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Chemical ozone loss in the Arctic vortex in the winter 1995–96: HALOE measurements in conjunction with other observations

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Abstract. Severe chemical ozone loss has been detected in the Arctic in the winter and spring of 1995–96 by a variety of methods. Extreme reductions in column ozone due to halogen catalysed chemistry were derived from measurements of the Halogen Occultation Experiment (HALOE) on board the Upper Atmosphere Research Satellite in the Arctic vortex. Here, we discuss further aspects of the HALOE observations in the Arctic over this period. Potential problems, both in the data themselves and in the methodology of the data analysis are considered and the reason for the differences between the Arctic ozone losses deduced from HALOE data version 17 and 18 is analysed. Moreover, it is shown that HALOE measurements in the Arctic in winter and spring 1995–96 compare well with observations by other ground-based and satellite instruments.

Key words. Chemical ozone loss (Arctic vortex 1995–96; halogen chemistry).

Introduction

Since 1989, chemical ozone loss in the Arctic vortex has been inferred from ozonesonde and aircraft measurements (e.g. Hofmann *et al.*, 1989; Proffitt *et al.*, 1990, 1993; Hofmann and Deshler, 1991; Koike *et al.*, 1991; Kyrö *et al.*, 1992; Browell *et al.*, 1993; von der Gathen *et al.*, 1995; Rex *et al.*, 1998a), albeit to a lesser extent than over Antarctica. More recent observations that also included satellite data (Larsen *et al.*, 1994; Manney *et al.*, 1994a, 1996a; Donovan *et al.*, 1995; Bojkov *et al.*, 1995; Wirth and Renger, 1996; Müller *et al.*, 1996; Goutail *et al.*, 1998a; Rex *et al.*, 1998b) have indicated particularly strong chemical ozone loss in the Arctic vortex for early 1993 and 1995.

Chemical ozone change in the Arctic is difficult to quantify, since dynamical processes cause substantial ozone variations (see e.g. Manney *et al.*, 1994a; von der Gathen *et al.*, 1995; Müller *et al.*, 1996, and references therein). Total ozone is a quantity that is especially problematical in this respect, as it is both subject to significant short term reductions due to high-pressure systems in the troposphere and characterised by a strong seasonal cycle in high latitudes. Climatological values for Northern Hemisphere high-latitude stations show an increase of total ozone between November and March by more than 100 DU (e.g., Hansen *et al.*, 1997).

In March 1996 and 1997, record low values of total ozone were measured in the Arctic vortex (Newman *et al.*, 1997); an observation which triggered further detailed investigations (e.g. GRL, 1997). While for both winters substantial chemical ozone depletion is inferred (e.g. Donovan *et al.*, 1996, 1997; Manney *et al.*, 1996b, 1997; Müller *et al.*, 1997a, b) there is consensus that direct dynamical effects contribute significantly to the unusually low ozone columns observed in March 1997 (Donovan *et al.*, 1997; Manney *et al.*, 1997). Consistent with these findings, the proxy ozone derived from HALOE tracer observations, which reflects the dynamical situation in the absence of chemistry, indicates much less descent (and thus lower total ozone) in the vortex in March 1997 than in March 1996 (Müller *et al.*, 1997a, b).

Here, we focus on the situation in the polar vortex in the Arctic winter and early spring of 1995–96. During this winter, temperatures were both extremely low and more persistently low than usual, if compared to long term records (Manney *et al.*, 1996b; Naujokat and

Pawson, 1996). This caused frequent polar stratospheric cloud (PSC) formation and subsequent chlorine activation of large vertical extent and uncharacteristically long duration (Hansen and Hoppe, 1996; Santee *et al.*, 1996a; Müller *et al.*, 1997a). Indeed, formation and sedimentation of ice particles as well as dehydration of stratospheric air were observed for the first time in the Arctic vortex in January 1996 (Vöemel *et al.*, l997; Hintsa *et al.*, 1998). Consistent with the continuing large-scale chlorine activation, anomalously low ozone levels were observed in the vortex throughout the lower stratosphere (Donovan *et al.*, 1996; Manney *et al.*, 1996b; Müller *et al.*, 1997a), large chemical ozone loss rates were derived from ozonesonde observations (Rex *et al.*, 1997), and unusually low ozone columns were observed by ground-based (Hansen *et al.*, 1997; Bojkov *et al.*, 1998; Goutail *et al.*, 1998b) and satellite (Müller *et al.*, 1997a) instruments in the Arctic. Further, total ozone column measurements of the GOME instrument (see later), indicate that the whole stratospheric vortex in March 1996 is characterised by extremely low ozone levels. Moreover, the expected enhancement of ground level UV-B radiation was observed (under clear sky conditions) at two midlatitude stations: Glasgow, 56°N (Moseley and MacKie, 1997) and Garmisch-Partenkirchen 47.5°N (Seckmeyer *et al.*, 1997); UV-B levels significantly higher than normal for the time of the year were measured during the first days of March 1996.

We concentrate here on the analysis of observations by the Halogen Occultation Experiment (HALOE) (Russell *et al.*, 1993) on the Upper Atmosphere Research Satellite (UARS) inside the Arctic vortex in winter and spring 1995–96. The relationship between an effectively inert trace substance (CH_4) and chemically more active compounds (HCl and O_3) in this data set have been used previously to deduce chlorine activation and chemical ozone loss (Müller *et al.*, 1996, 1997a). The purpose of this contribution is to provide further details of the observations in 1995–96 and to investigate questions that could not be fully discussed in Müller *et al.* (1997a) in particular the following questions:

1. May the O_3/CH_4 relation from the November 1995 HALOE observations be used as a reference for the conditions inside the incipient vortex ?
2. What is the reason for the differences between the Arctic ozone losses deduced from HALOE data version 17 and 18 (Müller *et al.*, 1996, 1997a) ?
3. How well do HALOE measurements compare with other observations in the Arctic in winter and spring 1995–96 ?

Ozone loss in the arctic vortex in 1995–96

HALOE observations

The HALOE instrument on UARS measures O_3, H_2O, NO_2, NO, HCl, HF, and CH_4, usually achieving 15 sunrise and 15 sunset (solar occultation) measurements each day along two approximately constant latitude

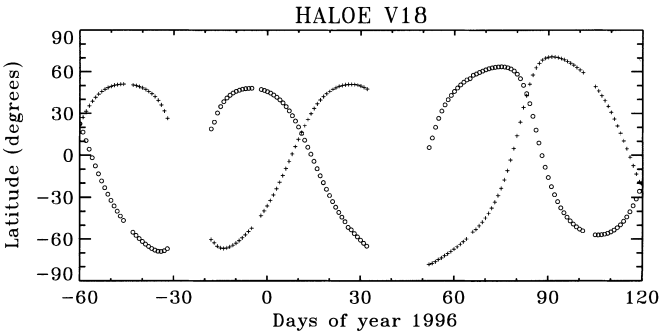

Fig. 1. Latitude coverage of HALOE observations from November 1, 1995 to April 30, 1996. *Circles* indicate sunrise observations, *plus signs* sunset observations

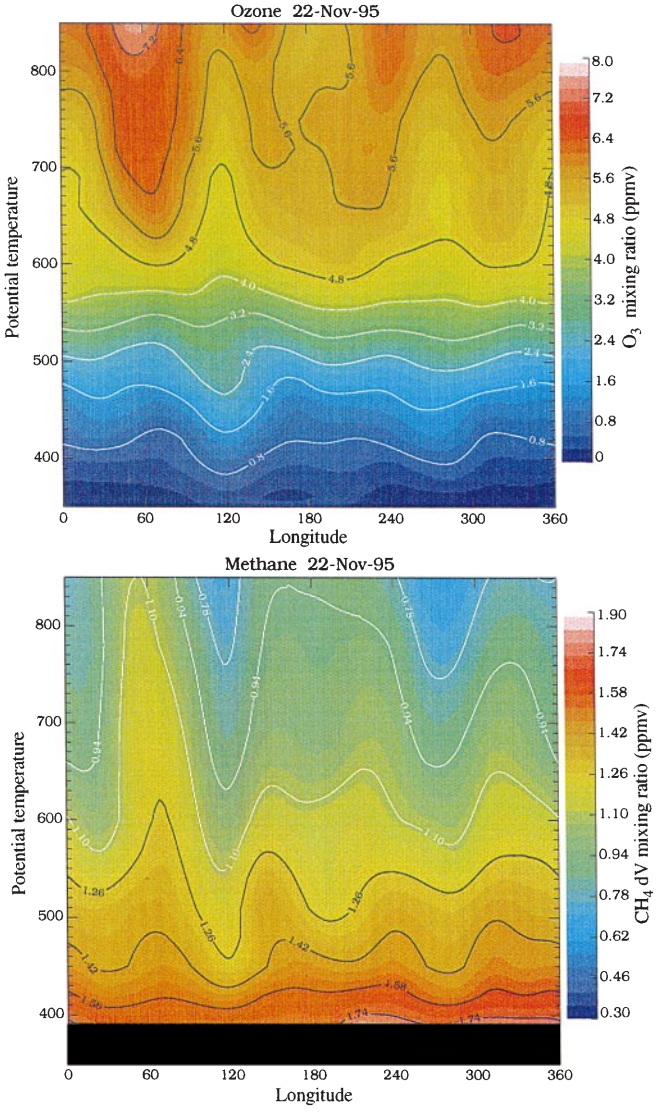

Fig. 2. Observations of the Halogen Occultation Experiment (HALOE) at sunset on November 22, 1995 (uarsday 1533), at 47°N. O_3 (*top panel*) and CH_4 (*bottom panel*) mixing ratios, measured by HALOE, are shown versus longitude and potential temperature (as a vertical coordinate). Vortex air is discernible over the whole altitude range at 118°E. The HALOE data were processed with software version 18

