**Atmospheric
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Technical note: A stratospheric climatology for O_3, H_2O, CH_4, NO_x, HCl and HF derived from HALOE measurements

J.-U. Grooß[1] and James M. Russell III[2]

[1]Institut für Chemie und Dynamik der Geosphäre I: Stratosphäre (ICG I), Forschungszentrum Jülich, Jülich, Germany
[2]Center for Atmospheric Sciences, Hampton University, Hampton, VA, USA

Abstract. The Halogen Occultation Experiment (HALOE) on board the Upper Atmosphere Research Satellite (UARS) has observed mixing ratios of important trace species in the stratosphere for more than a decade since 1991. Here we present a climatology for the stratosphere compiled from HALOE O_3, H_2O, CH_4, NO_x, HCl, and HF data for the period from 1991 to 2002. In this approach, the data are averaged over equivalent latitude instead of latitude in order to correctly reproduce the gradients at the transport barriers like the polar vortex edge. The climatology is compiled for 5 degree equivalent latitude bins. Seasonal dependence is taken into account by choosing intervals of one month. The climatology is available as an electronic supplement.

1 Introduction

Global atmospheric models such as Global Circulation Models (GCMs) and Chemistry Transport Models (CTMs) need information about the spatial distribution of the chemical composition for some key tracer species, e.g. ozone or water vapor, in order to achieve realistic results especially for the simulation of radiation. Both the calculation of photodissociation rates of chemical species and the calculation of diabatic heating rates are needed to achieve realistic changes in chemical composition and to achieve realistic vertical transport, respectively. There are two different approaches to using the distribution of chemical species for these calculations. Either the information about the key chemical species is prescribed and stems from a climatology compiled from different data sets or it is derived from other models. These simulations are often called *off line* simulations. The second approach is to use the model's own simulated values of the key species in the radiation code taking into account the feedback be-

tween chemistry and radiation. These simulations are often called *coupled* simulations. This approach would benefit from a realistic compilation of a climatology from observations to assure the quality of the derived ozone and water vapor fields on which the radiation simulations are based. Both approaches, *off-line* and *coupled*, can use climatologies of chemical tracers such as CH_4 or HF to validate the large-scale transport and other key chemical species to validate the chemistry of the simulation.

Here we present a climatology based on the satellite observations made by the Halogen Occultation Experiment (HALOE) on board the Upper Atmosphere Research Satellite, which has taken measurements of stratospheric trace species since it was first turned on in orbit on 11 October 1991 (Russell et al., 1993). The measured trace gases are O_3, CH_4, H_2O, HCl, HF, NO, and NO_2. Here we concentrate on O_3, CH_4, and H_2O observations, but we also provide information for the other chemical species. The HALOE instrument uses the solar occultation technique and daily observes up to 15 sunrise and 15 sunset profiles of these measured trace species at about constant latitudes for both groups.

The period covered of about 11 years (1991–2002) corresponds to one period of the solar cycle. We do not include the most recent data since September 2002, since in 2002 there was a very unusual major warming in the Antarctic (Newman and Nash, 2005) and also the observations have been less frequent after 2002.

Other available climatologies are also used in GCMs and CTMs. For example, Fortuin and Kelder (1998) compiled an ozone climatology for the period 1980–1991 from ozone sonde and satellite data. The main difference is that during that period the Antarctic ozone hole was not yet fully developed. Randel et al. (1998) also combined H_2O and CH_4 measurements from HALOE with other UARS experiments (MLS, CLAES) to build a climatology with an equivalent latitude mapping from the observation of the period November 1991 to March 1997. Chiou et al. (1997) compiled a water

Correspondence to: J.-U.Grooß
(j.-u.grooss@fz-juelich.de)

vapor climatology from 5.5 years version 6.1 SAGE II data. However, comparison with correlative data and an update of the retrieval (Thomason et al., 2004) indicated a significant low bias for this climatology. Previous, more abbreviated climatologies of stratospheric water vapor from HALOE have been published by Remsberg et al. (1990) and Russell and Swider (1991).

The UARS reference atmosphere project (http://code916.gsfc.nasa.gov/Public/Analysis/UARS/urap/home.html) also provides climatologies for key chemical species derived from UARS observations for a combination of different instruments on board UARS and also for HALOE measurements only. Depending on species, they use up to the first 7 years of UARS observations. The URAP data are generated by the method after Randel et al. (1998) in which the seasonal cycles are fit using a harmonic regression analysis. Besides a longer time range, we also include NO_x in the climatology as an additional chemical species.

To our knowledge no climatology of NO_x (=$NO+NO_2$) has yet been published. Beaver and Russell (1998) provide a climatology of HCl and HF from HALOE for the first 4.5 years of HALOE observations for 5 latitude bins and four seasons, which is a lower resolution and time range of the HALOE data set compared to the presented climatology.

The climatology presented here is already used in simulations. In the 3-D version of the Chemical Lagrangian Model of the Stratosphere (CLaMS) it has been used to calculate the diabatic heating rates that introduce vertical motion across the isentropic levels (Konopka et al., 2004, 2005). It was also used as input for the radiative transfer calculation from which the chemical photolysis frequencies are determined (Grooß et al., 2002; Grooß et al., 2004). A previous version of this climatology, compiled from 6 years of HALOE data, two-month averages and latitude bins instead of equivalent latitude, is used in the ECHAM-5 model (C. Brühl, pers. comm., 2004).

2 HALOE data uncertainties

The HALOE instrument was validated against a variety of correlative measurements. Also various remote sensors have been compared to HALOE more recently. We use HALOE data from version 19, which is the third public release. We briefly summarize these results for the stratosphere. Generally, the accuracy of the HALOE data decreases near the tropopause.

