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Validation of hydrogen fluoride measurements made by the Halogen Occultation Experiment from the UARS platform

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Abstract. The hydrogen fluoride (HF) molecule is important as a tracer and for study of chlorine input to the stratosphere due to CFC's. This paper describes the characteristics of and data from the Halogen Occultation Experiment (HALOE) HF channel, including steps taken to validate the results. The on-orbit precision of the HF measurements is shown to be better than 0.04 parts per billion by volume (ppbv) to 0.06 ppbv throughout the stratosphere. The estimated accuracy is 14% to 27% depending on altitude. The internal consistency of the HF measurements is excellent as judged by sunrise/sunset differences and comparison with HALOE CH₄ distributions. The mean difference between HALOE HF and correlative balloon underflight measurements is <7% from 5 mbar to 50 mbar. Comparisons with the shuttle ATLAS 1 Atmospheric Trace Molecules Observed by Spectroscopy (ATMOS) data are not as good and there is a systematic difference between HALOE (smaller) and ATMOS (larger) ranging from 10% to 20% at altitudes above the 10-mbar pressure level. Differences with ATMOS reach as much as 40% or more below the 10-mbar level. The larger differences in this region are believed to be due to dynamical influences on HF coupled with wide separations in space and time between HALOE and ATMOS measurements. Analysis of HALOE HF pressure versus longitude cross sections shows that obtaining close space and time coincidence can be very important in comparing tracer distributions. Typical characteristics of a pressure versus latitude cross section and polar orthographic projection are also discussed. Comparisons with latitudinal distributions of tracer measurements from previous experiments show similar features like the tropical double minimum due to the semiannual oscillation. All comparisons and analyses conducted provide good confidence in the validity of the HALOE HF results.

1. Introduction

The Halogen Occultation Experiment (HALOE) was launched on board the Upper Atmosphere Research Satellite on September 12, 1991, into a 57° inclination, 585 km nearly circular orbit. HALOE is a satellite solar occultation experiment which uses gas filter and broadband radiometry to measure vertical profiles of HCl, HF, CH₄, H₂O, NO, NO₂, O₃, aerosol extinction, and temperature versus pressure [see *Russell et al.*, 1977; *Russell*, 1980]. Measurements are made nearly globally over the latitude range from 80°N to 80°S. Approximately 15 sunrises and 15 sunsets are observed each day, usually in separate hemispheres, providing coverage of the full longitude range over narrow latitude

bands. Details of the experiment, geographic coverage of the measurements, instrument techniques, orbital performance, error mechanisms, and initial results are provided by *Russell et al.* [1993a]. The experiment has operated essentially without flaw from the time it was turned on, October 11, 1991, to the present and has provided a detailed data set for study of the photochemistry and dynamics of the stratosphere and mesosphere.

Stratospheric hydrogen fluoride (HF) is a crucial molecule because of its role as an indicator of chlorine input to the stratosphere due to human activities involving the chlorofluorocarbons (CFCs). When HF measurements are combined with hydrogen chloride (HCl) data, another HALOE molecule described in a companion paper in this issue, the relative importance of anthropogenic and natural sources of chlorine and changes with time can be studied. This is true because for chlorine, both natural and anthropogenic chlorine origins are important (e.g., CH₃Cl from the oceans and CFCs used industrially), but fluorine-bearing chemicals released into the stratosphere are believed to have negligible natural sources [*Cicerone*, 1981]. Consequently, HF, which is a postdissociation product of CFCs in the stratosphere, becomes an indicator of the anthropogenic chlorine input [e.g., *Molina and Rowland*, 1974; *Stolarski and Rundel*, 1975]. It has also been pointed out by *Sze* [1978] and *Sze and Ko* [1981] that since HF has a very long lifetime and while HCl is more reactive, particularly with OH, the combination of HF and HCl observed simultaneously should be useful in indirectly studying the OH chemistry. Finally, its very long lifetime makes HF an excellent

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tracer of atmospheric motions. This has been demonstrated in HALOE observations by the strong anticorrelations with HALOE CH_4 , which has an opposite vertical gradient to HF [Russell et al., 1993b; Luo et al., 1994a, b].

The vertical distribution and column amount of stratospheric HF have been measured by a number of balloon, aircraft, and ground-based instruments [Zander, 1981; Mankin and Coffey, 1983; Park et al., 1984; Zander et al., 1987a, b; Carli and Park, 1988; Coffey et al., 1989; Toon et al., 1989; Mankin et al., 1990; Wallace and Livingston, 1991; Toon et al., 1992]. Hydrogen fluoride has also been observed from space by the atmospheric trace molecule spectroscopy (ATMOS) experiment on Spacelab 3 [Raper et al., 1987; Zander et al., 1990] during late April to early May 1985 and in three subsequent ATMOS flights on the space shuttle ATLAS payload in late March/early April 1992, April 1993, and October/November 1994. The Spacelab 3 flight provided two zonally averaged profiles: a sunset at 29°N and sunrise at 48°S . The latter flights covered much broader latitude ranges. All of these HF observations have provided important information on the HF column amount, its seasonal and latitudinal dependence, and temporal trends. They also served to characterize the vertical profile very well, but the collected data set lacks good global coverage. This gap has now been filled by the HALOE experiment on UARS. Coverage over the full latitude range from 80°N to 80°S exists simultaneous with the other HALOE molecules. The HF data set now includes five Antarctic springs, four Arctic springs, and four summers in each polar region. Discussion of the HCl/HF ratio measured by HALOE will be the subject of a later paper.

In this paper we will focus on HF validation, data quality, and data limitation studies that have been performed. The performance of the HF channel has been excellent. The only known problem, as discussed later, is the effect of sunspots on the data, which affect less than 5% of the measurements. After a brief description of the measurement method we will describe the HF channel characteristics and signal profiles, error estimates and orbital precision values, internal data consistency analyses, comparisons with ground-based, balloon, and ATMOS observations, comparisons with model calculations, and finally, we will include some examples of latitude cross section, longitude cross section, and polar orthographic projection data products.

2. HALOE Measurement Approach, Channel Characteristics, and Signal Profiles

All HALOE measurements are made in the solar absorption mode, while the Sun is being occulted by the Earth limb during spacecraft sunrise and sunset. The HF measurement is made using the gas filter correlation radiometer instrument technique. This technique is powerful because it provides very high sensitivity (the ability to measure incoming radiance signal differences to 1 part in 10^5) and high spectral resolution (of the order of 0.07 cm^{-1}). This capability is achieved without dispersing the light, as is done with grating spectrometry, and without the complication of moving mirrors and high data rates, which come with interferometry. Energy from the Sun, after passing through the atmosphere, enters the instrument and is split into two paths: one contains a gas cell filled with HF and nitrogen at a total pressure of 0.2 atmospheres and 43% HF mixing ratio by volume and the second is a vacuum path. A detector is located in each path (see Russell et al. [1993a], for details). The signal used for data processing and atmospheric HF retrieval is the

difference signal in volts (ΔV) between the gas path and the vacuum path divided by the vacuum path signal (V). Processing the data in this way has the great advantage that the modulation signal ($\Delta V/V$) used for HF retrieval is virtually independent of atmospheric aerosol extinction. This is very important in light of the fact that Mount Pinatubo in the Philippines erupted June 12, 1991, and significantly increased the stratospheric aerosol loading.

