Airborne heterodyne measurements of stratospheric ClO, HCl, O₃, and N₂O during SESAME 1 over northern Europe

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Abstract. Vertical distributions of ClO, HCl, N₂O, and O₃ have been retrieved from airborne observations of pressure-broadened emission spectra in the frequency range of 620 to 690 GHz. Observations were made in February 1994 in the Arctic stratosphere above northern Europe with the Airborne Submillimeter SIS Radiometer (ASUR) during the Second European Stratospheric Arctic and Mid-latitude Experiment (SESAME) 1 campaign. ASUR is the first airborne submillimeter experiment to employ the new superconductor-insulator-superconductor (SIS) receiver technology for stratospheric ozone research. Owing to meteorological conditions, all observations were made outside the polar vortex. The retrieved volume mixing ratio (VMR) profiles show a good agreement with observations made by the submillimeter limb sounder (SLS) operated by the Jet Propulsion Laboratory (JPL) (Pasadena) and the Kern Forschungs Anlage (KFA) (Jülich). A comparison between retrieved VMR profiles and profiles obtained from the SLIMCAT three-dimensional stratospheric chemistry model also shows a good agreement. Two ClO emission lines, at 649 and 686 GHz, respectively, are shown to be equally adequate lines for observation purposes. An anticorrelation has been found between the N₂O and HCl VMR values, and also between the N₂O and ClO VMR values. The correlations between N₂O and HCl do not show the relatively low HCl VMR values correlated to relatively low N₂O values as shown by *Webster et al.* [1994].

Introduction

Reactive chlorine is known to deplete the stratospheric ozone concentration significantly at high-latitudes during the spring period. [*Solomon* 1990; *Anderson et al.*, 1991; *Webster et al.*, 1993]. Earlier observations have shown an anticorrelation between the ClO concentration (a reactive form of chlorine) and the O₃ concentration [*Anderson et al.*, 1989; *de Zafra et al.*, 1989]. Also, at midlatitudes the chlorine chemistry affects the O₃ concentration both at high altitudes and, by heterogeneous chemistry, at low altitudes [*Fahey et*

al., 1993]. However, details of O₃ depletion by chlorine species are still not fully quantitatively understood by lack of data. For example, some observations show an anomalous low chlorine budget at midlatitudes [*Webster et al.*, 1994].

To acquire more knowledge about the relevant processes of O₃ depletion, the Submillimeter Atmospheric Sounder (SUMAS) was employed during the European Arctic Stratospheric Ozone Experiment (EASOE) winter campaign 1991/1992 during which observations of ClO and HCl in the stratosphere were obtained [*Wehr et al.*, 1995]. The SUMAS instrument is a passive submillimeter heterodyne radiometer developed by the University of Bremen. It senses remotely the thermal emission of the rotational vibrations of molecules in the 620-650 GHz frequency band where detectable lines of ClO, O₃, HCl, and N₂O reside.

Subsequently, in 1994, the SUMAS instrument was significantly improved for the SESAME 1 campaign. For the first time the new superconductor-insulator-superconductor (SIS) technology was employed in an

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aircraft for stratospheric ozone research. The SIS junction, produced by the Space Research Organisation Netherlands (SRON), replaces the formerly used Schottky diodes. The new sensor, named Airborne Submillimeter SIS Radiometer (ASUR), accommodates a liquid helium-cooled Nb tunnel-junction detector.

The SIS junction was among the best in existence with respect to sensitivity and low single sideband (ssb) noise temperature, at the time of the campaign. The low ssb noise temperatures allow a better temporal and spatial resolution through shorter integration times and, consequently, shorter flight distances. It enables sequential detection of all molecular lines of HCl, ClO, N_2O, and O_3 during one single flight with a horizontal resolution better than 100 km.

In this paper we present data taken during the 1994 campaign as observed in the Arctic region and above northern Europe. The data were taken outside the polar vortex under conditions of undisturbed chemistry. This study is part of a long-term measurement series which was started by the EASOE campaign in 1992 [*Crewell et al.*, 1994].

Measurement Technique

Implementation of the SIS junction has lowered the mean system noise temperature (ssb) to values of less than 2500 K in the ASUR system [*Mees et al.*, 1994]. (Recently, during the 1995 campaign the ASUR ssb noise temperature was further lowered to a value of of approximately 800 K.) The SIS junction mixer is pumped by a solid state local oscillator source. The local oscillator consists of an InP Gunn oscillator with subsequent cascaded Schottky diode multipliers, increasing the generated frequency by a factor of 6. Frequency stability of $\Delta\nu/\nu < 5 \cdot 10^{-8}$ is attained by means of a phase-lock loop. A quasi-optical coupling scheme combines the observed signal and the local oscillator (lo) signal (see Figure 1).

Signals in both sidebands $\nu_{usb} = \nu_{lo} + \nu_{if}$ (upper sideband) and $\nu_{lsb} = \nu_{lo} - \nu_{if}$ (lower sideband) can be detected, where ν_{lo} is the tunable local oscillator frequency and ν_{if} is the first intermediate frequency of the receiver. A single sideband filter allows tuning of the receiver to either the upper, lower, or both sidebands.

The signals are spectrally analyzed by the back-end spectrometer filterbank. The filterbank consists of 28 channels with varying bandwidths (ten 8-MHz channels centered, and at each side of the line center are four 40-MHz channels at the inner wings and five 80-MHz channels at the outer wings). This configuration is a trade-off between high resolution in the center and a large overall bandwidth (1.2 GHz).

