

Evolution of stratospheric ozone during winter 2002/2003 as observed by a ground-based millimetre wave radiometer at Kiruna, Sweden

U. Raffalski¹, G. Hochschild², G. Kopp², and J. Urban^{3,*}

¹Swedish Institute of Space Physics, Kiruna, Sweden **please give more detailed affiliation**

²Institute of Meteorology and Climate Research, Forschungszentrum Karlsruhe and Universität Karlsruhe, Germany

³Observatoire Aquitain des Sciences de l'Univers, L3AB, Floirac, France,

* now at: Radio and Space Science Department, Chalmers University of Technology, Sweden

Received: 2 April 2004 – Published in Atmos. Chem. Phys. Discuss.: 11 January 2005

Revised: 8 April 2005 – Accepted: 23 April 2005 – Published: 10 June 2005

Abstract. We present ozone measurements from the millimetre wave radiometer installed at the Swedish Institute of Space Physics (Institutet för rymdfysik, IRF) in Kiruna (67.8° N, 20.4° E, 420 m asl). Nearly continuous operation in the winter of 2002/2003 allows us to give an overview of ozone evolution in the stratosphere between 15 and 55 km.

In this study we present a detailed analysis of the Arctic winter 2002/2003. By means of a methodology using equivalent latitudes we investigate the meteorological processes in the stratosphere during the entire winter/spring period. During the course of the winter strong mixing into the vortex took place in the middle and upper stratosphere as a result of three minor and one major warming event, but no evidence was found for significant mixing in the lower stratosphere.

Ozone depletion in the lower stratosphere during this winter was estimated by measurements on those days when Kiruna was well inside the Arctic polar vortex. The days were carefully chosen using a definition of the vortex edge based on equivalent latitudes. At the 475 K isentropic level a cumulative ozone loss of about 0.5 ppmv was found starting in January and lasting until mid-March. The early ozone loss is probably a result of the very cold temperatures in the lower stratosphere in December and the geographical extension of the vortex to lower latitudes where solar irradiation started photochemical ozone loss in the pre-processed air.

In order to correct for dynamic effects of the ozone variation due to diabatic subsidence of air masses inside the vortex, we used N₂O measurements from the Odin satellite for the same time period. The derived ozone loss in the lower stratosphere between mid-December and mid-March varies

between 1.1±0.1 ppmv on the 150 ppbv N₂O isopleth and 1.7±0.1 ppmv on the 50 ppbv N₂O isopleth.

1 Introduction

Millimetre wave measurements were performed at IRF between 1996 and 2000 on a campaign basis by the Institute of Meteorology and Climate Research (Institut für Meteorologie und Klimaforschung, IMK), Forschungszentrum Karlsruhe and Universität Karlsruhe, using the 268–280 GHz radiometer MIRA 2 (Berg et al., 1998). Since January 2001, the Swedish Institute of Space Physics operates its own ground-based millimetre wave radiometer KIMRA (Kiruna millimetre wave radiometer), performing measurements continuously throughout the year.

KIMRA has been built in collaboration with IMK. The instrument was designed to observe thermal emission lines of stratospheric trace gases between 195 and 225 GHz and to measure O₃, ClO, N₂O and HNO₃ in a continuous mode 24 h a day throughout the year. Additionally, it carries out routine measurements of the tropospheric transmission to estimate the tropospheric water vapour column. IMK performs the inversion of the measured spectra to retrieve profiles and columns of the atmospheric constituents.

For this study we have only used measurements of the twin peak ozone line at 195 GHz. This signature is much stronger than the disturbance due to baseline effects caused by instrumental problems. Spectra of the other trace gases are much weaker and therefore suffer more strongly from these effects and require further instrumental optimization.

Correspondence to: U. Raffalski
(uwe.raffalski@irf.se)

KIMRA at IRF has an obvious advantage in its location north of the Polar Circle with only a short polar night. This is favourable for studying the evolution of the polar stratospheric winter chemistry both inside and outside the polar vortex and for investigating early ozone loss. With a detailed study of the potential vorticity of the polar vortex using the Equivalent Latitude Method (Nash et al., 1996) it is possible to identify whether measurements have been taken inside or outside the Arctic polar vortex. Since ground-based measurements over a longer time-span such as a week observe a substantial part of the air mass inside the vortex, they provide some kind of vortex-averaged data.

A continuous time series can be obtained due to the fact that millimetre wave observations are scarcely impaired by changing weather conditions. Moreover, the Kiruna winter troposphere is usually very cold and dry. For instance, the mean tropospheric transmission at around 195 GHz from January to March 2002/03 during the measurements presented in this paper was around $60\% \pm 14\%$. The continuous measurements provide detailed ozone data that can be used not only for monitoring purposes, but also for process and case studies. Here we present examples of observations of ozone mini-holes in the beginning of December, the estimation of the chemical ozone loss during the winter, as well as mixing processes during the warming events in the course of the Arctic winter 2002/03.

2 Measurement method and instrumental setup

In the microwave region, the received power from faint thermal emission lines in the atmosphere is very small. In order to detect spectral lines of this low intensity with microwave radiometers there are two observation methods available, the total power method and the reference beam method. The millimetre wave radiometer KIMRA deploys the reference-beam method as described by Parrish et al. (1988). However, rather than using a second signal from the atmosphere at a higher elevation angle as reference signal (as suggested by Parrish), KIMRA deploys a rotatable wire grid which blends the brightness temperature of two calibration loads at high and low temperature. This results in a higher contrast in the resulting spectra compared to a reference beam from the atmosphere (Krupa et al., 1998).

