Profiles of Stratospheric ClO and O_3 Simultaneously Retrieved from Millimeter Wave Radiometry at Ny-Ålesund (Svalbard) during March 1997

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Ground-based millimeter wave observations of stratospheric trace gases have been carried out by IMK at the NDSC station Ny-Ålesund (78.9° N, 11.9° E) during March 1997. This measurement campaign was our contribution to the international intercomparison of chlorine monoxide (ClO) radiometers [1] coordinated by IUP, University Bremen, during which 5 individual instruments have been installed at the same site.

The radiometer of IMK has been operated continuously from 12 March to 31 March. Within this timespan there were coincident measurements in parallel with 1 or 2 of the other ClO radiometers on a few days, allowing a direct comparison of the measured spectra and the retrieved profiles. For the entire period the location was inside the polar vortex.

RADIOMETER SYSTEM

The radiometer used for these measurements was the improved version of the MIRA2 radiometer, which was previously used during winter 1996 at IRF, Kiruna. The instrument, which is based on a frontend manufactured by RPG, uses a Schotty mixer cooled with a 20 K closed cycle refrigerator. The receiver noise temperature is about 1100 K. With respect to the latest measurement campaign in winter / spring 1998 at Kiruna major improvements have been completed.

The radiometer is continuously tunable in the frequency range 268 - 279 GHz. Thereby it is capable of measurements of ClO, N_2O, HNO_3 and of several O_3 lines, as was shown during other campaigns [2]. Nevertheless at this occasion the system has been kept at a fixed tuning for a stable baseline in order to ease the retrieval.

The quality of the resulting spectra originates mainly from the advanced balanced calibration method [3], based on the internal adjustable reference load, and from the associated optimized measurement procedures. With this instrument spectral data with high time resolution have been measured continuously over 19 days, except for 2 days because of unfavorable weather.

The measured spectra include the 278.6 GHz signature of ClO, the 278.5 GHz signature of ozone and a few smaller neighboring O_3 lines. To reduce the influence of the thermal noise the spectra were integrated over typically 3 - 4 hours resulting in an almost continuous sequence of 100 individual spectra.

PROFILE RETRIEVAL

The inversion procedure used is an enhanced variant of the Optimal Estimation Method [4]. One essential improvement is the joint retrieval [5], which is the simultaneous profile retrieval of several constituents contributing to an observed spectrum. Thus there is no need to calculate day-night differences to eliminate the ozone lines for the retrieval of ClO profiles. The motivation for a further improvement are sinusoidal undulations in the spectra caused by parasitic resonances inside the radiometer. The periods of these standing waves were determined via Discrete Fourier Transform of the residuals of the measured spectra and a forward calculation. The resulting periods are used for the fit of the standing waves within the inversion process [6].

This direct inversion of spectra containing overlapping signatures of different constituents and standing waves allows the separate processing for any timespan. Thus, the achievable time resolution is only restricted by the noise temperature of the radiometer.

RESULTS

From the measurements up to 8 individual profiles per day have been inverted resulting in a series of 100 volume mixing ratio profiles of ClO and O_3, respectively, which provides an unique synopsis of the variations of both constituents over the entire period. For each profile a vertical resolution of 3 - 4 independent layers between 17 - 55 km has been achieved.

As is presented in Fig. 1 O_3 has a maximum in mixing ratio at 30 - 35 km which shows long-term variations, especially a decay after 20 March indicating O_3 loss. The ClO profiles illustrate both diurnal and long-term variations, which are interesting for further interpretation.

The high time resolution allows the visualization of the diurnal profile variations, exemplified for 17 March in Fig. 4, showing in particular the strong variation of the volume mixing ratio of the lower ClO layer in contrast to the weak variation of the upper ClO and the O_3 layer.

