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► **To cite this version:**

A. D. Robinson, G. A. Millard, F. Danis, M. Guirlet, N. R. P. Harris, et al.. Ozone loss derived from balloon-borne tracer measurements and the SLIMCAT CTM. *Atmospheric Chemistry and Physics Discussions*, 2004, 4 (6), pp.7089-7120. hal-00327920

HAL Id: hal-00327920

<https://hal.science/hal-00327920>

Submitted on 18 Jun 2008

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Ozone loss derived from balloon-borne tracer measurements and the SLIMCAT CTM

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Received: 16 August 2004 – Accepted: 24 September 2004 – Published: 3 November 2004

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Abstract

Balloon-borne measurements of CFC-11 (on flights of the DIRAC *in situ* gas chromatograph and the DESCARTES grab sampler), ClO and O₃ were made during the 1999/2000 winter as part of the SOLVE-THESEO 2000 campaign. Here we present the CFC-11 data from nine flights and compare them first with data from other instruments which flew during the campaign and then with the vertical distributions calculated by the SLIMCAT 3-D CTM. We calculate ozone loss inside the Arctic vortex between late January and early March using the relation between CFC-11 and O₃ measured on the flights, the peak ozone loss (1200 ppbv) occurs in the 440–470 K region in early March in reasonable agreement with other published empirical estimates. There is also a good agreement between ozone losses derived from three independent balloon tracer data sets used here. The magnitude and vertical distribution of the loss derived from the measurements is in good agreement with the loss calculated from SLIMCAT over Kiruna for the same days.

1. Introduction

Measurements of long-lived tracers such as N₂O, CH₄ and CFC-11 can be used to study a number of aspects of stratospheric transport and chemistry. In the Arctic region, these include the descent of air in the polar vortices (Schoeberl et al., 1990; Bauer et al., 1994) and mixing processes both at the vortex edge (Waugh et al., 1997; Pyle et al., 1994) and in the lowermost stratosphere (Bregman et al., 2000). Further, changes in the observed tight relations between long-lived tracers and ozone have been used to estimate the magnitude of chemical ozone loss in the Arctic vortex (Proffitt et al., 1990, 1993; Müller et al., 2001 and references therein). Other studies of ozone loss use different techniques (Rex et al., 1999; Goutail et al., 1999; Manney et al., 1997; Knudsen et al., 1998; Lucic et al., 1999 and references therein). Brief descriptions of these techniques, including the tracer relation approach, can be found in Harris

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et al. (2002), together with a comparison of some of their results. A discussion of the results obtained in this study with other estimates of ozone loss in the 1999/2000 winter is discussed in Sect. 5 Conclusions.

In situ measurements of tracers have often involved large grab samplers which can be flown only infrequently. In response to this limitation two new lightweight instruments have been developed recently by the University of Cambridge to measure CFCs: the in situ GC instrument DIRAC (Robinson et al., 2000) and the grab sampling instrument DESCARTES (Danis et al., 2000). These instruments are easy to operate and can be flown frequently on small, inexpensive balloons as part of a co-ordinated payload. The instruments were flown on several occasions into the vortex core during the SOLVE-THESEO 1999/2000 campaign (Newman et al., 2002) with other lightweight instruments including the SAOZ experiment (Pommereau and Piquard, 1994) or as part of the HALOZ project to measure ClO and related species (Vömel et al., 2001). In this paper we present results from nine balloon flights of these instruments made during SOLVE-THESEO 2000: four flights of the DIRAC in situ gas chromatograph and five flights of the DESCARTES grab-sampler.

In the first part of the paper, the instruments and the model are described. The next part presents the measured vertical profiles of CFC-11 and ozone (measured by electrochemical concentration cell (ECC) ozonesondes). The vertical profiles are then compared both with data from other instruments which flew during the campaign and to profiles calculated by the SLIMCAT 3-D chemical transport model (CTM), used to study ozone loss in previous Arctic winters (e.g. Guirlet et al., 2000). Measurements of ClO made using a resonance fluorescence instrument (Pierson et al., 1999) on some of the flights are also compared with the ClO profiles calculated in SLIMCAT. Changes in the CFC-11 and ozone correlations during the winter are then used to make estimates of the ozone loss that occurred in the vortex during 1999/2000. These estimates are compared to the chemical ozone loss calculated using the ozone and CFC-11 correlations above Kiruna for the same days, as calculated by SLIMCAT, in order to assess the representativeness of the balloon observations and to validate the

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model calculated ozone losses. In the final section we discuss the derivations of ozone loss in the vortex from balloon profiles of long-lived tracers and the consistency of the CFC-11, O₃, and ClO profiles between the measurements and SLIMCAT.

2. Measurements

5 The measurements featured in this study were made by three instruments flown on balloons from Kiruna during the SOLVE-THESEO 2000 campaign. The balloon flight details are given in Table 1. The instruments are now briefly described.

2.1. Halocarbons

10 DIRAC (Determination In situ by Rapid Analytical Chromatography) is a lightweight gas chromatograph for use on stratospheric balloons developed by the University of Cambridge. The operation and performance of DIRAC are described by Robinson et al. (2000). The instrument weighed 21 or 24 kg, depending on configuration (although recent instruments with superior performance now weigh <10 kg). A range of halocarbons is detected and can be measured quantitatively. The measurement with the high-
15 est precision is CFC-11, measured with a time resolution of about 2 min. DIRAC uses a Carboxen adsorbent to pre-concentrate samples of known volume before injection onto a separation column and electron capture detector. The accuracy for CFC-11 based on the uncertainty of the calibration gas used during the campaign is estimated to be 2.5%, this calibration gas is linked to NOAA-CMDL working standards by comparison
20 to an Aculife treated cylinder containing Niwot Ridge air (calibrated by NOAA-CMDL in July/August 2000).

