

FTIR and UV-Visible Measurements of Stratospheric Trace Species at Harestua, Norway during THESEO and Comparison with a 3-D Model.

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Abstract

Groundbased high resolution Solar FTIR and zenith sky UV-Vis measurements have been conducted at the complimentary NDSC site Harestua in southern Norway (60°N, 11°E, altitude 590 m), since 1994. This latitude is far enough north to frequently encounter polar air-masses during winter and spring, but still far enough south to make possible high quality Solar measurements all year around. Here are presented results from the THESEO (Third European Stratospheric Experiment on Ozone) campaign 1998-1999. Total columns of HCl, HF, HNO₃, ClO, ClONO₂, COF₂, O₃ and NO₂ were measured and compared with the 3-D CTM model SLIMCAT. The measurements show elevated ClO and OClO columns on Dec. 7 and Feb. 9 indicating chlorine activation in qualitative agreement with the model. However, during the period Dec 98 - Feb 99 the measurements show no significant ozone loss in contradiction to the model. This indicates that the chlorine activation seen was local in time and space, possibly caused by lee-waves. In March and April the ozone columns shows some chemical loss, but still less than the model.

1. Experimental

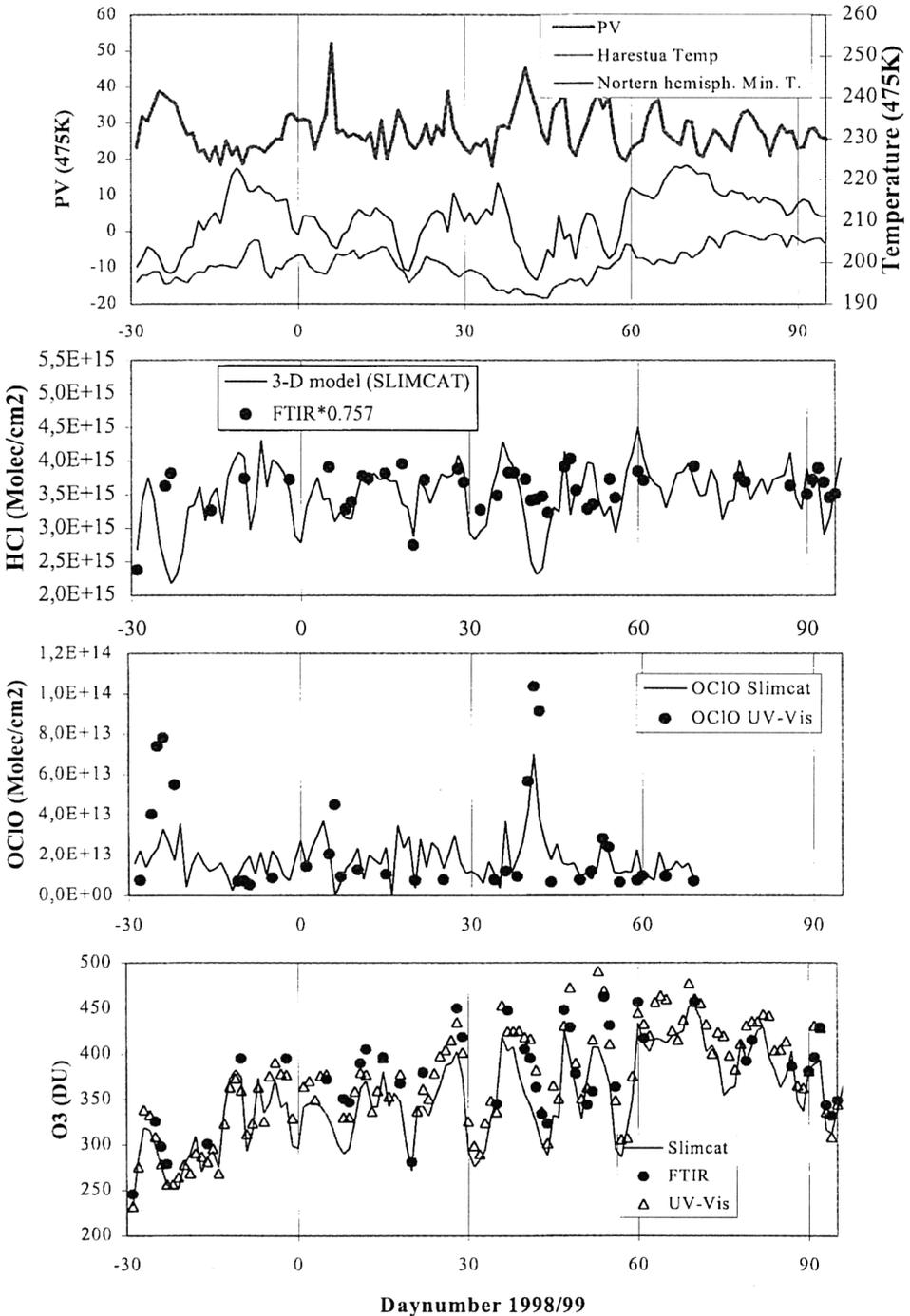
The FTIR measurements are performed using a Bruker 120 M high resolution FT spectrometer connected to a coeliostat. Total columns are retrieved from the Solar spectra using the softwares SFIT and MALT [1]. In addition vertical concentration profiles of some species are derived from the pressure broadened absorption lines, as presented in a companion poster [2].

