A-1(2) Experimental Study of Airborne Infrared Remote-Sensing for the Ozone-layer Chemistry (Final Report)

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Abstract

A Fourier-transform infrared spectrometer with resolution of 0.02 cm^{-1} was used for experiments to measure the solar spectra on board aircraft. The spectra, which were obtained through a ZnS or ZnSe infrared window, were analyzed with a new developed least square fitting method to deduce the equivalent width of HCl and HF absorption lines at 2925.9 cm^{-1} and 4038.95 cm^{-1}, respectively, and normalized for the vertical column density. The measured line shape was under constraint of the limited resolution of the instrument, i.e.,0.02cm^{-1}. The latitudinal distribution of the equivalent width showed increasing feature of total HCl and HF toward the higher latitudes. The diurnal change was large at the northern part of Japan. The results of this work showed no indication of the lack of stratospheric HCl over the mid-latitudes due to the heterogeneous chemical processes occurred in the polar stratosphere.

Key Words Aircraft Experiment, Infrared Spectroscopy, Ozone-Layer, Stratospheric HCl, Stratospheric HF

1. Introduction

Stratospheric ozone depletion, firstly detected as the Antarctic Ozone Hole, is now noticed globally over the arctic and even over mid-latitudes. The global ozone observations such as GO_3OS(WMO Global Ozone Observing System), satellite, the arctic campaign and NDSC (the Network for the Detection of Stratospheric Change) are on going, and Japanese researching group should focus their activity especially on the ozone layer over the eastern Asia. In recent years, decreasing of total ozone over the northern part of Japan has been often found in winter to spring. To make clear the state of ozone layer and the dynamical-and chemical-processes over Japan, an infrared remote sensing technique with high resolution Fourier-transform spectrometer (FTIR) has been developed in this work.

2. Research Objective

The chlorofluorocarbons(CFCs), artificially produced and emitted into the atmosphere, have long lived-times in the troposphere and are transported into the stratosphere where CFCs are destructed by strong solar UV radiation to emit free chlorine atoms(Cl). The Cl atoms react with ozone(O_3) and produce ClO. These radicals are converted finally into reservoirs such as HCl or ClONO2, which could reproduce free radicals. The stratospheric hydrogen-fluoride (HF) is produced in a similar process like HCl, however it is hard that HF reproduce fluorine atoms. It is expected that the stratospheric amounts of HCl and HF increase as the result of CFCs' emission into the atmosphere and the ozone depletion possibility with increase of stratospheric HCl also should increase.

To make clear the behavior of these species, HCl and HF, over Japan, development of airborne infrared remote-sensing technique and observational experiments have been made.

3. Research Method

An airborne FTIR (BOMEM model DA5) with a sun-follower has been used for measuring solar infrared radiation at wavenumber region of 500-5,000cm^{-1}. Many atmospheric species have their infrared absorption bands in the mid-IR wavenumber region.

The water vapor and carbon-dioxide have strong absorption bands in the mid-IR region. To reduce these interference, aircraft offers benefit for IR remote sensing at the height above the atmospheric boundary-layer. Handicap of airborne solar experiment is the effect of mechanical vibration due to the rotation of propellers and engines, therefore an anti-vibrational facilities were studied. The change of attitude or orientation of the aircraft also make difficulty to introduce the solar radiation exactly into the interferometer optics and this has resolved with performance of the sun-follower.

The absorption lines centered at 2925.9cm^{-1} for HCl and 4038.95cm^{-1} for HF are used for quantifying their column density in the atmosphere.

To assess the contribution of tropospheric HCl to the solar absorption in the atmosphere, bulk air sampling and an ion-chromatography analysis were adopted to measure amount of HCl.