25 During SOLVE-THESEO 2000, the in-flight precision using the onboard calibration gas was 0.5 pg of CFC-11, taken as the standard deviation of the linear fit (peak area versus mass of CFC-11 trapped) to all in-flight calibrations made during the campaign (Fig. 1). On this basis, the precision is dependent on sample size so, for samples

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taken at low altitudes (below 400 K) where CFC-11 mixing ratios are high and sample volumes are large, the precision is typically 5–8 pptv (corresponding to a typical sample mass in the range 10 to 20 pg of CFC-11). However, at higher altitudes (above 400 K), where both the sample volumes and CFC-11 mixing ratios are lower, the typical precision is in the range 10–20 pptv (corresponding to a typical sample mass in the range 0.2 to 7 pg of CFC-11). The calibration data collected during these balloon flights were based on a typical calibration flow of 22 ± 7 sccm. However, sample flow rates during this campaign were in the range 1.5 to 22 sccm with the lowest flow being at the highest altitude. Laboratory response curves for CFC-11 using calibration gas over a range of flows indicate an additional uncertainty in the measurement of 0.2 pg of CFC-11.

The sample flow rates at high altitude were up to 3 times lower than in previous DIRAC flights as a result of a new switching valve with an unexpectedly high flow resistance which could not be changed during the campaign. The low flow rates had a number of consequences resulting in higher uncertainties above about 400 to 420 K. For example, corrections to account for the time delay between sample air entering the inlet and arriving at the adsorbent were larger than usual, being in the range 20 to 60 s. Also, low pressure tests with DIRAC-B show that at low sample flows a small but significant fraction of air from lower altitude remains in the dead volume of the sample pathway (DIRAC-A made the SOLVE-THESEO 2000 measurements but was lost on a later flight in the campaign). This impacts on the measurements above 400 K, causing a small systematic bias toward higher reported CFC-11 values (on ascent). Measurements reported here are not corrected for this effect as any such correction, based on laboratory tests, would be of significant uncertainty in itself. The implementation of a new pumping system has now minimised this problem on the instrument. In the plots which compare DIRAC and DESCARTES CFC-11 data (presented later) we assume that this caused a constant offset as there is no evidence to suggest that the magnitude of this effect changed during SOLVE-THESEO 2000. Indeed, the good agreement in the ozone loss estimates with data from other instruments (presented later) indicates that there was no significant change in the relationship between the

DIRAC and DESCARTES CFC-11 measurements during the campaign.

The measurements are analysed here from four flights of DIRAC during SOLVE-THESEO 2000 (Table 1). Three of these (19 January, 27 January and 8 March 2000) were made as part of the HALOZ project (Vömel et al., 2001) and the other was part of the OMS in situ flight (5 March 2000). All DIRAC data presented here were taken during the balloon ascent. A fifth DIRAC flight was made in late March, but unfortunately the balloon and payload flew astray over Finland and were not recovered. We have no data for this flight as DIRAC was flying without telemetry.

DESCARTES (Détermination et Séparation par Chromatographie lors de l'Analyse des Résultats des Traceurs Echantillonnés dans la Stratosphère) is a lightweight (17 kg) balloon-borne grab sampling instrument developed by the University of Cambridge in which known volumes of stratospheric air are trapped on sample tubes containing Carboxen. A range of trapped halocarbons is subsequently measured in the laboratory using a gas chromatograph and electron capture detector. For a detailed discussion see Danis et al. (2000). The precision (1 sigma) of the CFC-11 measurements made by DESCARTES during this campaign was typically <15 pptv for samples taken below 400 K and typically 5–20 pptv for samples above 400 K. The accuracy is estimated to be about 4% for CFC-11 based on the uncertainty of the calibration gas. All measurements from DESCARTES presented here were analysed by IRF, Kiruna, enabling data to be obtained soon after instrument recovery. A total of 10 DESCARTES flights (including vortex edge flights) were made during the 1999/2000 winter. In this paper, measurements are analysed from five vortex core flights of the instrument, made with a range of host payloads (Table 1). As with DIRAC, all the DESCARTES CFC-11 data presented here were taken during the balloon ascent.

2.2. ClO

The ClO measurements used in this paper were made using an instrument based on the technique of chemical conversion/resonance fluorescence (Pierson et al., 1999). The balloon flights were made in the HALOZ project whose aim was to investigate

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ozone loss in the Arctic winter 1999/2000 (Vömel et al., 2001). A total of five balloon flights of the CIO instrument were made during the 1999/2000 winter. In this paper, measurements are presented from the HALOZ flight on 8 March 2000 (Table 1).

2.3. Ozone

- 5 The ozonesonde observations for all the flights used in this study (including those associated with the OMS in situ and Bonbon flights) were made by ECC sondes operated by the University of Wyoming and Esrange.