The instrument operates on board a high-flying aircraft (typical flight altitude approximately 10-12 km) which reduces signal absorption by tropospheric water vapor. The measurement geometry is defined by a zenith angle of 76°-78° with an antenna beam width of approximately 1.3° (full width at half maximum). The instrument observes the atmosphere through a polyethylene window in the fuselage in a direction perpendicular to the flight direction. To avoid Fabry-
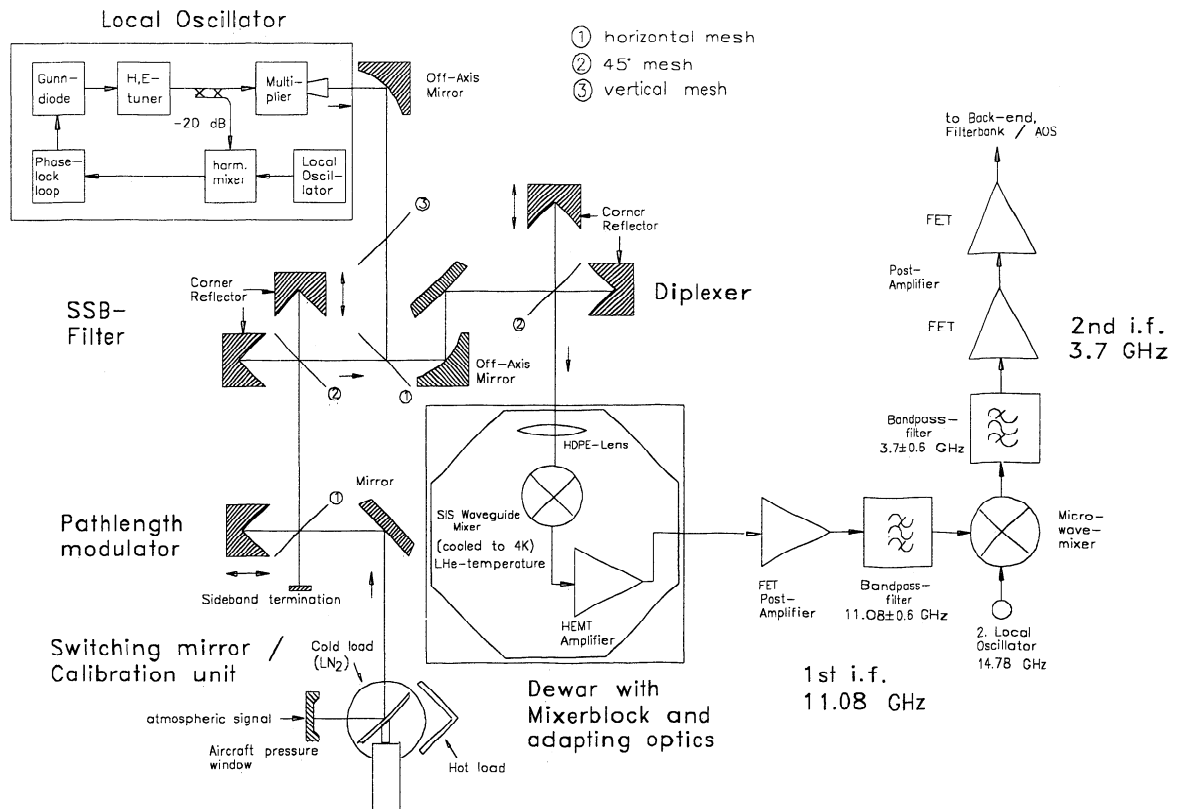

Figure 1. Quasioptics and IF-chain of the ASUR receiver.

Table 1. List of Flights Performed During the SESAME 1 Campaign in February 1994

Flight	Direction	Date	Time/UTC	Molecules
1	Munich–Stockholm	Feb. 9, 1994	0817/1044	ClO, N_2O, O_3
2	Stockholm–Kiruna	Feb. 9, 1994	1137/1415	ClO, HCl
3	Local flight Kiruna	Feb. 13, 1994	1210/1510	ClO, HCl
4	Kiruna–Hamburg	Feb. 16, 1994	1010/1315	ClO, HCl, N_2O, O_3
5	local flight Hamburg	Feb. 18, 1994	0957/1330	ClO, O_3 (686/664 GHz)

Perót interference by the window, the window is wedge-shaped, with an angle of $0.47°$. This arrangement successfully suppresses the baseline modulation observed during the EASOE campaign [*Wehr et al.*, 1995].

Calibration is accomplished by comparing the atmospheric signal alternately with the signals of two blackbodies at different temperatures (ambient "hot" temperature and liquid nitrogen "cold" temperature) inside the aircraft. A calibration cycle consists of four observations obtained in about 12 s. The hot and cold blackbodies are alternately observed. After each blackbody observation an atmospheric signal is obtained. This method provides high accuracy by removing gain and offset variations with timescales longer than 24 s disabling these variations to contribute to the measurement error. Additional technical information may be obtained from *Wehr et al.* [1995] and *Crewell et al.* [1994].