The HF channel spectral filter 5% relative response points are 4025 cm^{-1} and 4135 cm^{-1} (see Russell et al. [1993a] for a plot). This interval includes the R1, R2, and R3 HF lines centered at 4038.96 cm^{-1} , 4075.29 cm^{-1} , and 4109.93 cm^{-1} , respectively. The spectral range also contains atmospheric absorption lines due to CH_4 and H_2O , which have mixing ratios approximately 1000 times greater than HF. These interference absorbers contribute only a small amount to the measured signal, however, with the largest effect on retrieved HF being $\sim 10\%$ at $\sim 16\text{ km}$ and smaller at higher altitudes (see Table 1 discussed below). This is because of the ability of the gas filter method to substantially reject such effects. Solar CO lines are also present inside the HF spectral band pass, and these lines are included in the forward model used in the retrieval process. One problem that has occurred is the effect of sunspots on the HF signal, which is greater than for any other HALOE channel. It is believed that the effect is due largely to spectral lines in the sunspot regions which are not included in the processing software, and it is very difficult to correct the data. An example of sunspot effects and further discussion is included in a later section of this paper.

The normalized instantaneous vertical field of view (IFOV) function in the HF channel (Figure 1) approximates a Gaussian shape with a full width at half maximum of ~ 2 arc min or $\sim 1.6\text{ km}$ at the Earth limb. Off-axis rejection performance in orbit meets or exceeds requirements at all IFOV positions and by more than an order of magnitude ($<0.05\%$ of full signal) when the IFOV center is 6 arc minutes from the solar edge. The achievable vertical resolution of the HF measurement is $\sim 2\text{ km}$ when instrument noise and retrieval errors are considered, but the current archived data set (version 17) was retrieved at a vertical point spacing of 3 km giving an effective vertical resolution of about 4.5 km . Future data versions will be retrieved at 0.6-km intervals allowing the full vertical resolution of 2 km to be realized.

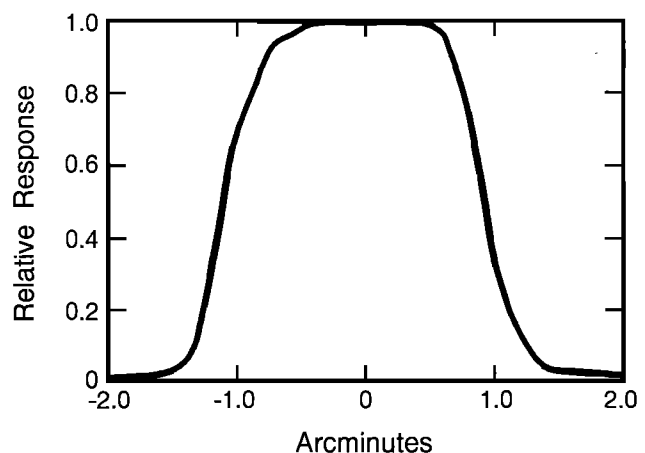


Figure 1. Halogen occultation experiment (HALOE) normalized instantaneous vertical field-of-view function in the hydrogen fluoride (HF) channel.

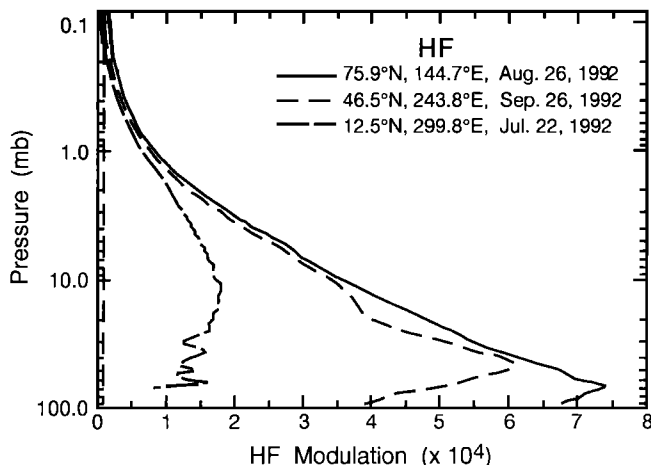


Figure 2. HALOE HF channel pressure versus modulation signal (unitless) at low, middle, and high latitudes in summer and early fall of 1992. The vertical dashed line is the instrument noise level.

Typical HF modulation signal versus altitude (pressure) profiles for low, middle, and high latitudes for northern summer and early fall of 1992 are shown in Figure 2. The vertical dashed line represents the modulation noise level. Note that since modulation is ΔV divided by V , the signal value is unitless. These profiles reflect the latitudinal distribution in HF, which has generally low mixing ratios in the tropics, due to tropical upwelling bringing air poor in HF up from the troposphere and high values of HF at high latitudes due to descent bringing HF-rich air downward. Note that the signal to noise (S/N) reaches a value of 1 at about the 0.2-mbar level. Generally, single-profile HF retrievals are not usable at altitudes any higher than the 0.5-mbar level. The HF signals, measured by the gas filter method, are affected by the magnitude of the relative velocity between the spacecraft and the atmosphere. This is due to Doppler shifting of atmospheric HF lines relative to HF lines in the gas filter cell. When the beta

angle (angle between the orbit plane and the Earth-Sun line) is small, the relative velocity, and hence Doppler shift, is greatest, causing a signal reduction due to the loss of full atmospheric/gas cell line correlation. This effect is taken into account in the data reduction, but it is quite small (maximum signal reduction of 10%) since the HF lines in the gas cell are quite broad. Even taking the Doppler shift into account, there appears to be a slight (<5%) beta angle dependence of unknown origin at the highest altitudes ($p < 1$ mbar), but this effect makes only a small change to the total error bar.

3. HALOE HF Channel Error Estimates

We have performed a detailed study of expected errors in the HF channel based on a combination of ground testing, orbital measurements, forward model uncertainties, and retrieval effects. The results of the study for selected pressure levels are presented in Table 1. The random and systematic error entries in the table provide both estimated precision and accuracy information needed for science studies. The estimated total error, obtained by calculating the root-sum-square of all individual errors, varies from a high of 27% at 100 mbar to as low as 14% at 5 mbar. These error estimates were obtained by calculating a signal profile based on realistic temperature-pressure, HF, CH₄, and H₂O profiles and the forward radiance calculation model used in the retrieval algorithm. Next, various kinds of errors were added to the signal. The true signal with errors added was then used to retrieve the HF profile. Error entries in the table are standard deviations obtained by comparing the assumed true HF profile used in the signal simulation with profiles retrieved from the “realistic” signal. The Monte Carlo technique and the formal method of Rodgers [1990] were used to estimate errors.

The instrument noise used in the simulations was obtained from statistical analysis of exoatmospheric data. It includes therefore digitization noise as well as detector and electronics noise. The tracker noise arises from solar pointer/tracker jitter effects which were also obtained from analysis of flight data. The H₂O

Table 1. HALOE HF Channel Mixing Ratio Error Estimates 1 σ in Percent

Error Parameter	Pressure Level, mbar						
	1	3	5	10	30	50	100
<i>Random Errors</i>							
Instrument noise	6	6	6	5	5	3	1
Tracker noise	1	1	1	1	4	7	10
H ₂ O	2	2	2	2	2	2	2
CH ₄	0	0	1	2	3	4	5
Temperature	1	1	1	3	5	5	5
<i>Systematic Errors</i>							
Forward model	9	9	9	10	11	12	18
Electronics	8	8	7	6	5	5	5
H ₂ O bias	4	4	4	4	4	4	4
CH ₄ bias	0	0	1	3	5	7	10
Temperature bias	3	3	3	4	6	9	9
Registration altitude	3	3	3	3	3	3	3
Total error, root-sum-square	15	15	14	15	18	21	27

and CH₄ entries reflect uncertainties in the HF retrieval due to errors in these interfering gases. These latter errors were based on error estimates for the HALOE H₂O and CH₄ channels. The HF errors due to temperature are based on error estimates for HALOE-retrieved temperature at levels above 10 mbar and errors in National Meteorological Center temperature fields below the 10-mbar level. The electronics uncertainties we included refer to errors arising due to electronics and field-of-view (FOV) deconvolution operations used in data processing, FOV mismatch effects, signal drift during an occultation event, gas channel balance offset [see *Russell et al.*, 1993a], linearity errors, and gain uncertainties. The forward model errors included uncertainties in calibration parameters, optical filter properties, gas cell conditions, absorption line strengths, spectral response characterization, and Doppler shift correction. The altitude registration uncertainty used is about 150 m. This value was determined from analysis of flight data.