3. SLIMCAT 3 dimensional chemical transport model

10 The SLIMCAT 3-D CTM (Chipperfield et al., 1996; Chipperfield, 1999; Guirlet et al., 2000) is an off-line model driven here with daily U.K. Met. Office winds and temperatures (Swinbank and O'Neill, 1994) on 12 isentropic potential temperature surfaces from 335 K to 2700 K at a resolution of 3.75° latitude by 3.75° longitude. The model uses the MIDRAD radiation scheme (Shine, 1987) to calculate the cross-isentropic flow and a second-order moments scheme (Prather, 1986) for tracer advection. The
15 Prather scheme has low numerical diffusion and maintains strong gradients in tracer distribution well, an important consideration in polar winter time simulations. A detailed stratospheric chemistry scheme is included in the model with 49 chemical species integrated over a 15 min timestep. In general, photochemical data are taken from DeMore et al. (1997) and photolysis rates are calculated using a scheme based on Lary and
20 Pyle (1991). The model also contains an equilibrium treatment of reactions on liquid sulphuric acid aerosols and on solid nitric acid trihydrate (NAT) and ice particles. This particular version of the model has been used in studies of Arctic ozone loss in three other cold winters in the late 1990s (Guirlet et al., 2000) and in a detailed comparison to ozone loss rates derived by the Match technique (Millard et al., in preparation,

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2004¹). The winter 1999/2000 simulation was initialised on 30 November 1999 from a low resolution multiannual simulation provided by Chipperfield (1999) with a total chlorine loading of 3.6 ppbv and a total bromine loading of 20 pptv. The winter simulation was run until May 2000 with daily output of the chemical fields.

5 4. Results

In the first part of this section the CFC-11 vertical profiles from DIRAC and DESCARTES are compared with CFC-11 measurements from two other instruments: the Bonbon whole air cryo-sampler (Schmidt et al., 1991) which flew as part of the TRIPLE payload; and the LACE instrument (Ray et al., 1999; Moore et al., 2003) which
10 was part of the NASA-SOLVE OMS in situ payload. The agreement between CFC-11 measured by DIRAC and DESCARTES is also examined for samples above the 360 K potential temperature level (approximating to the lower boundary of the vortex). A change in the relation between CFC-11 and ozone over the winter below this level is unlikely to be the result of chemical ozone loss. A good agreement between the
15 instruments, above the 360 K potential temperature level, would justify merging the two data sets in order to improve the estimate of ozone loss within the vortex core.

In the second part of this section the measurements from DIRAC and DESCARTES as well as the ClO and O₃ measurements are compared with the output of the SLIMCAT model to assess its performance. This is achieved by making direct comparisons of the
20 measured vertical profiles of CFC-11, O₃ and ClO with those from the model.

In the third and last part of this section the relation between measured CFC-11 and ozone is compared with the equivalent relation from the model. We also compare the measured CFC-11 ozone relations from our instruments with those from Bonbon

¹Millard, G. A., Pyle, J. A., Chipperfield, M. P., Shreibel, M., Frieler, K., Rex, M., and von der Gathen, P.: Capturing lagrangian 'Match' diagnosed ozone loss rates in the Arctic and Antarctic polar vortices, in preparation, 2004.

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and LACE. Estimates of ozone loss in the vortex can be derived from the change in the measured $O_3/CFC-11$ relation through the winter using the approach of Proffitt et al. (1990, 1993) and Müller et al. (2001 and references therein). A tight correlation between two tracers, such as exists between O_3 and CFC-11 at the start of the winter, should be maintained if it is assumed that there is no mixing or chemistry operating on the tracers. Changes in the correlation indicate that this assumption is not valid. Within the polar vortex, mixing is usually small and any change in the correlation is most likely due to chemistry. For the specific case of O_3 and CFC-11, the chemistry of the latter is very slow with small seasonal changes and the effect of mixing would be small since the relationship between O_3 and CFC-11 was effectively linear in this winter (Müller et al., 2003) so that changes in the expected correlation can be directly attributed to ozone destruction. Later in this section we use the measurements to estimate ozone loss by this method. Finally, the ozone loss estimates based on the measurements are compared to the losses derived using exactly the same methodology from the change in the $O_3/CFC-11$ correlations in the model.

4.1. Measurement comparison

Here we first compare our CFC-11 measurements with those from the Bonbon and LACE instruments and relate them to the vortex situation. Figure 2 shows the vertical profiles of CFC-11 and O_3 from the four DIRAC flights and the five DESCARTES flights together with CFC-11 and O_3 data from the LACE flights of 19 November and 5 March and the Bonbon flights of 27 January and 1 March. Potential vorticity (PV) maps from UKMO and the T21 MIMOSA model (Hauchecorne et al., 2002) indicate that the polar vortex between 19 November (LACE flight) and 3 and 15 December (DESCARTES flights) was well formed above 450 K. Below 450 K it strengthened significantly but is still best classified as a weak vortex. This can explain much of the structure apparent in both CFC-11 and O_3 on these flights particularly below 450 K. On the DESCARTES flight of 3 December (Fig. 2b) there was an elongated area of lower PV air above Kiruna at 475 K which could explain the higher CFC-11 values found at this level in the data.

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Maps of PV from MIMOSA show that all other flights featured here were essentially inside the Arctic vortex over the altitude range sampled (taking the vortex edge to be the zone of maximum PV gradient) and the CFC-11 profiles show the expected rapid decrease with altitude. The change in the profiles suggests that considerable descent across potential temperature surfaces occurred throughout the winter, particularly between early December and late January. On 19 January a notch in the DIRAC CFC-11 profile at 360 K is anticorrelated with the O₃ profile which suggests a dynamical feature; air at this potential temperature level is likely to be at or below the base of the vortex where such dynamical features are to be expected.

On the 27 January DIRAC flew from the same launch site about 2 hours after the Bonbon whole air cryo-sampler. Assuming that the two instruments flew through essentially the same air mass this provided an opportunity for comparison. CFC-11 measurements from DIRAC and Bonbon agree well within uncertainty limits up to about 400 K. Above this level the DIRAC measurements are between 20–30 pptv higher than those of Bonbon although the uncertainty of the DIRAC measurements is much higher in this region. It is likely that this difference between the two instruments is due to the lower than expected sample flow through the DIRAC inlet system, as discussed earlier. On 5 March DIRAC was deployed on the OMS in situ gondola which included the LACE instrument. Figure 2 shows that the DIRAC CFC-11 data compare well with LACE up to about 400 K. Above this level the DIRAC CFC-11 values are again up to 30 pptv higher than those from LACE. The evidence from the comparison with Bonbon and LACE indicates that DIRAC appears to systematically over-estimate CFC-11 above the 400 K level. However, such a systematic bias in the CFC-11 data does not affect the use of the tracer O₃ technique to estimate loss of ozone throughout the winter provided the bias is consistent over the campaign period (we later report good agreement in ozone loss estimates from Bonbon/LACE).