The UV-Vis measurements are conducted by observations of the light scattered at zenith, using two spectrometers covering the UV and visible spectral ranges respectively.

SLIMCAT is an off-line 3D chemical transport model (CTM) which uses meteorological analyses to specify the wind and temperature fields [3]. The model uses an isentropic vertical coordinate. The transport model is coupled to a detailed stratospheric chemistry model with 35 chemical tracers and around 100 chemical reactions. The model also has a treatment of heterogeneous reactions on polar stratospheric clouds and sulphate aerosols.

2. Results

In Fig.1 are shown the potential vorticity and temperature at 475 K above the site as well as the minimum temperature at this potential temperature in the northern hemisphere (NHM). It can be seen that the NHM temperature just barely reached 195 K level for PSC type I formation twice during the winter. On both these occasions Harestua was close to or in the vortex.



Figures 1 – 4. In Fig. 1 the potential vorticity (as an indicator of polar air) and temperature at the 475 K potential temperature level above Harestua are shown. Also shown in Fig 1 is the Northern hemisphere minimum temperature at 475 K. In Fig. 2 – 4 are shown time series of total columns measured by ground based FTIR or UV-vis spectroscopy, compared with total columns derived from the 3-D global CTM model SLIMCAT.

In Fig. 2 - 4 are shown examples of comparisons between total columns measured by the ground-based instruments during the THESEO winter, and total columns obtained by the SLIMCAT model at 12.00h for the Harestua coordinates. It should be noticed that the exact location of the air-mass probed by the ground-based instrument varies over the day due to the motion of the Sun. As the site often is located close to the vortex edge, large variations in total columns may be seen over the day. This difference in sampling has to be taken into account when comparing measured and modelled data.

- ◆ For HCl (Fig 1.) the modelled columns are 25% lower than the measured columns. This is partly due to the fact that the model only samples the air above about 12 km, while the measurements include the whole column down to the ground. These results are consistent with results from previous winters. The strong loss in HCl column in the model around day -23 and day 42 is not found in the measurements.
- ◆ For ClONO₂ (not shown) the agreement is better. Previous years the model has consistently underestimated the ClONO₂ columns, but not so this year. Some discrepancies on a finer scale are likely to be due to the complex atmosphere resulting from the early stratospheric warming this year.
- ◆ HNO₃ is underestimated with 20% by the model, but still follows the finer variations to a high degree (not shown).
- ◆ OCIO shows strong diurnal variation complicating the model comparison on an absolute scale. From Fig 3 is seen that significantly elevated concentrations was seen on two occasions around Dec. 7 and Feb. 9, coincident with elevated PV and low stratospheric temperatures as was shown in Fig. 1.
- ◆ Elevated columns of ClO was measured on Dec. 7 and Feb. 9. This is in qualitative agreement with the model, and indicates chlorine activation.
- ◆ For ozone the model is compared with both the FTIR and the UV-vis measurements (Fig.4.). The FTIR measurements also have been compared with a Brewer instrument located in Oslo and showed an excellent agreement ($0.7\% \pm 5\%$) over the period 1995 – 1999 [3]. In Table 1 below the average ozone column measured by FTIR, UV-Vis and Brewer for each month, is compared with the model results with and without (passive) ozone loss chemistry. In December, March and April the measurements shows good agreement, while in January and February the model underestimates the column.