Measurement Campaign 1994

The campaign covered the period from February 7 to 18, 1994. The research aircraft FALCON operated by the German Air and Space Research Establishment (DLR) Oberpfaffenhofen performed several flights over Germany and Scandinavia. A typical flight took 2.5 hours in which a distance of approximately 2000 km was covered.
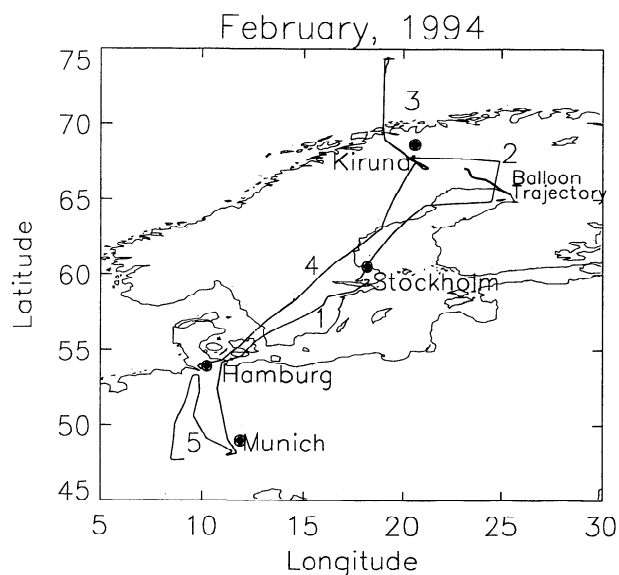

Figure 2. Flight tracks as flown during the 1994 campaign. The dashed line denotes the flight track of the Submillimeter Limb Sounder balloon.

Table 1 summarizes the flights. The actual flight tracks are shown in Figure 2. Initially, the ASUR instrument made observations in the 625-650 GHz frequency range. On February 18, a new local oscillator at 675 GHz was integrated into the receiver which enabled measurements of O_3 and ClO at 664 and 686 GHz, respectively.

Data Analysis

The signal emitted by the species under study needs to be extracted from the measured signals. For this purpose the measured spectrum is calibrated and corrected for the window transmission [*Crewell*, 1993]. Individual spectra are averaged in a straightforward manner to obtain a sufficiently high signal-to-noise ratio. Spectra containing spikes or large baseline offsets are omitted in the averaging procedure.

Further correction of the measured signal is required for the emission by other molecules, mainly ozone and water vapor. In accordance with the procedure applied by *Wehr et al.* [1995], the O_3 correction for ClO spectra is based on calculated emissions from O_3 profiles obtained from balloonborne observations. The O_3 absorption of the ClO signal is assumed to be negligible. For HCl the O_3 correction is derived from direct O_3 measurements by ASUR. The O_3 absorption of the HCl signal is calculated and corrected for. In the case of N_2O, the spectra had to be corrected for the emission of the oxygen isotope ($O^{18}O^{16}$). The continuum emission in the frequency range of 620 to 690 GHz is mainly due to water vapor. It yields a roughly constant offset in the observed spectra with a variability of about 10 K. The water vapor amount is unknown since it is not observed simultaneously with the spectra. Therefore the offset is treated as a free parameter in the retrieval model, neglecting absorption effects. In Figure 3, a typical example of a measured spectrum of ClO is shown.

In the frequency range considered, the observed spectral line shape can be related to the vertical volume mixing ratio (VMR) distribution of the molecule of interest by calculating radiative transfer in a nonscattering and nonrefracting atmosphere. The applied retrieval method to obtain VMR profiles from the pressure broadened emission spectra is based on the optimal estimation method by *Rodgers* [1976]. This method uses a priori data. Tests showed that stable solutions were found after two iterations. The a priori profiles for ClO and HCl were the same as used by *Crewell et al.* [1994]. The a priori profile for N_2O at midlatitudes is obtained
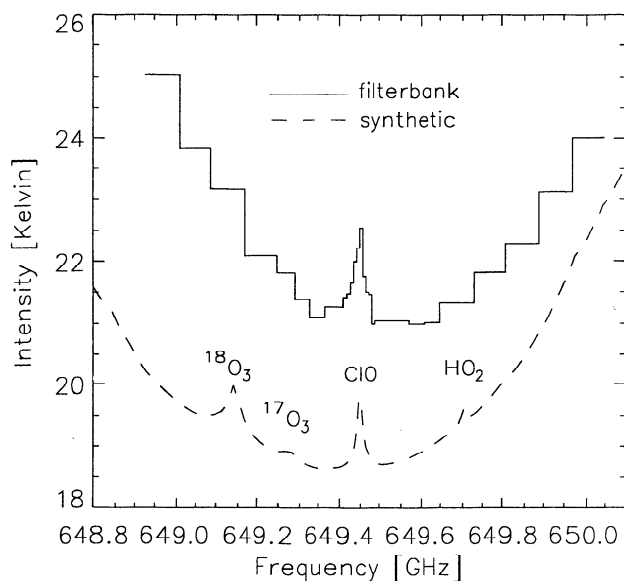
Figure 3. An emission spectrum of ClO (upper line) observed at 649 GHz on February 16, 1994, flight track 1. Averaged position 57°N, 12°E. The width of the steps in the upper curve indicates the width of the filter bank channels. The lower curve is a theoretical calculation of the expected atmospheric spectrum over this frequency range. The lower curve is given an offset to facilitate the intercomparison. The curvature in the baseline is caused by strong emission lines of O_3 at each side of the ClO line.

from *Gidel et al.* [1983]. The a priori profile for O_3 is a combination of a balloonborne observation on February 9, 1994, extended with a standard profile given in Figure 7b. The error on the spectrum as used in the optimal estimation method is determined by instrumental noise and by the estimated error of the correction for contaminating emissions.

The accuracy of a retrieved VMR profile is determined by line strength, signal-to-noise ratio, and spectral resolution. A detailed error analysis of the applied technique is given by *Wehr et al.* [1995]. Within the retrieval procedure, a vertical resolution of 8-10 km is preferred to a significantly smaller resolution (e.g., 1-2 km) because it minimizes possible artifacts caused by error amplification of the retrieval technique [*Rodgers* 1990; *Wehr et al.* 1995]. A constant VMR is assumed inside each layer. A horizontal resolution better than 100 km is achieved for the ASUR system. This horizontal resolution depends on aircraft speed and the integration time required to obtain a spectrum with a sufficiently high signal-to-noise ratio.