KIMRA has been developed in collaboration with IMK. It has been designed and built for measurements of O_3 , ClO , HNO_3 and N_2O in the frequency range from 195 to 225 GHz. In addition the retrieval provides the tropospheric transmission and an estimate for the tropospheric water vapour column.

A conventional Schottky diode mixer is deployed, cryogenically cooled to about 35 K. The receiver noise temperature in Single Side Band (SSB) mode is about 1800 K.

The radiometer is equipped with a periscope-like mirror system enabling measurements in any direction above the

horizon. However it pointed northward for all measurements presented in this paper.

A first technical description of the instrument can be found in Raffalski et al. (2002).

3 Data analysis

The measured spectra are integrated in order to reduce the noise and thus to improve the S/N ratio. The integration time depends on the tropospheric conditions and is typically about 0.5–2 h for a single profile of stratospheric ozone. In situations of high tropospheric transmission an even shorter integration time of 5 to 10 min is sufficient for a successful retrieval.

The varying content of water vapour in the troposphere leads to absorption of the atmospheric signal while it produces an offset in the background signal. These two effects have to be taken care of in the data retrieval. The contrast between line signal and background in the measured spectra varies between 5 K and 25 K, depending on the tropospheric conditions.

The radiative transfer model described in Kopp (2001) uses merged HITRAN 96 (Rothman et al., 1998) and JPL (Pickett et al., 1998) spectral data as supplied by the Bernese Atmospheric Multiple Catalog Access Tool (Beamcat) (Feist, 2004). Daily pressure and temperature profiles of the National Centers for Environmental Prediction, NCEP, (Kanamitsu, 1989) are merged with ground temperature data as measured at IRF (available on <http://tempo.irf.se/cgi-weather/weather-page>) and are then used in the forward model. For the retrieval a modified “Optimal Estimation Method” (Rodgers, 1976) is used.

The retrieval yields vertical profiles of the volume mixing ratio (vmr) of ozone in the altitude range between 15 and 55 km. Using the full width at half maximum (FWHM) of the averaging kernels as a criterion we can obtain a vertical resolution of the ozone profiles of at best 7 km (at 25 km altitude). As an example, the lefthand side of Fig. 1 shows the vertical resolution of the ozone profiles for 20 January 2003 as calculated from the averaging kernels. The righthand side of Fig. 1 shows the sensitivity of the instrument for ozone as estimated from the sums of the averaging kernels at given altitude levels. The instrument has a sensitivity for ozone of at least 75% in the vertical range from about 15 to 55 km. The uncertainty in the retrieved profiles due to standing waves and systematic errors amounts to at least 1 ppmv (Kopp, 2001); errors due to thermal noise are mostly negligible due to the integration of the measured spectra.

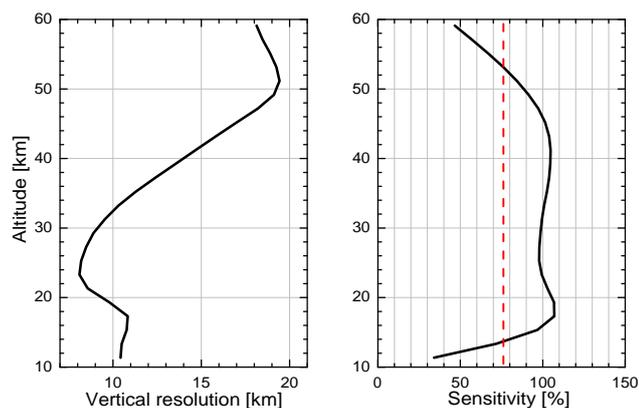


Fig. 1. Vertical resolution (left) and sensitivity of the ozone measurements as calculated from the averaging kernels of the measurements of 20 January 2003. The dashed red line depicts the 75% sensitivity threshold defining the altitude range where the measurements provide significant information.

4 Evolution of the Stratosphere over the Northern Hemisphere in winter 2002/2003

4.1 Methodology

To analyse the evolution of ozone during the winter period we have only used measurements well inside the polar vortex. The definition of the vortex edge follows the Equivalent Latitude Method as suggested by Nash et al. (1996). The equivalent latitude is obtained by calculating the area of the vortex enclosed by a certain PV-isoline and plotting areas of different PV values as concentric circles around the pole. The equivalent latitude is a measure for the size of the vortex and should not be confused with geographical latitudes. When considering Ertel's potential vorticity against the equivalent latitude, the mean vortex edge can now be defined as the steepest gradient in the resulting curve. The points of strongest curvature in this function yield the outer and inner vortex edge, respectively. This calculation leads to strong oscillations, spikes and even to no results if the vortex is not well confined. In particular this is the case if the transition area from inside to outside vortex is very large or totally undefined, as during the vortex formation or break-up phase and during events of strong distortion. In these cases no steepest gradient can be calculated and/or the points of the strongest curvatures result in an unrealistic width of the edge area. On some days no edge can be identified at all.