There were two occasions during SOLVE-THESEO 2000 on which DESCARTES flew within 24 h of Bonbon. On 27 January, Bonbon flew first with DESCARTES following 24 h later and on 1 March Bonbon flew just 5 h earlier than DESCARTES. All four

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of these flights were into the vortex core. Examination of the vertical profiles of CFC-11 for these flights reveals that the DESCARTES measurements are systematically higher than those from Bonbon at any given altitude. At low altitude (300 to 350 K) the DESCARTES mixing ratios are typically 20 pptv higher than those from Bonbon and at higher altitude (>400 K) mixing ratios are typically 10–20 pptv higher than Bonbon. Examination of the CFC-11 O₃ relation for the comparison flights shows that for any given O₃ mixing ratio the corresponding DESCARTES CFC-11 value is higher than that from Bonbon. However, this difference is within uncertainty limits for high O₃ mixing ratios (>2000 ppbv). As with the DIRAC CFC-11 comparison with Bonbon and LACE we argue that the systematic bias in the DESCARTES CFC-11 data does not affect the use of the tracer O₃ technique to estimate loss of ozone throughout the winter provided the bias is consistent over the campaign period.

To assess the relative agreement between vortex core measurements of CFC-11 from DIRAC and DESCARTES we examine the difference in the data for the late January and early March flights when the instruments flew within one or two days of each other (Fig. 3). The differences are based on linear interpolation of the DIRAC CFC-11 data to the potential temperature levels of the DESCARTES data. Only those data collected above 360 K are used in the assessment which approximates to the lowermost extent of the vortex circulation system. For both the January and March DIRAC and DESCARTES flight pairs there were only small differences beyond measurement uncertainties between the data points and the line $y=x$ (a 1:1 agreement) and so a merged data set was produced, based on the DIRAC and DESCARTES measurements, to estimate ozone loss using the tracer ozone relation technique.

4.2. Measurement and model agreement

Here we compare our measurements of CFC-11, O₃ and ClO directly with the SLIMCAT model output data. Comparisons between the measured CFC-11 profiles and those calculated by the SLIMCAT 3-D CTM for the same location and day are shown in Fig. 4. They show generally good agreement for CFC-11 values greater than 50 pptv (below

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about 450 K). There is also good agreement between O_3 measured by the ECC sondes and the O_3 calculated in SLIMCAT although the model generally underestimates the amount of ozone during March by about 200 ppbv. On certain flights, structure in the CFC-11 measurements, whilst not picked up by the model at this resolution, is seen anticorrelated in the ozonesonde data (e.g. 3 December 1999, 27 January and 8 March 2000) confirming that the instruments are able to capture these dynamical features.

The overall good agreement between the CFC-11 measurements and the model gives confidence in the modelled available chlorine Cl_y , a principal influence on the model ClO. To compare ClO observations (8 March) with model calculations, the output from the SLIMCAT simulation of the 1999–2000 northern hemisphere winter (stored daily at 12:00 UT) was used to initialise a set of stacked photochemical box models. The stacked box models were then integrated over a diurnal cycle for comparison with the observations, following the method used by Solomon et al. (2000). The stacked box models used an identical chemistry scheme to the full 3-D model except that all 49 chemical species in the model were integrated separately with a 1 min time step; no family or photochemical steady-state assumptions were made. To avoid prohibitive computational costs SLIMCAT uses certain family approximations and assumes photochemical steady-state partitioning for the ClO_x family (Cl, ClO and Cl_2O_2), so it cannot itself provide ClO predictions accurate enough for comparison with the data.

Figure 5 shows a comparison between the ClO measurements and results from the ClO- Cl_2O_2 model for 8 March 2000. The agreement between measurements and model is good in terms of both the position and magnitude of the ClO maximum, lending further confidence in the ability of the full three-dimensional model to reproduce observed ozone in the vortex. The structure at 425 K in the measurements is not picked up in the model. The analysed temperature fields do, however, indicate that air masses at this altitude had been recently processed on PSCs but this feature is between two model levels at 400 K and 450 K. The in situ ClO measurements represent an excellent opportunity to test the ability of the model to reproduce one of the most important radicals in springtime, lower stratospheric polar photochemistry (Vömel et

al., 2001).

All the flights considered were inside the polar vortex where the model performance is good; we caution however that there may be regions, especially at the vortex edge, where detailed comparisons between model and observations are less satisfactory.

5 4.3. Estimated ozone loss in the vortex

As discussed above, chemical ozone loss can be estimated from changes in the relation between the mixing ratios of O_3 and a long-lived tracer, assuming that mixing of air between the two measurements does not significantly change the initial relationship. Preliminary examination of other tracer data sets in SOLVE-THESEO 2000 indicates that there was little mixing into the vortex between the second half of January and early March 2000 (Rex et al., 2002; Richard et al., 2001). Here, we merge the measurements of CFC-11 and ozone (for measurements made above 360 K) from the DIRAC and DESCARTES flights and use this merged data set to estimate ozone loss. First, the observed O_3 /CFC-11 relations from the merged DIRAC and DESCARTES flights are compared with relations from the Bonbon and LACE flights and also the equivalent relations from the SLIMCAT model. Secondly, these relations, for both the merged measurements and the model, are used to estimate ozone loss as a function of altitude in the vortex for two periods through the winter.