Results of VMR Retrieval

The retrieved VMR profiles of ClO and HCl all show characteristics typical of undisturbed chemistry. ClO and HCl profiles both show an increase with altitude reaching maximum values of 0.5 ppb and 3.0 ppb, respectively, in the upper layer; see Figure 4. Analysis of the potential vorticity pattern during the campaign in

the Arctic confirmed that no flight took place inside the polar vortex. These results are in contrast with earlier measurements with the SUMAS instrument inside the Arctic vortex (February 1992 and February 1993) where ClO VMR values up to 1.0 ppb at 20 km altitude were observed over northern Scandinavia [*Wehr et al.*, 1995, *Crewell et al.*, 1995].

The flight from Stockholm to Kiruna on February 9 was coordinated with the balloonborne submillimeter limb sounder (SLS) experiment [*Stachnik et al.*,
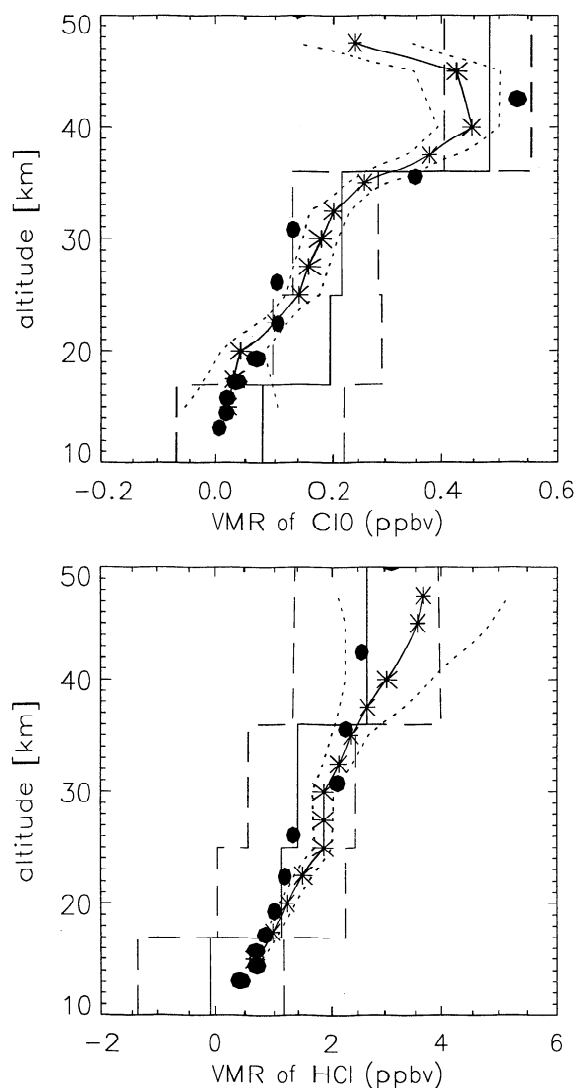

Figure 4. Retrieved VMR profiles of ClO and HCl observed by the ASUR instrument (boxes, solid line) in comparison with the Submillimeter Limb Sounder observations (solid line with asterisks) on February 9, 1994, flight track 2 in Figure 2. The 1σ uncertainty limits for ASUR on February 9, 1994, are designated by a dashed line and those for SLS by a dotted line. The ASUR 1σ uncertainty limits are determined by the sum of the diagonal elements of the covariance matrix as described by *Wehr et al.*, [1995]. ASUR ClO: averaged position 63°N, 20°E. ASUR HCl: averaged position 67°N, 21°E. SLS: averaged position 67.5°N, 22°E. The three-dimensional chemical transport model results are denoted by the solid dot.
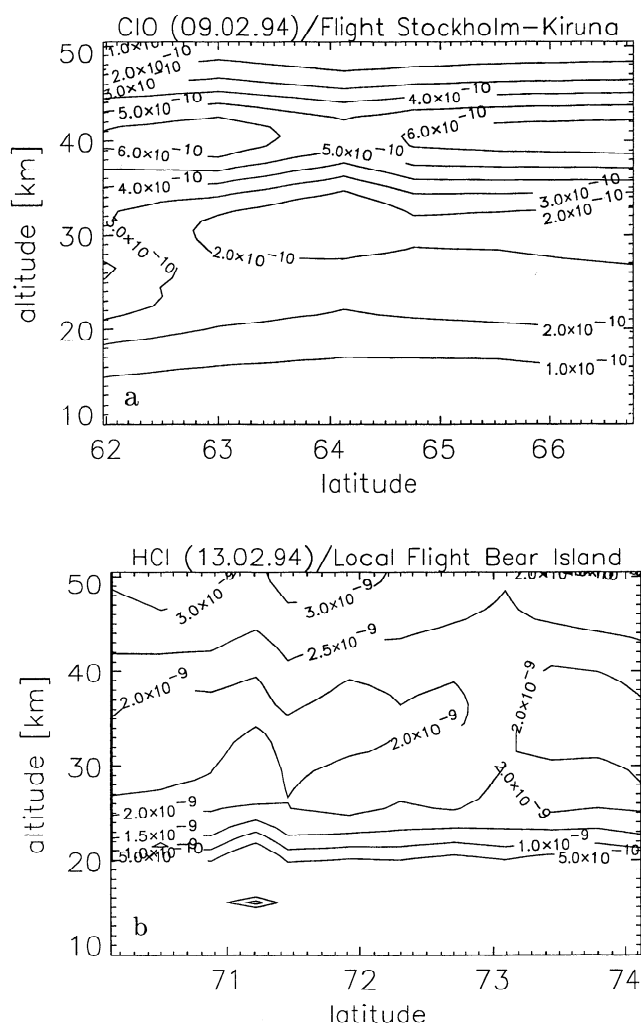
CIO (09.02.94)/Flight Stockholm—Kiruna

HCl (13.02.94)/Local Flight Bear Island

Figure 5. Contour plots of VMR values as a function of latitude and altitude (a) for ClO on February 9, 1994, during the flight Stockholm-Kiruna (flight track 2) and (b) for HCl on February 13, 1994, during the round flight to Bear Island (flight track 3).

