NASA/CR-97- 207551 7N-45-CR

Title: An investigation of aerosol and ozone measurements from the Cryogenic Limb Array Etalon Spectrometer: Validation and relation to other chemical species.

Final Report

PI: Terry Deshler, CoPI: Derek Montague

Period: 1 December 1994 - 30 November 1997

Dept. of Atmospheric Science, University of Wyoming, Laramie, WY 82071

Grant Number: NAG 5-2801

Throughout this study we focused on comparisons of CLAES and in situ measurements of ozone and aerosol extinction. Thus the comparison is between satellite data representative of large spatial regions and in situ data representative of nearly point samples. Both instruments provide vertical profiles, but the region of overlap is limited to between approximately 10 and 100 mb.

CLAES Version 7 ozone measurements have been compared to electrochemical cell ozonesonde measurements over McMurdo Station, Antarctica (78°S, 167°E), Dumont d'Urville, Antarctica (66.7°S, 140°E), Laramie, Wyoming (41°N, 106°W), and Bear Island, Norway (74.3°N, 19°E). Comparisons were made between vertical ozone profiles, and between integrated column ozone over the region of overlap of the measurements. Comparisons using CLAES Version 8 data are underway. CLAES Version 8 aerosol extinction measurements at all wavelengths have also been compared to University of Wyoming aerosol extinctions over McMurdo Station, Antarctica, and over Laramie, Wyoming. Coincidences in all cases were determined by minimizing the distance between the CLAES observations and the surface station, and the time separation between the satellite and in situ measurements.

The ozone comparison used only overlapping portions of the CLAES Version 7 data and ozonesonde profiles. This comparison indicated that CLAES and the sondes displayed the same temporal trends over all stations; however in most cases, the CLAES column ozone was larger than the integrated ozonesonde measurements. The comparisons are summarized in Table 1. These results suggest a systematic bias in CLAES ozone measurements, causing them to be, on average, 4 to 22 % larger than in situ measurements. This bias is consistent with Bailey et al. (1996), and is suggested by Bailey et al. to be due to calibration, instrument, and a priori components. The bias is altitude dependent. At both Dumont d'Urville and Laramie the CLAES and ozonesonde measurements differ by less than 10% at altitudes above 60 mb. At Bear Island the comparison deteriorates above 40 mb while CLAES and sonde differ by less than 10 % below this altitude. At McMurdo the agreement is poor at all altitudes. Sharon Hauer, the graduate student who worked on this data, is still writing her M.S. thesis. These ozone comparisons, along with new comparisons with Version 8 data, make up part of that work.

Table 1. Comparison of CLAES and in situ column ozone over the region of vertical overlap of the two measurements. Mean differences and standard deviations (in Dobson units) and number of comparisons for all four stations.

Station	Mean Difference (CLAES-Sonde)	Standard Dev.	No. of Comparisons
McMurdo Station, Ant.	35.57	25.33	15
Dumont d'Urville, Ant.	17.71	35.86	7
Laramie, Wyoming	15.57	24.79	7
Bear Island, Norway	9.28	22.79	15

The CLAES aerosol measurements were compared to in situ aerosol measurements made by the University of Wyoming using a condensation nucleus counter ($r > 0.01$ μm) and an optical aerosol counter ($r > 0.15$ - 2.0 μm in 8 size bins). The measured cumulative particle size concentrations are fit with either unimodal or bimodal lognormal size distributions. Aerosol extinctions were calculated from the measured aerosol size distributions using a Mie scattering algorithm previously developed for the HALOE wavelengths (Hervig et al., 1996). Errors on aerosol extinction calculated from the in situ measurements were estimated, using a Monte Carlo simulation, to be on the order of 50%. The HALOE algorithm was expanded to include the CLAES wavelengths.

The CLAES and HALOE data were used to derive straight-forward, easily used conversions between aerosol absorption coefficient and surface area and volume. Conversions are developed for stratospheric sulfate and for polar stratospheric cloud (PSC) particles assumed to be composed of either nitric acid trihydrate (NAT) or liquid ternary aerosol (LTA). The cross section-moment relationships were determined by considering the extensive record of in situ aerosol size distribution measurements to calculate the aerosol absorption coefficients, surface areas, and volumes.