4. Result

Three campaigns were carried out during three years of 1993-1995. The first one started on December 16, 1993 at Yao-airport to Asahikawa, Hokkaido with an infrared window of ZnS (Figure 1). The FTIR experiments were done successfully on December 17 between Asahikawa to Sendai, on December 18 between Sendai to Kochi, on December 19 between Kochi to Amami and on December 20 between Amami to Tokasiki Is. The second one was done on the similar course as the first one on January 26 to February 1 in 1995. In this campaign, because of the weak intensity of solar radiation through IR window made of ZnSe, the sun-follower could not work sufficiently to introduce the sun light into the optics of FTIR.

The third one was started on February 7,1996 at Yao to Memanbetsu, Hokkaido and focused on the temporal and spatial processes over the northern part of Japan (Figure 2). The FTIR experiments were done successfully with the infrared window of ZnSe on February 10 from Memanbetsu to Sendai, on February 11 from Memanbetsu to Aomori, on February 12 from Memanbetsu to Sendai and on February 13 from Sendai to Yao.

An aircraft, Fair-child Marlin IV of Showa Avi.Co., was chartered as the special purpose of the experiments, and the FTIR system, sun-follower, ozone-meter and air sampling bottles were installed on board the aircraft. The position of the aircraft was detected with GPS navigation system and recorded every minute.

The measured solar spectra were analyzed with a least square method developed for deciding the zero-absorption base line. The shape of HCl and HF absorption lines were assumed to have the Voigt type, which is produced with mixture of the Doppler and Lorentz types and mixture of the temperature and pressure broadening of the absorption line. The examples of the success of the least square fitting for HCl and HF absorption lines in solar spectra were shown in Figure 3 and 4. The widths of Doppler and Lorentz were 0.01-0.02cm^{-1} and 0.001-0.01cm^{-1} for HCl 2925.9cm^{-1} lines, respectively. The estimated values for actual temperature effect show 0.002-0.003cm^{-1}, and the fitted value for Doppler (Gaussian) type is too large. This means the fitted width is effectively the width of the instrumental function, i.e., the resolution of the spectrometer (0.02cm^{-1}) limited the Gaussian type of the components.

The latitudinal distribution of the equivalent widths for the absorption lines of HCl(2925.9cm-1) and HF(4038.95cm-1) , normalized for the vertical column, are shown in Figure 5 and 6, respectively. The markers of cross show the results from the campaign of February 1996. It is clear that the latitudinal dependency of increasing amounts of total HCl and HF toward the higher latitudes were seen for all campaign. For the case of February 1996 campaign, diurnal variation for both HCl and HF were larger over northern part of Japan, especially for HF. The latitudinal dependence is thought as a common feature for the stratospheric species which have a longer life time in the lower stratosphere or higher latitudes. In fact, total ozone measured over Japan shows the same latitudinal distribution and these species should be controlled effectively by the dynamical transport processes.

The results of chemical analysis of the air at the summit of Mt. Fuji shows the tropospheric density of the atmospheric HCl, 0.09ppbv.

5. Discussion

An airborne measurement of stratospheric HCl and HF is possible with a higher resolution FTIR, however for the case of longer wavelength using MCT detector it is required to improve the signal to noise ratio in future.

With this work, it was shown that the spatial or temporal change of both HCl and HF was similar and it is hard to conclude that the stratospheric HCl was affected by the heterogeneous chemical reactions occurred in the cold polar stratosphere. If it was affected in the polar vortex, it should revive or recover during the advection from higher to mid latitudes. The main mechanism, which controls the diurnal or seasonal variation of stratospheric HCl and HF over Japan, is thought the transport process.

Figure 1. Flight course of the FTIR solar experiment in December 1993.