The advantage of this method over a fixed PV value for a certain isentropic level to determine the vortex edge is the more realistic interpretation of the potential vorticity as given by the European Centre for Medium-Range Weather Forecast (ECMWF). Given the potential vorticity over a particular site, we can determine more precisely whether a measurement at this site on a particular day was performed inside the polar vortex, in the vortex edge region, or outside the polar

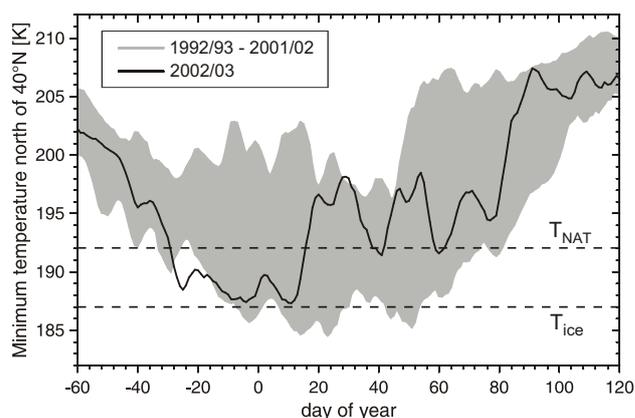


Fig. 2. Minimum temperatures north of 40° N at the 475 K isentropic level (ECMWF-data). The shaded area depicts the variability for the winters 1992/1993–2001/2002. The dashed lines depict PSC formation temperatures for T_{NAT} and T_{ICE} .

vortex (Karhu et al., 2003). Thus, measurements can be identified as observations inside the polar vortex which otherwise (with a fixed PV value as vortex edge) would have been misinterpreted as outside-vortex or vortex-edge observations.

4.2 Meteorology of the winter

The temperature evolution during the winter was quite peculiar. Figure 2 shows the temperature variation of this winter compared to the 10 winters prior to 2002/03 for the 475 K isentropic level (ECMWF-data). The ECMWF analysis shows an exceptionally low minimum temperature at the beginning of December 2002. Throughout December temperatures stayed below the formation temperature for PSCs. In mid-January a major warming occurred. After that the temperature did not reach the low early winter values again even if the values dropped to near-formation temperature for some short periods in February and March.

During the whole of December PSCs have been observed by Lidars at Ny-Ålesund (78.9° N, 11.9° E) and Alomar (69.3° N, 16° E) (EORCU, 2003). Also early ClO activation was reported (Urban et al., 2004), though most of the ClO remained in the dimer due to the lack of sunlight in December (EORCU, 2003).

Figure 3 presents the evolution of the polar vortex in equivalent latitudes as calculated from ECMWF T213 data for different isentropic levels. The plates clearly show the formation of the vortex as early as in November 2002 at the 950 K isentropic level, and the downward evolution towards lower isentropic levels during the cold winter period. The minor warming at the end of December appears only as some weakening of the vortex at 950 and 675 K, respectively, i.e., at higher levels.

Also the major warming in mid-January barely affected the lower stratosphere. While the vortex at 950 K isentropic