belts (Russell *et al.*, 1993). Because of the UARS orbit, the location of these latitude belts varies with season. In Fig. 1, the latitude coverage of HALOE in late 1995 and early 1996 is shown. Therefore, regular HALOE measurements inside the Arctic vortex are not available; nevertheless, vortex observations were obtained in January, March, and April 1996. Vortex air was furthermore observed in mid and late November 1995, shortly after the formation of the Arctic vortex. A coherent vortex is formed when the maximum average wind speed at ≈450 K along PV isolines reaches about 15 m s^{-1} (Nash *et al.*, 1996), which occurred in early November 1995 (Coy *et al.*, 1997).

Compared to mid-latitude air, vortex air is characterised by enhanced potential vorticity (PV) and, due to diabatic descent, substantially lower CH_4 mixing ratios. Especially towards the vortex edge, in a region of strong gradients of both PV and tracer mixing ratios, however, the resolution of PV computed from meteorological analyses is not sufficient to yield a good correlation with tracer observations, which show more fine scale structure (Fairlie *et al.*, 1997; Tuck and Proffitt, 1998). Here, we use the HALOE tracer (CH_4) observations to discriminate between mid-latitude and vortex air and discuss PV as a corroborating evidence. Four examples of occasions when HALOE sampled Arctic vortex air in the winter 1995–96 are shown for CH_4 and O_3 in Fig. 2 for November 22, 1995, at 47°N, in Fig. 3 for January 30, at 49°N, in Fig. 4 for March 16, at 64°N, and in Fig. 5 for April 1, 1996, at 70°N. CH_4 and O_3 mixing ratios are shown versus longitude and potential temperature (as the vertical coordinate). Owing to downward transport through diabatic descent in the vortex, CH_4 mixing ratios inside the vortex are lower on a specific isentropic surface than outside. On all four days, vortex air characterised by lower CH_4

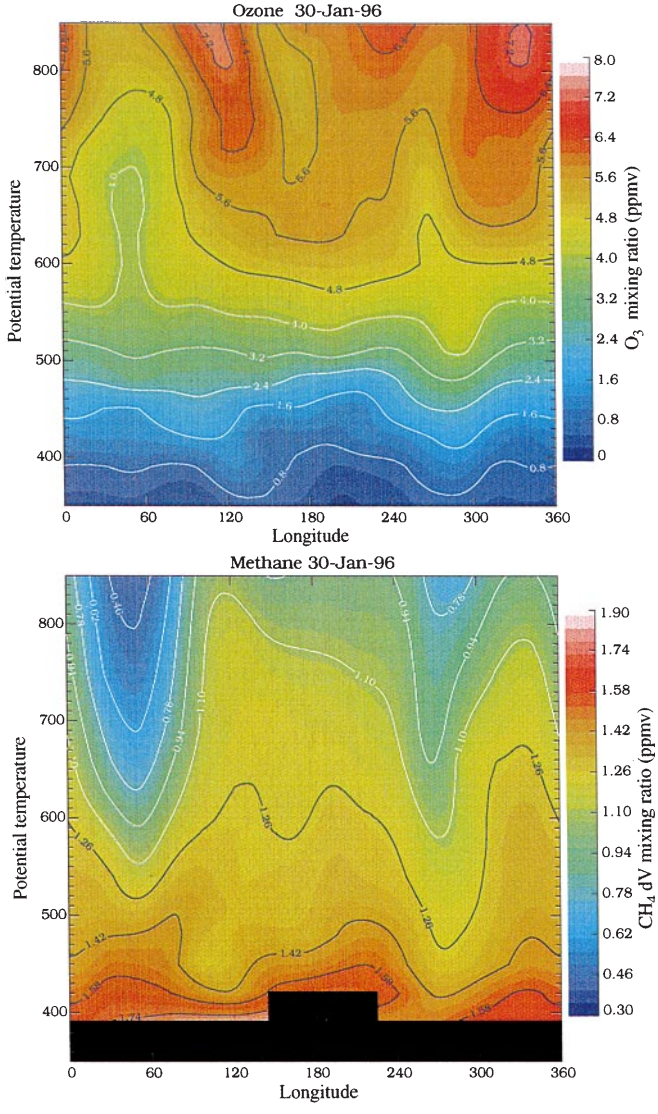

Fig. 3. As Fig. 2 but for sunset observations on January 30, 1996 (uarsday 1602), at 49°N. Vortex air is discernible over the whole altitude range at 267°E

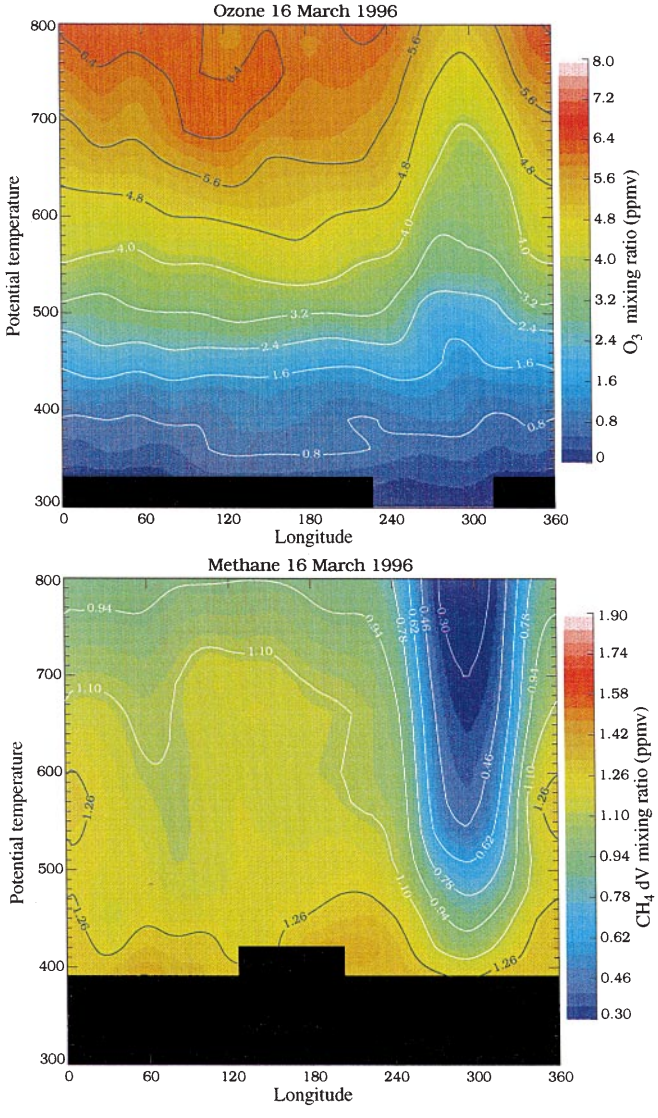

Fig. 4. As Fig. 2 but for sunrise observations on March 16, 1996 (uarsday 1648), at 64°N. Vortex air is discernible over the whole altitude range at 294°E

