# OBSERVATIONS OF AEROSOL BY THE HALOE EXPERIMENT ONBOARD UARS: A PRELIMINARY VALIDATION

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Abstract. The HALOE experiment measures vertical profiles of aerosol extinction at five infrared wavelengths. Four of these observations are obtained using a combination of gas filter and broadband radiometer measurements in bands of HF, HCl, CH4, and NO centered at wavelengths of 2.45, 3.40, 3.45, and 5.26  $\mu m$ , respectively. The fifth is obtained using broadband radiometer measurements of  $CO_2$  transmission at 2.79  $\mu m$ . Error analysis shows that the random extinction uncertainties are generally less than 10% in the aerosol layer, increasing to over 20% at the profile tops. HALOE spectral extinction measurements are shown to be consistent with predicted spectral extinction for stratospheric sulfate aerosol. Profile comparisons between HALOE and independent sources result in generally good agreement in the shape and magnitude of peak extinction and the altitude where the peak extinction occurs. In addition, global aerosol distributions obtained from the data are consistent with expected aerosol morphology. Although the validation is preliminary, the HALOE aerosol data appear to be of excellent quality and to accurately represent optical characteristics and distribution of the aerosols.

#### Introduction

Increased aerosol loading from the eruption of Mt. Pinatubo could have important effects on stratospheric temperatures and chemistry. In addition, the aerosol can have serious effects on remote sensing experiments by causing contaminant absorption or emission which can saturate measurements or be incorrectly interpreted as nonaerosol signal. Measurements made by the Halogen Occultation Experiment (HALOE) onboard the Upper Atmosphere Research Satellite (UARS) are used to infer profiles of limb-path aerosol transmission and extinction coefficient at five wavelengths. HALOE is a solar occultation experiment which measures attenuation of solar intensity by the atmosphere in eight infrared bands. The experiment is described in detail by Russell et al. [1993]. HALOE uses four gas filter channels and four broadband radiometer channels. Additionally, each gas filter measurement is accompanied by a broadband radiometer measurement at the same wavelength. The broadband radiometer measurements are seriously affected by attenuation due to aerosol, and signal corrections are essential in order to obtain useful mixing ratios in the presence of The gas filter measurements however, are essentially unaffected by spectrally flat aerosol attenuation and they allow accurate measurements to be made of attenuation by the primary gas in each channel. After the gas mixing ratio is retrieved in each gas filter channel, the broadband portion can be used to infer the aerosol

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transmission. Aerosol transmission is also obtained using one of the broadband radiometer channels centered on the 2.79 µm CO<sub>2</sub> band. This is done assuming a constant CO<sub>2</sub> mixing ratio and using independent temperature and pressure data provided by the National Meteorological Center (NMC). The aerosol transmission profiles are then used to retrieve vertical distributions of aerosol extinction coefficient. The physical, vertical resolution provided by the instrument is 2 km, and profiles are over sampled at 0.3 km vertical increments. The current algorithm retrieves aerosol extinction at a vertical spacing of 3 km for the gas filter channels and 1.5 km for the CO2 channel. Although finer resolution is obtainable for the gas filter channels, computational considerations favor the coarse vertical spacing. The aerosol extinction profiles serve both as useful physical measurements and to correct the broadband radiometer measurements for aerosol effects. The method for removal of aerosol effects from the radiometer signals will be presented in a later paper. In this paper, we will describe measurements of spectral aerosol extinction by HALOE, the measurement error analysis, global aerosol observations, and comparisons with theory and independent measurements as a preliminary validation of the aerosol measurements.

#### Aerosol measurement method

As previously mentioned, two different methods are used to infer limb path aerosol transmissions. The first uses gas filter measurements of HF, HCl, CH4, and NO at center wavelengths of 2.45, 3.40, 3.45, and 5.26  $\mu$ m, respectively. A gas filter measurement is made by splitting the incoming solar ray into two paths, one passing through a vacuum and the other through a cell containing the gas to be measured. An electronic gain adjustment is used in one detector circuit to adjust the signal output so that the two electro-optical paths are matched when the target gas is not present in the intervening atmosphere (i.e., "balanced" outside the atmosphere). When the target gas (e.g. HCl) is present in the intervening atmosphere, a spectral content is introduced to the incoming energy which is correlated to the absorption line spectrum in the gas cell. This causes a signal difference between the two paths ( $\Delta V$ ) which is strongly correlated to the amount of target gas in the intervening atmosphere. In the simplest form (neglecting instrument terms), the gas mixing ratio can be inferred from the ratio of  $\Delta V$  to the vacuum path signal, V, according to:

