the Tropical Ozone Transport Experiment/Vortex Ozone Transport Experiment (TOTE/VOTE) in late 1995/early 1996. This mission afforded the opportunity to compare the DIAL system's stratospheric ozone measuring capability with other remote-sensing instruments through correlative measurements over a latitude range from the tropics to the Arctic. These instruments included ground-based DIAL and space-based stratospheric instruments: HALOE; MLS; and SAGE II. The ozone profiles generally agreed within random error estimates for the various instruments in the middle of the profiles in the tropics, but regions of significant systematic differences, especially near or below the tropopause or at the higher altitudes were also found. The comparisons strongly suggest that the airborne UV DIAL system can play a valuable role as a mobile lower-stratospheric ozone validation instrument.

Introduction

The TOTE/VOTE project, designed to study transport of filaments of air transported into or out of the arctic polar vortex and the tropical stratospheric reservoir and compare the measurements with the model calculations using the contour advection scheme [Schoeberl and Newman, 1995], involved airborne measurements onboard the NASA DC-8 between December 8, 1995 and February 19, 1996, and provided an opportunity for ozone measurement comparisons.

The airborne UV DIAL system has been used in a number of aircraft campaigns since 1980 [Browell, 1989; 1995], and was updated with current state-of-the-art Nd:YAG lasers and Nd:YAG-pumped dye lasers shortly before the TOTE/VOTE mission [Richter et al., 1996]. The airborne UV DIAL system measures ozone above the

aircraft from approximately 2 km above the aircraft altitude up to about 16 km above the aircraft. The spatial resolution used is 1.3 km in the vertical and 70 km in the horizontal.

Measurements

Data from two ground-based UV DIAL systems (STROZ LITE and the Jet Propulsion Laboratory's (JPL's) system, both at Mauna Loa Observatory) and three space-based instruments (HALOE, MLS, and SAGE II) were compared.

Ground-based DIAL

The comparisons with the Goddard Space Flight Center (GSFC) Stratospheric Ozone Lidar Trailer Experiment (STROZ-LITE) [McGee et al., 1993, 1995] at the Mauna Loa Observatory (MLO) were closest in time and space. The intercomparison turned up an error of a few percent in the choice of ozone absorption coefficients used by the airborne UV DIAL¹. Once the Bass and Paur [1985] absorption coefficients were adopted, the airborne UV DIAL and STROZ-LITE ozone measurements agreed very well.

Detailed comparisons were made on December 11 and February 13 using the nearest STROZ-LITE profile and a 5- or 9-minute airborne UV DIAL measurement near the MLO. The profiles are shown in Figures 1 and 2. The mean difference (the average of the normalized differences at each altitude) between the two data sets from 18.5 to 25.5 km is 3.1% on December 11 and 5.8% on February 13, which compares favorably with the combined standard (random) error of the measurements over this interval (5.0% on December 11 and 2.9% on February 13). The larger difference on February 13 was likely due to the ozone structure near 19-20 km, which changed with time. There are larger differences in the upper troposphere/lower stratosphere, where the ozone number densities are lower (the absorption coefficients used are optimized for higher ozone number densities) and the atmospheric ozone variability is greater. Note, however, that there is a 100-200 m altitude uncertainty for the DC-8 platform. If altitude adjustments are made, using, for example, the best fit to the vertical ozone structure, the agreement improves.

Comparisons were also made with the JPL DIAL at MLO [McDermid et al., 1991, 1997] for the same dates. Four JPL profiles were combined for the December 11 compari-

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¹The Molina and Molina (MM) coefficients are several percent higher than an average of several measurements in the 290-320-nm spectral region. Compared with 7 other measurements made at 4 mercury lines between 1953 and 1988, as tabulated in Yoshino et al. [1988], the MM coefficients near 295 K are 1.4%-5.3% higher, averaging 4.2% higher.