O_3: The combined systematic and random uncertainty of single ozone profiles in the lower stratosphere (up to 10 hPa) is between 9 and 25% and between 9 and 20% in the upper stratosphere. The agreement with correlative measurements is typically better than 10% down to 200 hPa in the extra-tropics and down to 100 hPa in the tropics and subtropics (Brühl et al., 1996; Bhatt et al., 1999). HALOE O_3 agrees with SAGE (v6.1) within

about 5% (Randall et al., 2003). The differences between HALOE and POAM III are within 10% and often within 5% (Randall et al., 2003).

H_2O: The combined systematic and random uncertainty of single water vapor profiles in the lower stratosphere is between 14 and 24% and up to 30% in the upper stratosphere. The agreement with correlative measurements is typically better than 10% (Park et al., 1996; Kley et al., 2000). The differences between HALOE and ATMOS are generally better than 10% and of the order of 5% (Michelsen et al., 2002). The agreement with SAGE II water vapor measurements (version 6.2) is better than 5% (Taha et al., 2004).

CH_4: The combined systematic and random uncertainty of single CH_4 profiles in the lower stratosphere is between 11 and 19% and between 6 and 27% un the upper stratosphere. The agreement with correlative measurements is typically better than 15% (Park et al., 1996).

NO_x: The combined systematic and random uncertainty of single NO profiles in the lower stratosphere is between 14 and 21% and up to 30% in the upper stratosphere. For NO_2 profiles it is between 9 and 21% throughout the stratosphere. The mean differences from correlative observations for NO_2 data are about 10–15% and for NO data about 15% (Gordley et al., 1996). The HALOE NO_2 data between 20 and 33 km agree with POAM III within about 0.2 ppbv (Randall et al., 2002). Above about 1 hPa level, mostly no NO_2 is detected. As we are interested in the sum $NO+NO_2$, which is dominated by NO at these levels, we set NO_2 to zero above the 1 hPa level in the case that no NO_2 is detected.

HCl: The combined systematic and random uncertainty of single HCl profiles in the lower stratosphere is between 14 and 24% and between 12 and 15% in the upper stratosphere. The mean difference between HALOE and correlative measurements ranges from 8 to 19% (Russell et al., 1996b).

HF: The combined systematic and random uncertainty of single HF profiles in the lower stratosphere is between 15 and 27% and about 15% in the upper stratosphere. The mean difference between HALOE and correlative balloon measurements is below 7% between 5 and 50 hPa. Above 10 hPa, HALOE HF is systematically lower than remote observations by ATMOS by 10–20% (Russell et al., 1996a).

The vertical resolution of the data is between 2 km for O_3, H_2O, and N_2O and 4 km for CH_4, NO, HCl, and HF.
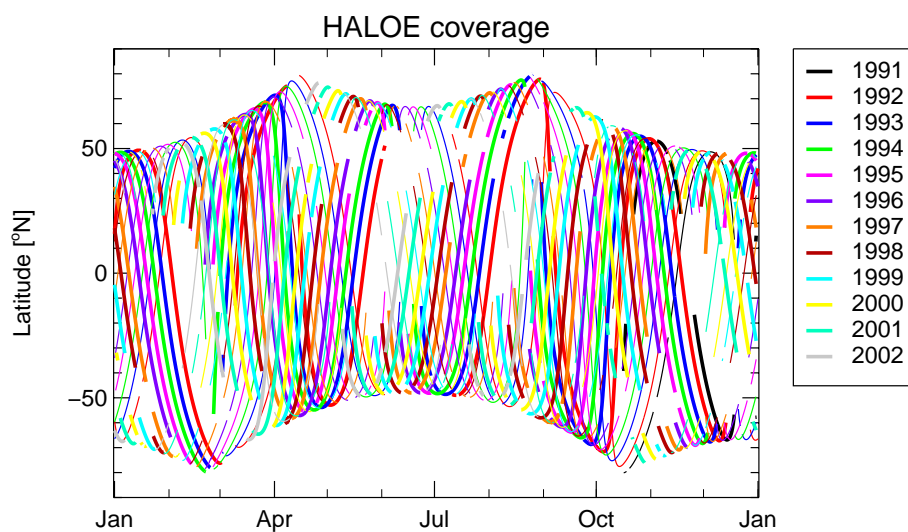
Fig. 1. Coverage of the HALOE instrument. Shown is the latitude of the HALOE observation as function of time of year for all years indicated by different colors. Sunrise observations are depicted by thick lines and sunset observation by thin lines.
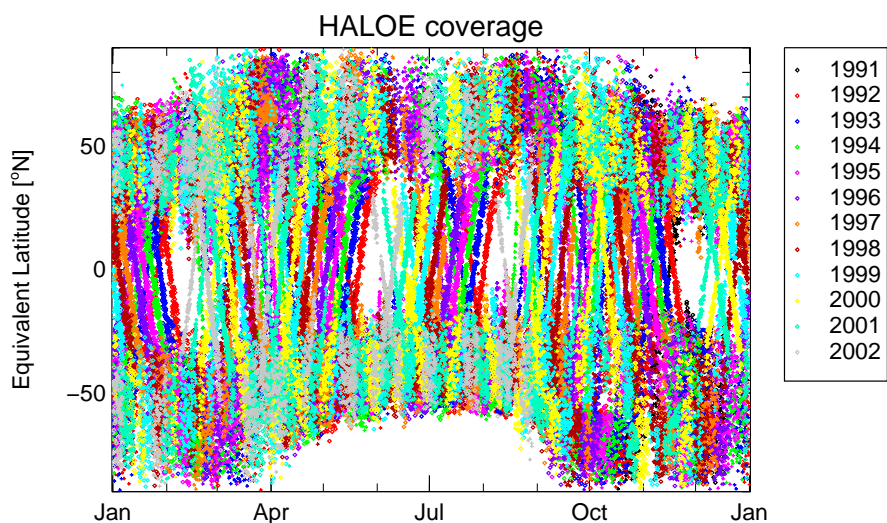

Fig. 2. As Fig. 1, but for equivalent latitude derived from UKMO PV on the 475 K potential temperature level. Sunrise observations are depicted by diamond symbols and sunset observation by crosses.