To examine the changes in the vertical distribution of ozone throughout the winter, flights made within a few days of each other are treated as a merged ensemble (Table 2). CFC-11 data for each flight within each ensemble are interpolated to the potential temperature measurement levels of the later flight of the ensemble. The CFC-11 data at each potential temperature level are then averaged to obtain a single CFC-11 profile. The uncertainty for each averaged CFC-11 measurement is the average of the quadrature addition of the fractional uncertainties of each individual measurement used in that average. An average ozone profile is then generated in exactly the same way as the CFC-11 average profile for each ensemble. The date of each averaged ensemble is taken as the average of all the individual flight dates used to generate the

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ensemble. The ozone change is finally obtained by taking the difference in ozone on equivalent CFC-11 levels between paired ensembles.

Figure 6a shows the relations between CFC-11 and O_3 for the merged DIRAC and DESCARTES data between the early December and late January flight ensembles.

5 There is little difference in the relation between the December and January data (average dates 9 December and 25 January, respectively, 47 days separation), suggesting little loss during this period. Figure 6a also shows the O_3 /CFC-11 relations for the LACE flight on 19 November and the Bonbon flight on 27 January 2000. There is a good agreement between the merged early December data and the LACE November data for all values of ozone apart from the region between 2600 and 3300 ppbv where the LACE data shows lower ozone for a given CFC-11 value by up to 500 ppbv. This could be attributed to a dynamical feature in the November flight data in the earlier stages of the vortex formation (Salawitch et al., 2002). The late January O_3 /CFC-11 relation for the merged data is typically between 10 and 20 pptv higher in CFC-11 for a given value of ozone when compared to the 27 January Bonbon flight. This is in part due to the difference in the measured vertical profiles of CFC-11 and part due to the earlier average date of the merged data (25 January).

10 In contrast to the December to January period, a clear split is seen in the O_3 /CFC-11 relations between January and March (average dates 25 January and 5 March, respectively, 39 days separation) as shown by the solid and dashed regression lines in Fig. 6b. Note that the uncertainty in the relation is large for the high altitude points at the top of the depleted ozone layer as the sample volumes and flow rates here were low and also this region was only sampled by DIRAC. From the merged data we diagnose ozone loss throughout the profile for CFC-11 mixing ratios less than about 175 pptv (corresponding to about 370 K). There is a 1100 ppbv change in ozone at a CFC-11 mixing ratio of 50 pptv (about 430 K), a 700 ppbv ozone change for 100 pptv CFC-11 (about 405 K) and a 300 ppbv O_3 change for 150 pptv of CFC-11 (about 385 K). Also evident from Fig. 6b is the excellent agreement in early March between the O_3 /CFC-11 relation for the merged data and that from the 1 March Bonbon flight and the 5 March LACE flight

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(strengthening our argument for only a systematic bias between DIRAC/DESCARTES and the other instruments).

A SLIMCAT calculation was performed for this winter (1999/2000) and the ozone evolution agreed well with data (Sinnhuber et al., 2000; Millard et al., in preparation, 2004¹). To investigate ozone change based on SLIMCAT, the model data for each flight date are grouped into ensembles and treated in the same way as the measurements. Investigations into the possible impact of vortex inhomogeneity on model ozone loss calculations show that the maximum variability in ozone loss along 5° PV equivalent latitude bands reached 27% in mid January 2000. This ozone loss variability then remained below 18% from the beginning of February onward, between 440 K and 500 K potential temperature (Millard et al., 2002). The low variability in vortex ozone loss makes the use of balloon based measurements as indicators of vortex average ozone loss particularly relevant in this winter. SLIMCAT diagnosed ozone loss rates in 1999/2000 agree well with 'Match' diagnosed ozone loss until March 2000 (Millard et al., 2004¹). Thereafter, SLIMCAT underestimates the rate of ozone loss by ~2 ppbv per sunlit hour due to insufficient model denitrification in the presence of large NAT PSCs. We note also that the model may not have reproduced the observed chlorine activation accurately because of a negative temperature bias in the forcing fields; nevertheless, modelled and observed ClO do compare well (Fig. 5). Figure 7 shows the equivalent plots to Fig. 6 for the vertical distributions of O₃ and CFC-11 from SLIMCAT above Kiruna for the DIRAC/DESCARTES merged flight dates. Little change in the relation is seen between the December 1999 and January 2000 dates, consistent with the observations, but a large shift is again seen between the January and March dates. The SLIMCAT CFC-11/O₃ relation for early March clearly shows the top of the depleted ozone layer in common with the measurements.

Figure 8 shows the altitude distribution of ozone change using the merged data for the period 9 December to 25 January obtained by plotting the ozone difference at fixed CFC-11 values against the relevant potential temperatures for the 25 January flight ensemble. Uncertainty estimates (indicated by horizontal bars) are the addition in

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quadrature of the standard errors for the ozone mixing ratios in the regressions defining the $O_3/CFC-11$ relations (shown in Fig. 6). The measurements show no ozone loss in the 400–450 K region between December and January. Also shown in Fig. 8 is the altitudinal ozone change for the period from 25 January to 5 March obtained by plotting the ozone difference for fixed CFC-11 values against the potential temperatures for the 5 March flight ensemble. During this period there was substantial ozone loss with a maximum of about 1200 ppbv in the 440–470 K region with ozone loss to a lesser extent down to about 370 K. Figure 8 also shows the altitude distribution of ozone change using the Bonbon and LACE data for the period 27 January to 5 March calculated using the same methodology as with the DIRAC and DESCARTES data. There is an excellent agreement in both the magnitude and position of the ozone loss maximum and both data sets show ozone loss extending down to the 370–380 K region.