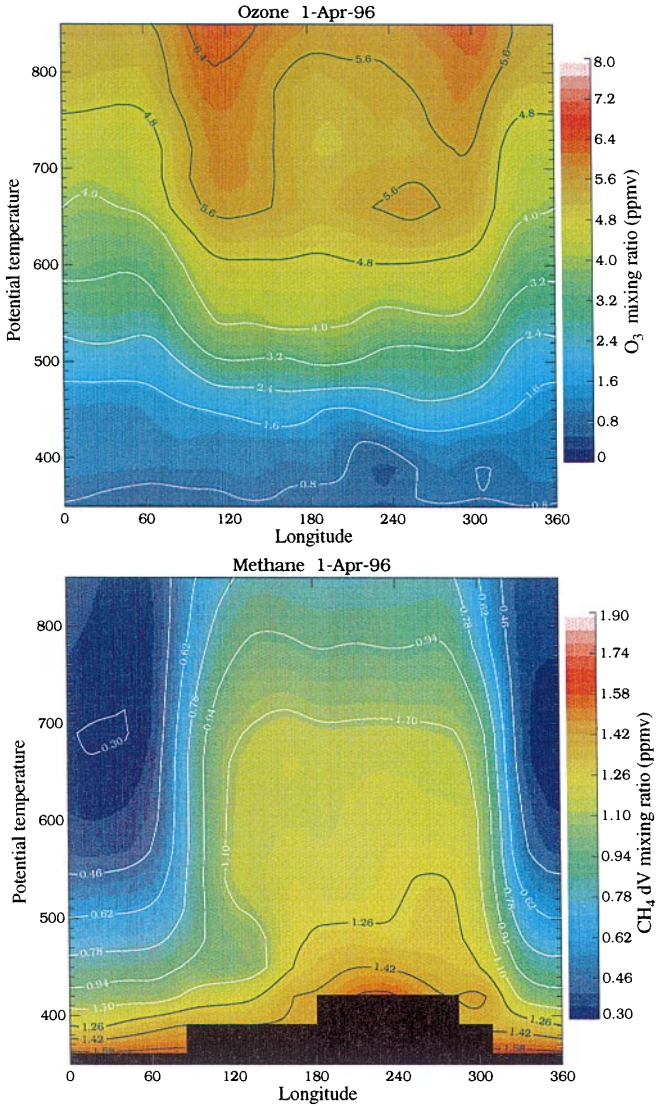

Fig. 5. As Fig. 2 but for sunset observations on April 1, 1996 (uarsday 1664), at 70°N. Vortex air is discernible over the whole altitude range at 8°W–64°E

mixing ratios is discernible (though not always over the whole altitude range), much as observed in previous years (Müller *et al.*, 1996). To use the relationship between an effectively inert trace substance and chemically more active compounds has been established as a method to discriminate chemical change from large variations due to dynamical processes (e.g., Proffitt *et al.*, 1993; Müller *et al.*, 1996). Compact relationships are expected for species with sufficiently long photochemical lifetimes (Plumb and Ko, 1992). In particular, neglecting mixing across the vortex boundary, an unchanging relationship between ozone and chemically inert tracers (such as N_2O, CH_4 or HF) is predicted for the air mass inside the polar vortex, if no chemical ozone loss would occur (Plumb and Ko, 1992). This is actually observed in HALOE measurements of the O_3/CH_4 relation between November 1995 and late January 1996 (Müller *et al.*, 1997a), i.e. over a time period when no substantial chemical ozone loss is expected due to the lack of sunlight. Although significant ozone loss *rates* (in ppb per hour of sunlight) have been observed for January 1996, the accumulated ozone loss then is still small (Rex *et al.*, 1997). Furthermore, the relationship of the inert compounds HF and CH_4 in the Arctic vortex in 1995–96 is conserved between November 1995 and April 1996 (Fig. 6). Any deviation from the initial O_3/CH_4 relation in the vortex over winter and spring is therefore an indication of chemical ozone change.

From this analysis, Müller *et al.* (1997a) concluded that local chemical O_3 destruction has led to reductions of about 60% over the height range of 400–480 K (with

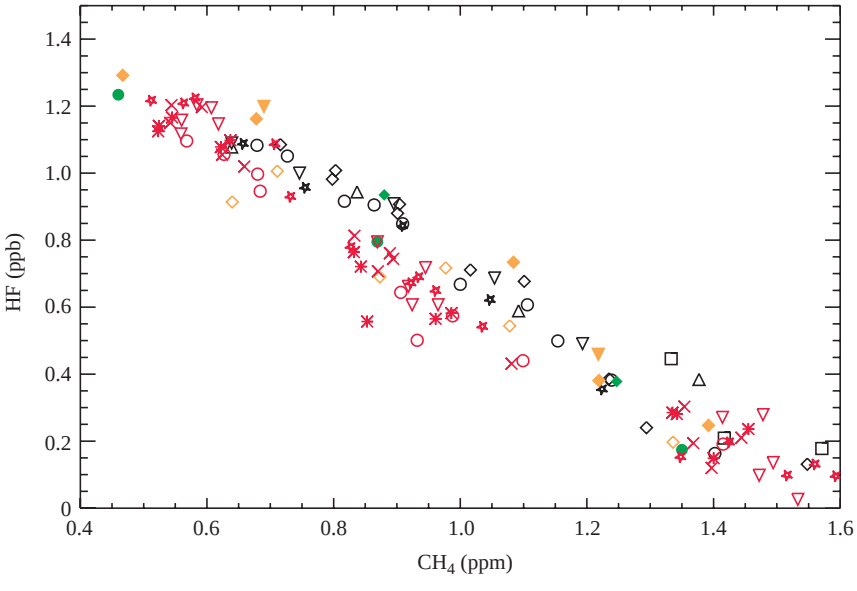

Fig. 6. The relation between CH_4 and HF volume mixing ratios in the Arctic vortex over the winter 1995–96. *Black* symbols indicate measurements in November 1995, *orange* symbols in January 1996, *green* symbols in February 1996, and *red* symbols in April (1–5) 1996. The conservation is valid for methane mixing ratios of 0.4–1.6 ppmv, corresponding to a potential temperature range (in March and April) of 350–600 K. (The *symbols* indicate individual days: □ 14-Nov-95; △ 15-Nov-95; ◇ 20-Nov-95; ○ 21-Nov-95; ▽ 22-Nov-95; ⋆ 23-Nov-95; ▼ 29-Jan-96; 30-Jan-96; ◇ 31-Jan-96; 1-Feb-96; ● 28-Feb-96; ○ 1-Apr-96; ∗ 2-Apr-96; × 3-Apr-96; 4-Apr-96; ⋆ 5-Apr-96)

Table 1. HALOE observations inside the polar vortex in March and April 1996. In the table location, potential vorticity (in potential vorticity units, at 550 K potential temperature), calculated ozone loss (for the altitude range 350–550 K; 150–25 hPa) and the measured ozone column (above 100 hPa) (both in Dobson units (DU)) of all HALOE observations in the vortex in March and April 1996 is listed. (The date is given as uarsday, e.g. uarsday 1635 is March 3, 1996 and uarsday 1664 is April 1, 1996)

Uarsday	Latitude	Longitude	Ozone loss	Observed column
1635.	51.9	24.3	135.5	196.6
1636.	53.7	23.4	126.2	183.5
1641.	60.5	281.3	131.7	184.7
1642.	61.4	280.0	139.6	183.8
1642.	61.4	304.1	126.5	214.2
1643.	62.2	278.5	172.2	170.9
1643.	62.1	302.6	167.4	189.2
1644.	62.8	277.0	159.8	173.7
1644.	62.7	301.1	182.2	183.6
1645.	63.3	275.4	161.4	184.4
1645.	63.2	299.5	163.5	182.2
1645.	63.2	323.6	182.2	189.5
1646.	63.5	273.7	158.2	185.6
1646.	63.5	297.8	163.9	185.4
1646.	63.5	321.9	188.8	191.5
1647.	63.6	272.0	166.6	177.2
1647.	63.6	296.1	153.9	181.7
1647.	63.6	320.2	172.9	187.1
1647.	63.6	344.3	157.4	216.3
1648.	63.5	294.3	171.6	182.1
1649.	63.0	292.5	152.9	190.6
1658.	63.5	13.6	120.9	179.9
1658.	63.3	37.6	103.2	181.7
1658.	63.0	61.6	106.3	192.7
1659.	66.9	350.2	109.6	197.0
1659.	66.7	14.1	108.6	183.6
1659.	66.5	38.1	111.7	187.6
1659.	66.4	62.1	120.8	193.6
1660.	68.9	350.8	100.3	201.3
1660.	68.8	14.8	109.4	188.1
1660.	68.7	38.7	122.6	184.3
1660.	68.6	62.7	111.8	194.1
1661.	70.1	351.4	124.4	197.4
1661.	70.0	15.4	137.2	186.9
1661.	70.0	39.4	139.0	188.0
1661.	69.9	63.3	121.2	198.5
1662.	70.7	351.9	123.1	195.4
1662.	70.6	15.9	124.2	188.3
1662.	70.6	39.8	136.4	193.2
1662.	70.6	63.8	113.6	206.1
1663.	70.8	352.1	138.8	191.3
1663.	70.8	16.1	123.8	190.4
1663.	70.8	40.1	124.8	196.5
1663.	70.8	64.1	117.8	207.2
1664.	70.5	352.0	136.7	192.5
1664.	70.5	16.1	133.4	191.3
1664.	70.6	40.1	114.1	193.9
1664.	70.6	64.1	125.5	197.3
1665.	70.0	351.7	135.6	196.2
1665.	70.0	15.8	134.8	192.6
1665.	70.1	39.8	143.3	193.5
1665.	70.1	63.8	128.7	192.3
1666.	69.2	351.2	139.8	194.1
1666.	69.3	15.3	143.8	191.0
1666.	69.3	39.3	133.8	192.8
1666.	69.4	63.3	122.9	194.2
1666.	69.4	87.4	134.6	200.4
1667.	68.2	350.5	123.5	206.1
1667.	68.3	14.5	120.9	191.6
1667.	68.4	38.6	133.9	189.0
1667.	68.4	62.6	136.4	193.1
1667.	68.5	86.7	129.5	203.2
1668.	67.0	349.6	135.9	199.3
1668.	67.1	13.6	113.0	195.8
1668.	67.2	37.7	133.9	190.5
1668.	67.3	61.8	119.9	194.3
1668.	67.4	85.8	116.2	206.3
1669.	65.8	12.6	119.8	185.0
1669.	65.9	36.7	123.4	199.3
1669.	65.9	60.7	112.5	203.1
1669.	66.0	84.8	123.4	198.1
1670.	64.3	35.5	130.3	197.0
1670.	64.4	59.6	136.1	214.5
1670.	64.6	83.6	128.1	212.4
1671.	62.7	34.2	134.9	197.7
1671.	62.8	58.3	144.1	229.3
1671.	62.9	82.4	146.9	237.4
1671.	63.0	106.4	125.2	230.3
1672.	60.8	32.8	125.5	197.2
1672.	60.9	56.9	140.6	233.2
1673.	58.8	31.3	133.8	198.6
1673.	58.9	55.4	140.8	235.0