The magnitudes of the various errors used in the simulations, the effects individual errors cause on the total estimated uncertainty, discussion of how the errors were applied to obtain the estimates in Table 1, and comparison of results from the Monte Carlo and *Rodgers* [1990] techniques are provided in the paper by *Deaver et al.* [1995].

4. Sunspot Effects on HF Retrievals

The HALOE HF retrieval can be heavily corrupted by sunspots when a sunspot or sunspot cluster occurs within the instantaneous field-of-view (IFOV). Because all HALOE channels share the same IFOV, sunspots can affect all channels, but by far the largest effects are seen in the HF data. Sometimes the entire retrieved profile is invalidated by sunspots, e.g., when the exoatmospheric gas/vacuum channel balance occurs in the presence of a sunspot or when the lockdown point on the Sun is overlapped by a sunspot [see *Russell et al.*, 1993a]. The problem is caused in part by the fact that the sunspot can have a spectrum of unknown absorption lines that are not considered by the retrieval algorithm. Sunspots are rich in molecular and atomic spectral absorption which can correlate and anticorrelate with atmospheric spectral features and the HALOE instrument gas cell spectra. In this case, totally unrealistic values of HF are retrieved, e.g. >1.5 parts per billion by volume (ppbv) to 2 ppbv through the entire vertical profile. At other times, during the limb scan, solar rotation can cause a sunspot to rotate into the IFOV that was not present when the exoatmospheric balance occurred. In this case, only certain altitude ranges are affected. An example of a profile like this is shown in Figure 3. Another effect occurs when atmospheric refraction causes the IFOV to effectively scan the solar disk during an occultation and possibly encounter sunspots [e.g., *Russell et al.*, 1993a]. We have found that the only time sunspots cause a data problem is when we observe a localized depression in the HF ΔV signal solar limb darkening curve obtained during the exoatmospheric solar scans which is $\sim > 5$ noise equivalent modulation (NEM) units. When the depression from the normal limb darkening curve is smaller than this, our experience has shown that any change in retrieved HF is not discernible in the results. We believe this kind of "step function" effect is related to spectral content in the sunspot. Studies are under way to better estimate the error due to sunspots using parameters in the HALOE data stream. It appears now that the effect is either so large that the data are unusable or so small that there is no problem and for this reason no entry appears in the error summary

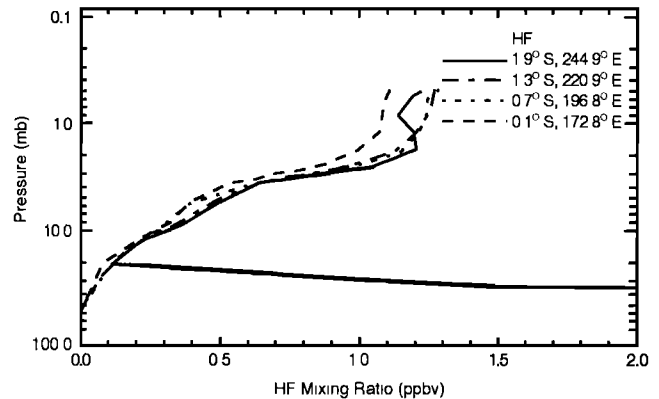


Figure 3. HALOE HF tropical sunset profiles measured on April 2, 1993. The sunspot contamination effect is clearly seen in the profile measured at 1.9°S.

of Table 1. Less than 5% of the HALOE events collected up to July 1993 have sunspot effects, and we list them in Table 2. The days and corresponding instrument modes (sunrise or sunset) where sunspots have obviously invalidated at least some of the HF data for the first 2 UARS years are included in the table. The data user is cautioned to review the data quality file released with the data and to note any events that have been flagged for sunspot contamination. Also, the data set itself should be carefully reviewed to look for any more subtle sunspot effects. Further, when a sunspot effect is flagged for HF, the other HALOE channels should be carefully evaluated. Because of the reasons discussed above, there is very little promise for future improvement of sunspot corrupted data.

5. On-Orbit Precision and Internal Data Consistency

We have determined an indicated precision for HALOE HF measurements by calculating the standard deviation about the daily zonal mean for various time periods and locations, e.g. tropics and summer middle and high latitudes. This is only an indicated upper limit precision, because atmosphere variability contributes to the standard deviation value in addition to random changes due to instrument noise. Retrieved zonal mean HF profiles at 14°N, 42°N, and 76°N during the summer of 1992 are shown in Figure 4 along with the standard deviations. These plots indicate a nearly constant or slightly increasing standard deviation with altitude at all three latitudes. The value is about 0.03 to 0.04 ppbv over the 100-mbar to 3-mbar range growing to ~ 0.1 ppbv near 0.4 mbar. The tropical standard deviation profile shows a peak in the lower stratosphere of about 0.07 ppbv. This is indicative of a general variability in the data at these altitudes, which could be due to atmospheric dynamics effects or aerosol effects on the pointer tracker. Work is currently under way to better understand results in this region. The estimated precision determined from a Monte Carlo retrieval analysis using an orbit-observed NEM for the HF channel is shown by the solid circles in the figure. Agreement between the estimated precision (σ_e) and the observed standard deviation (σ_o) is close in the 2-mbar range, and it is expected that σ_o would be greater than σ_e in the region below 10 mbar where dynamics effects are more important. Above about 2 mbar it appears that the estimated precision is too large. This is probably due to further signal

Table 2. HALOE HF Data Indications of Sunspot Contamination

Date	UARS Day	Latitude	Mode
Nov. 16, 1991	0066	35°N	rise
Jan. 10, 1992	0121	47°N	rise
Jan. 27, 1992	0138	30°N	set
Jan. 28, 1992	0139	26°N	set
Jan. 31, 1992	0142	11°N	rise
Feb. 1, 1992	0143	6°N	rise
Feb. 5, 1992	0147	15°S	rise
Feb. 7, 1992	0149	25°S	rise
Feb. 23, 1992	0165	73°S	set
Feb. 24, 1992	0166	72°S	set
Feb. 25, 1992	0167	73°S	set
April 24, 1992	0226	43°S	rise
July 12, 1992	0305	48°S	rise
Aug. 16, 1992	0340	59°N	set
Aug. 17, 1992	0341	62°N	set
Oct. 22, 1992	0407	43°N	rise
		69°S	set
Nov. 9, 1992	0425	9°S	set
Nov. 28, 1992	0444	45°S	rise
Nov. 29, 1992	0445	48°S	rise
Nov. 30, 1992	0446	50°S	rise
Dec. 11, 1992	0457	66°S	rise
Jan. 9, 1993	0486	49°S	set
Feb. 6, 1993	0514	52°N	set
March 10, 1993	0546	27°N	rise
April 2, 1993	0569	3°N	set
June 11, 1993	0639	60°N	rise
June 12, 1993	0640	53°N	rise
July 11, 1993	0669	41°N	set

smoothing which was not included in the Monte Carlo analysis. In general, the agreement is reasonable, indicating that the random behavior of the instrument is well understood.