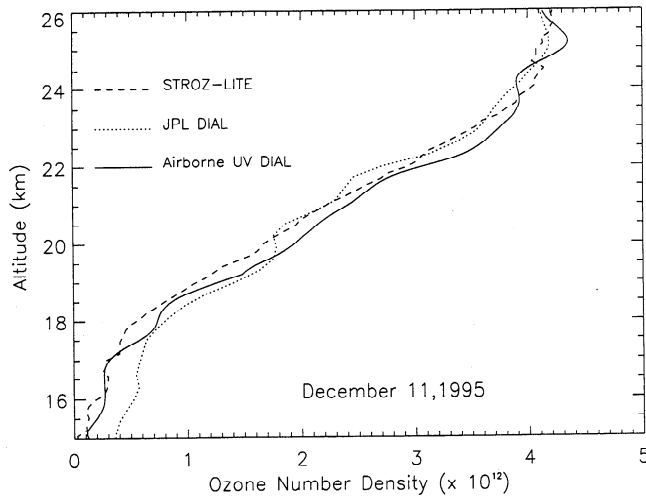


Figure 1. Comparison of the airborne UV DIAL, STROZ LITE, and the JPL UV DIAL system at the Mauna Loa Observatory, December 11, 1995.

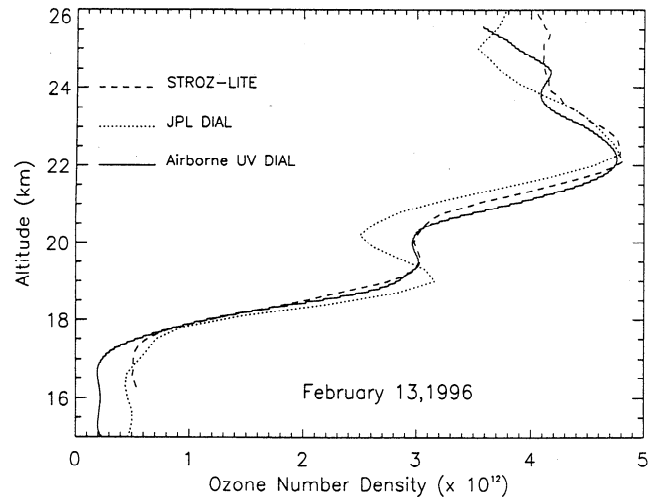


Figure 2. Same as Figure 1, but for February 13, 1996.

son, but there was only one JPL profile to use on February 13, taken approximately 4-5 hours prior to the overflight. The mean difference between the two (airborne DIAL - JPL DIAL) in the 20-25.5 km range was -6.4% on December 11 and -8.4% on February 13, compared with combined standard errors of 5.7% on December 11 and 5.6% on February 13. The measurement difference was larger on February 13 because there was an ozone dip near 20 km which was more pronounced during the time of the JPL profile than during the overflight, and because JPL uses 900 m averaging interval while the airborne DIAL uses 1300 m. The data below 20 km don't agree well due to reasons given for STROZ-LITE. In addition, since the JPL DIAL system has a larger receiver and a more energetic laser, it has a problem with saturation of electronics elements in the detector chain, many of which have current limits.

SAGE II

SAGE II [Mauldin et al., 1985] was launched October 5, 1984, and continues to operate today. Comparisons were made on December 8, 20, and 22. Time differences in the measurements varied from 6 to 13 hours, while latitude differences were minimal except for December 22 when it was 3 degrees. Longitudinal differences varied from 1.8 to 7.4 degrees. As shown in Table 1, the agreement is very good from 21 to 27 km, ranging from 3.8 to 6.3% with a mean difference based on absolute values of 5.3% in this range. The combined estimated error in this region is 3.9%. (Estimated error is determined from the stated measurement error for each instrument.) This agreement is comparable to the agreements seen with ROCOZ-A [Cunnold et al., 1989; Barnes et al., 1991], ECC sondes [Barnes et al., 1991], and several instruments onboard UARS [Cunnold et al., 1996]. In the region from 13-18 km, the difference between the

Table 1. Comparison of SAGE II with the airborne UV DIAL. The ozone values are given in units of number density ($\times 10^{12}$).