3 Method

In the polar winters of both hemispheres, large differences of mixing ratios of chemical tracers inside and outside the polar vortices are a typical feature as well as large gradients at the vortex edge. HALOE does not sample the polar vortex on a regular basis. However, often the polar vortices are displaced from a circumpolar symmetry and large fractions of the polar vortex reach latitudes at which HALOE does observe. These air masses contain information from the polar vortex and typically have high potential vorticity (PV) values. Therefore the PV information was used in compiling the climatology presented here. The observations were sorted

and combined with respect to equivalent latitude (Lary et al., 1995). The equivalent latitude was derived from PV using the analyses provided by the United Kingdom Meteorological Office (UKMO).

Figure 1 shows the latitude versus the time of year of the observations covered by the HALOE instrument between 1991 and 2002. Due to the solar occultation geometry, no observations are made during the polar night. Figure 2 shows the HALOE coverage with respect to equivalent latitude. Clearly, the range of equivalent latitudes covered by HALOE is larger than the range of latitudes, especially during the polar winter in both hemispheres.

During the polar summer, there is also a larger coverage of equivalent latitude compared to latitude. However, due to the weak PV gradient in the polar summer, there may be no tight correlation between chemical mixing ratios and PV. Therefore the concept of equivalent latitude is limited for the polar summer. For completeness, we also include a similar climatology in the supplement that is binned with respect to geographical latitude instead of equivalent latitude. This climatology is not explicitly discussed here.

The exact determination of the potential vorticity of a HALOE observation (required to determine equivalent latitude) was performed in the following way. First the observation location was transformed to a time for which UKMO analyses were available, i.e. to 12 h UT of the day of observation. This was done by (backward or forward) trajectory calculations from the time of observation until 12 h UT of the day of observation for potential temperatures between 400 K and 2000 K using the CLaMS trajectory module (McKenna et al., 2002) and wind data from UKMO analyses. The equivalent latitude was then calculated from the PV at the respective potential temperature level for the synoptic location determined in this way.

Between October 1991 and August 2002, a total of 78 600 HALOE profiles of O_3, CH_4, H_2O, HCl, and HF mixing ratio were then combined into bins of 5 degree equivalent latitude for 22 pressure levels between 316 and 0.1 hPa and 12 months. Each data point was assigned a weight equal to the inverse of its given accuracy. In doing so, data with low accuracy (e.g. at the bottom of some measured profiles or due to high aerosol content) only have a low contribution to the determined climatological average value. Since the area of the first and last equivalent latitude bin ($\pm 87.5°$) is rather small and thus does not contain many data points, these bins were combined with the next bins at $\pm 82.5°$.

The HALOE data points that have a retrieved estimated precision of more than 2 times their mixing ratio are rejected from this compilation. Also data points that show unexpectedly large mixing ratios, for example more than 15 ppm ozone and more than 2.0 ppm CH_4, were rejected. Further, it was required that at least 5 HALOE observations were needed in a bin.

In the tropical tropopause region below 100 hPa, often high clouds interfere with the observations, resulting in unreliable results. For example, NO_2 data and HF data in the tropics show a significant increase below the 100 hPa level which is not expected. Therefore NO_2 data and HF data below 100 hPa that increase by more than twice their mixing ratio at 100 hPa are not used for this climatology.

For NO_x, a slightly different procedure was chosen. At the observation time (sunrise or sunset) there is a fast conversion from NO_2 to NO and from NO to NO_2, respectively. The sum $NO_x \bar{N}O + NO_2$ is approximately constant. However, during the night time, also N_2O_5 is produced from NO_x which is not photolyzed by the time of the sunrise observation. Therefore, the sunrise NO_x observations should be corrected by the frac-

tion that is still in the form of N_2O_5. To avoid this correction, we use only sunset profiles for compiling the climatology of NO_x.

Since HALOE does not observe air masses during the polar night, there are gaps in the data sets, even when equivalent latitude mapping is used. To achieve a complete global dataset with the best estimate of the annual dependence, these gaps were filled. Typically, the air within the polar vortex is isolated from mid-latitudes and descends diabatically during the polar night. This causes, for example, the CH_4 mixing ratios at a given height to decrease with time. Thus, the latitudinal gradient increases through the polar nights. Therefore an extrapolation in latitude may not lead to realistic mixing ratios. In this study, we interpolated in time between the observations at the formation and at the end of the polar vortex at high equivalent latitudes, which should yield more realistic mixing ratios. In the following figures, the regions of these filled data gaps are hatched in white.

The advantage of this approach is that the climatology is compiled from a single data source, therefore no biases between different sources have to be taken into account. This method is also possible because of the long-term stability of the HALOE observations. The experiment has been very stable over the 13 plus years of operation thus far. The absolute change in signal level when viewing the sun outside the atmosphere has been no more than 0.5% in the H_2O channel and less than 2–3% in the CH_4 and O_3 channels. Even so, since the measurement is made by ratioing the signals obtained while viewing through the atmosphere to the signals measured outside the atmosphere, any change in the absolute signal level is removed. Also, other parameters such as spectral filter stability and field of view (FOV) mismatch effects have been carefully examined and no significant drifts in the instrument have been found.