VMR of N20 (ppbv)

Figure 6. Retrieved VMR profiles of N_2O as observed by ASUR on February 9, 1994, (solid line with triangle) and on February 16, 1994, (solid line with crosses). Flight tracks 1 and 4 in Figure 2. For comparison, an SLS measurement taken near Kiruna on February 9, 1994, is shown by solid line with asterisks. The data are also compared to the observations made at Thule outside the polar vortex during the EASOE campaign by *Emmons et al.*, [1994] (solid line). The 1σ uncertainty limits for ASUR on February 9, 1994, are designated by dashed line and those for SLS by dotted line. The 1σ uncertainty limits for ASUR on February 16, 1994, are similar to the limits of February 9, 1994. To improve the clarity of the graph the uncertainty limits of February 16, 1994, are not shown. N_2O on February 9, 1994: averaged position 56 °N, 13°E, N_2O on February 16, 1994: averaged position 56 °N, 11°E, SLS on February 9, 1994: averaged position 67.5°N, 22°E. The three-dimensional chemical transport model results are denoted by the open diamond (February 9, 1994) and the solid dot (February 16, 1994).

1992]. The SLS operated by Jet Propulsion Laboratory (JPL), Pasadena, California, on a balloon of the Kern Forschungs Anlage (KFA), Jülich, Germany, was launched from Esrange near Kiruna. The SLS observed HCl and ClO using the same spectral region as ASUR. On February 9, SLS data were also obtained outside the polar vortex. In Figure 4, the profiles obtained with the ASUR instrument are compared to the SLS profiles. There is good agreement between the ASUR profiles and the SLS profiles.

The high sensitivity of the ASUR instrument allows short integration periods which enables the measurement of a large number of profiles during one flight. In Figure 5, contour plots of ClO and HCl versus altitude and latitude are shown. These contour plots were produced using an optimal estimation method with a vertical resolution of 1 km. As described above such a representation can cause artifacts. Therefore the reader should keep in mind that these results should be smoothed vertically over layers of about 8 km. There is
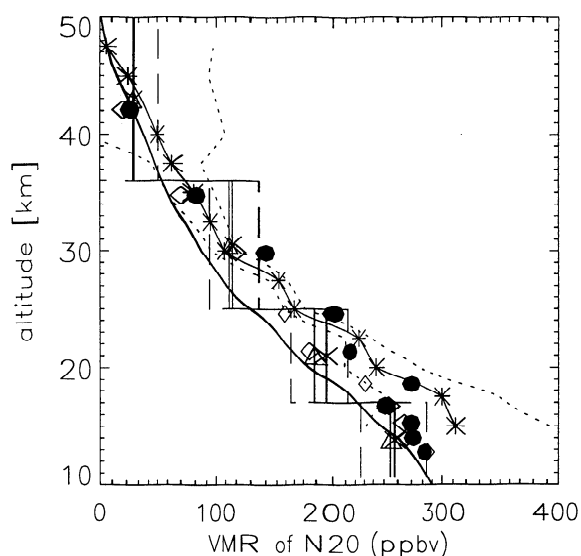
no significant indication of perturbed chemistry due to heterogeneous chemistry on aerosol particles.

N_2O has been measured at two different dates during the 1994 campaign of February 9 and 16. These two measurements were performed at nearly the same latitude. Figure 6 shows that the N_2O VMR remains approximately the same over this period of time. The N_2O results were also compared to the SLS data. There is again good agreement between the SLS data and the ASUR data despite the fact that there are substantial differences in place and time of the observations. The N_2O VMR profiles in Figure 6 are also in agreement with N_2O profiles observed outside the polar vortex by *Emmons et al.* [1994].