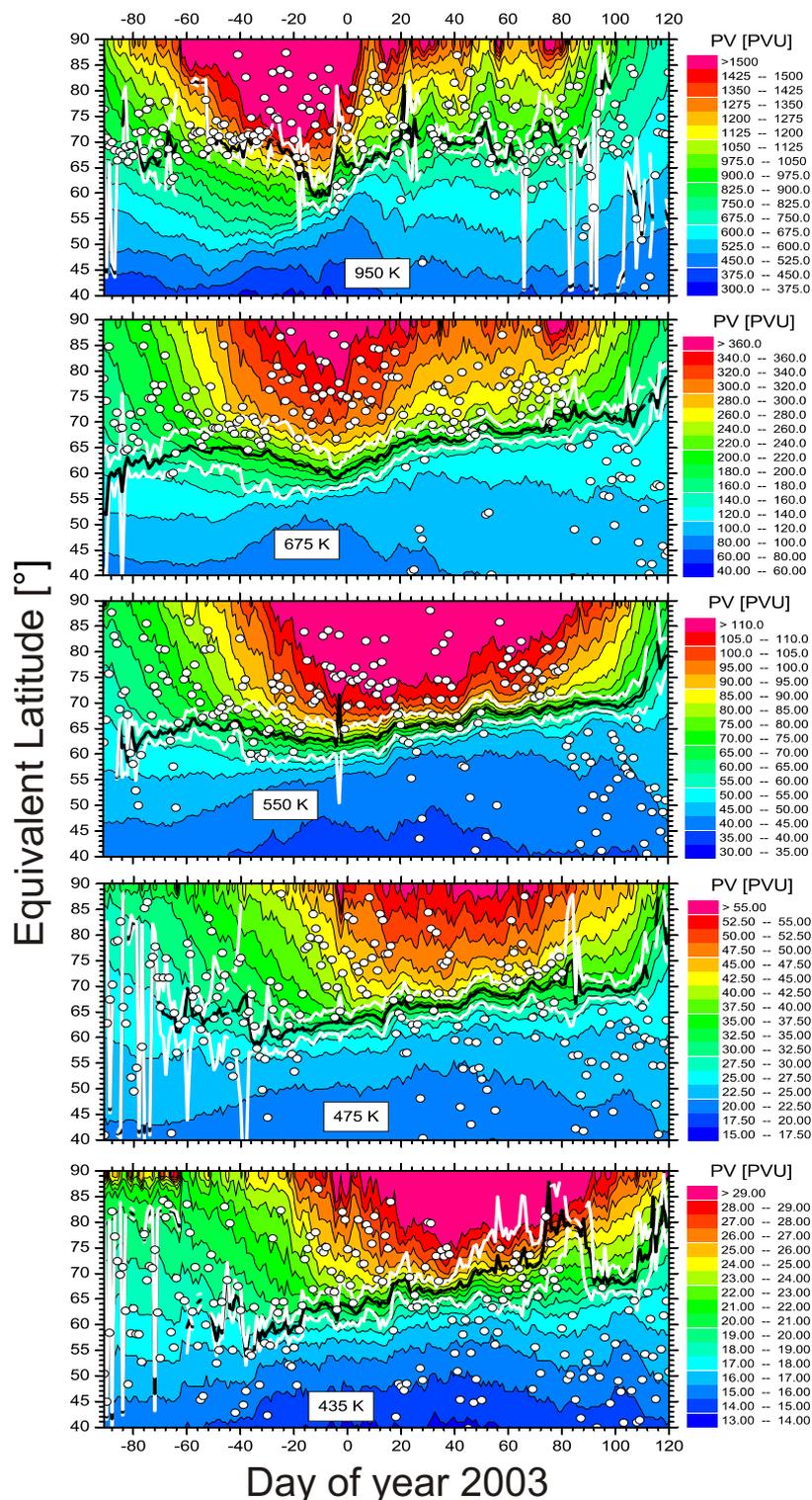


Fig. 3. Evolution of the polar vortex in terms of equivalent latitudes. The white lines describe the inner and outer edge of the vortex, respectively, while the black line describes the mean vortex edge, i.e. the strongest gradient in PV for the particular day. The colour coding shows the strength of the vortex in PVU. The open circles depict the PV over Kiruna on the particular measurement days and refer to the underlying PV colour coding, showing how far away from or how deep inside the vortex a measurement has been taken.

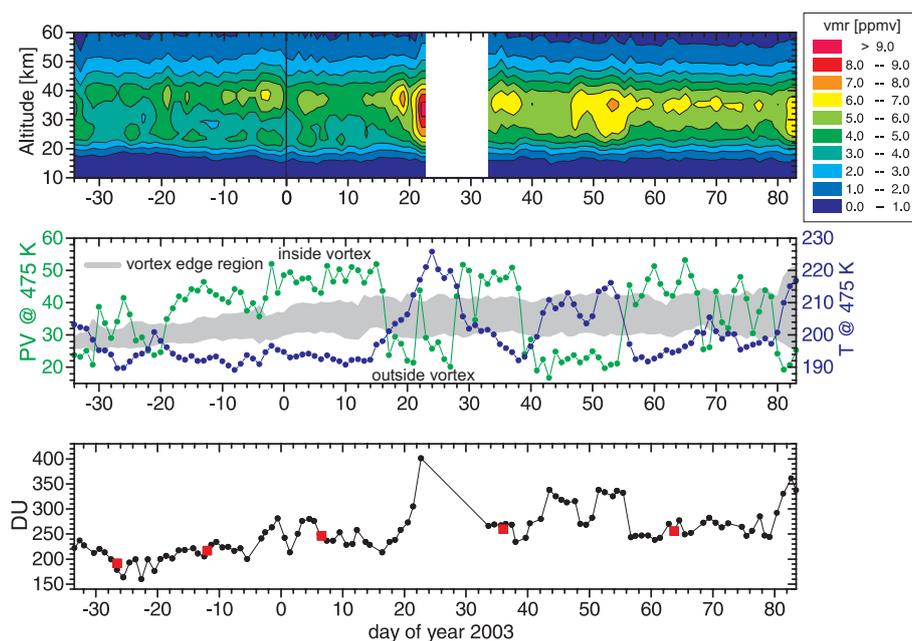


Fig. 4. Evolution of ozone between 28 November 2002 and 25 March 2003. The upper panel shows the time series of ozone vmr profiles. Between 23–31 January there was no data due to technical problems. The middle panel shows the potential vorticity and temperature at the 475 K isentropic level (ECMWF-data). The grey shaded area gives the PV of the vortex edge area. The lower panel shows the stratospheric ozone columns above an altitude of 10 km. The red squares give the mean values for the periods when Kiruna was well inside the vortex.

level was weakened substantially by that warming event, this evolution does not reach down into the lower stratosphere. Although the minimum temperature of the vortex at the 475 K level (as shown in Fig. 2) increases, no substantial area loss or weakening of the vortex can be seen in Fig. 3 at 550 K isentropic level and below.

The two minor warmings in February and March can best be seen at 550 K and 675 K. The final warming over all altitudes in the stratosphere starts at the end of March and the vortex has disappeared by the end of April.

This analysis of the vortex evolution based on the equivalent latitude method is consistent with the EORCU meteorological analysis (EORCU, 2003).

The white and black lines in Fig. 3 show the inner, outer, and mean vortex edge, while the open circles depict the position of Kiruna relative to the Arctic vortex. As can be clearly seen, Kiruna was well inside the vortex for long periods in the course of the Arctic winter 2002/03.

5 Ozone

In this study we used ozone profiles as measured by the IRF-radiometer between 28 November 2002 and 25 March 2003. The resulting time series of ozone profiles is shown in the upper panel of Fig. 4. While a few sporadic days with no measurements were interpolated in this graph, no measure-

ments were taken during the entire period between 23 and 31 January due to technical problems.

The middle panel of the same figure shows the potential vorticity and temperature at the 475 K isentropic level (ECMWF-data). The grey shaded area gives the PV of the vortex edge area between the inner and outer vortex edge as described in Sect. 4.1. Very low temperatures can be seen in December when Kiruna was well inside the polar vortex and PSCs were observed (EORCU, 2003).