peak losses of about 70%) between late January and early April. This is consistent with the results of Rex *et al.* (1997), who infer an ozone loss rate of 27–34 ppb per day in the lower stratosphere over this period from ozonesonde measurements that corresponds to an accumulated O_3 loss at about 470 K of $\approx$ 64%. Somewhat lower ozone loss rates (22 ppb per day for February 1996) are derived from MLS observations (Manney *et al.*, 1996b), the temporal behaviour of O_3, however is very similar.

The local chemical ozone loss in the vortex in the lower stratosphere that is reflected in a change in the relation of O_3 and CH_4 mixing ratios over winter and early spring, may be integrated to calculate the chemical change in column ozone: We use the "early vortex" O_3/CH_4 relationship (Table 3) to compute a proxy for the "unperturbed" ozone (O_3), expected for March and April in the absence of chemical change. The separation between the observed and the proxy ozone profiles is a measure of the accumulated chemical ozone loss over winter and early spring (Müller *et al.*, 1996, 1997a). Therefore, vertically integrating over the difference between measured and proxy ozone, yields a diagnostic measure of the deficit in column ozone in March/April in the lower stratosphere (between about 150 and 25 hPa). Note that this calculation is not affected by complications of the seasonal change in total ozone (see earlier) as we focus here solely on the situation in March/April and use CH_4 as a reference frame, thereby taking the effect of diabatic descent in the polar winter latitudes into account. In Table 1, the ozone column loss for each individual HALOE profile in the vortex during March and April 1996 is listed. All vortex measurements show severe chemical ozone loss exceeding 100 DU, where typical values range between 120–160 DU. In Table 1

also noted are the column ozone (above 100 hPa) observed by HALOE as a reference and the PV at the 550 K isentropic level (calculated from UKMO meteorological analyses) as an approximate indication of the location of the observation relative to the vortex boundary. Moreover, column ozone and the PV at 550 K is listed in Table 2 for the HALOE vortex observations in November 1995 and in January 1996 for completeness.

GOME observations

The Global Ozone Monitoring Experiment (GOME) is a new passive remote sensing instrument launched by ESA aboard the second European Research Satellite (ERS-2) on 21. April 1995 (Burrows *et al.*, 1993, and references therein). The total column amount is retrieved from GOME measurements in the wavelength range between 325 and 335 nm of the upwelling radiance from the atmosphere and the extra-terrestrial irradiance. The technique known as differential optical absorption spectroscopy (DOAS) is used (Eisinger *et al.*, 1997, and references therein).

GOME measurements of total ozone in high northern latitudes in late winter and early spring 1996 show very low values in March 1996 (Fig. 7) in accordance with total ozone derived from SBUV-2 (Newman *et al.*,

Table 2. Location and potential vorticity (in potential vorticity units, at 550 K potential temperature) of HALOE observations inside the vortex in November 1995 and January 1996. The measured ozone column above 100 hPa (in DU) is listed as well. (The date is given as uarsday e.g. uarsday 1525 is November 14, 1995 and uarsday 1601 is January 29, 1996). For January 1996 MLS measurements for the column ozone above 100 hPa are available for comparison. The mean MLS ozone column (Version 4) close to the HALOE measurement locations (within 10° latitude and 20° longitude) is 314 DU, 307 DU, and 299 DU for uarsdays 1601, 1602, and 1603 respectively (G. Manney, personal communication, 1998)

Uarsday	Latitude	Longitude	Potential vorticity	Observed O_3 column
1525	50.9	27.7	52.9	270.4
1526	51.0	338.2	50.8	202.8
1529	50.3	359.2	66.0	260.0
1531	49.3	94.2	55.7	244.6
1531	49.1	22.1	60.5	242.2
1532	48.4	117.8	60.7	268.5
1532	48.2	21.7	59.5	243.1
1533	47.3	117.6	67.1	265.2
1534	46.0	117.6	65.9	252.7
1601	50.1	267.3	67.7	325.1
1602	49.3	267.4	53.9	292.4
1603	48.2	267.7	59.8	256.3

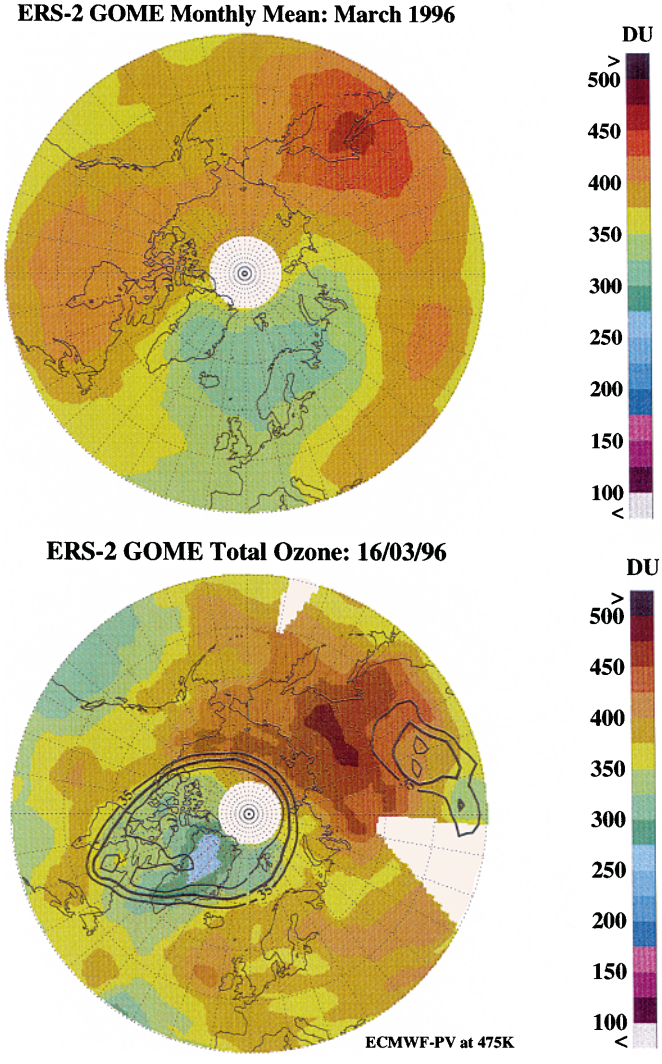

Fig. 7. GOME total ozone measurements in March 1996. *Top panel*: monthly mean total ozone in the Northern Hemisphere for March 1996. *Bottom panel*: total ozone on 16 March 1996. *Black lines* indicate the edge of the vortex; *contour lines* for 35 (*thick line with label*), 42 (*thick line*) and 48 (*thin line*) 10^{-6} K m^2 (kg s)$^{-1}$ are shown

Table 3. The early vortex CH_4/O_3 relation

Year	CH4 /O_3 relationship
1991	$O_3 = 19.52 \cdot CH_4^4 - 81.63 \cdot CH_4^3 + 119.84 \cdot CH_4^2 - 75.43 \cdot CH_4 + 21.38$
1992	$O_3 = 5.69 \cdot CH_4^3 - 19.25 \cdot CH_4^2 + 15.96 \cdot CH_4 + 0.75$
1993	$O_3 = 6.47 \cdot CH_4^3 - 22.17 \cdot CH_4^2 + 19.40 \cdot CH_4 - 0.33$
1994	$O_3 = 5.08 \cdot CH_4^3 - 17.31 \cdot CH_4^2 + 14.61 \cdot CH_4 + 0.60$
1995	$O_3 = 2.98 \cdot CH_4^3 - 11.20 \cdot CH_4^2 + 9.52 \cdot CH_4 + 2.14$

The empirical CH_4/O_3 relation (for O_3 and CH_4 in ppmv) from early vortex measurements for the five winters considered (based on version 18 HALOE data). The relations for 1991 and 1992 are valid for $CH_4 = 0.5 - 1.5$ ppmv, those for 1993 and 1994 for $CH_4 = 0.6 - 1.5$ ppmv, and the one for 1995 for $CH_4 = 0.5 - 1.6$ ppmv. (Updated version of Table 5 of Müller *et al.*, 1996)

1997). Throughout this region, monthly mean total ozone values between 300–475 DU are observed for March 1996, while the corresponding March monthly mean values between 1971 and 1980 range from 360–520 DU (Newman *et al.*, 1997). Typical long-term climatological values for March total ozone from the Dobson station Tromsø (69.4°N) are $\approx 440 \pm 50$ DU (Hansen *et al.*, 1997) and the March average over the polar region (from 63°N to 90°N) from BUV and Nimbus 7 TOMS total ozone measurements during the eighties (1971–80), is about 470 DU (Newman *et al.*, 1997).