An important test of data validity and instrument performance is comparison of sunrise and sunset profiles at the crossover points when the measurements overlap in latitude. This crossover occurs on the order of 10 days a year at various latitudes ranging from the tropics to high latitudes. Since HF is a long-lived gas in the stratosphere, no diurnal variability is expected and sunrise/sunset measurements should yield close to the same

values. The only differences expected would be due to space and time separations of the sunrise/sunset points, which would lead to mixing ratio differences due to dynamical effects. An example of these differences for the tropics is shown in Figure 5. This figure shows zonal mean HF profiles for sunrise (dotted) and sunset (solid) averaged for crossovers which occurred between November 19, 1991, and July 24, 1993, in the range of 20°N to 20°S. The means and root-mean-square (RMS) differences are also plotted. A total of 35 profile pairs separated by $\leq 1^\circ$ latitude, $\leq 15^\circ$ longitude, and ≤ 30 hours time were used. Note that mean differences are no more than 0.01 ppbv throughout most of the

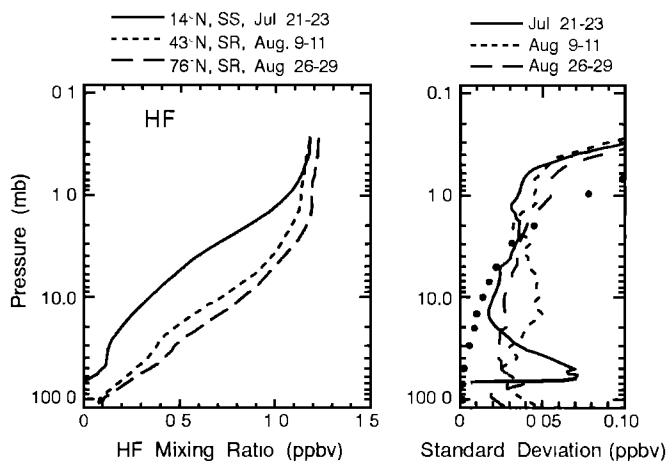


Figure 4. HALOE HF zonal mean mixing ratio and standard deviation about the mean at low, middle, and high latitudes during the summer of 1992. The circles are estimated precision values determined from retrieval simulations based on observed on-orbit instrument noise.

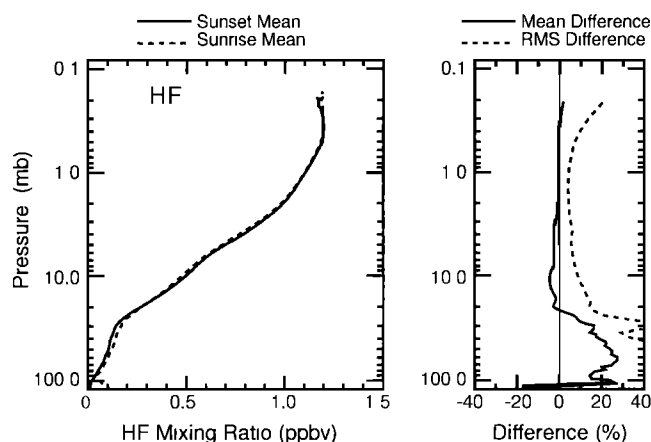


Figure 5. HALOE HF zonal mean profiles near 5°N and sunrise minus sunset mixing ratio differences in percent based on 35 profile pairs measured during the period November 19, 1991, to July 24, 1993.

vertical range from 100 mbar to 0.2 mbar with the largest differences occurring below 20 mbar and reaching a value of ~ 0.04 ppbv or ~ 20 to 30%. This larger difference could well be due to space and time differences between sunrise and sunset observations, which can cause larger mixing ratio differences at the lower altitudes where HF spatial gradients are larger. This figure is typical of sunrise/sunset differences seen at other latitudes. The other differences are small and indicate that no problems exist with the measurement or data processing due to things such as Doppler shift application and spectroscopic uncertainties; that is, no rise/set differences are expected and none of any consequence are observed.

We also performed another internal data consistency check by comparing pressure versus latitude and pressure versus longitude cross sections of HF and CH₄. Both are long-lived tracer molecules in the stratosphere, which have opposite vertical gradients. Therefore anticorrelation of cross-section features is expected. A typical example of this is the pressure versus longitude comparison in Plate 1 for 72°N on April 4, 1992, which shows a vortex structure. Note that at approximately 90°E and 200°E and at altitudes below the 20-mbar level there is a sharp gradient where HF increases from 0.4 to 0.8 ppbv and, at the same time, CH₄ sharply decreases from 1.2 ppbv to about 0.4 parts per million by volume (ppmv). Each parameter remains uniformly high (HF) or low (CH₄) in between these limits. Independent United Kingdom Meteorological Office (UKMO) wind information in the UARS database on this day shows that the HALOE measurement samples crossed well inside the northern vortex (poleward of the maximum wind line) in the 90°E–200°E range. Such CH₄ and HF behavior has been shown previously to be caused by vertical descent inside the vortex, bringing air rich in HF and poor in CH₄ down from above [e.g., Russell et al., 1993b]. Careful examination shows close qualitative anticorrelation in all features of the two cross sections. The broad anticorrelation inside the vortex has been noted; but in addition, the smaller-scale features inside the vortex, e.g., at 150°E and 30 mbar, is another example, as well as the deep HF minimum and CH₄ maximum centered at $\sim 40^\circ$ E and 30 mbar. The envelope of sharp HF and CH₄ gradients across the entire longitude range is also clearly correlated. A more quantitative verification of this is provided in Figure 6, which is a plot of HF versus CH₄ over the 50-mbar to 1-mbar-pressure range and for the time period from August 26 to 29, 1992, at 76°N. The least squares fit of a straight line through the data for HF values of ≤ 1.1

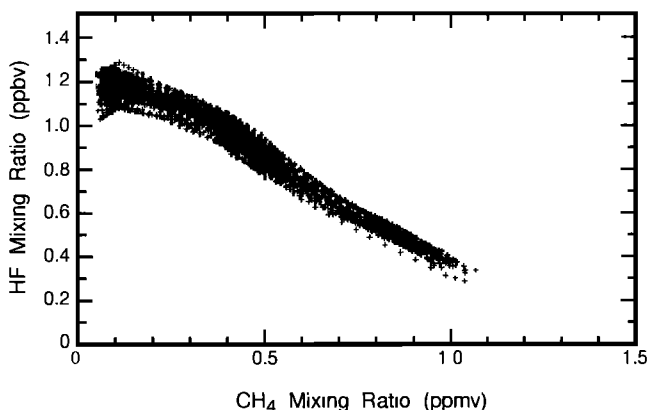


Figure 6. HALOE HF versus CH₄ mixing ratio over the 50-mbar to 1-mbar range and for sunrise events at 76°N, August 26, 27, 29, 1992.

ppbv gives a slope of -10^{-3} or a normalized mixing ratio slope of -1.0 (i.e., anticorrelated). Note that there is a slight “S” shape to the plot, which is most pronounced at the largest HF values. This is most likely due to chemistry combined with general circulation effects, which change the CH₄ and HF relationship at the low and high altitudes. When a plot like this is made for vortex conditions, the regression is very different due to chemistry-dynamics interactions. This is the subject of a paper by Luo et al. [1994b]. The anticorrelation between HALOE, CH₄, and HF retrievals is both qualitatively and quantitatively robust and shows a high degree of internal data consistency between these two very different channels.