Altitude (km)	13.5	14.5	15.5	16.5	17.5	18.5	19.5	20.5	21.5	22.5	23.5	24.5	26.5	27.5	28.5
December 8															
DIAL (48N, 117.7W)	1.65	1.74	1.86	1.68	2.15	3.07	3.71	4.26	4.97	4.55	4.12				
SAGE (48N, 110-160N)	1.25	1.95	1.86	1.86	2.54	3.14	3.15	4.53	4.64	4.45	4.45				
Estimated combined error (%)	20	12.2	10.6	9.5	6.6	4.2	4.1	4.4	4.7	5.3	5.0				
(DIAL-SAGE)/DIAL (%)	24	-21	0	-7.1	-18	-2.3	15	-6.3	6.6	2.2	-8.0				
December 20															
DIAL (4.5S, 152W)			0.051	0.093	0.597	1.27	1.75	2.38	3.04	3.34	3.55	3.94	4.29	3.88	3.38
SAGE (4S, 154W)		0.015	0.026	0.062	0.213	0.689	1.40	1.94	2.53	2.95	3.15	3.38	4.16	4.56	4.47
Estimated combined error			366	82	22	10.6	4.5	3.9	4.0	3.4	2.7	3.1	4.0	4.2	4.8
(DIAL-SAGE)/DIAL (%)			-20	-129	-15.4	-10.2	-10.9	-6.3	3.0	5.7	4.8	-5.6	-6.3	-15.2	-29
Average absolute difference (%)*	46	12	18	10	22	10	14	9	4.7	3.8	6.3	4.8	6.3	15.2	29

* The average includes data for December 22, when the UV DIAL was near 24N, 163W and SAGE II was near 21N, 170W.

Table 2. Comparison of HALOE (50N, 113W at 0007 UT) with the airborne UV DIAL (50N, 108W at 0425 UT) on January 24, 1996. The ozone values are given in units of number density ($\times 10^{12}$ molec./cm³).

Altitude	13.5	14.5	15.5	16.5	17.5	18.5	19.5	20.5	21.5	22.5
Airborne DIAL	3.68	2.93	2.66	5.08	7.18	7.07	6.88	6.65	5.59	4.73
HALOE	3.33	2.84	2.97	4.60	6.62	7.48	7.84	7.29	6.41	5.64
Difference (DIAL-HALOE)/mean (%)	10	3	-11	10	8	-6	-13	-10	-14	-18

DIAL and SAGE measurements is generally in accordance with the estimated errors of the two measurements. SAGE error increases at lower altitudes due to errors that propagate in the onion-peeling technique used to determine ozone number densities at lower altitudes. The differences at 18 and 19 km (10.2-27%) are larger than the estimated errors (4.2-16%), and also vary in sign. The SAGE team is aware of about a 10% over-reporting of ozone in this region due to a problem with the data-processing algorithm's altitude determination in regions where the ozone profile is varying rapidly, such as near the tropopause (J. M. Zawodny, private communication), which could explain the difference on Dec. 20, where DIAL and SAGE were closest in time and space. (The SAGE instrument team is developing a new algorithm to better handle the altitude.) The differences on Dec. 8 and 22 likely arose from measuring different air masses, since the differences in latitude or longitude were 3-5 degrees.

Halogen Occultation Experiment

There was only one intercomparison possible between the airborne UV DIAL and HALOE [Russell et al., 1993], at 50°N near 110°W on January 24. The difference in time and space was 4:20 and 5 deg. longitude, respectively. The agreement between 13 and 20 km, 0 to 13%, (Table 2) is about the same as between STROZ-LITE and HALOE as presented in Brühl et al. [1996] and between a UV absorption photometer and HALOE as presented in Grose et al. [1997]. However, the difference increases at higher altitudes. Version 18 HALOE data were used. It has been noticed that the airborne UV DIAL data are systematically lower than those of other instruments at the higher altitudes, such as HALOE and SAGE II. This effect probably arises from the presence of signal-induced signal, which reduces the measured ozone values because the absorbing wavelength is more strongly affected. A more thorough investigation of the systematic bias between DIAL and HALOE or SAGE II is being investigated using back-trajectory calculations to ensure that the same air mass is being sampled by both instruments (R. B. Pierce, private communication; see, also, Pierce et al. [1997]). The large variability of ozone concentrations in the lower midlatitude stratosphere makes comparisons difficult unless the same air mass is being compared.