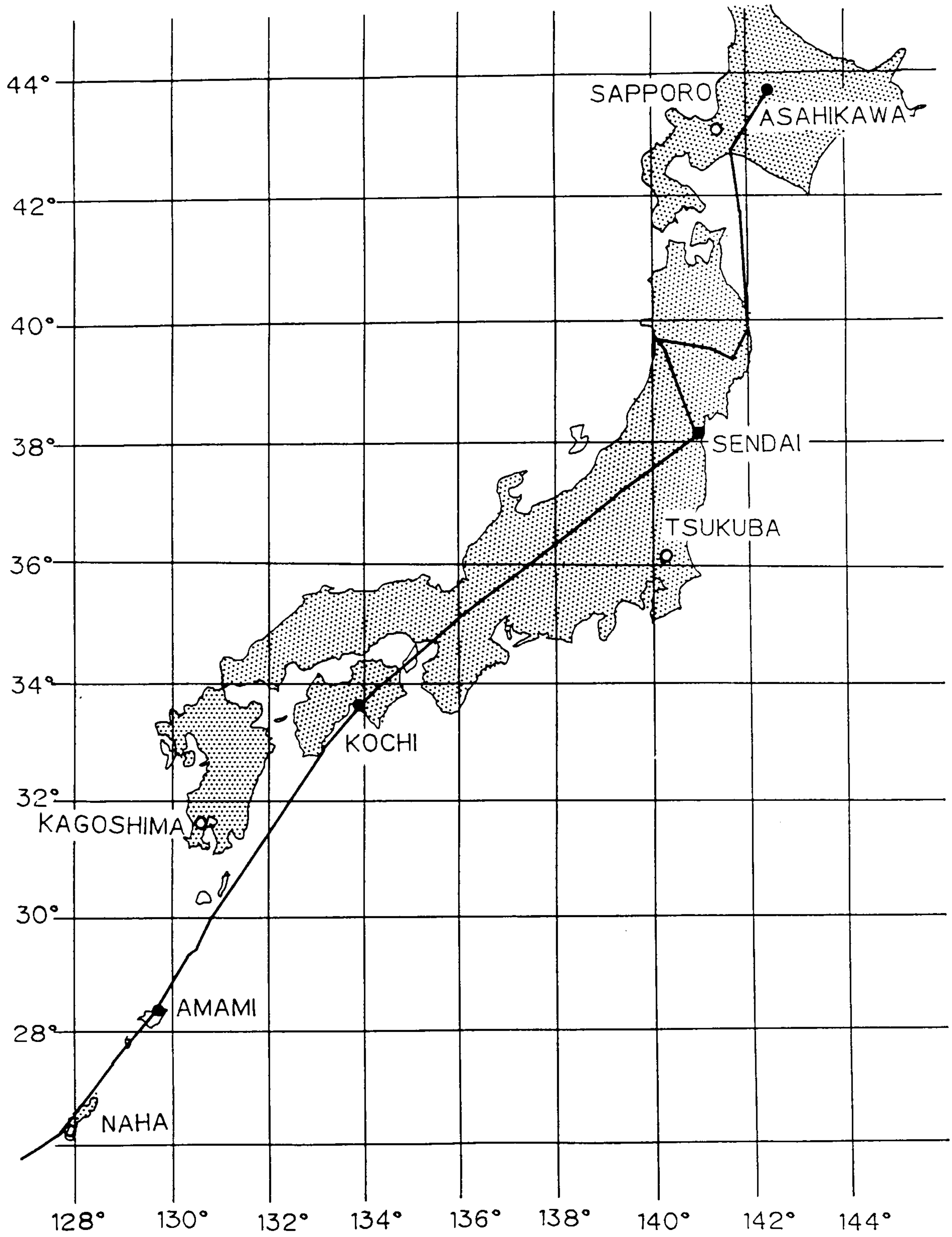

Figure 2. Flight course of the FTIR solar experiment in February 1996.