The limit of this approach is that data must be interpolated through periods with no observations, e.g. the polar night. Also species with a significant diurnal variation (e.g. ozone in the mesosphere) cannot be covered as the data always correspond to local sunrise or sunset. HALOE does not measure trace species in the troposphere regularly. Therefore this approach of compiling a climatology is best suited for the altitude range of the stratosphere.

4 Results

Figures 3 to 7 show examples of the compiled climatology for O_3, H_2O, CH_4, NO_x, HCl, and HF, respectively. Shown are the months of January, April, July, and October between 330 and 0.33 hPa. The other months are not shown here.

The ozone data displayed in Fig. 3 show a maximum of ozone mixing ratios of about 10 ppm in the tropics at 10 hPa. Also visible is the ozone depletion in the Antarctic vortex in October below 30 hPa, the so-called ozone hole, which was already well established during the period 1991–2002.
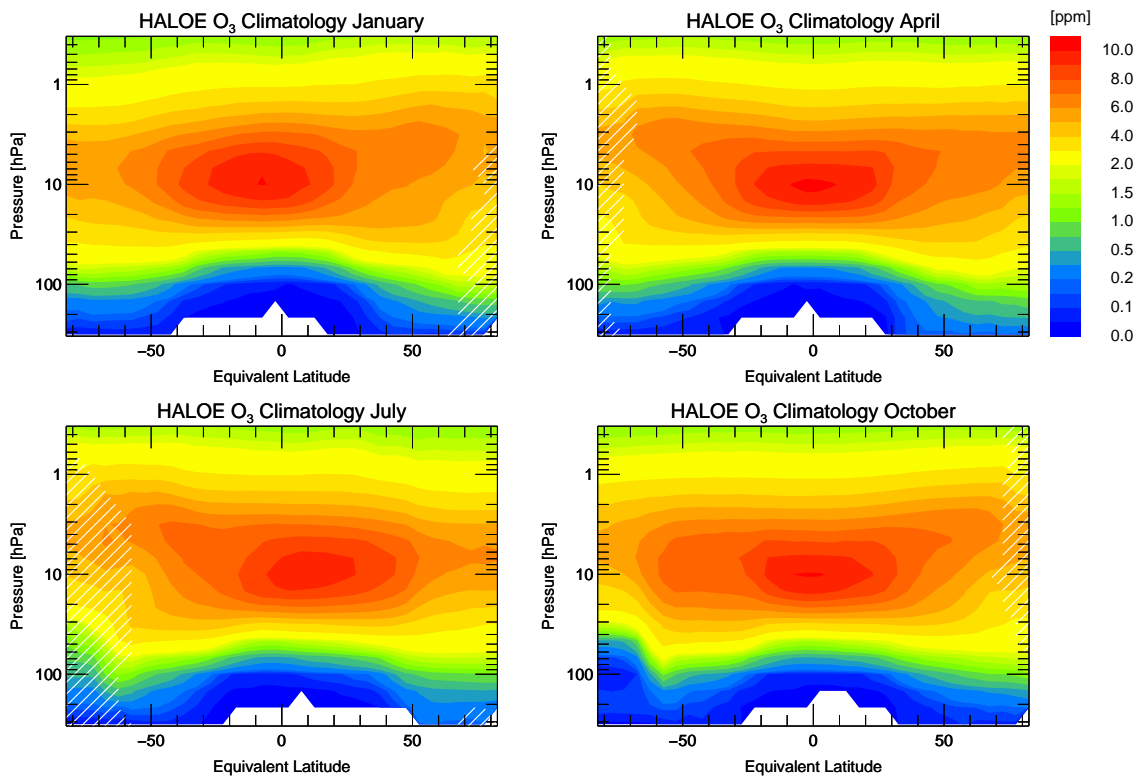
Fig. 3. Climatology for ozone for the months January, April, July, and October. Areas hatched in white indicate data interpolated in time.
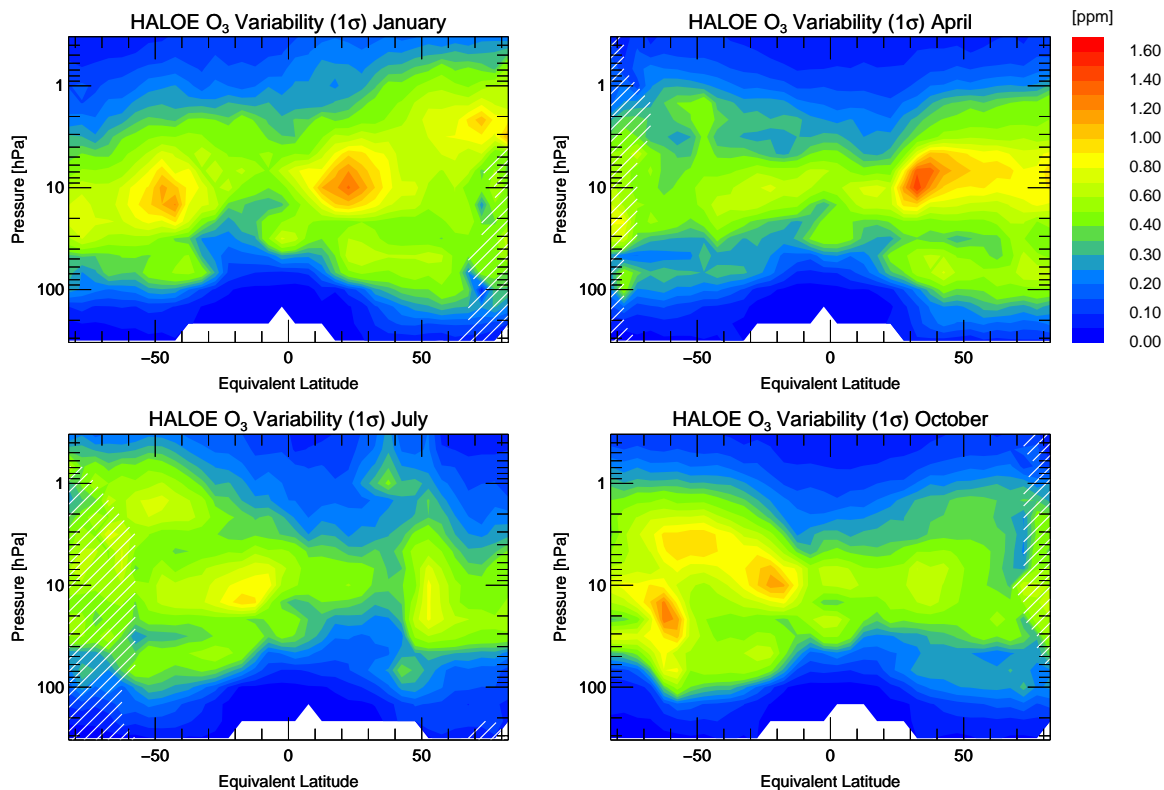