6. HALOE HF Comparisons With Correlative Measurements and Model Results

HALOE data have been compared with a series of balloon measurements made using solar occultation [e.g., Toon et al., 1989; Toon, 1991] and far-infrared emission [Traub et al., 1991; Nolt, private communication, NASA, Langley Research Center, 1992]. Measurements have also been compared with the ATLAS 1 ATMOS data mentioned earlier and with ground-based infrared measurements of column amounts made from the Jungfraujoch Observatory in Switzerland [e.g., Zander et al., 1987a]. All of the balloon comparisons occurred at $\sim 34^\circ$ N, and ATMOS comparisons were made over a range of latitudes from the equator to $\sim 47^\circ$ S.

6.1. Balloon Comparisons

Table 3 provides the names of the investigators, measurement approach, the error bars, underflight dates, locations of the flights, and space and time differences between HALOE and balloon observations. Because coverage is limited with satellite solar occultation, it was usually not possible to obtain very close coincidence in space and time between HALOE and the correlative balloon observations. The protocol adopted by the HALOE science team was to strive for the closest possible coincidence in space first and time second, since HF is a slowly changing tracer molecule. Consequently, some comparisons are off by as much as 1 to 2 weeks in time. The composite error bar for the correlative set, obtained by computing the mean error weighted by the number of flights in the comparison set, is $\sim 7\%$. This is the best mean profile agreement that can be expected between HALOE HF and the balloon data. The mean profiles for the set as well as the mean and RMS differences are plotted in Figure 7. The largest mean difference anywhere in the range of observations is $\sim 53\%$. This occurs at the lowest altitude where dynamics effects and spatial/time coincidence are more important. Over the 5-mbar to 50-mbar range the mean differences are $<7\%$ which is within the error bar overlap of the balloon and HALOE data. The RMS differences range from 12% to 40% over most of the profile, and they reach 82% near 100 mbar where HF values are very small. Table 4 shows the calculated mean differences, the RMS differences, and the standard deviation at selected altitudes in both percent and mixing ratio units. Representative individual profile comparisons with balloon Fourier transform interferometer (FTIR) occultation [e.g., Toon, 1991] and far-infrared [e.g., Traub et al., 1991] observations are shown in Figures 8, 9, and 10, respectively. Note that the error bars of the HALOE data overlap with the Toon and Traub data (Figures 8 and 9, respectively) which is the best agreement that can be expected. Comparisons with independent balloon observations have been of great value in

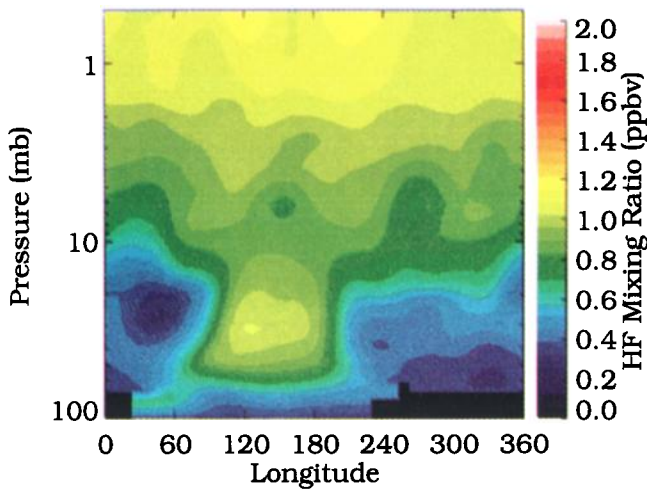


Plate 1. HALOE HF and CH₄ pressure versus longitude cross sections at 72°N, April 4, 1992.

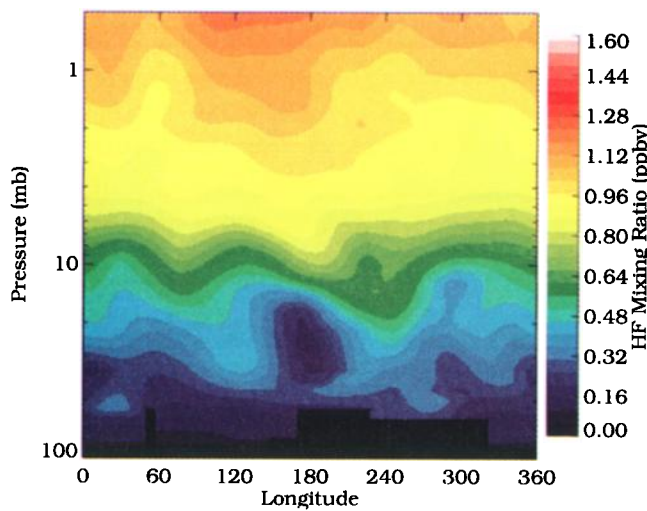


Plate 2. HALOE HF sunset pressure versus longitude cross section, May 7, 1992, at 34°N.

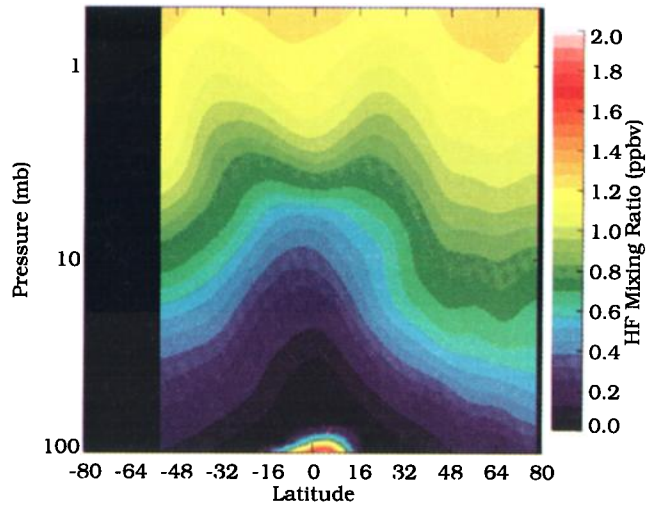


Plate 3. HALOE HF pressure versus latitude cross section (sunrises and sunsets combined) for April 12 to May 24, 1993.

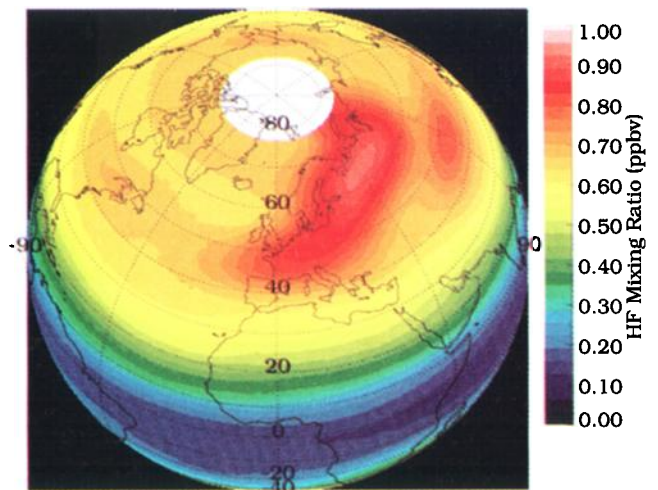


Plate 4. HALOE HF polar orthographic projection on the 20-mbar surface (sunrises and sunsets combined) for April 12 to May 24, 1993.