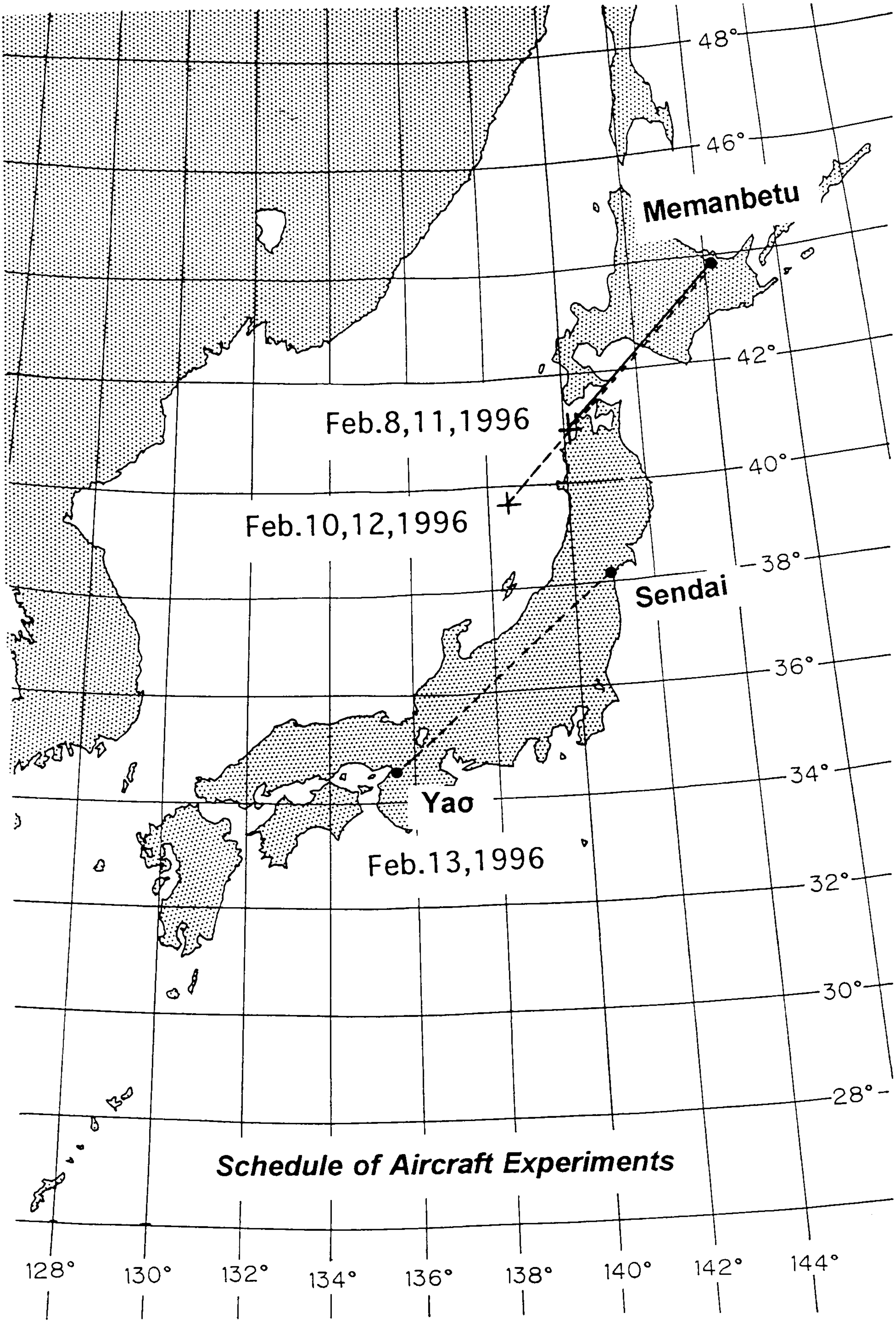

Figure 3. An example of the least square fitting for HCl absorption at 2925.9cm^{-1}.