Microwave Limb Sounder (MLS)

The MLS [Barath et al., 1993] performs well near and above the ozone number density peak near 70 mb as seen by the results at high latitudes [Froidevaux et al., 1996]. The

values at 100 and 46.3 mb are direct measurements using the averaging kernels; those at 68.3 and 31.6 mb are interpolated from the two adjacent averaging kernels. Our comparisons with MLS for December 9 near 77°N are within MLS error values for the four lower pressure levels (11-45%) (Table 3). Part of the difference between the DIAL and MLS values undoubtedly comes from the fact that stratospheric ozone in the high latitudes has high spatial variability, and that the DIAL and MLS were not sampling the same air masses.

In comparisons with the MLS in the tropics on February 17, good agreement is found only at 31.6 mb (23.6 km), where the 6% is within the 9-10% errors given for the MLS measurements at that altitude. The differences at 68.3 and 46.3 mb (47% and 58%, respectively) are approximately the

Table 3. Comparison of the MLS with the airborne UV DIAL. Units of ozone are 10^{12} molecules/cm³.

MLS Pressure level (mb)	100	68.3	46.3	31.6
December 9, 1995				
Altitude (km)	15.2	17.5	19.7	21.9
DIAL (78.5N, 106.5W)	4.53	6.00	4.95	3.20
MLS (77.3N, 100.3W)	5.41	5.25	4.56	3.31
DIAL precision (%)	1.9	0.6	1.4	5.0
MLS precision*	0.55	0.34	0.22	0.19
MLS estimated error (%)	10	6.5	4.8	5.7
Difference/DIAL (%)	19	13	8	3.4
Altitude (km)				
Airborne DIAL (75.3N, 168.7W)	4.10	4.42	4.52	3.53
MLS (75.30N, 164.17W)	6.22	5.03	3.90	2.97
DIAL precision (%)	3.8	1.8	1.9	5.7
MLS estimated error (%)	39	24	12.7	10
Difference/DIAL (%)	41	13	15	17
February 17, 1996				
Altitude (km)	16.6	18.8	21.2	23.6
Airborne DIAL (1.2 N, 207 E)	0.43	1.32	2.67	3.21
MLS (1.21 N, 214.4 E)	6.10	2.40	0.45	3.43
MLS estimated error (%)	42	53	123	10
Airborne DIAL (20.3 N, 204 E)				
MLS (20.3 N, 200.4 E)	1.06	2.42	3.58	4.27
MLS stated error (%)	2.93	2.88	2.54	4.09
Difference/DIAL for Feb. 17**	86	47	21	9
Difference/DIAL for Feb. 17**				
	88	63	59	9

* Table 2a in Froidevaux et al. [1996].

** Includes comparisons at 1.2 and 7.5N

same as the stated MLS errors in the data files at those pressure levels (63% and 59%). The errors for the 100 mb pressure level are somewhat larger than would be expected from the reported MLS errors. MLS has large errors in the tropics at lower altitudes, probably due, in part, to the fact that the tropopause is so high there, giving low ozone values at the altitudes where comparisons were made. It is also possible that the differences could be attributed to the airborne DIAL measuring in regions of low ozone due to convective outflow and MLS measuring in regions not so affected.

Summary and conclusion

The participation of the airborne UV DIAL system during the TOTE/VOTE mission afforded the opportunity for validation of the DIAL system for making stratospheric ozone measurements by intercomparisons with several ground-based and space-based instruments. Over the regions of best comparison, the differences, which were similar to the estimated combined errors for the airborne UV DIAL and other instrument, were as follows: for ground-based UV DIAL systems, 3-8%; for HALOE, <10%; for MLS, 6% at the highest altitude; and SAGE II, 4-9% from 20-27 km. In addition, when differences were found, they often highlighted problems with either the airborne DIAL (incorrect ozone absorption coefficients at first, systematic lower ozone measurements at higher altitudes), or the other instruments: JPL UV DIAL (saturation of detector electronics at lower altitudes); MLS (difficulty in measuring low ozone mixing ratios below the ozone peak); SAGE II (height-determining algorithm, affecting data near the tropopause). Instruments such as the airborne UV DIAL system can, thus, operate as mobile ozone-profile validation instruments, reaching places inaccessible to ground-based instruments, and can find significant systematic errors.