Fig. 4. Standard deviation (1σ) in ppm of the climatology for ozone (Fig. 3) for the months January, April, July, and October. Areas hatched in white indicate data interpolated in time.
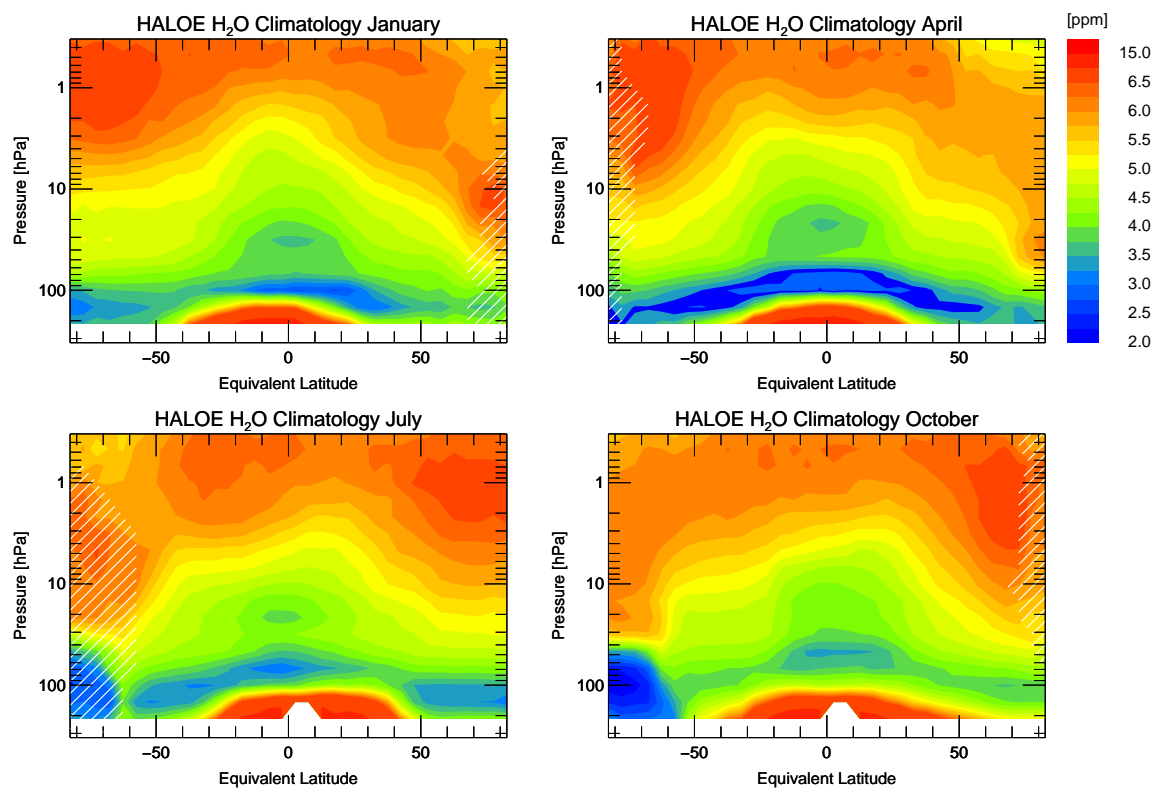
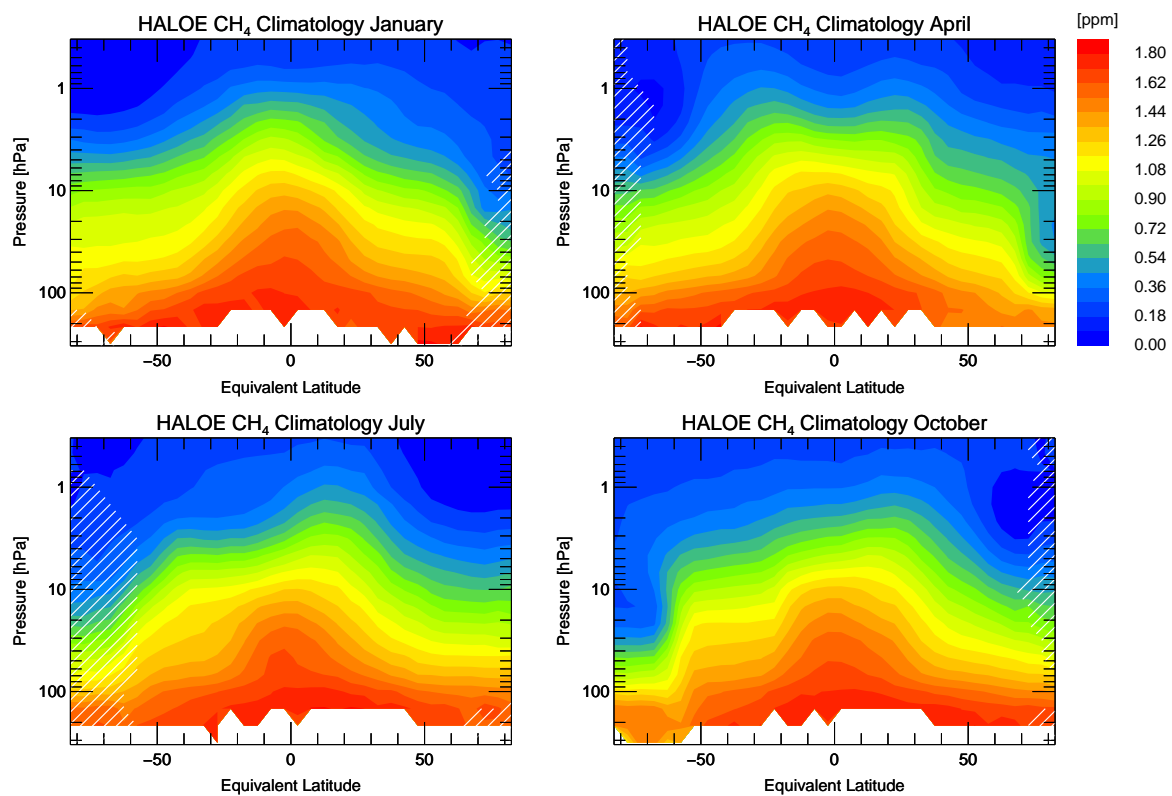**Fig. 5.** As Fig. 3, but climatology for H_2O.