The low ozone regions in Fig. 7 are related to the location of the polar vortex, which was generally centred off the pole towards northern Europe (Naujokat and Pawson, 1996; Manney *et al.*, 1996b) in early 1996. Indeed, the region of low total ozone in early 1996 is in general colocated with the position of the polar vortex. In Fig. 7 the ozone distribution on March 16, 1996 (see also Fig. 4) is shown; defining the edge of the vortex at $\approx$ 35 PVU (Rummukainen *et al.*, 1994; Rex *et al.*, 1997), which corresponds to the maximum gradient in potential vorticity, clearly, the low total ozone values occur within the confines of the polar vortex. This finding is consistent with MLS observations which show that the region of very low ozone mixing ratios in the lower stratosphere (at $\approx$ 465 K) in early 1996 is situated inside the polar vortex (Manney *et al.*, 1996b) and with HALOE measurements of the ozone vertical profile that show very low mixing ratios inside the polar vortex in March and early April 1996 (Müller *et al.*, 1997a). These observations are in accordance with the notion that substantial chemical ozone loss occurred inside the vortex in early 1996.

Discussion of uncertainties in the analysis of the HALOE observations

The O_3/CH_4 relation in the incipient vortex in 1995

The reliability of a method employing ozone-tracer relationships to deduce chemical ozone loss, depends on the quality of the initial ozone-tracer relation, which is used as a reference state for the conditions before the onset of chemical ozone destruction. Müller *et al.* (1997a) have used an empirical O_3 versus CH_4 relation

$$O_3 = 2.98(CH_4)^3 - 11.20(CH_4)^2 + 9.52(CH_4) + 2.14 \quad (1)$$

(for O_3 and CH_4 in ppmv and valid for 0.5 ppmv $< CH_4 <$ 1.6 ppmv), derived from HALOE vortex observations in late November 1995 as a reference. One needs to demonstrate that this relation in the vortex, shortly after its formation phase in early November (Coy *et al.*, 1997), is accurately known and is not influenced substantially by out of vortex air. The joint occurrence of low methane mixing ratios, indicating diabatic descent, and high PV, both characteristics of vortex air, is used as a criterion to discriminate vortex from out

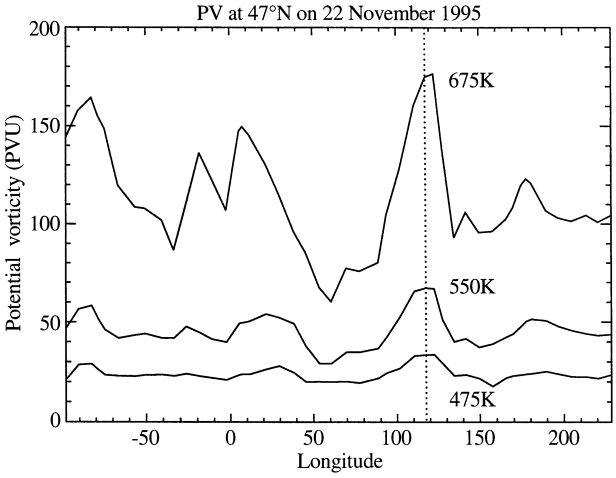

Fig. 9. Potential vorticity on November 22, 1995 at 47°N at three potential temperature surfaces against longitude. *Dotted line* indicates the location of the HALOE observation inside the vortex. Potential vorticity is derived from UK-Meteorological Office (UKMO) meteorological analyses

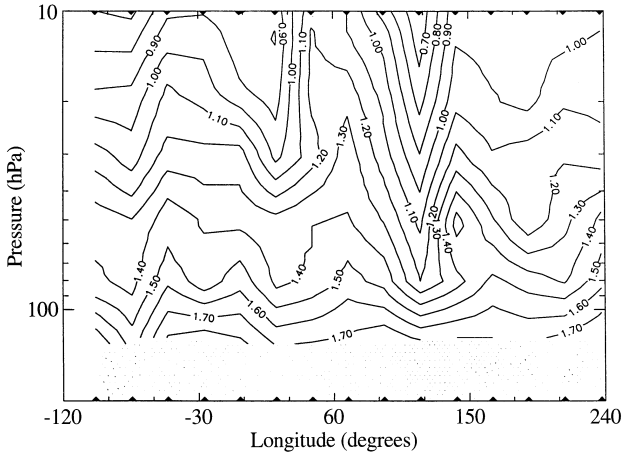

Fig. 8. Observations by HALOE at sunset on November 22, 1995 at 47°N. CH_4 mixing ratios (in ppmv) are shown against pressure and longitude. The *black triangles* indicate the longitude of the HALOE measurements

Table 4. Location and potential vorticity (in potential vorticity units, at 550 K potential temperature) of HALOE observations at 47°N on November 22, 1995 (uarsday 1533)

Latitude	Longitude	Potential vorticity
46.6	−98.4	46.1
46.7	−74.4	46.0
46.8	−50.5	44.1
46.8	−26.4	47.7
46.9	−2.4	39.9
47.0	21.6	54.0
47.1	45.6	37.9
47.2	69.6	34.8
47.3	93.6	42.2
47.3	117.6	67.1
47.4	141.6	41.6
47.5	165.6	42.3
47.6	189.6	50.6
47.7	213.6	44.0
47.7	237.6	41.0

of vortex observations. However, in Müller *et al.* (1996), where this issue has been discussed in some detail, only examples of an application of the methodology for the winters 1991–92 and 1992–93 are shown. Therefore, we present here a typical example of a HALOE observation inside the "early vortex" in 1995–96, namely on November 22, 1995 (uarsday 1533).

In Fig. 8, the HALOE observations of CH_4 mixing ratios on November 22, 1995 at 47°N are shown against pressure and longitude. Note that the longitude-height pattern is very similar to a presentation where potential temperature is used as a vertical coordinate (Fig. 2). The air inside the vortex at 118°E clearly stands out as a region of low CH_4 mixing ratios, which are caused by diabatic descent. Indeed, the potential vorticity at this location is enhanced throughout the lower stratosphere (Fig. 9; Table 4); an observation which corroborates the classification of this air mass as vortex air.

As observed in earlier winters (Müller *et al.*, 1996), the O_3/CH_4 relationship inside the vortex in mid and late November 1995 is clearly distinct from that observed outside (Fig. 10). Further, the outside vortex relations show much more variability than those inside the vortex and the ozone mixing ratios for a given CH_4 mixing ratio are larger outside than inside the vortex (Fig. 10). Moreover, the ozone vertical profiles measured inside the vortex in November 1995 are distinct from the profiles measured outside (Fig. 11); on constant potential temperature (Θ) surfaces, they generally show higher ozone mixing ratios inside the vortex than outside. This observation is consistent with the notion of an ozone increase on Θ-surfaces caused by the subsi-

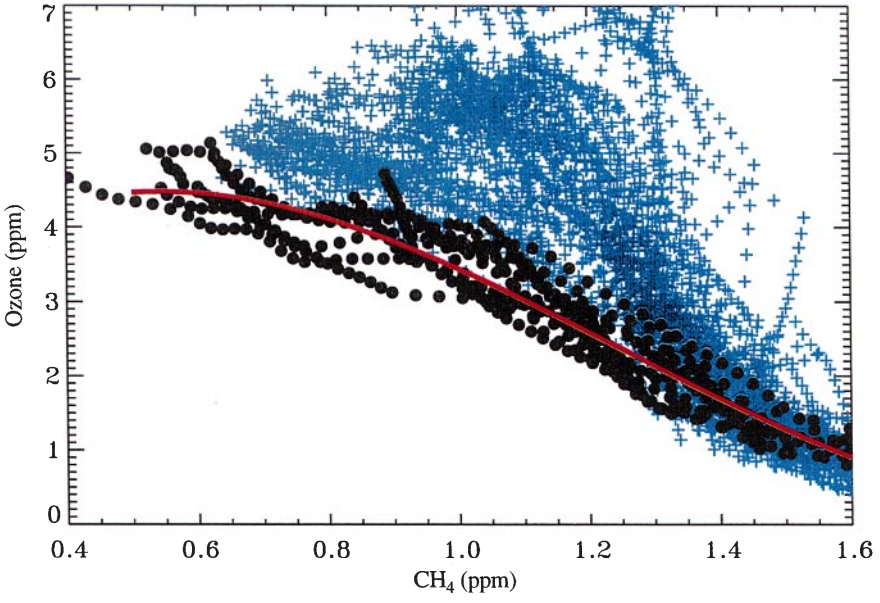

Fig. 10. HALOE measurements of the O_3/CH_4 relationship in early November 1995. The complete set of sunset data for all November observations listed in Table 2 is shown. Measurements inside the vortex are shown as *black circles,* outside measurements are shown as *blue plus* signs. Overplotted in *red* is the early vortex relation (Eq. 1)