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References

- Barath, F. T., et al., The Upper Atmosphere Research Satellite Microwave Limb Sounder instrument, *J. Geophys. Res.*, **98**, 10,751-10,762, 1993.
- Barnes, R. A., et al., Stratospheric Aerosol and Gas Experiment II and ROCOZ-A ozone profiles at Natal, Brazil: A basis for comparison with other satellite instruments, *J. Geophys. Res.*, **96**, 7515-7530, 1991.
- Bass, A. M. and R. J. Paur, The ultraviolet cross-sections of ozone: I. Measurements, in *Atmospheric Ozone, Proc. Quad. Ozone Symposium*, Halkidiki, Greece, C. Zerefos and A. Ghazi, eds., D. Reidel, Hingham, Ma., 606-610, 1985.
- Browell, E. V., Differential absorption lidar sensing of ozone, *Proc. IEEE*, **77**, 419-432, 1989.
- Browell, E. V., Airborne lidar measurements, *Rev. Laser Engineering*, **23**, 135-141, 1995.
- Brühl, C., et al., Halogen Occultation Experiment ozone channel validation, *J. Geophys. Res.*, **101**, 10,217-10,240, 1996.
- Cunnold, D. M., et al., Validation of SAGE II ozone measurements, *J. Geophys. Res.*, **94**, 8447-8460, 1989.
- Cunnold, D. M., et al., Overview of UARS ozone validation based primarily on intercomparisons among UARS and Stratospheric Aerosol and Gas Experiment II Measurements, *J. Geophys. Res.*, **101**, 10,335-10,350, 1996.
- Froidevaux, L., et al., Validation of UARS Microwave Limb Sounder ozone measurements, *J. Geophys. Res.*, **101**, 10,017-10,060, 1996.
- Grose, W. L., et al., Intercomparison of ozone measurements in the lower stratosphere from the UARS Halogen Occultation Experiment and the ER-2 UV absorption photometer, *J. Geophys. Res.*, **102**, 12,135-13,140, 1997.
- Malicet, J., et al., Ozone UV spectroscopy. II. Absorption cross-sections and temperature dependence, *J. Atmos. Chem.*, **21**, 263-273, 1995.
- Mauldin, III, L. E., et al., Stratospheric Aerosol and Gas Experiment II instrument: a functional description, *Opt. Eng.*, **24**, 307-312, 1985.
- McDermid, I. S., et al., Differential absorption lidar systems for tropospheric and stratospheric ozone measurements, *Opt. Eng.*, **30**, 22-30, 1991.
- McDermid, I. S., et al., NDSC lidar intercomparisons and validation: OPAL and MLO3 campaigns in 1995, in A. Ansmann, R. Neuber, P. Rairoux, and U. Wandinger (eds.), *Advances in Atmospheric Remote Sensing with Lidar*, Springer-Verlag, Berlin, 525-528, 1997.
- McGee, T. J., et al., Raman DIAL measurements of stratospheric ozone in the presence of volcanic aerosols, *Geophys. Res. Lett.*, **20**, 955-958, 1993.
- McGee, T. J., et al., Improved stratospheric ozone lidar, *Opt. Eng.*, **34**, 1421-1430, 1995.
- Molina, L. T., and M. J. Molina, Absolute absorption cross-sections of ozone in the 185-350 nm wavelength range, *J. Geophys. Res.*, **91**, 14,501-14,508, 1986.
- Pierce, R. B., et al., Photochemical calculations along air mass trajectories during ASHOE/MAESA, *J. Geophys. Res.*, **102**, 13,153-13,167, 1997.
- Richter, D. A., et al., Advanced airborne UV DIAL system for stratospheric and tropospheric ozone and aerosol measurements, *Advances in Atmospheric Remote Sensing with Lidar*, A. Ansmann, R. Neuber, P. Rairoux, and U. Wandinger, eds., 395-398, 1996.
- Russell, J. M., et al., The Halogen Occultation Experiment, *J. Geophys. Res.*, **98**, 10,777-10,797, 1993.
- Schoeberl, M. R., and P. A. Newman, A multiple-level trajectory analysis of vortex filaments, *J. Geophys. Res.*, **100**, 25,801-25,815, 1995.
- Yoshino, K., et al., Absolute absorption cross-section measurements of ozone in the wavelength region 238-355 nm and the temperature dependence, *Planet. Space Sci.*, **36**, 395-398, 1988.
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