The vertical distribution of ozone change using the SLIMCAT results was examined for the periods of early December to late January and late January to early March (both using the same dates as the merged DIRAC/DESCARTES data). As with the measurement data, the ozone differences are plotted against the potential temperature of the later flight date for each period. These model ozone change distributions are shown in Fig. 8 together with the measurements. The model shows slight ozone loss (<300 ppbv) in the 450–550 K region in the early December to late January period. The model shows a maximum ozone loss of 1300 ppbv at 480 K for the late January to early March period, very similar to the loss derived from the measurements.

5. Conclusions

The ascent data from DIRAC and DESCARTES provide a set of CFC-11 profiles from a total of nine balloon flights made during the 1999/2000 Arctic winter. These vertical profiles compare well with SLIMCAT CFC-11 profiles for the same dates and location. Observations of ClO on the 8 March flight are in good agreement with ClO output for the SLIMCAT model in the region where we derive ozone loss, giving confidence in the

ability of the full 3-D model to reproduce observed ozone in the vortex.

Using ECC ozonesonde data from the balloon flights inside the vortex (or from small balloons launched soon after) changes in the O₃/CFC-11 relationship are observed throughout the winter. Grouping the flights into ensembles and merging the DIRAC and DESCARTES data provides a means of estimating the vertical change in ozone concentration from early December to late January and from late January to early March. SLIMCAT data generated for the same dates and location as the measurements are grouped into the same ensembles and treated identically to the measurement data. The merged flight ensembles show no ozone loss between early December and late January, the model shows slight ozone loss (<300 ppbv in the 450–550 K region). Between late January and early March the merged measurements show a maximum ozone loss of 1200 ppbv in the 440–470 K region which compares excellently with the maximum loss calculated by the model (1300 ppbv at 480 K).

A thorough comparison of ozone loss rates requires a detailed examination of the conditions (period, altitude, location) over which the ozone loss occurs (Harris et al., 2002) and is beyond the scope of this study. However simple comparisons with the other studies in the 1999/2000 winter (Hoppel et al., 2002; Klein et al., 2002; Müller et al., 2003 and discussion therein; Rex et al., 2002; Richard et al., 2001; Salawitch et al., 2002; Schoeberl et al., 2002; summary in Table 8 of Newman et al., 2002) show good agreement. We find that the peak loss occurred in the 440–470 K region, which is the same as Müller et al. (2003) at the low end of the range. Our peak loss of 1200 ppbv is less than those of some other studies (e.g. 2000 ppbv of Rex et al., 2002, and 1800 ppbv of Richard et al., 2001), but this is at least partly related to the earlier end to the period we consider. The cumulative ozone loss reported by Match on about 5 March (the average date of our March flights) is about 1500 ppbv (Fig. 6 in Rex et al., 2002), within the estimated uncertainties of the two techniques.

Finally, the study shows that in winters when the polar vortex is relatively undisturbed, tracer ozone relationships measured from a single location (and model output also) can be used to provide ozone loss estimates which are in good agreement with the

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modelled ozone loss for the vortex core as a whole.

Acknowledgements. We gratefully acknowledge the following for their financial support: the Research DG of the European Commission through the THESEO O₃ loss (ENV4-CT97-0510), THESEO-EuroSOLVE (EVK2-CT-1999-00047) and CRUSOE (EVK2-1999-00252, EVK2-CT-2001-20012) projects; the UK Natural Environment Research Council through its UTLS Ozone programme; the NERC UK Universities Global Atmospheric Modelling Programme (UGAMP) and the NERC Centre of Atmospheric Science (NCAS); the UK Department of Environment, Transport and Regions through its Global Atmospheres Division (EPG 1/1/83); and US National Science Foundation. We would also like to acknowledge CNES, Esrange and NASA for provision of the flights we had during SOLVE-THESEO 2000 and W. Brune for his assistance during the OMS in situ flight. G. Millard thanks the EPSRC for a studentship. We thank A. Hauchecorne for provision of MIMOSA PV maps.

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Table 1. Balloon flights in the 1999/2000 winter from Esrang, Kiruna (67.9 N, 21.1 E) which carried the DIRAC/DESCARTES instruments featured in this work. * ECC ozonesonde flown as part of the main payload. † ECC ozonesonde flown soon after the main balloon.

Date	Launch hour (UTC)	Guest instrument	Host payload
3 Dec. 1999	10	DESCARTES	OMS in situ*
15 Dec. 1999	11	DESCARTES	SKERRIES†
19 Jan. 2000	11	DIRAC	HALOZ (CIO)*
27 Jan. 2000	12	DIRAC	HALOZ (CIO)*
28 Jan. 2000	11	DESCARTES	SAOZ†
1 March 2000	10	DESCARTES	HALOZ (CIO)*
5 March 2000	15	DIRAC, DESCARTES	OMS in situ*
7 March 2000	14	DESCARTES	SAOZ†
8 March 2000	12	DIRAC	HALOZ (CIO)*

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Table 2. Grouped DIRAC and DESCARTES flight ensembles used to examine ozone change from December to January (a) and from January to March (b) in the 1999/2000 Arctic winter.