Table 3. HALOE HF Correlative Measurement Overpass Locations and Investigators

Investigator	Date of Underflight	Location of Underflight	Latitude/ Longitude	Time Difference	Measurement Displacement From HALOE Tangent Point	
					Latitude	Longitude
Traub far-infrared emission, accuracy 9%, precision 5%	May 29, 1992	Fort Sumner, N.M.	36.5°N/104.8°W	7 hours	1.3°S	12.1°E
	Sept. 29, 1992	Fort Sumner, N.M.	36.9°N/100.4°W	<1 day	0.5°N	19.5°E
	March 24, 1993	Daggett, Calif.	37.1°N/107.4°W	12.5 day	3.5°N	3.5°E
	May 23, 1994	Fort Sumner, N.M.	36.3°N/104.7°W	2.6 days	1.5°S	13.0°E
Toon infrared FTIR occultation, precision 5% at 40 km to 15% at 20 km	Sept. 14, 1992	Fort Sumner, N.M.	35.2°N/103.9°W	14 days	0.5°N	160.5°E
	April 3, 1993	Daggett, Calif.	34.8°N/111.1°W	2 days	2.2°N	0.4°E
	Sept. 26, 1993	Fort Sumner, N.M.	34.0°N/109.4°W	<1 day	0.4°N	0.5°E
	May 22, 1994	Fort Sumner, N.M.	36.6°N/109.7°W	1.5 days	0.9°N	16.3°W
Nolt far-infrared FTIR emission, accuracy 15% at <25-30 km, 8% at 3038 km, precision typically half of accuracy	May 4, 1992	Fort Sumner, N.M.	34.0°N/103.0°W	2.4 days	2.2°S	12.9°W

gaining confidence in the validity of the HF results. The HALOE error bars also overlap in the upper portion ($p \leq 10$ mbar) of the comparison with the Nolt et al. profile (Figure 10), but in the lower stratosphere they overlap for only one of the two HALOE profiles shown. The two HALOE profiles (taken at 245°E and 269°E) bracket the longitude of the balloon measurement but were taken three days later on May 7. Note the difference in the two HALOE profiles measured on the same day. These differences are much greater than the upper limit precision estimated using Figure 4. Plate 2 shows the HALOE HF pressure versus longitude cross section on May 7, which clearly indicates the degree of longitudinal variability, probably resulting from stratospheric wave activities. This longitudinal behavior also varies from day to day. Space and time coincidence therefore is very important in these comparisons, and we believe this is the reason for most of the differences noted in the lower stratosphere.

6.2. ATMOS HF Comparisons

Even though the shuttle ATMOS/ATLAS 1 flight in 1992 was in the same inclination orbit as UARS, differences in the orbital altitude and the time of day for insertion coupled with ATMOS optical filter usage strategy caused poor HALOE/ATMOS space and time coincidence. A window of 2° in latitude and 60° in longitude was selected for coincidence as a goal. Out of 11 comparisons, however, only 4 were within 1 or 2 days. The remaining were separated in time by 1 to 2 weeks. Consequently, in view of this large time separation the HALOE/ATMOS comparisons should be considered semiquantitative. The mean profiles, the mean difference profile, and the RMS difference are shown in Figure 11. The mean mixing ratio profiles do not have much geophysical significance because the coverage includes the 25°N to 47°S range. This includes HF profiles with very low values up

Table 4. Correlative Minus HALOE HF Measurement Comparison Statistics

Pressure Level, mbar	Mean Difference*		RMS Difference		Standard Deviation	
	%	pptv	%	pptv	%	pptv
5	1.4 ± 1.9	12.3 ± 17.3	17.3	156.9	5.7	52.1
7	-0.5 ± 1.2	-4.0 ± 10.6	11.7	95.6	3.9	31.8
10	4.2 ± 1.6	29.2 ± 11.5	15.4	107.9	4.9	34.6
15	4.1 ± 2.0	23.8 ± 11.9	18.9	109.6	6.1	35.7
20	0.9 ± 2.2	4.5 ± 11.3	20.2	101.6	6.7	33.8
30	2.8 ± 2.4	12.4 ± 10.5	21.6	94.9	7.1	31.3
50	-6.8 ± 6.5	-18.8 ± 17.9	39.5	108.9	15.9	43.8
70	16.9 ± 11.3	-23.9 ± 16.0	59.0	83.5	25.3	35.8
100	-53.0 ± 20.8	-44.4 ± 17.4	81.8	68.5	35.9	30.1

Percents are determined by dividing by the HALOE measurements; parts per trillion by volume (pptv).

*Second entry is the standard deviation of the mean difference obtained from dividing the standard deviation by the square root of the number of samples.

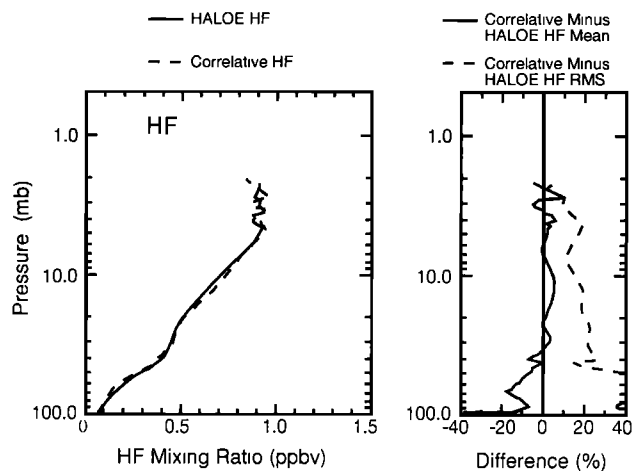


Figure 7. HALOE HF balloon comparison statistics based on nine correlative measurement underflights.

to 2 mbar (tropical profiles) and profiles with much higher mixing ratios for these altitudes at the higher latitudes. The mean profiles are useful, however, in giving the sense of differences between HALOE and ATMOS. Differences at altitudes above the 15-mbar level are $\sim < 10\%$ which is within the error bar overlap of the two experiments. Differences grow to $\sim 36\%$ at lower altitudes where dynamical changes over a 1- to 2-week time period could be very important with HALOE systematically lower in mixing ratio than ATMOS. Considering these factors, this level of agreement is reasonable.

6.3. Ground-Based HF Column Amount Comparisons

We have also performed comparisons with total HF column amount measurements made by the Liège group [e.g., Zander *et al.*, 1987a] from the International Scientific Station of the Jungfrauoch (Switzerland, 46.5°N, 8.0°E, 3580 m altitude). That team used high-resolution solar spectra obtained with both a grating instrument and two Fourier transform spectrometers covering the HF-related spectral interval used by HALOE. The estimated error in the ground-retrieved column amounts of HF is typically $\pm 4\%$.

We used a HALOE/Jungfrauoch spatial coincidence criterion of $\pm 3^\circ$ latitude and we applied HALOE daily zonal mean profiles

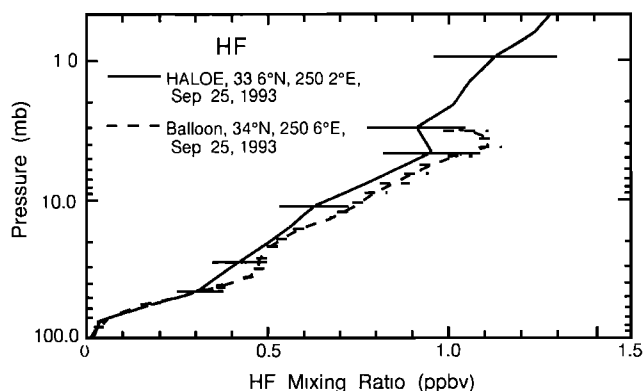


Figure 8. HALOE HF mixing ratio on September 25, 1993, compared with balloon solar occultation infrared measurements on September 26, 1993, at 34°N (G. C. Toon *et al.*, private communication, 1994).

in the comparison. The HALOE column amounts were obtained by integrating the zonal mean profile vertically above the lowest reliable HALOE measurement altitude, which we determined to be 50 mbar. We recognize that important column amount values between ~ 200 mbar and 50 mbar are not included, but we found occasional, rather small HALOE profile features of ~ 0.2 ppbv maximum magnitude at the lowest altitudes which caused the column sum to change significantly. We are not certain if these features are real and a longer HALOE time base is needed to study this. Figure 12 shows the data comparisons for 2 1/2 years of HALOE operations. Note that there is the expected column amount offset between the ground-based and the 50-mbar column amount data, but both show a similar annual increase. Also, a clear annual cycle can be seen. We conducted a fit to both data sets using annual, semiannual, and linear trend terms. The annual rate of increase based on the linear trend term is 4.79%/yr for the ground-based data and 5.77%/yr for HALOE. The reference column amount values used for the percentage calculations were the values of the linear terms (straight lines in the figure) at the time that the HALOE time series started. The lower altitude limit of the HALOE data is constrained by aerosol and cloud effects on the solar pointer/tracker.