Fig. 6. As Fig. 3, but climatology for CH_4.
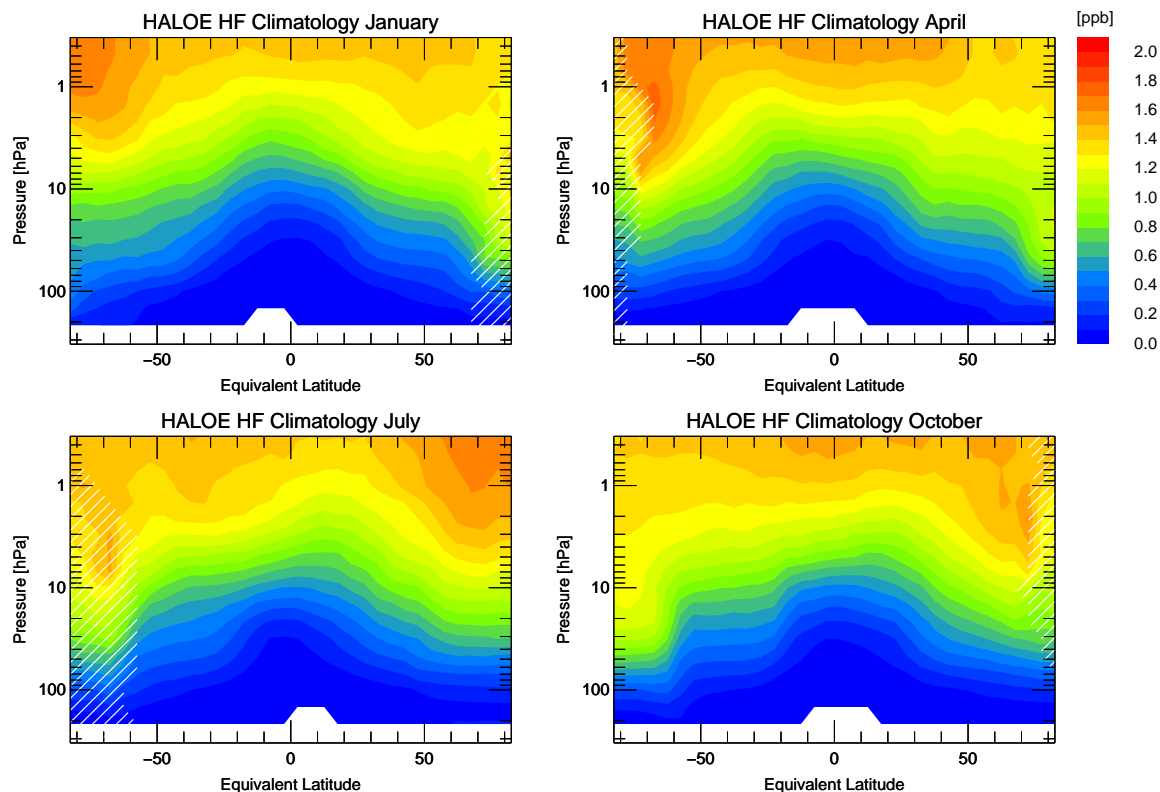
Fig. 7. As Fig. 3, but climatology for HF.

Figure 4 shows the standard deviation (1σ) of the ozone mixing ratio of each bin. It is typically 5–15% in the mid to upper stratosphere and larger below about 50 hPa. Large variability of over 1 ppm is seen at the polar vortex edge in the southern hemisphere in October at the equivalent latitude of 65° S. This is mainly caused by the variability of the size of the polar vortex and is also seen in the variability of CH_4 (not shown here).