$$\frac{\Delta V}{V} = \frac{\int (\tau_A \tau_{aer} \tau_g G - \tau_A \tau_{aer}) d\gamma}{\int \tau_A \tau_{aer} d\gamma}$$
(1)

where  $\gamma$  is wave number and the integral is over the spectral bandpass,  $\tau_A$  is the atmospheric gas transmission,  $\tau_{acr}$  is the aerosol transmission,  $\tau_g$  is the gas cell transmission, and G is the electronic gain. The refractive index of a stratospheric aerosol particle, typically composed of 75% H<sub>2</sub>SO<sub>4</sub> and 25% H<sub>2</sub>O [Rosen, 1971], at the NO channel bandpass center wavelength (5.263  $\mu m$ ) is 1.342 - 0.144i [Palmer and Williams, 1975]. Over the bandpass, the difference in refractive index is  $\pm 0.005$  in the real part and  $\pm 0.009$  in the imaginary part. Since the refractive index determines the

optical properties, such an aerosol is essentially spectrally flat across the bandpass. The optical properties of the aerosol are similarly flat spectrally for the other channels. Transmission by sulfate aerosol may, therefore, be treated as constant in the bandpass, and removed from the integrals. Equation 1 then becomes:

$$\frac{\Delta V}{V} = \frac{\int (\tau_A \tau_g G - \tau_A) d\gamma}{\int \tau_A d\gamma}$$
 (2)

which contains only terms involving gas transmission and therefore, allows accurate retrieval of the principal gas mixing ratio in each channel. Once the gas mixing ratio has been retrieved, the V signal may be simulated without aerosol attenuation so that the ratio of measured (V<sub>meas</sub>) to simulated (V<sub>sim</sub>) vacuum path signals will give the aerosol transmission:

$$\frac{V_{meas}}{V_{sim}} = \frac{\tau_{aer} \int \tau_A d\gamma}{\int \tau_A d\gamma} = \tau_{aer}$$
 (3)

The second method of inferral uses broadband radiometer measurements of CO2 transmission centered at a wavelength of 2.79 µm. A measured CO<sub>2</sub> transmission profile is used to register the measurement altitudes in pressure space by matching the measured profile shape with a simulated CO2 transmission profile [Park et. al., 1979]. The profile shapes are matched above the aerosol layer. The simulated CO2 transmission is calculated using the temperature/pressure profile assuming a constant CO2 mixing ratio profile (352 ppmv). Although the measured signal in the CO<sub>2</sub> channel is due to CO<sub>2</sub> and aerosol attenuation, the simulated signal includes only CO2 attenuation. As in the other HALOE channels, aerosol transmission is essentially spectrally flat across the CO<sub>2</sub> bandpass and may be removed from the integral over wavenumber. The transmission due to aerosol, therefore, may be inferred from the ratio of measured to simulated CO2 signals, as in equation 3. The reader is referred to Finger et. al. [1965] and Gelman et. al. [1986] for discussions of stratospheric NMC temperature and pressure data.

Transmission and extinction coefficient are related by the expression:

$$\tau_{qer} = \exp(-\int \beta_e dL) \tag{4}$$

where L is optical path length along the limb, and  $\beta_e$  is the

aerosol extinction coefficient (km-1) The transmission profiles are used to retrieve profiles of aerosol extinction coefficient. In the retrieval, an extinction profile is modified from the top down in an "onion-peeling" fashion until the calculated transmission profile matches the measured transmission profile.