The lower panel in Fig. 4 shows the stratospheric ozone columns above an altitude of 10 km for all days when measurements were performed. The red squares give the mean columns for those periods when Kiruna was well inside the polar vortex.

The column density can be calculated starting at an altitude as low as 10 km due to the fact that KIMRA still is sensitive below 15 km but with less than 75% as can be seen in Fig. 1. Furthermore, the vertical resolution amounts to 10 km at an altitude of 15 km which implies that significant contribution to the retrieved volume mixing ratios at 15 km comes even from 5 km above and below. This information has been used in the calculation of the column density.

5.1 Ozone mini-holes in early December

In early December we find extraordinarily low ozone columns in our measurements. These low ozone columns coincide with the ozone mini-holes between 4 and

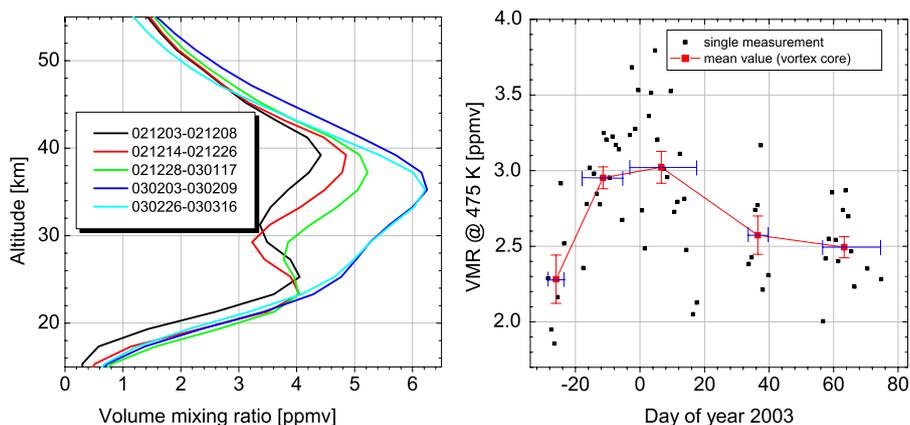


Fig. 5. Evolution of the ozone vmr inside the polar vortex during the winter of 2002/03. The plot on the lefthand side shows averaged profiles for periods when Kiruna was well inside the vortex. The volume mixing ratios at the 475 K isentropic level are presented on the righthand side. The red error bars presented here are solely due to the data averaging over certain evaluation periods, that is, the statistical error or the standard error of the mean. Blue bars depict the periods used for the mean. All measurements are taken from the vortex core.

11 December as reported by EORCU (2003). The partial column density reaches the lowest values of 163 and 160 DU respectively on December 5 and 8, something which was not observed again later during this winter. Although PSCs have been observed and ClO_x levels were elevated (though mostly in form of the dimer) we believe that the observed low ozone is caused by dynamical effects as described by EORCU (2003): a raise in the tropopause height, coinciding with a strong vortex and reduced ozone levels above 20 km, led to the exceptionally low ozone levels.

5.2 Ozone during the warming events

The strong upper stratospheric minor warming at the end of December led to increasing ozone values at those altitudes over Kiruna, which during that time was well inside the polar vortex. Temperatures and potential vorticity at the 475 K isentropic level remained unaffected by this warming event.

After 18 January, the volume mixing ratios of ozone in the middle stratosphere increased drastically, leading to extremely high values of more than 9 ppmv above an altitude of 30 km on 23 January. At the same time, the measured stratospheric ozone columns and the temperatures at the 475 K isentropic level increased significantly. All these findings are the result of the major warming which occurred in January and transported ozone rich and warm air masses from lower latitudes into the Arctic stratosphere. The minor warmings in February and March are not so prominent in the time series presented in Fig. 4.

5.3 Ozone loss

In order to investigate the evolution of ozone inside the polar vortex, mean profiles were calculated for periods when Kiruna was well inside the vortex (lefthand side of Fig. 5). From early December to early January, the diabatic subsi-

dence in the middle and lower stratosphere can be seen. The profiles in February and March show enhanced ozone vmr values in the middle stratosphere, remnants of the mixing processes during the major warming in January. However no such ozone enhancement can be observed in the lower part of the profiles. On the contrary, these profiles show less ozone in the lower stratosphere below an altitude of 22 km than the profiles from late December and mid January. We believe this is the result of chemical ozone depletion.

The righthand side of Fig. 5 shows the measured vmr inside the polar vortex at the 475 K isentropic level. The black dots are the individual measurements and the red squares are the mean values for the different periods when Kiruna was well inside the vortex. The vertical error bars give the standard error of the mean and the horizontal error bars give the periods which were used for the respective average. First an increase of the volume mixing ratios can be observed due to the diabatic subsidence which brings ozone-rich air masses from higher altitudes to the level considered. After January the vmr values decrease due to chemical ozone depletion. Without considering the effect of subsidence in the polar vortex, the cumulative ozone loss at the 475 K isentropic level is about 0.5 ppmv or 16% until mid-March. After 16 March Kiruna was never again well inside the polar vortex at the 475 K isentropic level (see Fig. 3).