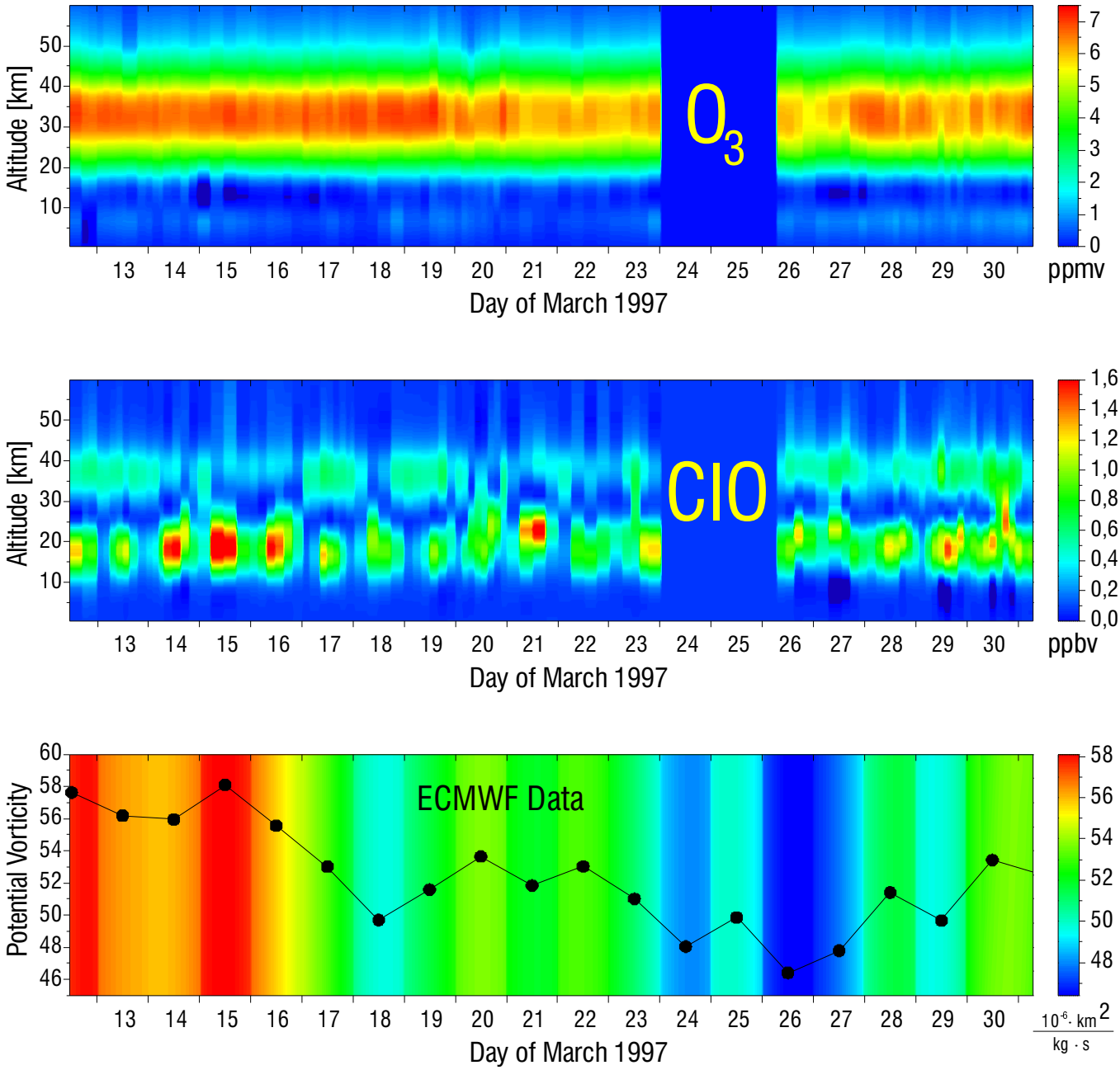

Fig. 1-3: Contour plots of series of volume mixing ratio profiles of O_3 (Fig. 1) and ClO (Fig. 2), respectively, that have been retrieved during 19 days (due to unfavorable weather no measurements could be taken on 24 and 25 March). In Fig. 3 the IPV at 475 K and 80° N is plotted (ECMWF data).