O_3 and ClO were also measured during one flight over Germany using a different local oscillator frequency at 675 GHz, allowing to measure a ClO resonance line at 686 GHz. Derived ClO and O_3 profiles are shown in Figure 7. We have again compared ASUR profiles with the SLS data described above, and find good agreement
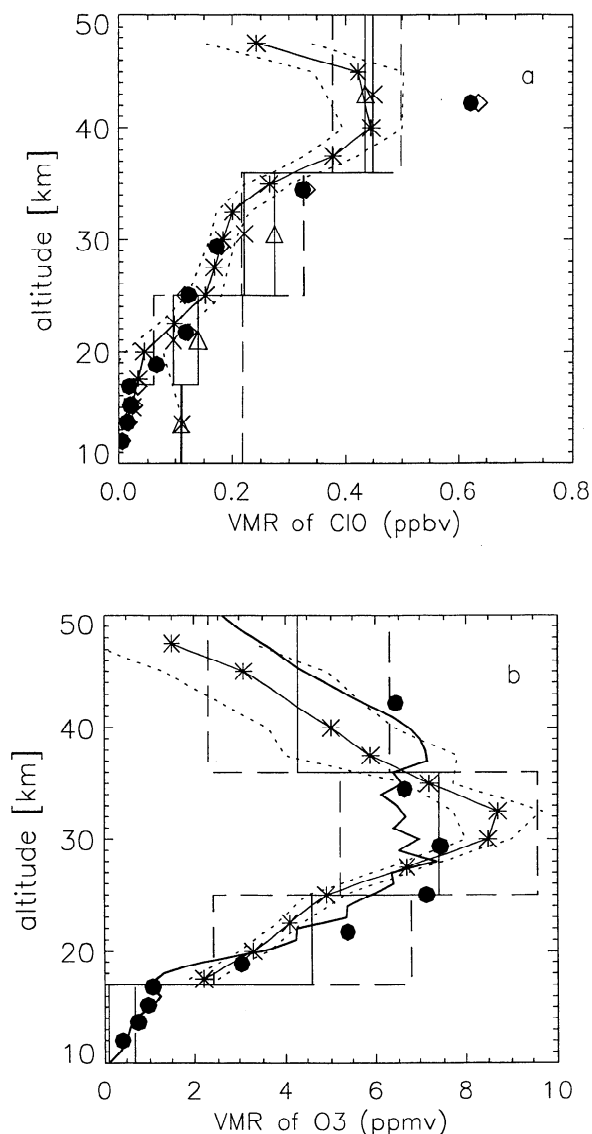
despite differences in time and location. This is probably due to the fact that both ASUR and SLS measurements were taken outside the polar vortex. There is an apparent difference between the ClO values observed by ASUR at the latitudes of 48° and 52° (see Figure 7), however this difference is within the 1 σ uncertainty limits. Any intrusion of air processed by polar stratospheric clouds at the higher latitude of 52° could not be detected. The obtained O_3 VMR values served a technical purpose in the correction of the spectra of HCl as described in the data analysis.

The Three-Dimensional Model

We have compared our measurements with results from the SLIMCAT three-dimensional offline chemical transport model (M. P. Chipperfield et al., Analysis of UARS data in the southern polar vortex in September 1992 using a chemical transport model, submitted to the *Journal of Geophysical Research*, 1996). The model uses meteorological analyses to specify the wind and temperature fields and is coupled to the TOM-CAT stratospheric chemistry model which integrates the standard O_x, NO_y, ClO_y, BrO_y, and HO_x species along with certain long-lived tracers (e.g., N_2O). The model has 31 chemical tracers and around 100 chemical reactions. The model also has a treatment of heterogeneous reactions on polar stratospheric clouds and sulfate aerosols [e.g., see *Chipperfield et al.*, 1995].

For the results presented here the model was integrated from November 26, 1993, until February 24, 1994, using United Kingdom Meteorological Office analyses [*Swinbank and O'Neill*, 1994]. The model has 11 isentropic levels between 350 K and 2100 K with a horizontal resolution of 2.5 degrees in latitude (in the northern hemisphere) and 5.6 degrees in longitude. Chemical initialization of a three-dimensional model is critical and we used a mixture of satellite observations and results from a two-dimensional latitude-height model. The initial O_3 field was taken from Microwave Limb Sounder Observations [*Froidevaux et al.*, 1995], and N_2O data for November 1992 from the Cryogenic Limb Array Etalon Spectrometer [*Roche et al.*, 1995] was used to constrain the initial long-lived tracer distributions.

The three-dimensional model results are included in Figure 4, 6, and 7 for the same location and time as the respective ASUR observations. This preliminary comparison shows generally good agreement between ASUR observations and the model calculations. The model ClO profile in Figure 4 confirms the absence of elevated ClO in the lower stratosphere. While the 40 km ClO maximum from the model agrees well with the data on February 9, on February 18 at lower latitudes (Figure 7) the model overestimates the observations. This discrepancy will be the subject of future research and will require a quantitative analysis of other model species (e.g., CH_4) which control the ClO:HCl partitioning in the upper stratosphere. The N_2O and O_3 profiles from the model agree well with the ASUR observations, although the model O_3 profile also tends to overestimate the observations around 40 km.
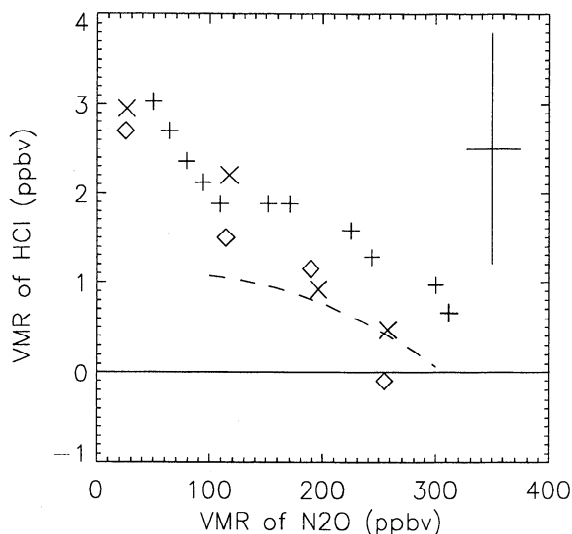
Figure 7. Retrieved VMR profile of ClO and O_3 as observed on February 18, 1994, with a local oscillator working at 675 GHz. Flight track 5 in Figure 2. (a) ClO of the first part of the flight at an averaged latitude of 48° shown by solid line with crosses, and ClO of the last part of the flight at an averaged latitude of 52° shown by solid line with triangles. The averaged longitude of both parts is 10°E. Only the 1σ uncertainty limits for the last part of the flight are shown to improve the clarity of the plot. The 1σ uncertainty limits for the first part were similar to the values of the last part. The SLS data are represented in a similar way as in Figure 4. The three-dimensional chemical transport model results are denoted by the open diamond (48°) and the solid dot (52°). (b) O_3 as retrieved at an averaged position of 50°N, 10°E compared to SLS on February 9, 1994: averaged position 67.5°N, 22°E. The 1σ uncertainty limits for ASUR are designated by the dashed line. The SLS data are represented in a similar way as in Figure 4. The three-dimensional chemical transport model results are denoted by the solid dot. The a priori line (solid line) is based on ozone sonde observations on February 9, 1994, obtained at the Lindenberg station (52.13°N, 14.07°E). The observations are extrapolated with a standard profile to 50 km altitude.