In order to calculate the ozone loss that is not masked by dynamical processes, we must match the ozone values presented in the lefthand side of Fig. 5 with a dynamic tracer. N_2O is widely used as a long-lived tracer to investigate dynamical effects such as the diabatic subsidence inside the polar vortex. N_2O has no stratospheric sources and has a lifetime in the lower stratosphere of about one year. Thus the abundance of N_2O can be considered to be constant for the time period of a winter/spring season.

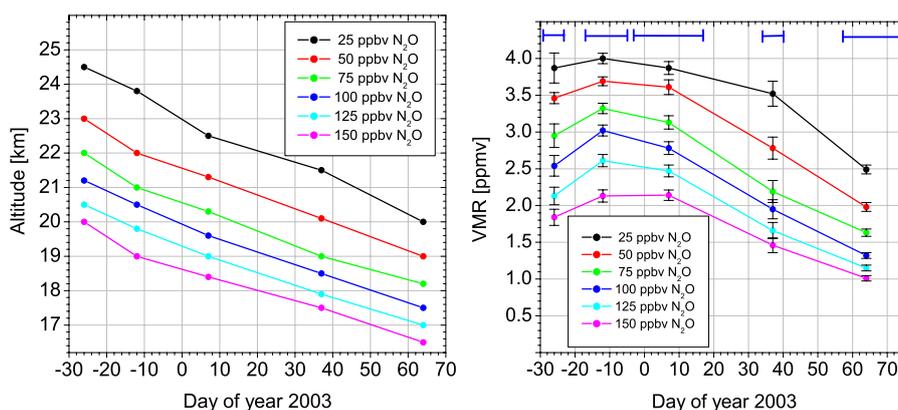


Fig. 6. The plot on the lefthand side shows the diabatic subsidence of vortex air at different N_2O levels as measured by Odin/SMR, averaged according to the periods when Kiruna was well inside the vortex. The righthand side shows mean ozone volume mixing ratios as measured by the ground-based microwave radiometer on Odin N_2O isopleths. Error bars depict solely the statistical error of the averaging process. Blue bars depict the periods used for the mean. All measurements are taken from vortex core.

Since the IRF millimetre wave radiometer does not yet provide N_2O -data, we have used vortex-averaged N_2O data from the Sub-Millimetre Radiometer (SMR) on board the Odin satellite (Urban et al., 2004) to calculate the diabatic subsidence for different altitude levels. The lefthand side of Fig. 6 shows the vortex-averaged N_2O data. For a better comparison these data are also averaged for the periods when Kiruna was well inside the Arctic vortex. The isopleths between 150 and 50 ppbv show a nearly linear subsidence after mid December. The subsidence from early December to the end of March amounts to between 3.5 and 4 km for the different N_2O isopleths. This steady subsidence gives evidence that no significant mixing processes took place at these levels during the warming events during the course of the winter. Only the 25 ppbv isopleth shows some deviation from the linear subsidence of the other isopleths between early January and early February. This is probably the result of some mixing processes at this level during the major warming in January.

Our ground-based measurements over a longer time period in principle give vortex-averaged values (as long as only data from inside the vortex are used). Thus, binning of Odin's N_2O in the same way as our ozone data yields an estimate for the vortex-averaged diabatic subsidence which we can use to estimate the chemical ozone loss during the winter period. The righthand side of Fig. 6 shows the ozone volume mixing ratios of the ground-based ozone measurements on the N_2O isopleths from Odin. The vertical error bars are again the standard error of the mean and the horizontal error bars give the periods used for averaging. In early December a slight increase of ozone vmr values can be seen on all N_2O isopleths. This increase cannot be explained by dynamical processes since they should have been removed by using N_2O as a dynamic tracer. Probably this increase is an artefact introduced by the limited vertical resolution of the ground-based

millimetre wave radiometer: when air masses subside, the ozone profile is compressed in the vertical direction. Thus, the higher ozone volume mixing ratios from higher altitudes are partly mapped into the considered (lower) level due to the limited resolution even if the considered level is also subsided.

Another explanation for the increasing ozone vmr values in December is that the ozone-tracer relationship is not very well established in the initial phase of the winter, giving rise to a quite large uncertainty since the selection of the early vortex profiles determines the early winter ozone-tracer relations (Harris et al., 2002).

In order to avoid the early winter uncertainties in the ozone-tracer relationship we use ozone values from mid December as initial values for an ozone-tracer relationship deploying Odin N_2O -data. Some ozone loss prior to mid-December which might have been excluded this way, however, should be quite limited due to the lack of sunlight. This assumption is supported by Tilmes et al. (2003), who found large column ozone loss starting in January 2003 using the tracer-tracer correlation technique.

After mid December, ozone concentrations show a general decrease due to chemical ozone loss. One cause for the early ozone loss even in January might be the geographical extension of the vortex to lower latitudes. Thus, the preprocessed stratospheric air masses were exposed to stronger solar irradiation than in the polar region, resulting in photochemistry and ozone depletion. The cumulative ozone loss between mid-December and mid-March amounts to 1.1 ± 0.1 ppmv on the 150 ppbv N_2O isopleth (20.0–16.5 km) and 1.7 ± 0.1 ppmv on the 50 ppbv N_2O isopleth (23.0–19.0 km).

However we can see two deviations from the overall evolution:

- The lowest altitude depicted by the 150 ppbv N_2O

isopleth shows no onset of ozone loss before mid January. This might be due to the only sporadic PSC occasions at these low altitudes (18–19 km) during late December and until mid January (see EORCU, 2003).