The ground-based data show large excursions at times, predominantly during the January to April months (see, for example, February/March 1992), but generally lasting only 1 to 2 days; these are associated with HF-rich masses of polar air having intruded to midlatitudes as evidenced by trajectory investigations (R. Zander, private communication, 1994). Similar large excursions are not seen in the HALOE results, very likely because of the latitudinal and longitudinal coincidence criteria applied to the HALOE data binning for the comparison.

6.4. Comparisons With Previous Measurements and Model Results

In Figure 13 we have compared HALOE data in May 1992 at 48°S with ATMOS Spacelab 3 results measured on May 5, 1985, and ATMOS/ATLAS 1 results measured at southern midlatitudes in March 1992 [Zander *et al.*, 1994]. Only the ATMOS data above 6 mb are plotted in Figure 13. This figure clearly shows the buildup of HF at all altitudes due to continued use of the CFCs during this period. Luo *et al.*, [1994a] have compared HF column amounts from HALOE with the 1985 ATMOS data and infer an increase rate of $\sim 4\%$ to 8% a year. This value is in good

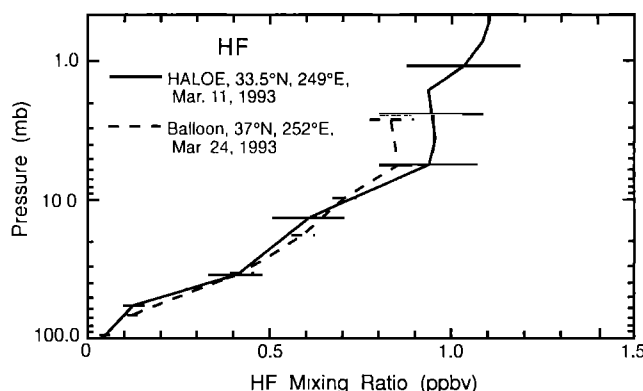


Figure 9. HALOE HF mixing ratio on March 11, 1993, compared with balloon far-infrared emission measurements on March 24, 1993, at 35°N (W.A. Traub *et al.*, private communication, 1994).

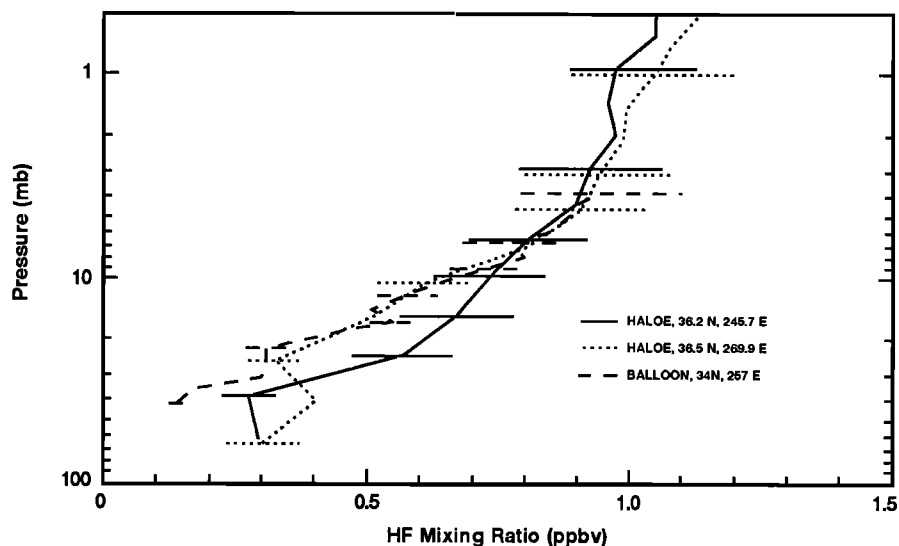


Figure 10. HALOE HF mixing ratio on May 7, 1992, at 36°N compared with balloon far-infrared emission measurements on May 4, 1992, at 34°N (I. G. Nolt, private communication, 1994).

agreement with the mixing ratio increases computed using various altitudes in Figure 13 and agrees reasonably well with HF annual increases indicated in Figure 12 and measurements by numerous other instruments prior to HALOE [see *Rinsland et al.*, 1991, Table 1].

While comparison with model results is not proof of HF validation, it is instructive and a good reasonableness test. *Luo et al.* [1994a] have made comparisons with the National Center for Atmospheric Research (NCAR) two-dimensional model for various times of the year both in terms of pressure versus latitude cross sections and in terms of vertical profiles. The reader is referred to that paper for details. In general, the shapes of the measured and modeled cross sections are similar and the absolute values agree well. Some of the dynamical features in the data, e.g., the spring double minimum near the subtropics, are not seen

in the model because tropical dynamics features are not yet included. Vertical profile comparisons for the 15° N and 45°–55° latitude range of the summer hemispheres (when dynamics effects are minimized) also show good agreement in profile shapes and absolute values when a unity CF_2O photodissociation quantum yield and the early 1990s tropospheric CFC boundary conditions are used. In summary the data versus model comparisons look quite reasonable and differences are well within expectations based on known model deficiencies.

7. HF Global Distribution and Data Limitations

The HF pressure versus latitude cross section for April 12 to May 24, 1993, shown in Plate 3 and the polar orthographic projection on the 20-mbar surface in Plate 4 for the same time period illustrate the main features of the HF global distribution. As discussed by *Russell et al.* [1993a], these are not synoptic plots but are assembled from a 6-week time series of observations made as the HALOE measurements spiral around the globe up and down in latitude. HF is driven by the dynamics, however, and the time scale of the general stratospheric circulation is of the order of several months, i.e., seasonal. Therefore the pictures in Plate 3 and 4 should reasonably represent the synoptically sampled latitudinal distribution. The expected upwelling in the tropics is clear in Plate 3 from the region of low HF values which extend to the upper stratosphere (note that the high HF at the lowest altitudes near the equator is an artifact). Also, the expected double minimum structure in the upper stratosphere observed in tracer measurements made by other experiments [see *Jones and Pyle*, 1984] is present in these data as well. One feature of note is the hemispheric asymmetry between the ~8-mbar and the 40-mbar levels which indicates some apparent residual vertical descent in the northern polar region for the time period we selected. Clear evidence exists for descent in the HF signature in the late winter/early spring data for both hemispheres and in results for the Antarctic vortex region [*Russell et al.*, 1993b]. These features, which either have been observed previously or are expected based on knowledge of the stratospheric general circulation, provide further confidence in the validity of the HALOE HF measurements.