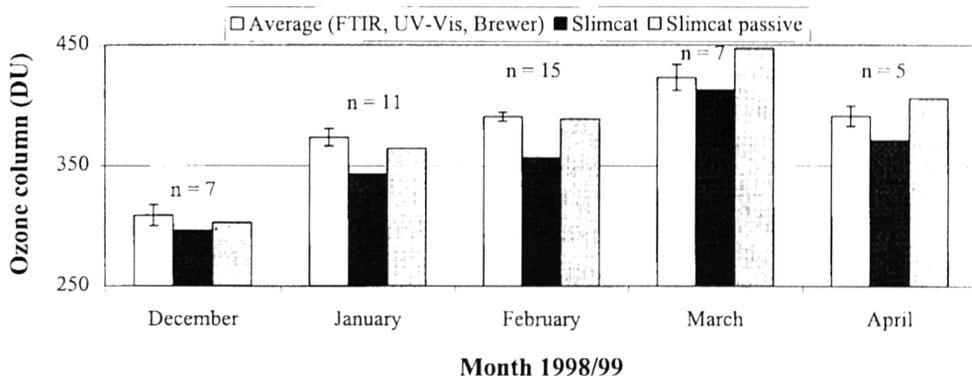


Table 1. Average ozone total column measured by FTIR, UV-Vis and Brewer compared with the model results with and without (passive) ozone loss chemistry. Also shown is the standard deviation of the measurements as well as the number of measurement days for each month.

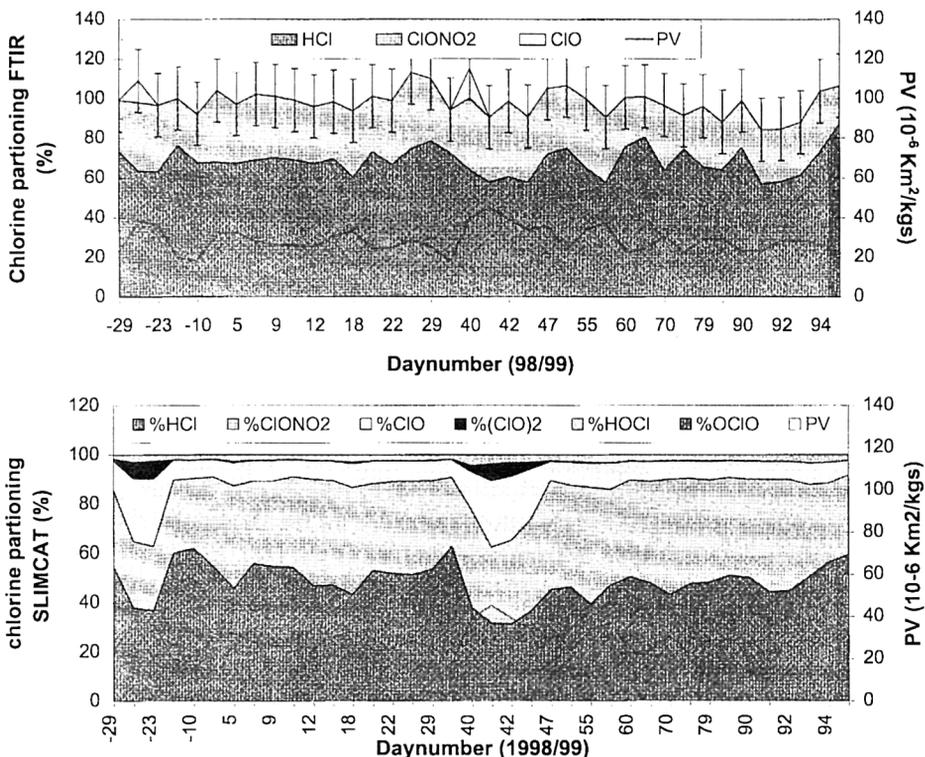


Figure 5. Chlorine partitioning in total columns measured by ground-based FTIR and modelled by Slimcat. The FTIR partitioning is obtained by ratioing the respective chlorine compounds HCl, ClONO₂ and ClO, with the total fluorine (HF + 2 · COF₂), and then normalise to the value during early fall and late spring. Also shown is the potential vorticity at the 475 K level. (Note the discontinuous x-axis values.)

In Fig 5. is shown the chlorine partitioning above the site as measured by FTIR and modeled by SLIMCAT. In both figures the chlorine activation associated with the elevated PV levels is clearly reflected in a drop in HCl column. In the model this loss is considerably deeper than in the measurement, and also results in twice as high ClO column ($1.6 \cdot 10^{15} \text{ mol/cm}^3$ on Dec 7 1998 as compared to $\approx 0.8 \cdot 10^{15} \text{ mol/cm}^3$ measured by the FTIR). It also seems that the model predicts a larger relative amount of ClONO₂ over HCl throughout the period.

3. Acknowledgements

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References

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