- The evolution of the ozone volume mixing ratio on the 25 ppbv N₂O isopleth (24.5–20.0 km) deviates slightly in that it shows a smaller decrease after the major warming event in January compared to the other levels. We think this is the result of the mixing processes during the warming event.

The ozone loss derived from ground-based measurements can be compared to Odin ozone data. Unfortunately Odin's ozone vmr on N₂O isopleths is only available on the 25 ppbv and 50 ppbv N₂O isopleths and only between mid December and mid February. For this comparison Odin N₂O data is deployed for a tracer correlation with the ground-based millimetre wave measurements at Kiruna. We find an ozone loss between mid December and mid February of 0.5 ± 0.2 ppmv on the 25 ppbv isopleth and 0.9 ± 0.2 ppmv on the 50 ppbv isopleth, respectively. The measured loss on the 25 ppbv isopleth is probably affected by the mixing processes during the warming event in January. Nevertheless, these values compare well to the vortex-wide ozone depletion measured by Odin in the same period (Urban et al., 2004). Unfortunately Odin data do not allow ozone loss calculation for lower altitudes due to the limited altitude range of the ozone measurements analysed so far.

The cumulative ozone loss in the lower stratosphere between mid-December and mid-March is of the same order as the ozone loss which was detected by microwave radiometry and FTIR measurements in Kiruna in the cold arctic winter of 1999/2000 (Kopp et al., 2002). The relatively high ozone loss in the winter of 2002/2003 fits well to the meteorological situation of this winter with very low temperatures in early December and the reported chlorine activation (Urban et al., 2004) and denitrification (EORCU, 2003) mainly in January and lasting to mid-March.

6 Summary and conclusions

The winter of 2002/03 was unusually cold in the lower stratosphere in early December, and minimum temperatures in the polar vortex stayed below PSC-formation temperature until mid-January. The minor warmings in December, February and March and the major warming in January weakened the Arctic vortex. Using the potential vorticity against equivalent latitudes and averaged Odin N₂O data we show that mixing processes during these warming events were only important above the 550 K isentropic level or above the 50 ppbv N₂O isopleth, respectively.

Almost continuous ozone measurements have been taken using the 195–225 GHz radiometer at Kiruna from the end of November 2002 to the end of March 2003. From the measurements we retrieved a time series of ozone vmr profiles and stratospheric columns.

In early December 2002 we found extraordinarily low ozone column densities over Kiruna. These low values coincide well with the ozone mini-holes reported by EORCU (2003) for this time span.

The mixing processes during the warming events in December and in particular in January led to increasing ozone mixing ratios in the middle and upper stratosphere. The maximum was reached in January with more than 9 ppmv above an altitude of 30 km; this is an extraordinarily high value. These mixing processes are also clearly visible in the measured ozone profiles which were averaged for periods when Kiruna was well inside the polar vortex.

In contrast, the ozone concentration in the lower stratosphere decreased after January due to chemical depletion. One reason for this early ozone loss might be the exposure of pre-processed stratospheric vortex air masses to solar irradiation at lower latitudes with subsequent photochemical ozone depletion.

At the 475 K isentropic level the cumulative loss amounted to about 0.5 ppmv, but part of the loss was masked by diabatic subsidence of vortex air. To account for this effect, we have used N₂O data as measured by the Odin satellite to calculate the vortex-averaged diabatic subsidence. The N₂O isopleths in the lower stratosphere show a subsidence between 3.5 and 4 km from early-December 2002 to mid-March 2003. We could find no evidence for mixing during the warming events in the course of the winter below the 25 ppbv N₂O isopleth. On the 25 ppbv isopleth we detected some indications for mixing during the major warming in January.

We used the diabatic subsidence as calculated from the Odin N₂O data to estimate the ozone loss as detected by the ground-based microwave measurements. The cumulative ozone loss between mid-December and mid-March amounts to between 1.1 ± 0.1 ppmv and 1.7 ± 0.1 ppmv, respectively, depending on altitude.

For comparison with Odin data during the period mid-December to mid-February we evaluated ozone loss on the 50 ppbv and 25 ppbv N₂O isopleths in detail. In this case the ozone loss calculated from ground-based measurements amounts to 0.9 ± 0.2 ppmv and 0.5 ± 0.2 ppmv, respectively. These values compare well to the ozone depletion measured by Odin/SMR. The losses are in the same order as those measured in the cold Arctic winter of 1999/2000 in Kiruna by microwave radiometry and FTIR measurements.

Acknowledgements. We would like to thank the ECMWF and NILU for providing the PV and temperature data. The authors also thank the NCEP for providing temperature and pressure data used for the retrievals via the Goddard automailer system. The authors are grateful to the Swedish-French team of the Odin/SMR experiment for providing the vortex mean N₂O data. We also thank the referees for their helpful comments and suggestions. This work has partially been funded by the BMBF/DLR (contract FKZ 50EE0011). KIMRA was partly funded by the Swedish Knut and Alice Wallenberg foundation and the Swedish Research Council (NFR).