Uncertainties in taer will come from errors in the measured and simulated V signals. For the gas filter channel aerosol inferral, errors in tager include errors in the retrieved mixing ratio of the principal gas, errors in the mixing ratios of interfering species in the broad-filter bandpass (e.g. H<sub>2</sub>O in the NO bandpass), and other experimental and model errors. Systematic and random errors in the HALOE measurements are discussed in detail by Russell et al. [1993] and will not be discussed here. Uncertainties in the retrieved mixing ratios are calculated operationally in the HALOE data processing. The effects of errors in  $V_{sim}$  and  $V_{meas}$  on the uncertainty in  $\tau_{aer}$  and retrieved aerosol extinction were incorporated in a Monte Carlo analysis with retrievals of actual observations to determine the random uncertainties of the gas filter channel aerosol measurements. Profiles of retrieved aerosol extinction for the gas filter channel measurements are shown in Figure 1 for observations made at 15.8°S, 161.6°E on October 6, 1992. The vertical spacing in the retrievals was 3 km. The calculated uncertainty profiles and uncertainty bounds (measurement ± uncertainty) are plotted with each extinction profile. Note that the four measurements obtain nearly identical profile shapes at pressures above 20 hPa. The maximum retrieved extinction values, roughly 2 x 10-3 km<sup>-1</sup> at 50 hPa, represent heavy aerosol loading. The random uncertainties are less than 10% of the measurement values in the aerosol layer (100 to 30 hPa) for all four profiles. Random uncertainties above the aerosol layer increase to 20 to 30% for the CH4, HCl, and NO channel measurements. The HF channel aerosol profile exhibits greater than 100% random uncertainty at 15 hPa, owing to a sharp decrease in extinction and a corresponding increase in the magnitude of the uncertainty. The calculated uncertainties discussed above represent the random errors in the measurements. The systematic uncertainties have not yet been rigorously determined, but are estimated to be less than 15%. This analysis applies to a specific set of measurements and different, but not greatly varied, results may be expected for other measurement conditions. For the CO2 channel aerosol inferral, errors in V<sub>sim</sub> include errors in the NMC pressure/temperature data and errors in the assumed CO<sub>2</sub> mixing ratio, in addition to other experimental and model errors. The uncertainty in this aerosol measurement was not rigorously determined at the time of this writing.

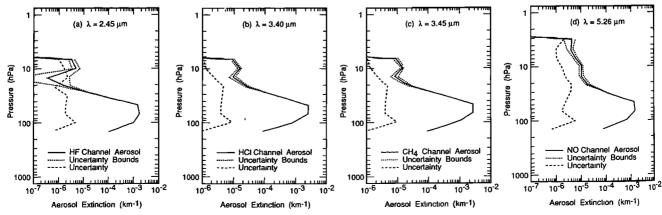


Fig. 1. Aerosol extinction profiles for the four gas filter channels. Each is shown with the calculated random uncertainty profile and uncertainty bounds.

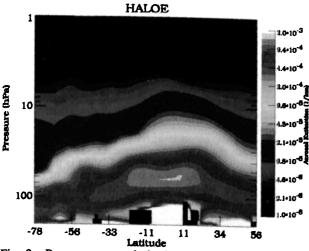


Fig. 2. Pressure versus latitude cross section of 5.26 µm aerosol extinction, sunsets from September 21, 1992, to October 15, 1992.

#### Global observations

HALOE sampling covers a near-global latitude range over periods of 20 to 30 days, obtaining two complete longitude sweeps each day, one at the latitude of sunsets and the other at the latitude of sunrises. Figure 2 is a pressure versus latitude cross section of zonal mean 5.26 µm aerosol extinction from sunset data. The data represent a type of time series, beginning at 56°N on September 21, 1992 and ending at 78°S on October 15, 1992. The black regions near 150 hPa represent missing data. The high extinction values at pressures above 150 hPa from 33°S to 50°N are likely due to

Pressure (hPa)

1000

0.0000

ALOE 39.7N 265.4E

0.0016 Aerosol Extinction (km-1)

0.0024

0.0008

cirrus cloud layers. The data show a layer of maximum extinction (about 1.5 x 10-3 km-1) near 60 hPa in the equatorial regions, owing to the eruption of Mt. Pinatubo (15.1°N, 120.4°W) 15 months prior to these observations. This layer decreases in magnitude and altitude towards higher latitudes due to transport of the aerosol by global circulations. Trepte and Hitchman [1992] have related global circulations and aerosol morphology following tropical volcanic eruptions and show aerosol distributions similar to those observed by HALOE. An interesting feature in Figure 2 is the relatively high extinction at pressures above 100 hPa between 55°S and 65°S. This feature corresponds with the location of the polar vortex where the temperatures (less than 195K) suggest polar stratospheric clouds.

### Comparisons

We report here on a comparison between HALOE aerosol extinction measurements and aerosol extinction calculated from a profile of aerosol size distribution measured by the University of Wyoming using balloonborne particle counters [Deshler et. al., 1993]. The detailed characteristics of these particle counters have been described by Hofmann and Deshler [1991]. The extinction profile calculated from 1 km averages of the measured aerosol size distributions at 41°N, 254°E on August 8, 1992, will be compared with HALOE profiles of aerosol extinction retrieved at 3 km vertical spacing at 40°N, 265°E and 40°N, 241°E on August 9, 1992. Although a time difference of one day exists, the spatial coincidence was good. The extinction coefficient for a population of aerosol may be calculated through Mie theory using the aerosol size distribution and refractive index for the wavelength of interest, assuming spherical particles, which is a valid assumption for the liquid sulfuric acid-water droplets. The bulk of stratospheric aerosol are a mixture of H<sub>2</sub>SO<sub>4</sub> and H<sub>2</sub>O and can generally be considered 75%