Figure 5 clearly shows the increase of H_2O mixing ratio with increasing altitude throughout the stratosphere, which is caused by the CH_4 oxidation. Also, the strong dehydration in the Antarctic vortex in winter and spring is well pronounced in October. The increase of H_2O with decreasing altitude at the tropical tropopause region may be underestimated, as for large tropospheric H_2O mixing ratios no precise HALOE observations are made since the lines employed for the HALOE H_2O retrieval are nearly optically saturated. Thus observations with low H_2O mixing ratios contribute more data points to the climatology.

The importance of CH_4 for radiation is less than ozone or H_2O. The major CH_4 sources are emissions at the earth's surface and the major sink is its oxidation in the stratosphere. Due to its long lifetime, it is a good dynamical tracer and may be used to verify the simulations of meridional transport and the diabatic descent in the polar vortices. In the CH_4 climatology (Fig. 6), especially the diabatic descent over the win-

ter in the polar vortices is clearly visible for both hemispheres yielding low CH_4 mixing ratios inside the vortex compared to mid-latitudes. The corresponding tracer gradient at the vortex edge is also clearly visible. Similarly, HF can be used as a dynamical tracer. Its major source is the photodissociation of CFCs in the stratosphere. The sink in the troposphere is rainout. Therefore the HF climatology (Fig. 7) does show features similar to CH_4 with the opposite sign of its vertical gradient.

The variability of the HCl climatology (Fig. 8) is caused by the increase over the investigated time period, e.g. from about 2.8 ppb to 3.35 ppb at 0.3 hPa between 1992 and 1997. Therefore, the climatology represents only the average HCl but not a value that is typical of the whole time considered. For completeness these data are left in this study.

As explained above, the NO_x climatology (Fig. 9) was compiled only from the sunset observations of HALOE to avoid the variability caused by nighttime conversion into N_2O_5. These NO_x mixing ratios should therefore correspond to daytime values. For the interpretation of the NO_x climatology one has to consider further that especially in the upper polar stratosphere, NO_x is significantly influenced by intermittent solar activity (Randall et al., 2005). Therefore the NO_x mixing ratios do show high variability in the upper polar stratosphere.
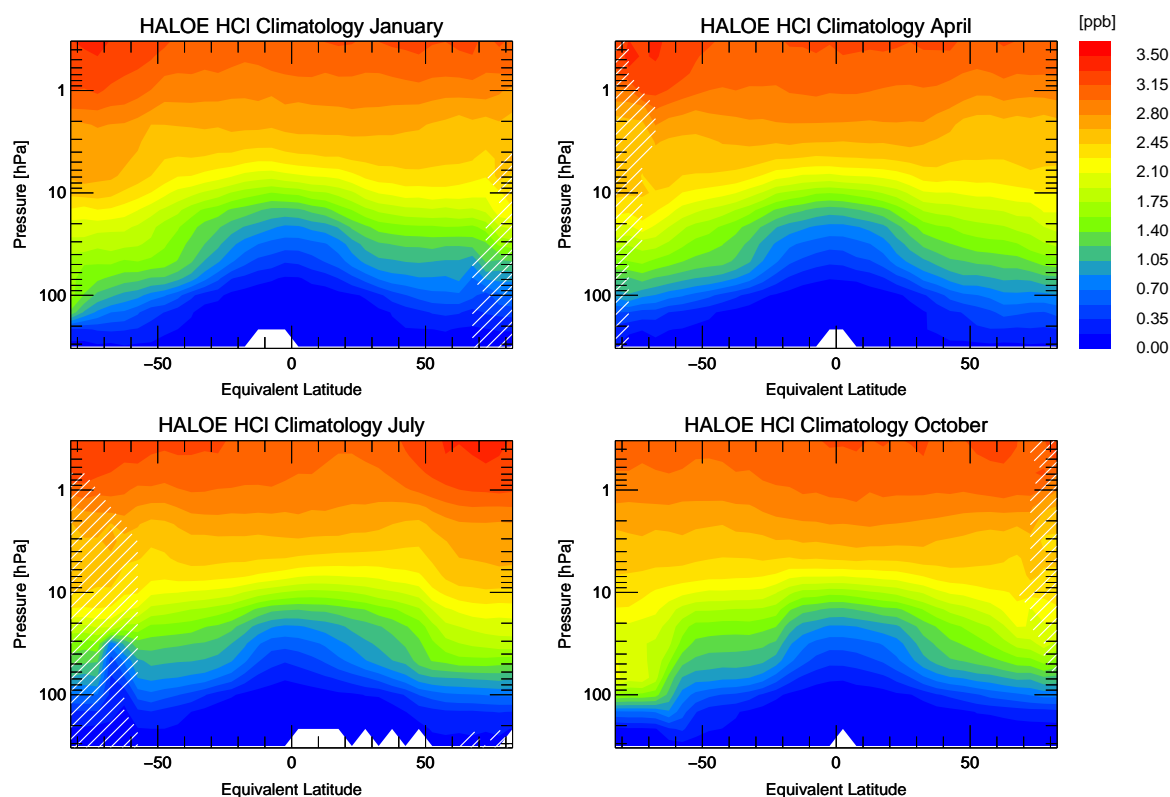
Fig. 8. As Fig. 3, but climatology for HCl.

The presented results for O_3, CH_4 and H_2O are comparable to the 7-year HALOE URAP climatology mentioned above. However, the URAP climatology was compiled with lower latitude resolution using a harmonic regression analysis to reconstruct the seasonal cycle as by Randel et al. (1998). Therefore the URAP climatology provides much smoother latitudinal gradients and lower values of 1-σ variability (O_3 about 20%, CH_4 about 30% and H_2O about 40% between 500 and 1000 K potential temperature).

If this climatology is used for model predictions, the long-term trends of the considered species may be important. As the variability in the climatology is caused by both long-term increase and year-to-year variability, we also include monthly averages of each month during the considered time period in the supplement.