(a): Ozone change: early December to late January

	December flight ensemble	January flight ensemble
Date 1	DESCARTES 3 Dec. 1999	DIRAC 19 Jan. 2000
Date 2	DESCARTES 15 Dec. 1999	DIRAC 27 Jan. 2000
Date 3		DESCARTES 28 Jan. 2000
Average date	9 Dec. 1999	25 Jan. 2000

(b): Ozone change: late January to early March

	January flight ensemble	March flight ensemble
Date 1	DIRAC 19 Jan. 2000	DESCARTES 1 March 2000
Date 2	DIRAC 27 Jan. 2000	DIRAC 5 March 2000
Date 3	DESCARTES 28 Jan. 2000	DESCARTES 7 March 2000
Date 4		DIRAC 8 March 2000
Average date	25 Jan. 2000	5 March 2000

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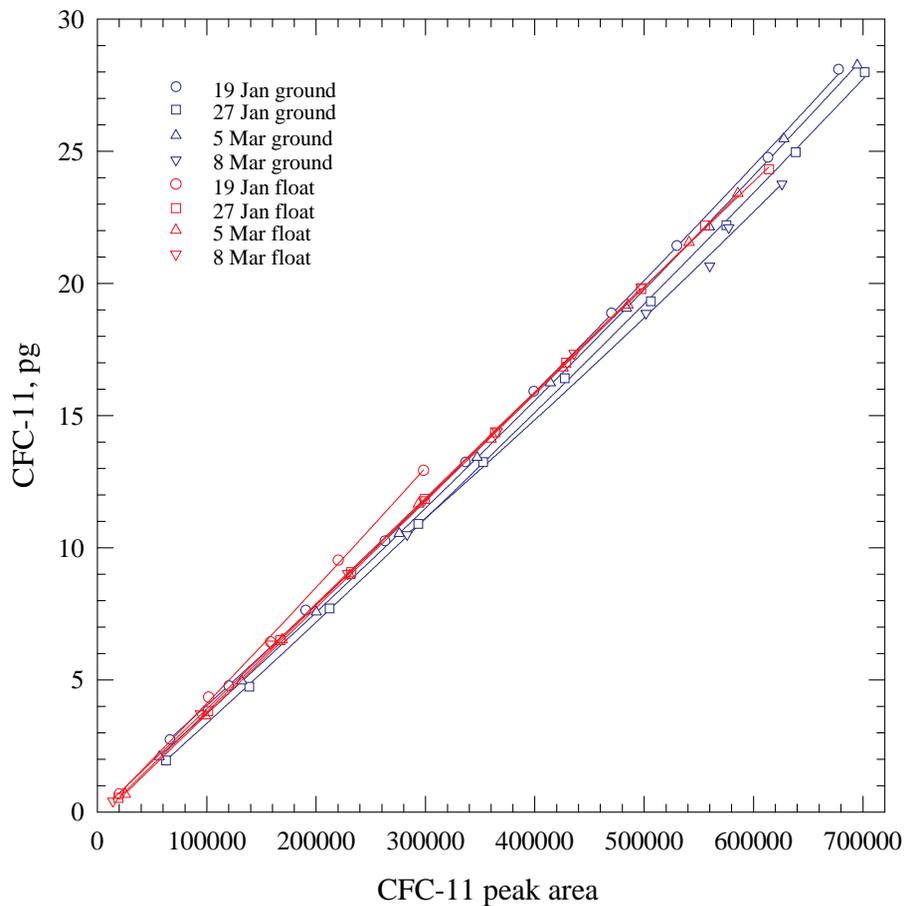


Fig. 1. DIRAC in-flight CFC-11 response curves for 19 January, 27 January, 5 March and 8 March used to calibrate the in situ CFC-11 measurements.

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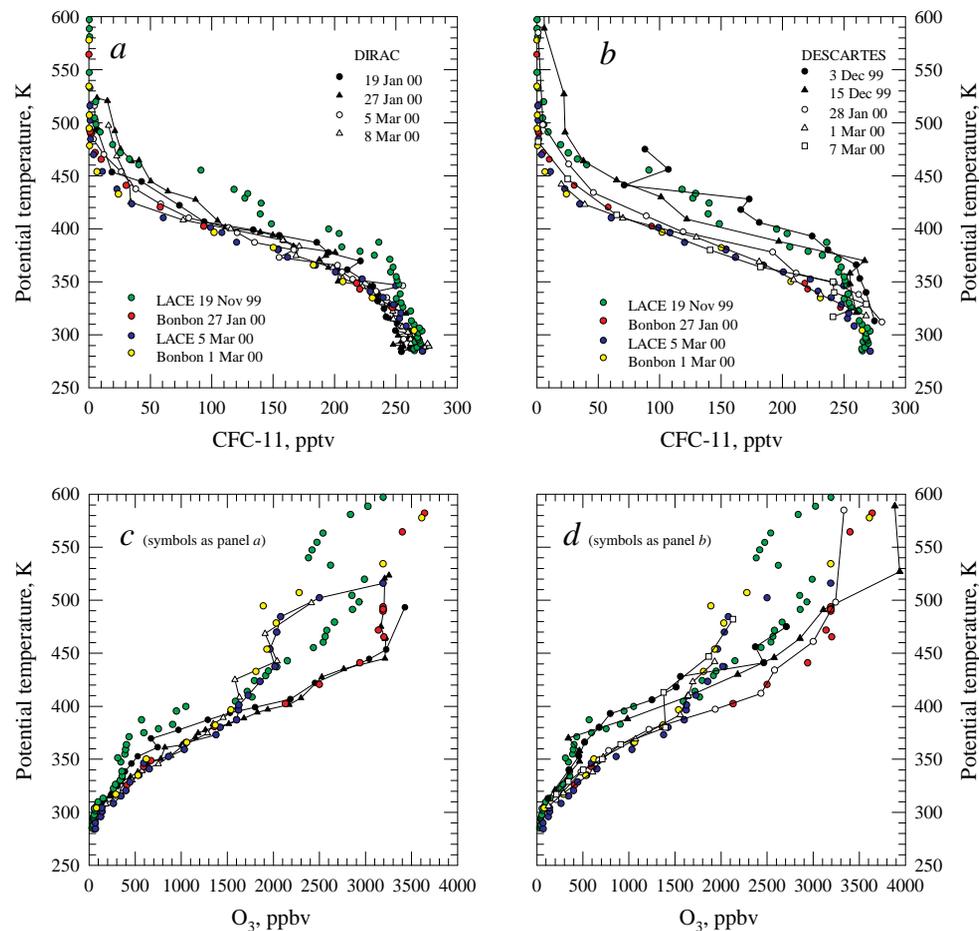


Fig. 2. Vertical profiles of: CFC-11 from the DIRAC flights (panel a); CFC-11 from the DESCARTES flights (panel b); ozone from ECC sondes for the DIRAC flights (panel c); and the DESCARTES flights (panel d) used as a basis for the ozone loss estimates.