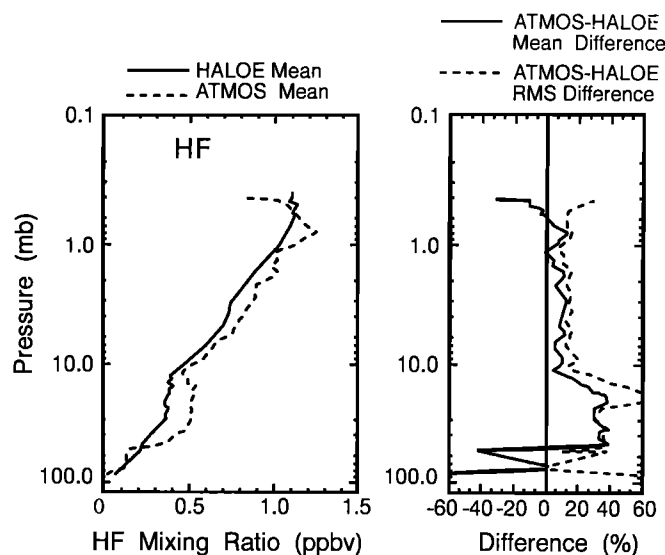


Figure 11. HALOE HF mixing ratio compared with data collected during the shuttle ATMOS/ATLAS 1 mission in late March/early April 1992 (M. R. Gunson et al., private communication, 1994) based on 11 comparisons covering the range from 25°N to 47°S.

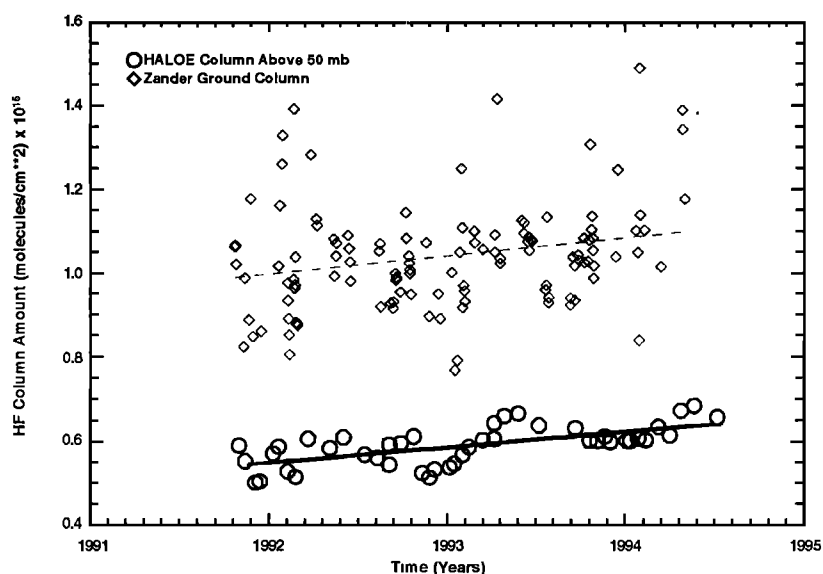


Figure 12. HALOE HF total column amount measurements made within 3° latitude of the Jungfraujoch Station (46.5° N, 8.0° E) compared with ground-based measurements made at that station by the University of Liège group (R. Zander, private communication, University of Liège, 1994).

The main data limitations are at the upper and lower altitudes for the most part. As noted earlier, the single-profile retrieval upper altitude limit is ~ 60 km (~ 0.5 mbar). Cross sections or daily zonal means can extend higher to perhaps 0.1 mbar because averaging provides some noise reduction. Sometimes in the lower stratosphere when the data cuts off due to aerosol effects on the pointer tracker, for example, false and usually increased HF mixing ratio features appear mostly at the lowest retrieved point. Similar problems occur when the cut-off is at or below the tropopause. Users should note this. The only other known problem with the data is the effect of sunspots described earlier in this paper (see Table 2 for dates).

8. Summary and Conclusions

The HALOE HF observations are among the most robust provided by HALOE. This channel is not affected very much by

Doppler shift or spectral interference phenomena, the S/N is excellent, and the orbital performance has been very stable. The data show a high degree of internal consistency when comparing sunrise and sunset profiles and when comparing geophysical features with CH_4 changes. The opposite vertical gradients for the HF and CH_4 profiles cause expected anticorrelations in the pressure versus latitude and longitude cross sections. The precision of the data observed in orbit is about 0.04 ppbv to 0.06 ppbv throughout the stratosphere and the estimated accuracy is $\sim 14\%$ to 27% depending on altitude. The largest errors in the profile occur at the highest altitudes (due to reduced S/N) and at the lowest altitudes (due to pointer/tracker and spectral interference effects). Data users should carefully review the data quality file accompanying the released results. The limitations of the data, most characteristics of the results as well as precision, and accuracy all appear to be well understood from the viewpoint of real atmospheric variations as opposed to instrument-induced changes.

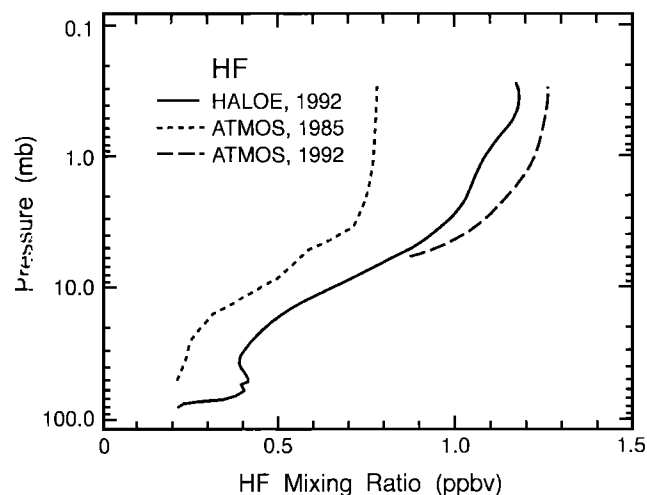


Figure 13. HALOE HF zonal mean mixing ratio profiles at 48° S on May 8-9, 1992, compared with ATMOS Spacelab 3 results at 48° S on May 5, 1985, and ATMOS/ATLAS 1 results at southern midlatitudes in late March 1992 [Zander et al., 1994].

Comparisons with balloon underflight data provide good confidence in HALOE HF measurements. While coincidences in space and time were not good because of occultation viewing geometry and balloon launch timing, results for the composite balloon data set and HALOE agree to within the error bars of the respective measurements. The mean difference is better than 7% through most of the range of balloon measurements. The RMS difference ranges from 12% to 40% through most of the comparison region. Comparisons with ATMOS, ATLAS 1 measurements were not so good, but the time coincidence was poor (mostly 1 to 2 weeks or more). There appears to be a systematic offset between HALOE and ATMOS HF measurements of about 10% at altitudes above the 15-mbar level growing to $\sim 36\%$ in the lower stratosphere. These differences are within the overlap of error bars for the two data sets above the 15-mbar altitude. Efforts are under way to better understand the lower-stratosphere differences, but they are probably related to the poor space and time coincidence and the fact that the comparisons occurred mostly in the southern hemisphere fall when dynamical activity was on the rise.

HALOE HF comparisons with 1985 ATMOS measurements are consistent with the annual increase rate due to CFC usage observed by ground-based experiments, and the data show the accumulated increase at all altitudes. Comparisons with the NCAR two-dimensional model during summer conditions when dynamical activity was minimal showed reasonable agreement in shapes and magnitudes for the vertical profile and pressure versus latitude cross section.

HALOE has provided the first global view of HF for all seasons. The global distribution shows features observed previously for other tracer molecules, e.g., the double minimum structure in the equinox tropics, tropical upwelling, and high-latitude downwelling features. The data further show evidence of strong vertical descent in the polar regions during winter/spring conditions. This feature is internally consistent with similar features in HALOE, CH₄, H₂O, NO, NO₂, and HCl data. All studies done to date lead to the conclusion that the HF observations, are of sufficient quality to be used with confidence in scientific investigations of the middle atmosphere.

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