Figure 8. Correlation between N_2O VMR values and HCl VMR values obtained at the same altitude. Data from flight on February 9 shown by open diamond. N_2O: averaged location 56 °N, 13°E, HCl: averaged location 59°N, 18°E. Data from flight on February 16 shown by crosses. N_2O: 56 °N, 11°E, HCl: 60°N, 16°E. The pluses denote data obtained with the SLS on February 9. The averaged $\pm1\sigma$ uncertainties are denoted by the large plus sign in the upper-right corner of the figure. The dashed line corresponds to the fit proposed by *Webster et al.* [1994] which approximates his observations.
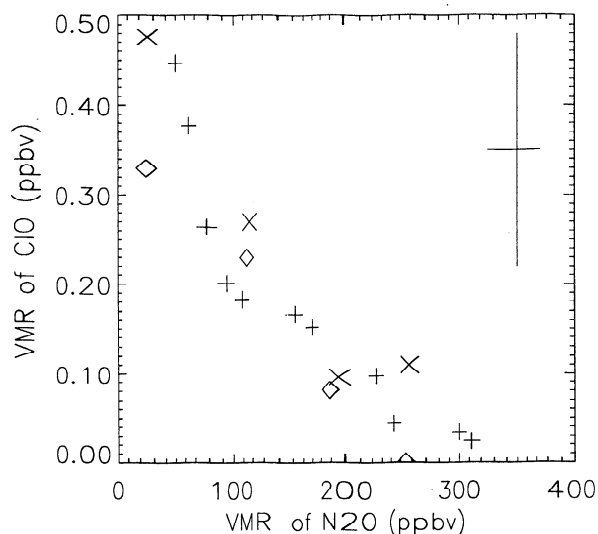
Discussion

In the stratosphere an anticorrelation has been found by ASUR between the N_2O and the HCl VMR. This anticorrelation is expected from photolysis of N_2O and CFCs in the middle stratosphere, where the Cl released from the CFCs enables the formation of HCl. Figure 8 shows N_2O VMR values at various altitudes correlated with HCl VMR values obtained at the same altitudes. The SLS data are also shown in Figure 8. The slope of the SLS HCl/N_2O ratio is different compared with the ASUR data. This is consistent with Figure 4 and 6 where the SLS data show higher values of HCl and N_2O compared with the ASUR data.

Previous in situ observations at 20 km altitude with the ER 2 have shown an anticorrelation between the N_2O and the HCl VMR outside the polar vortex, [*Webster et al.*, 1994]. Compared to the results of *Webster et al.* [1994] the ASUR results display a steeper slope, which implies a higher HCl VMR. The problem raised by *Webster et al.* [1994] of too low HCl VMR related to low values of the N_2O VMR, does not show up in our measurements. The ASUR results are in good agreement with the observations made by various other instruments on other platforms such as the space shuttle, balloons, and satellites also shown by *Webster et al.* [1994].

The photolysis process described, also yields an anticorrelation between N_2O and ClO up to 40 km altitude in air which is not affected by heterogeneous chemistry. In Figure 9 the N_2O VMR values are correlated to the

Figure 9. Correlation between N_2O VMR values and ClO VMR values obtained at the same altitude (caption as Figure 8). February 9, 1994, ClO: averaged location 54°N, 11°E. February 16, 1994, ClO: averaged location 57°N, 12°E.

corresponding ClO VMR values. The ClO VMR values obtained on February 16 are higher compared to the values obtained on February 9. Observations by the MLS on UARS showed that the polar vortex was moving toward Scandinavia in the period February 10 to 26 [*Waters et al.* 1995]. This may explain the higher ClO values on February 16.

Conclusions

From February 9 to 16, 1994, observations of chlorine monoxide, hydrochloric acid, nitrous oxide, and ozone were made with the ASUR system in the lower and middle stratosphere in Arctic and Subarctic regions outside the polar vortex. The ASUR measurements are in good agreement with the measurements of a balloon-borne limb sounder (SLS) and with three-dimensional chemical transport model calculations. Both ASUR and SLS show a steady increase with altitude of the VMR of HCl and ClO outside the polar vortex below 40 km.

Results obtained outside the polar vortex show an anticorrelation between the VMR of N_2O and HCl. The ratio N_2O / HCl observed here differs from the one observed by *Webster et al.* [1994]. The ratio is in agreement, however, with other observations also given by *Webster et al.* [1994]. Furthermore, the anticorrelation between N_2O and ClO is presented.

On February 18 a local oscillator working at a higher frequency was implemented in the ASUR instrument. During a local flight over Germany, comparative observations of ClO and O_3 were made at frequencies of 664 GHz and 686 GHz, respectively. The VMR profiles obtained for ClO at 686 GHz are comparable to results at 649 GHz, demonstrating the suitability of the ClO line at 686 GHz for atmospheric sounding.

Future analysis of the SESAME 1 data involves a more detailed comparison with model results. The cor-

relation between chlorine species and the N_2O enables comparison between measurements in different time periods. The differences found in these chlorine relations inside and outside the vortex as studied in the SESAME 3 campaign in 1995, and the detailed study of the SESAME 1 results will be the subject of a separate paper.