Edited by: J. Burrows

References

- Berg, H., Krupa, R., Hochschild, G., Kopp, G., and Kuntz, M.: Millimeter wave radiometer with adjustable internal calibration load for high resolution measurements of stratospheric constituents, *Proceedings of 2nd ESA Workshop on Millimetre Wave Technology and Applications: Antennas, Circuits and Systems*, pp. 372–377, Espoo, 1998.
- Feist, D. G.: The Bernese Atmospheric Multiple Catalog Access Tool (BEAMCAT): a tool for users of popular spectral line catalogs, *Journal of Quantitative Spectroscopy and Radiative Transfer*, 85, 57–97, 2004.
- Harris, N. R. P., Rex, M., Goutail, F., Knudsen, B. M., Manney, G. L., Müller, R., von der Gathen, P.: Comparison of empirically derived ozone losses in the Arctic vortex, *J. Geophys. Res.*, 107(D20), doi:10.1029/2001JD000482, 2002.
- Kanamitsu, M.: Description of the NMC global data assimilation and forecast system, *Weather and Forecasting*, 4, 335–342, 1989.
- Karhu, J. A., Taalas, P., Damski, J., Kaurola, J., Ginzburg, M., Villanueva, C. A., Piacentini, E., and Garcia, M.: Vertical distribution of ozone at Marambio, Antarctic Peninsula, during 1987–1999, *J. Geophys. Res.*, 108(D17), 4545, doi:10.1029/2003JD001435, 2003.
- Krupa, R., Hochschild, G., Fischer, H., and Wiesbeck, W.: Balanced Calibration Technique with an Internal Reference Load for Ground Based Millimeter Wave Radiometry, 1998 IEEE International Geoscience and Remote Sensing Symposium Proceedings; Vol. I; pp. 387–389; Seattle, July 1998.
- Kopp, G.: The Karlsruhe Millimeterwave Forward Model, *Proceedings of the second International Workshop on Millimeter and Sub-Millimeter Wave Radiative Transfer Modeling*, Osterholz-Scharmbeck, Germany, June 19–22, 2000, Logos Verlag Berlin, 2001, ISBN 3-89722-585-9, ISSN 1615-6862, 2001.
- Kopp, G., Berg, H., Blumenstock, Th., Fischer, H., Hase, F., Hochschild, G., Höpfner, M., Kouker, W., Reddman, Th., Ruhnke, R., Raffalski, U., and Kondo, Y.: Evolution of ozone and ozone related species over Kiruna during the THESEO 2000 – SOLVE campaign retrieved from ground-based millimeter wave and infrared observations, *J. Geophys. Res.*, 108(D5), 8308, doi:10.1029/2001JD001064, 2003.
- EORCU,
<http://www.ozone-sec.ch.cam.ac.uk/EORCU/Reports/wr0203.pdf>, European Ozone Research Coordinating Unit, 14 Union Road, Cambridge, CB2 1HE, UK, 2003.
- Nash, E. R., Newmann, P. A., Rosenfield, J. E., and Schoeberl, M. R.: An objective determination of the polar vortex using Ertel's potential vorticity, *J. Geophys. Res.*, 101, 9471–9478, 1996.
- Parrish, A., de Zafra, R. L., Solomon, P. M., and Barrett, J. W.: A ground-based technique for millimetre wave spectroscopic observations of stratospheric trace constituents, *Radio Science*, 23, 106–118, 1988.
- Urban, J., Lautié, N., Le Flochmoën, E., Murtagh, D., Ricaud, P., De La Noë, J., Dupuy, E., Drouin, A., El Amraoui, L., Eriksson, P., Frisk, U., Jiménez, C., Kyrölä, E., Llewellyn, E. J., Mégie, G., Nordh, L., and Olberg, M.: The Northern Hemisphere Stratospheric Vortex during the 2002–03 Winter: Subsidence, Chlorine Activation and Ozone Loss observed by the Odin Sub-Millimetre Radiometer, *Geophys. Res. Lett.*, 31, L07103, doi:10.1029/2003GL019089, 2004.
- Pickett, H. M., Poynter, R. L., Cohen, E. A., Delitsky, M. L., Pearson, J. C., and Müller, H. S. P.: Submillimeter, millimetre, and microwave spectral line catalog, *J. Quant. Spectrosc. Radiat. Transfer*, 60, 883–890, 1998.
- Raffalski, U., Berg, H., Hochschild, G., and Kopp, G.: Continuous ozone measurements over Kiruna during winter/spring 2002: A new millimeter wave radiometer operated at the Swedish Institute of Space Physics, Kiruna, Sweden, *Proceedings of the Sixth European Symposium on Stratospheric Ozone Research*, Gothenberg, pp. 369–372, 2002.
- Rodgers, C. D.: Retrieval of Atmospheric Temperature and Composition from Remote Measurements of Thermal Radiation, *Reviews of Geophysics and Space Physics*, 14, 609–624, 1976.
- Rothman, L. S., Rinsland, C. P., Goldman, A., Massie, S. T., Edwards, D. P., Flaud, E. J.-M., Perrin, A., Camy-Peyret, C., Dana, V., Mandin, J.-Y., Schroeder, J., McCann, A., Gamache, R. R., Wattson, R. B., Yoshino, K., Chance, K. V., Jucks, K. W., Brown, L. R., Nemtchinov, V., and Varanasi, P.: The HITRAN Molecular Spectroscopic Database and HAWKS (HITRAN Atmospheric Workstation): 1996 Edition, *J. Quant. Spectrosc. Radiat. Transfer*, 60, 5, 665–710, 1998.
- Tilmes, S., Müller, R., Groöf, J.-U., Höpfner, M., Toon, G. C., and Russell III, J. M.: Very early chlorine activation and ozone loss in the Arctic winter 2002–2003, *Geophys. Res. Lett.*, 30(23), 2201, doi:10.1029/2003GL018079, 2003.