For the period considered here, the ozone hole was already well established. Towards the end of the period, the maximum expected mixing ratio of total inorganic chlorine (Cl_y) in the stratosphere was reached, thus also the maximum of HCl in the upper stratosphere. The stratospheric Cl_y mixing ratio, the precursor of ozone depletion, is expected to decrease slowly over the next few decades, therefore the ozone mixing ratio averaged over the chosen period may be representative of the next few decades in this respect. Water vapor in the stratosphere has been reported to increase. Rosenlof (2002) suggested 0.45%/year over the last 45 years.

A water vapor increase was also reported for the first 5 years of HALOE observations (Randel et al., 1999). However, it has been reported that this has not continued over the following years (Randel et al., 2004). This behavior can partly be explained by the increase of stratospheric CH_4 oxidation, caused by various factors such as the increase of Cl_y until about 1995 and the 11-year solar cycle (Röckmann et al., 2004). It is not clear whether this water vapor increase will continue in the future.

5 Data access

The compiled climatologies for O_3, H_2O, CH_4, NO_x, HCl, and HF are available as a supplement (http://www.atmos-chem-phys.org/acp/5/2797/acp-5-2797-sp.tar). The supplement contains a NetCDF[1] file haloe_climat_eqlat.nc with the described climatology with equivalent latitude binning. The supplement contains both the average data for each bin (equivalent latitude, pressure, month) and its standard deviation. Also, the number of observations corresponding to one bin is included. For data interpolated in time the number is set to –1. The file haloe_lat_climat.nc contains a similar climatology but with latitude binning instead of equivalent

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[1]For details of this file format see http://www.unidata.ucar.edu/packages/netcdf.
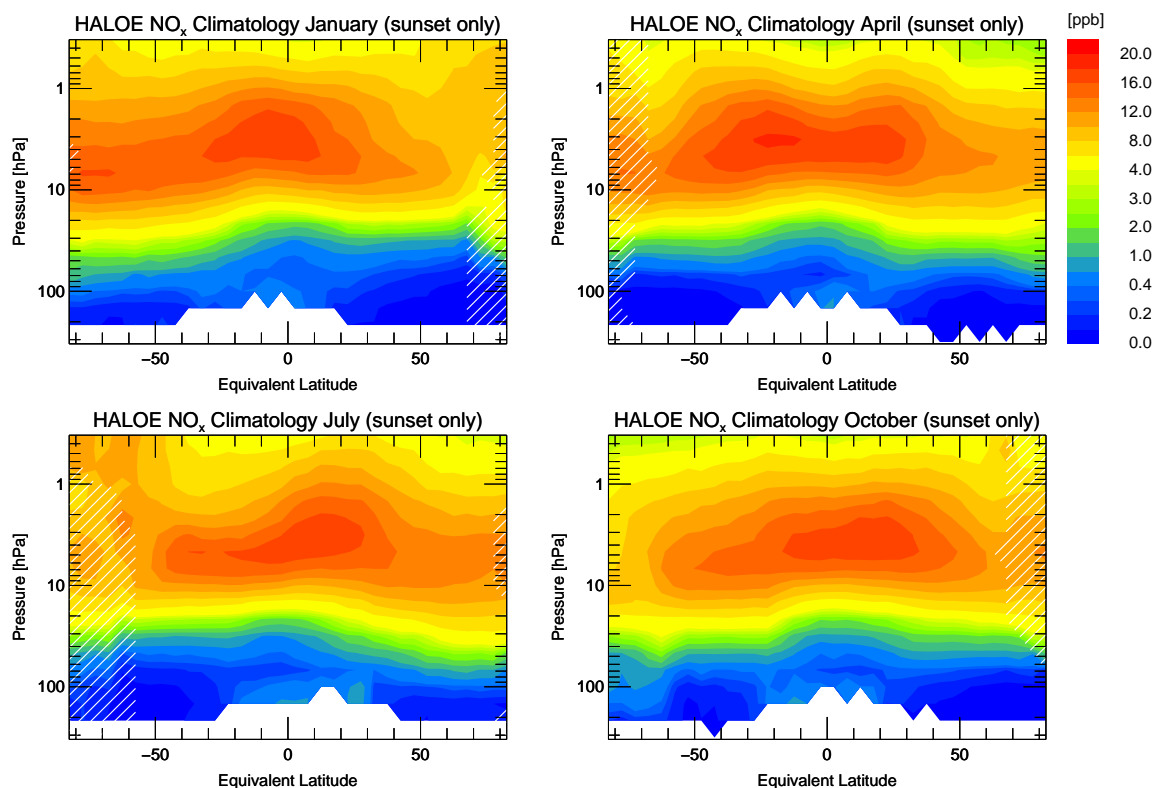
Fig. 9. As Fig. 3, but climatology for NO_x. Here only sunset observations have been used.

latitude binning. Moreover, file haloe_eqlat_monthly_avg.nc contains monthly averages of each month during the considered time period. The chemical mixing ratios and their standard deviation of files haloe_climat_eqlat.nc and haloe_climat_lat.nc are also added as ASCII tables (e.g. haloe_climat_eqlat_o3.dat, haloe_climat_eqlat_o3_stdev.dat).

6 Conclusions

The HALOE climatology presented here was compiled especially as input for GCM and CTM simulations representing the current status of the atmosphere. It uses a single stable data source and therefore avoids problems with biases between datasets. Despite some limitations discussed above, the climatology is well suited as input for model simulations for the 1990s. The climatology for the tracers CH_4 and HF may be used for the evaluation of meridional transport. The climatology for the other species may also be used for evaluation of simulated stratospheric chemistry.

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