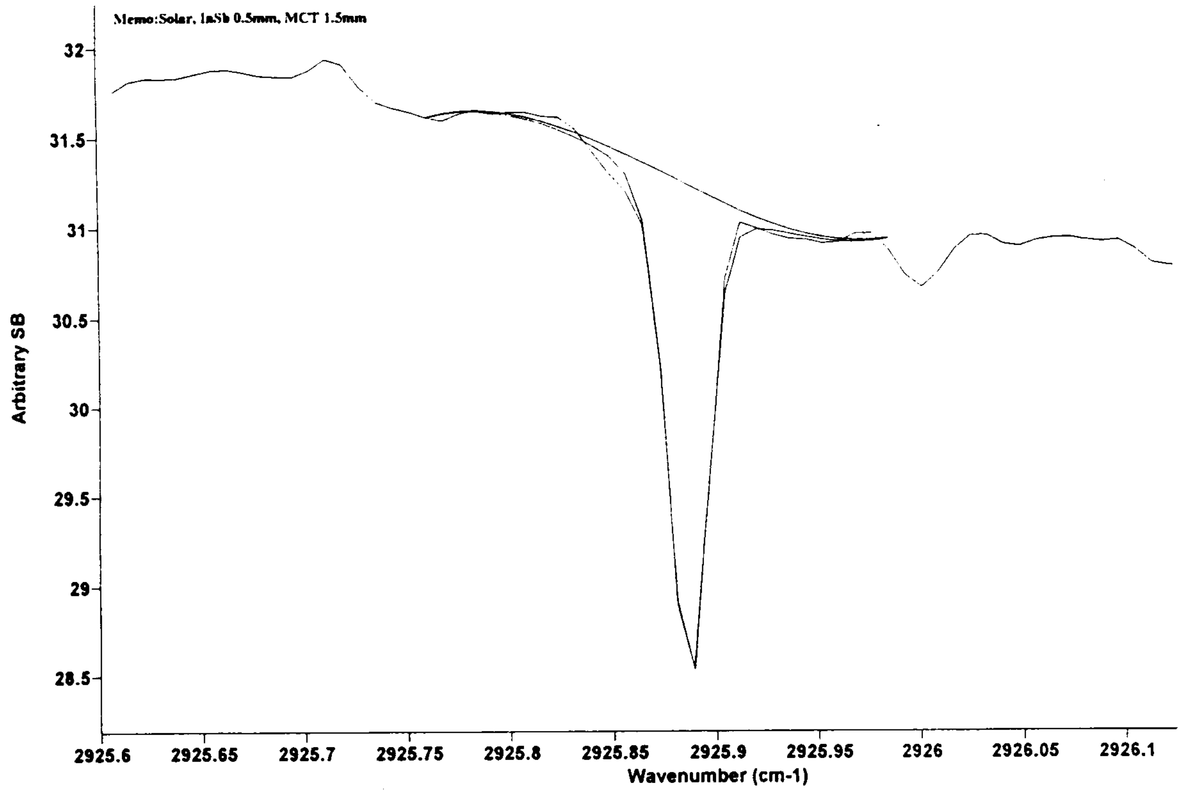

Figure 4. An example of the least square fitting for HF absorption line (4038.95cm^{-1})