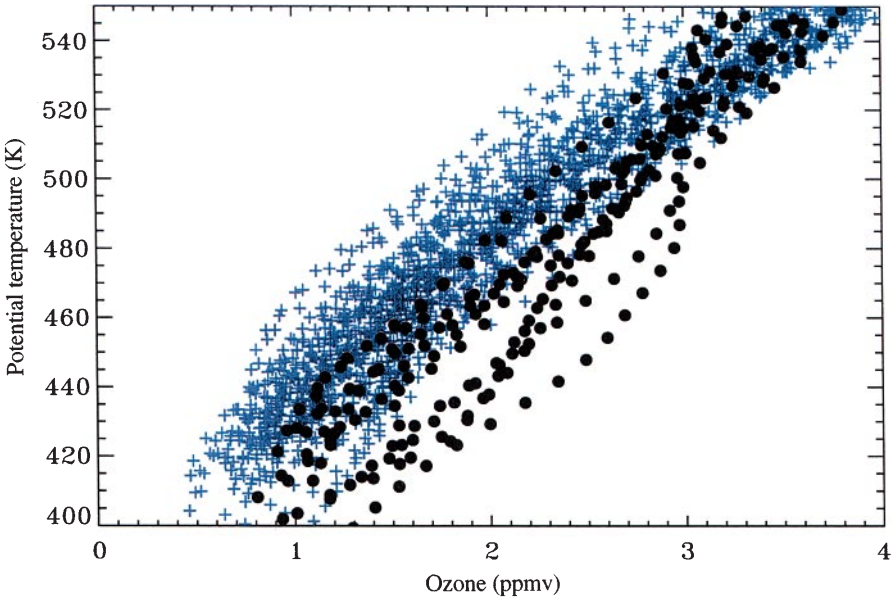

Fig. 11. As Fig. 10, but showing vertical profiles of O_3 mixing ratio (in ppmv) against potential temperature as the vertical coordinate. Measurements inside the vortex are shown as *black circles*, outside measurements are shown as *blue plus signs*

dence of air inside the vortex (as long as air is transported downwards from below the ozone mixing ratio maximum). Thus, the "early vortex" CH_4/O_3 relation (Eq. 1) may be considered as characteristic of the incipient vortex.

Differences in the ozone losses deduced from HALOE data versions 17 and 18

An examination of the O_3/CH_4 relationships between November/December and March/April for the first five winters of HALOE observations in the Arctic revealed reductions in ozone inside the vortex region, clearly manifested by significantly lower O_3 volume mixing ratios for equal values of the CH_4 mixing ratio (Müller *et al.*, 1996). This study was based on HALOE version 17 (V17) data. A reanalysis of the derived ozone loss for those winters based on version 18 (V18) data (Müller *et al.*, 1997a) yielded larger calculated ozone losses. The reason for the observed increase in the calculated ozone losses when V18 data are used instead of V17, is due to a combination of several smaller changes in the HALOE data, which sum up to the observed effect. Firstly, O_3 mixing ratios have increased from V17 to V18. Therefore, the empirical O_3/CH_4 relations derived from V18 data show larger ozone mixing ratios (Table 3). However, the increase is stronger at higher altitudes and for larger O_3 mixing ratios, so that the estimated ozone loss increases. (We have also verified this through sensitivity calculations.) Further, CH_4 mixing ratios at lower altitudes (below about 50–70 hPa) have decreased from V17 to V18, which also leads to an increase in the calculated ozone loss. Both effects together are responsible for the fact that the ozone loss calculated previously from V17 data was underestimated.

There is a remaining uncertainty in V18 data, however, regarding CH_4 measurements at sunrise (SR) at lower altitudes, below $\approx$ 50 hPa, namely that CH_4 mixing ratios are possibly underestimated below this

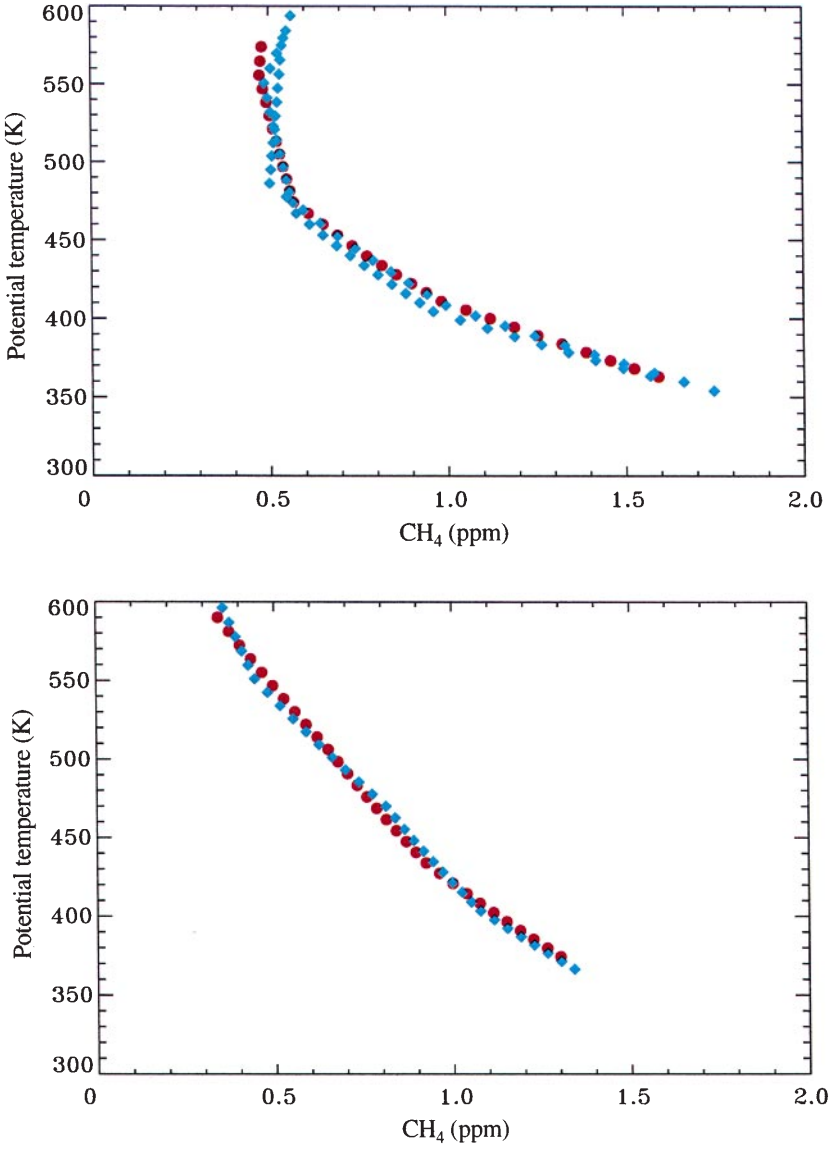

Fig. 12. Comparison of CH_4 vertical profiles observed at sunrise and sunset in the Arctic vortex. *Top panel:* observations on April 7, 1993 (uarsday 574); sunset observations (at 65.4°N, 11.2°E and 62.9°N, 105.8°E) are indicated by the *cyan diamonds*, sunrise observations (at 59.6°N, 353.2°E) are indicated by the *red circles*. *Bottom panel*: observations on April 2, 1994 (uarsday 934), sunset observations at 56.4°N, 288.9°E (*cyan diamonds*) and sunrise observations at 55.2°N, 282.6°E (*red circles*)

altitude by < 10–15%. Since the observations in the incipient vortex in November 1995 (and in all other winters as well), are taken at sunset (SS), this implies a possible overestimate of the calculated loss in the ozone column for SR observations. Sensitivity calculations, performed to derive an estimate of the consequences of this potential problem indicate that 35 DU is an upper limit for the possible overestimate. The SS observations in the vortex in early 1996 (26 March to 15 April), however, are not affected by this uncertainty in V18 data. Moreover, this question is related to an uncertainty in the accuracy of the HALOE sun tracking, a problem which is to a large extent caused by the stratospheric aerosol layer. In the polar vortex, however, the stratospheric aerosol is strongly reduced due to diabatic descent (Browell *et al.*, 1993; Wirth *et al.*, 1994). Indeed, the O_3/CH_4 relations inside the vortex derived on two occasions where SS and SR measurements overlap inside the Arctic vortex (Figs. 12, 13) show good agreement. Furthermore, preliminary V19 data available for SS/SR overlap, for which the problem with the HALOE sun tracking is expected to be solved, indicate that the CH_4/O_3 relation does not change substantially between V18 and V19 for SR vortex observations.