The strong variation of the lower ClO layer is caused by the formation and destruction of the ClO dimer [7]. In the morning after sunrise the ClO dimer is photolyzed and ClO is formed by the following reactions:

$$Cl_2O_2 + h\nu \rightarrow Cl + ClOO$$
$$ClOO + M \rightarrow Cl + O_2 + M$$
$$2(Cl + O_3 \rightarrow ClO + O_2)$$

After sunset nearly all ClO is bound again in the dimer:

$$ClO + ClO + M \rightarrow Cl_2O_2 + M$$

The long-term variation of the lower ClO layer seems to be correlated to the IPV at 475 K, which is shown in Fig. 3.

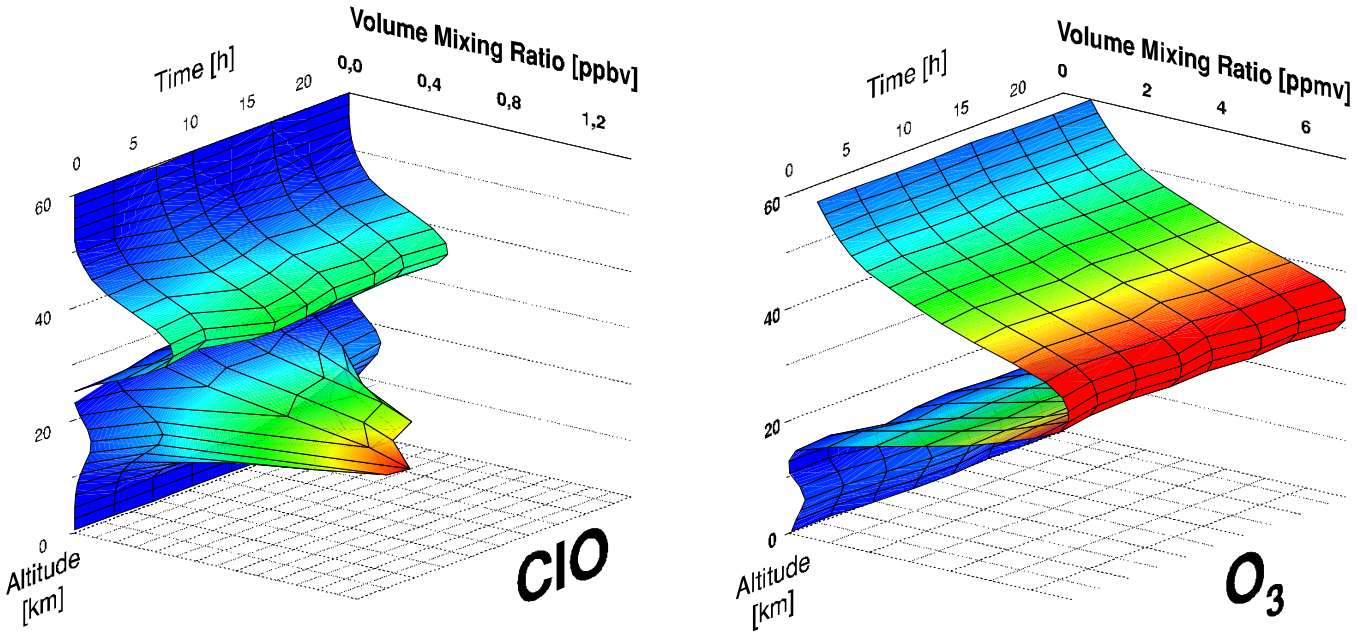

Fig. 4: Diurnal variation of ClO and O_3 profiles for 17 March 1997

CONCLUSIONS

From the millimeter wave spectra measured almost continuously from 12 March to 31 March 1997 up to 8 profiles of O_3 and ClO could be retrieved each day due to the good signal to noise ratio. In the resulting series of profiles with unprecedented high time resolution the diurnal variation of ClO is predominant. Nevertheless long term variations of ClO and O_3 are also discernible

The diurnal variation of the lower ClO layer is a result of the photolysis of the ClO dimer. The long term variation of ClO seems to be correlated with the IPV at 475 K. The comparison of the different millimeter wave measurements performed at the same site at Ny-Ålesund during 1997 will be concluded soon. The high time resolution of the spectra enables a comparison with the chemical model KASIMA [8] of the Research Center Karlsruhe, too. A complementary high resolution observation of the vortex edge is among our major goals for the next future.

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