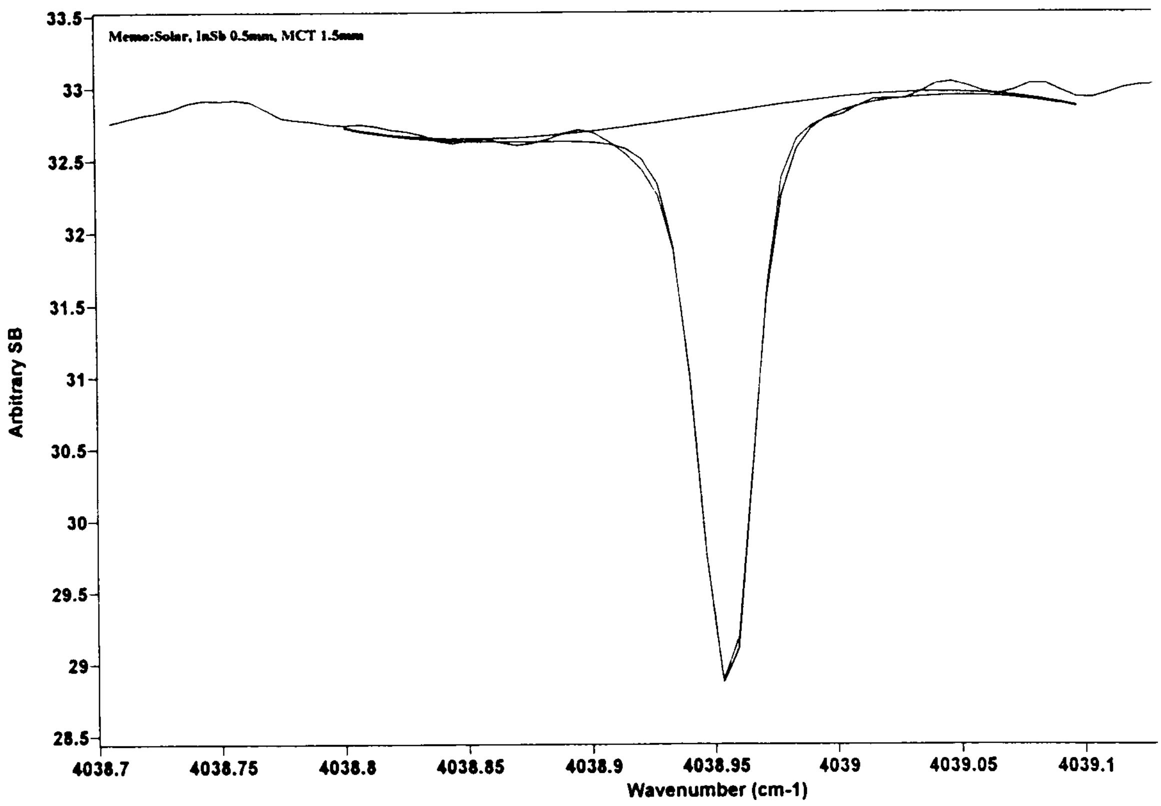

Figure 5. The latitudinal distribution of the stratospheric HCl. The unit is the equivalent width in $0.001 cm^{-1}$ and arbitrary for the total column density.

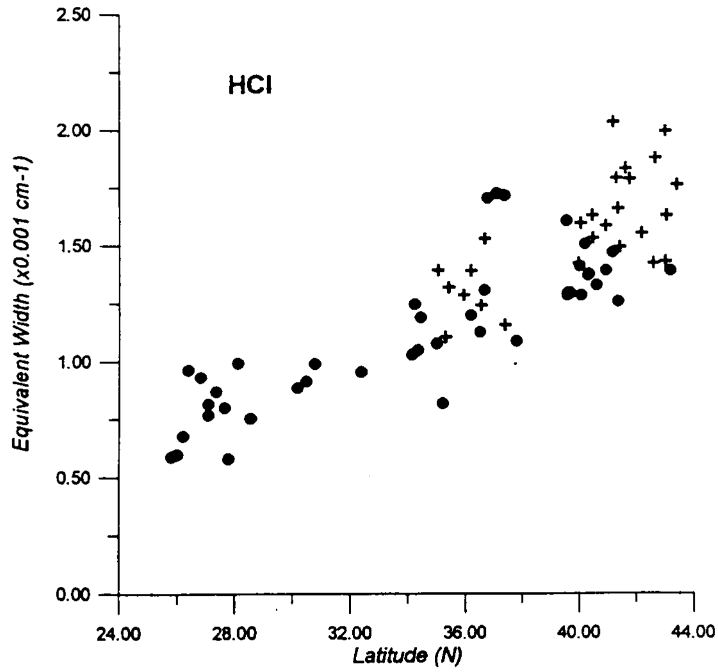

Figure 6. The latitudinal distribution of the stratospheric HF. The unit is same as in Fig.5.