References

Anderson, J. G., W. H. Brune, and M.H. Proffitt, Ozone destruction by chlorine radicals within the antarctic vortex: The spatial and temporal evolution of ClO-O_3 anticorrelation based on in situ ER 2 data, *J. Geophys. Res., 94*, (D11), 11,465-11,479, 1989.

Anderson, J. G., D. W. Toohey, and W. H. Brune, Free radicals within the antarctic vortex: The role of CFCs in antarctic ozone loss, *Science, 251*, 39-45, 1991.

Chipperfield, M.P., J.A. Pyle, C.E. Blom, N. Glatthor, M. Höpfner, T. Gulde, Ch. Piesch, and P. Simon, The variability of ClONO$_2$ and HNO$_3$ in the arctic polar vortex: Comparison of Transall Michelson interferometer for passive atmospheric sounding measurements and three-dimensional model results, *J. Geophys. Res., 100*, 9115-9129, 1995.

Crewell, S., Submillimeter-Radiometrie mit einem flugzeuggetragenen Empfänger zur Messung atmosphärischer Spurenstoffe, Verlag Shaker, Aachen, Germany, 1993.

Crewell, S., K. Künzi, H. Nett, T. Wehr, and P. Hartogh, Aircraft measurements of ClO and HCl during EASOE 1991/92, *Geophys. Res. Lett., 21*, (13), 1267-1270, 1994.

Crewell, S., R. Fabian, K. Künzi, H. Nett, T. Wehr, W. Read, and J. Waters, Comparison of ClO measurements by airborne an spaceborne microwave radiometers in the Arctic Winter Stratosphere 1993, *Geophys. Res. Lett., 22*, (13), 1489-1492, 1995.

de Zafra, R.L, M. Jaramillo, J. Barrett, L.K. Emmons, P.M. Solomon, and A. Parrish, New observations of a large concentration of ClO in the springtime lower stratophere over Antarctica and its implications for ozone-depleting chemistry, *J. Geophys. Res., 94*, (D9), 11,423-11,428, 1989.

Emmons, L.K., J.M. Reeves, D.T. Shindell, and R.L. de Zafra, N_2O as an indicator of Arctic vortex dynamics: Correlations with O_3 over Thule, Greenland, in February and March, 1992, *Geophys. Res. Lett., 21*, (13), 1275-1278, 1994.

Fahey, D.W., et al., In situ measurements constraining the role of sulfate in midlatitude ozone depletion, *Nature, 363*, 508-514, 1993.

Froidevaux, L., et al., Validation of UARS MLS O_3 measurements, *J. Geophys. Res.*, in press, 1995.

Gidel, L.T., P.J. Crutzen, and J. Fischmann, A two dimensional photochemical model of the atmosphere, 1, Chlorocarbon emissions and their effect on stratospheric ozone, *J. Geophys. Res., 88*, 6622-6640, 1983.

Mees, J., S. Crewell, H. Nett, G. de Lange, H. van de Stadt, J.J. Kuipers, and R.A. Panhuyzen, An airborne SIS-receiver for atmospheric measurements of trace gases at 625 to 760 GHz, *Proceedings Fifth International Symposium on Space TeraHertz Technology*, pp. 142-156, Ann Arbor, and *IEEE Trans. Microwave Theory Tech., 43*, 2543-2548, 1995.

Roche, A.E., et al., Validation of CH_4 and N_2O measurements by the CLAES instrument on UARS, *J. Geophys. Res.*, in press, 1995.

Rodgers, C. D., Retrieval of atmospheric temperatures and composition from remote measurements of thermal radiation, *Rev. Geophys., 14*, (4), 609-624, 1976.

Rodgers, C. D., Characterization and error analysis of profiles retrieved from remote sounding measurements, *J. Geophys. Res., 95*, (D5), 5587-5595, 1990.

Solomon, S., Progress towards a quantitative understanding of Antarctic ozone depletion, *Nature, 328*, 411-413, 1990.

Stachnik, R.A., J.C. Hardy, J.A. Tarsala, J.W. Waters, and N.R. Erickson, Submillimeterwave heterodyne measurements of stratospheric ClO, HCl, O_3, and HO_2; First results, *Geophys. Res. Let., 19*, 1931-1934, 1992.

Swinbank, R., and A. O'Neill, A stratosphere-troposphere data assimilation system, *Mon. Weather Rev., 122*, 686-702, 1994.

Waters, J.W., G.L. Manney, W.G. Read, L. Froidevaux, D.A. Flower, and R.F. Jarnot, UARS MLS observations of lower stratopheric ClO in the 1992-93 and 1993-94 Arctic winter vortices, *Geophys. Res. Lett., 22*, 823-826, 1995.

Webster, C.R., R.D. May, D.W. Toohey, L.M. Avallone, J.G. Anderson, P. Newman, L. Lait, M.R. Schoeberl, J.W. Elkins, and K.R. Chan, Chlorine chemistry on polar stratospheric cloud particles in the arctic winter, *Science, 261*, 1130-1133, 1993.

Webster, C.R., et al., Hydrochloric acid and the chlorine budget of the lower stratosphere, *Geophys. Res. Lett., 21*, 2575-2578, 1994.

Wehr, T., S. Crewell, K. Künzi, J. Langen, H. Nett, J. Urban, and P. Hartogh, Remote sensing of ClO and HCl over northern Scandinavia in winter 1992 with an airborne submillimeter radiometer, *J. Geophys. Res., 100*, (D10), 20,957-20,968, 1995.

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