Comparison of HALOE measurements with other observations in the winter 1995–96

Although for the winter 1995-96 no other direct measurements of the CH_4/O_3 relation are available, the comparison of the HALOE derived CH_4/O_3 relation for the early winter 1991, where such measurements are available, showed good agreement (Müller *et al.*, 1996). Moreover, the comparison of HALOE ozone profiles within the Arctic vortex in 1995–96 with observations by

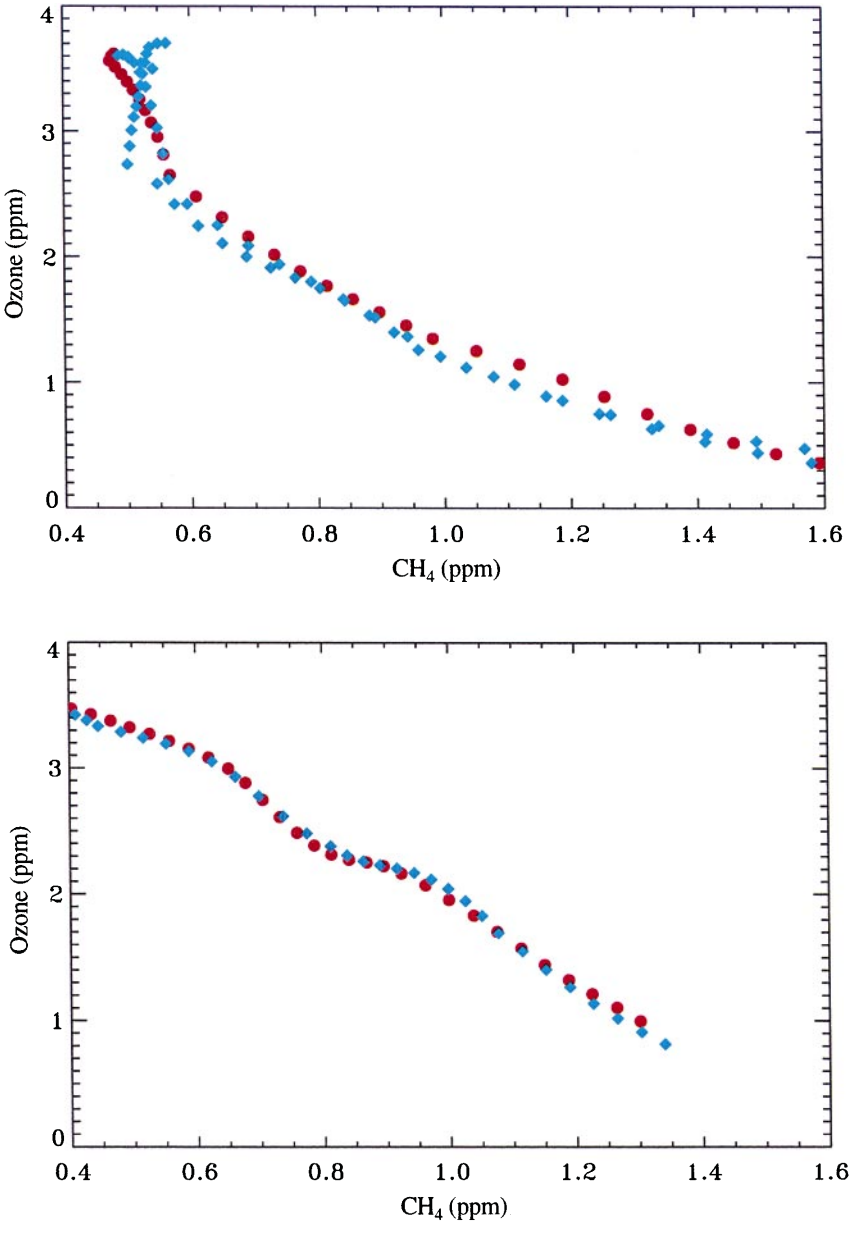

Fig. 13. As Fig. 12, but showing the relation of CH_4 versus O_3

the Microwave Limb Sounder (MLS) aboard UARS on March 3, 1996 (Fig. 14) and ground-based lidar observations (Donovan *et al.*, 1996) (Figs. 15, 16) both in March 1996 and November and December 1995 shows good agreement. Both this agreement as well as the relative homogeneity of the HALOE (Müller *et al.*, 1997a) and of the lidar ozone profiles (Donovan *et al.*, 1996) inside the vortex in March and April 1996, if potential temperature is used as the vertical coordinate, indicate that the ozone mixing ratios inside the Arctic polar vortex are fairly homogeneous at this time. The variation among individual ozone profiles observed by HALOE throughout the lower stratosphere in March and early April 1996 is typically ≈ 0.5 ppm. The clearly stronger variability seen in the total ozone observations (see also Fig. 7) are strongly correlated with lower stratospheric temperatures (Manney *et al.*, 1996b, Fig. 8). This is typically the case for short-term fluctuations in total ozone, which are caused dynamically by tropospheric disturbances (e.g. Dobson *et al.*, 1929; McKenna *et al.*, 1989).

A further important quantity which has an impact on the conclusions regarding chemical ozone loss, is the descent rate implied by the HALOE CH_4 observations. Unfortunately, to our knowledge, there are no studies of the descent rates in the Arctic vortex for the winter 1995–96. There are problems in comparing descent rates derived for different Arctic winters, which show a certain year to year variability (Müller *et al.*, 1996) and, even more, descent rates for different hemispheres (Manney *et al.*, 1994b; Rosenfield *et al.*, 1994; Lahoz *et al.*, 1996). Further, estimates of the descent rate in the vortex from the motion of tracer isopleths across isentropes constitute only a lower limit of the actual descent rate (Tuck and Proffitt, 1998). Figure 17 shows the methane vertical profiles measured by HALOE on April 7, 1993 at 64°N, which may be directly compared to Fig. 3 of Abrams *et al.* (1996) and which show very good agreement. Consequently, the implied lower limit of the descent rates (≈ 0.8 km/month at 20 km) are in accordance as well. Further, we show in Fig. 18 that the morphology of the CH_4 vertical profiles in the high latitudes in early April 1996 is not too distinct from what is observed by both ATMOS (Abrams *et al.*, 1996) and HALOE (Fig. 17) in 1993.

Moreover, HALOE HCl observations provide information on the extent of chlorine activation. Extremely low HCl mixing ratios were observed in early March 1996 (Müller *et al.*, 1997a), demonstrating the occurrence of chlorine activation in late winter 1996. This finding is consistent with the observation of a

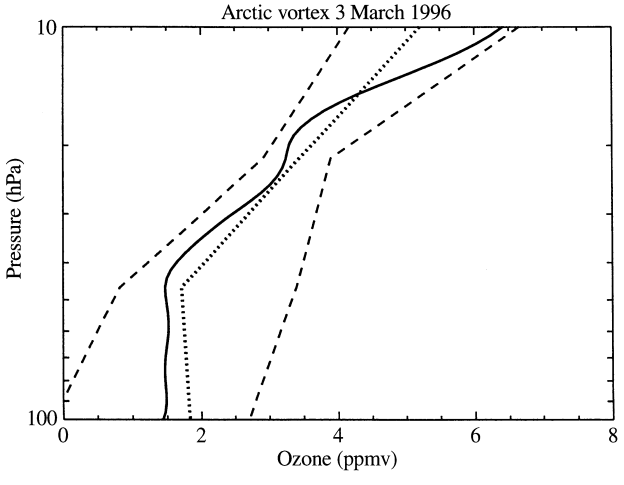

Fig. 14. The ozone profile observed by HALOE on March 3, 1996 at 51.9°N and 24.3°E inside the vortex (*solid line*) compared with 14 MLS (Version 4) profiles (G.Manney, personal communication 1997). The MLS data were selected to fulfil the following requirements: (1) latitude between 45° and 60°N: (2) longitude between 340° and 360° or 0° and 60°E: (3) UKMO PV at 465 K greater than 2.5 10^{-5} K m^2 (kg $s)^{-1}$, so that they are comparable with the HALOE measurements. The *two dashed lines* show the range of the MLS observations, their mean value is indicated by the *dotted line*. The comparable values for the column ozone above 100 hPa are 197 DU and 195 DU for HALOE and MLS respectively