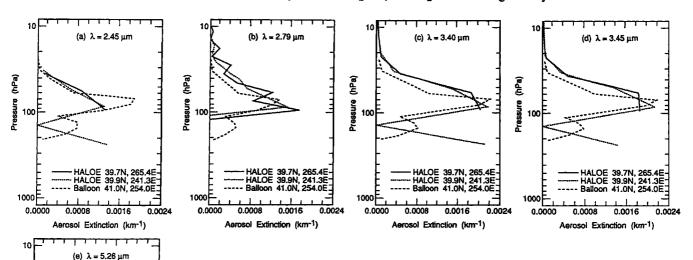


Fig. 3. Comparison of HALOE aerosol extinction profiles measured August 9, 1992, with aerosol extinction calculated from the in situ particle measurements on August 8, 1992. Comparisons are made for the five HALOE wavelengths.

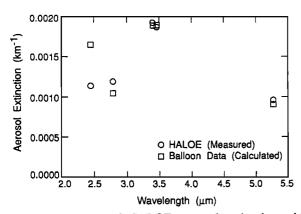


Fig. 4. Comparison of HALOE spectral extinction with values calculated from the in situ particle measurements. The values are averages in a layer from 60 to 100 hPa.

H<sub>2</sub>SO<sub>4</sub> in weight composition [Rosen, 1971; Toon and Pollack, 1976; Hofmann and Rosen, 1983]. Measurements of the refractive indices of sulfuric acid-water mixtures are given in Palmer and Williams [1975] at 300K for wavelengths from 2.5 to 25 μm and compositions from 25 to 95% H<sub>2</sub>SO<sub>4</sub> by weight. These indices were corrected for temperature using the Lorentz-Lorenz formula [Steele and Hamill, 1981]. Extinction was calculated for the HALOE measurement wavelengths using the log-normal size distribution at each level in the balloon profile, assuming 75% H<sub>2</sub>SO<sub>4</sub> weight composition of the aerosol.

Comparisons of the extinction profiles for the five aerosol measurement wavelengths are shown in Figure 3. balloon and satellite measurements agree on the altitude and magnitude of the peak extinction as well as the general shape of the aerosol layer, except for the 2.45 µm comparison where the HALOE measurements are 25% less than the calculated values. The offset in the 2.45  $\mu m\,$  comparison may be due to greater sensitivity to aerosol size as wavelength decreases. Some differences in the aerosol observed by each measurement may be expected given the spatial and temporal separation of the two measurements. At pressures less than 60 hPa, the HALOE measurements tend to be greater than the calculated values and at pressures above 100 hPa the HALOE results become much smaller than the calculated values. The uncertainty in the HALOE extinctions increases for altitudes below the tropopause. In the upper troposphere, the occurrence of cirrus clouds is possible. Given the temporal and spatial separation of the two measurements, such a phenomenon is unlikely to have been observed by both. Overall, the agreement between the two techniques is encouraging, especially when considering the very different nature of each.

The wavelength dependence of extinction measured by HALOE and that calculated from the in situ measurements are compared in Figure 4, serving as a check of the spectral consistency of each. In Figure 4, average extinction in a layer from 100 to 60 hPa (layer of peak extinction) is plotted as a function of wavelength. The agreement at the 2.79, 3.40, 3.45, and 5.26 µm wavelengths is excellent, while at 2.45 um, the difference is greater. The good spectral agreement between the measurements and calculations would suggest that the in situ measurements and HALOE both accurately represent the aerosol population and that the assumed aerosol composition, 75%  $H_2SO_4$ , is consistent with the true atmospheric aerosol composition.

## Summary

Measurements of aerosol extinction at five wavelengths by HALOE have been described. The calculated random

extinction uncertainties were less than 10% in the aerosol layer, increasing to over 20% above the aerosol layer, as the measured extinction decreased. The systematic uncertainties have not been rigorously determined at this time. The pressure versus latitude cross section of aerosol extinction shows morphology consistent with global circulations. The profile comparisons with calculations made using the University of Wyoming balloon data show good agreement in magnitude, shape, and altitude of the peak extinction layer. Additionally, the wavelength dependence of the HALOE extinction measurements agrees well with extinction calculated from the in situ measurements, assuming a 75% sulfate composition. Although the validation is preliminary, the HALOE aerosol measurements appear to be of excellent quality, offering a reliable global data set for scientific

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