This work used profiles measured in stratospheric sulfate aerosols over Laramie, Wyoming (41°N), covering the complete cycle of the Pinatubo volcanic aerosol cloud, and profiles measured in sulfate aerosols and PSCs over McMurdo Station, Antarctica (78°S), during each Austral spring from 1990 to 1995 (see for example, Deshler et al., 1993). The results have been accepted for publication (Hervig and Deshler, 1998). Some of the results involving the CLAES data are described below.

For the case of sulfate aerosols, size distribution profiles measured over Laramie between August 1991 and March 1995 were used (43 profiles, ~1100 distributions). Absorptions were calculated from these size distributions using Mie theory with the refractive indices for sulfate aerosols. The balloonborne sampling did not include water vapor, and a constant 5 ppmv was assumed to compute the sulfate aerosol composition (Steele and Hamill, 1981). The refractive index versus composition was determined according to Palmer and Williams (1975). Volume (V) and surface area (S) are determined directly from the size distributions and the cross sections versus moments were fit according to:

$$M = A\, b_{e,a}(1)^{B} \qquad (1)$$

where M is either S or V, $b_{e,a}(1)$ is either the extinction or absorption coefficient at wavelength 1, and A and B are constants. These relationships have standard deviations of about 5% for volume and 15% for surface area. The surface areas and volumes determined from CLAES using this method under middle latitude conditions agree with in situ measurements to within ±20%.

The size distributions and refractive indices of PSC particles are different than for sulfate aerosols and thus the cross section-to-moment conversions will change for measurements in PSCs. The moment-cross section relationships in PSCs were investigated using a six year record (1990 to 1995) of aerosol size distribution profiles measured in sulfate aerosols and PSCs during the Austral spring over McMurdo Station (52 profiles, ~1800 distributions). Calculations with these profiles were made using either the refractive indices for crystalline NAT, or for LTA. The balloonborne sampling did not include water or nitric acid vapors and constant mixing ratios were assumed (2 ppmv H_2O and 5 ppmv HNO_3) to determine the equilibrium temperature of NAT (T_{NAT}) (Hanson and Mauersberger, 1988) and the equilibrium composition of LTA (Carslaw et al., 1995). The calculations assuming NAT used all aerosol observations where the temperature was below T_{NAT}, with the NAT refractive indices reported by Toon et al. (1994). For LTA, the equilibrium composition was determined and the appropriate LTA index from Hervig et al. (1997) was used. The relationships for LTA were determined from data where the calculated equilibrium nitric acid content of the droplets was greater than 5 wt.% (typically temperatures less than about 194 K). The relationships for NAT and LTA have the same form as Equation 1. The results of these comparisons of CLAES and in situ data were not as encouraging as the comparisons at mid latitudes.

In addition to this work attempts were made to use the CLAES aerosol extinction data at several wavelengths at mid latitudes to infer aerosol size distributions. The technique follows that developed using HALOE extinction data (Hervig et al., 1998), which infers an effective radius from a single extinction channel and then uses the effective radius to constrain the determination of aerosol median radius and distribution width from extinction ratios at other channels. Although the number of extinction ratio measurements available from CLAES were sufficient to determine the problem, the results were disappointing and this approach has been abandoned.

Publications resulting from this award:

Hervig, M. E., and T. Deshler, Aerosol surface area and volume inferred from HALOE and CLAES measurements of stratospheric aerosols, *J. Geophys. Res.*, in press, 1998.

Abstract. Relationships are presented for converting remote measurements of stratospheric aerosols to surface area and volume densities. Measurements from the Halogen Occultation Experiment (HALOE), the Cryogenic Limb Array Etalon Spectrometer (CLAES), and the Improved Limb Atmospheric Spectrometer (ILAS) were considered. The relationships were derived using an extensive record of in situ aerosol size distribution measurements made in sulfate aerosols and in polar stratospheric clouds (PSCs). Conversions were derived for sulfate aerosols using the appropriate refractive indices, and for PSCs using the refractive indices for either nitric acid trihydrate or liquid ternary H2SO4-H2O -HNO3 aerosols. Coincident measurements from HALOE, CLAES, and in situ particle counters are in generally good agreement at middle latitudes, for surface area and volume densities. Analysis of CLAES measurements under cold polar conditions suggests that these data should be used with caution.

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