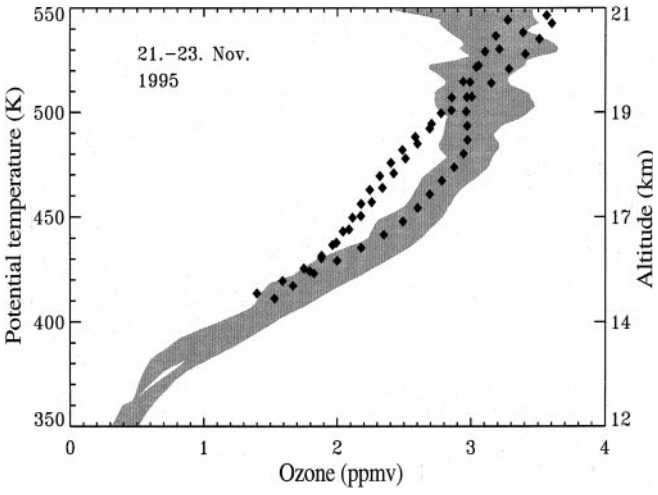

Fig. 15. The ozone profiles observed by HALOE in late November 1995 inside the vortex (*diamonds*) compared with ground-based lidar observations (at 80.0°N, 86.4°W) inside the vortex (*Donovan et al.*, 1996) between November 29, and December 29, 1995. The *grey area* indicates the variation of the lidar profiles by ± one standard deviation around the mean profile

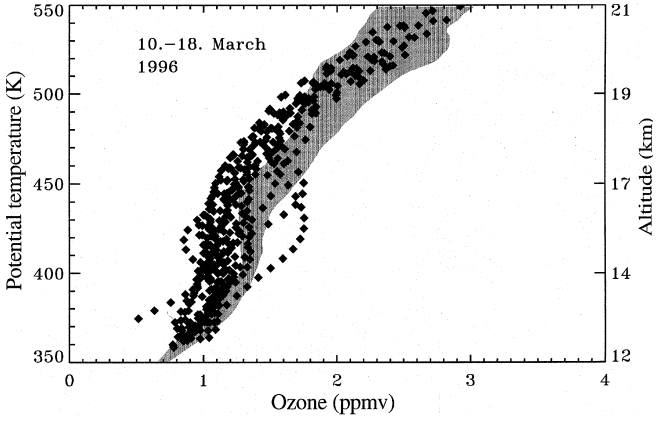

Fig. 16. As Fig. 15, but HALOE ozone profiles in March 1996 in the vortex (*diamonds*) compared with lidar observations (*Donovan et al.*, 1996) between March 10 and March 22, 1996

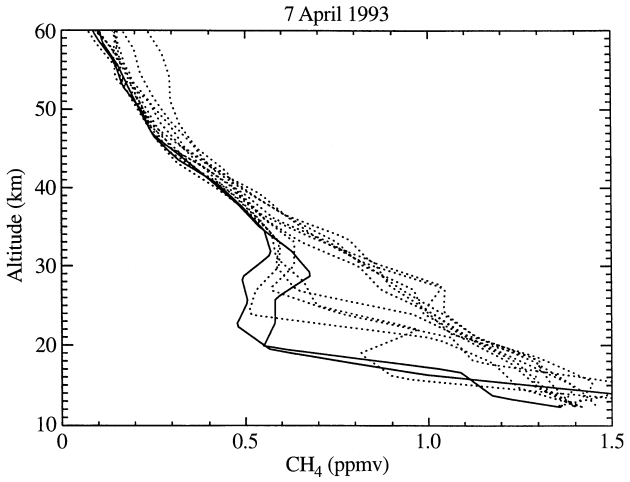

Fig. 17. HALOE measurements of CH_4 vertical profiles at 60°N on April 7, 1993. Measurements clearly inside the polar vortex are shown as *solid lines*, those outside and at the vortex edge by *dotted lines*

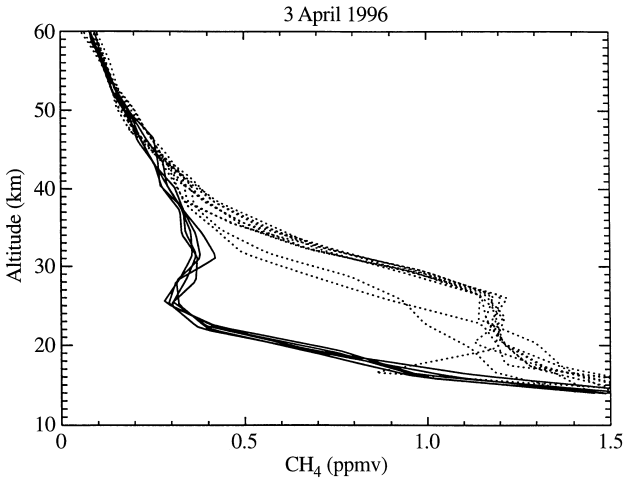

Fig. 18. As Fig. 17, but for HALOE measurements of CH_4 vertical profiles at 69°N on April 3, 1996

large scale, severe decrease in gas-phase HNO_3 by MLS (Santee *et al.*, 1996a) and the simultaneous observation of very large aerosol extinction by HALOE (Fig. 19) on March 3, 1996 (in high northern latitudes around 0°W), both indicative of extensive PSC formation. Because the deactivation of chlorine in the Arctic requires more than 2–3 weeks (Müller *et al.*, 1994; Douglass *et al.*, 1995; Santee *et al.*, 1996b) ozone loss must have continued during March 1996; a conclusion which is in accordance with the ozone loss rates deduced from the Match ozonesonde-campaign in 1995–96 (Rex *et al.*, 1997).

Conclusions

We have examined simultaneous HALOE observations of O_3 with a chemically inert tracer (CH_4), inside the Arctic vortex in the winter and spring of 1995–96 to detect chemical ozone depletion in the lower strato-

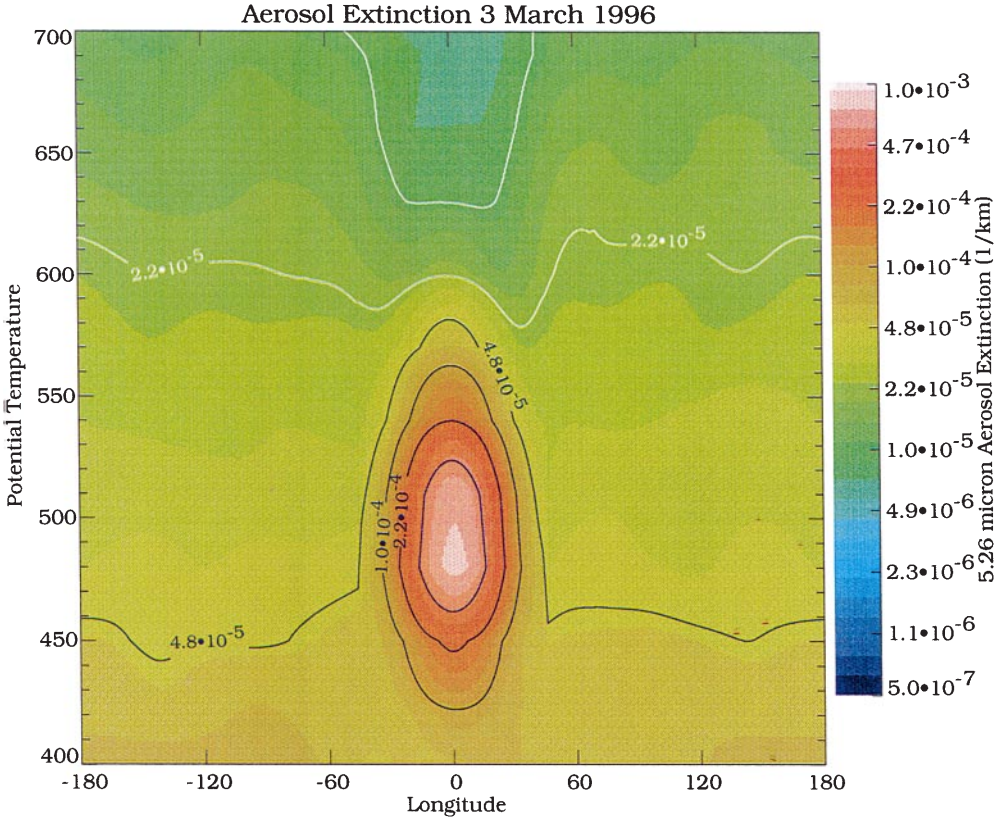

Fig. 19. HALOE measurements of aerosol extinction (5.26 μm) in km^{-1} at 52°N on March 3, 1996

sphere, despite strong variations caused by dynamical processes. Severe chemical ozone column loss of 120–160 Dobson units is derived for this period (see also, Müller *et al*., 1997a). Here, we have considered further aspects of the HALOE observations in the Arctic over this period. It was demonstrated that the November 1995 HALOE observations may be used as a reference for the chemically unperturbed conditions inside the incipient vortex. Further, it was shown that the reason for the observed increase in the calculated column ozone losses when V18 data are used instead of V17 (Müller *et al*., 1996, 1997a), could be understood as the effect of a combination of several smaller changes. Finally, it was concluded that there is good agreement between the relevant HALOE measurements in the Arctic in winter and spring 1995–96 with observations by other ground-based and satellite instruments.

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