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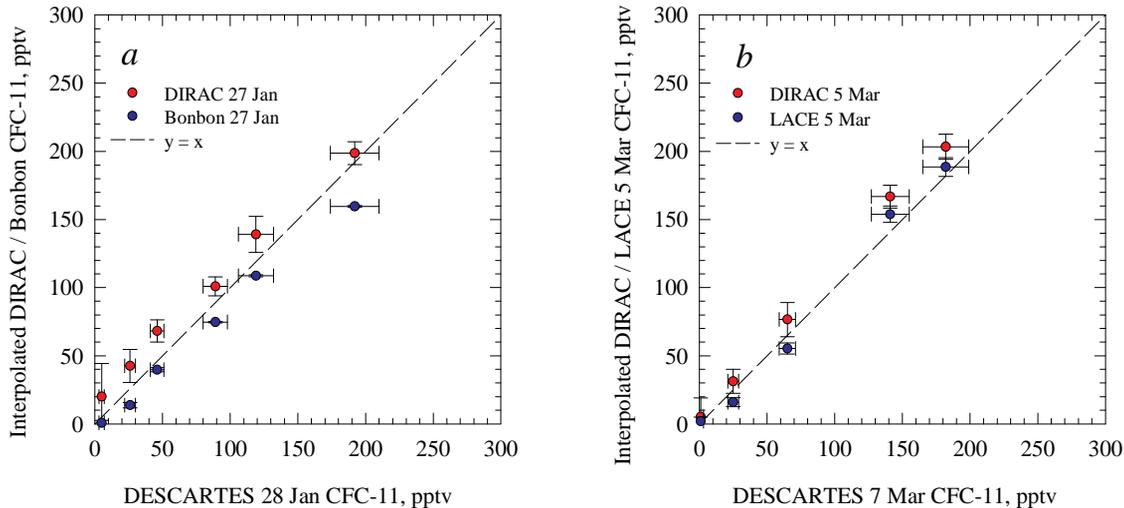


Fig. 3. Plots comparing DIRAC and DESCARTES measurements from late January (panel **a**) and early March (panel **b**). The data from both instruments are merged into a single data set on the basis of this agreement for measurements made above 360 K. Bonbon and LACE measurements interpolated to the potential temperature of the DESCARTES measurements are shown for comparison.

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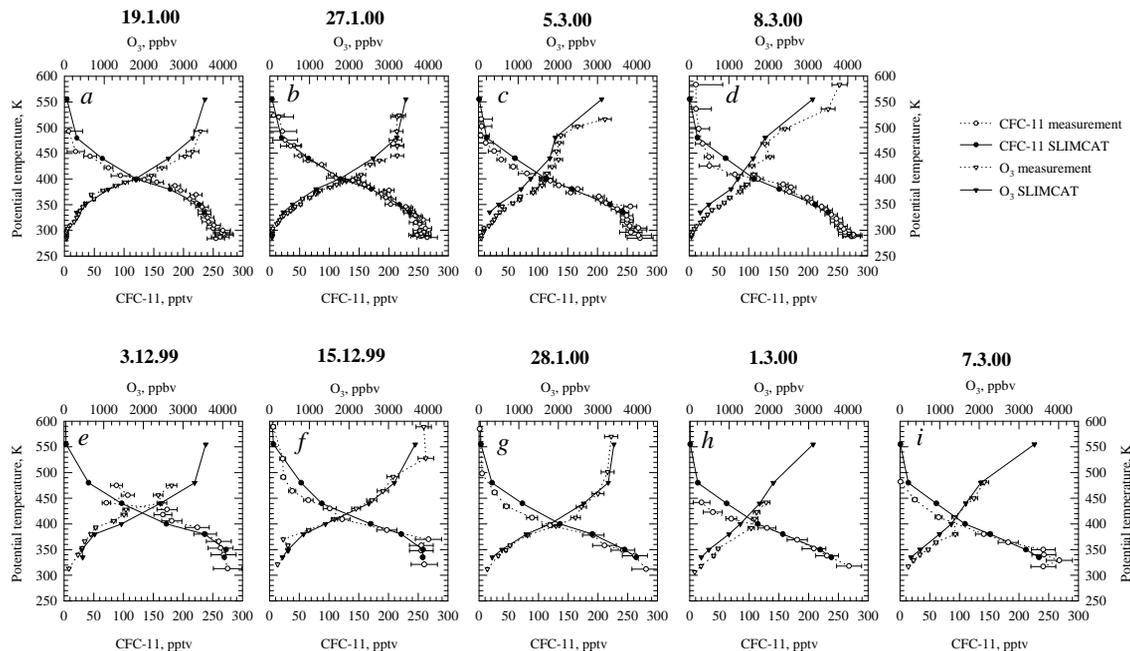


Fig. 4. Comparisons between the measurements and SLIMCAT data for CFC-11 and ozone (panels **a** to **d**: DIRAC flights; panels **e** to **i**: DESCARTES flights).

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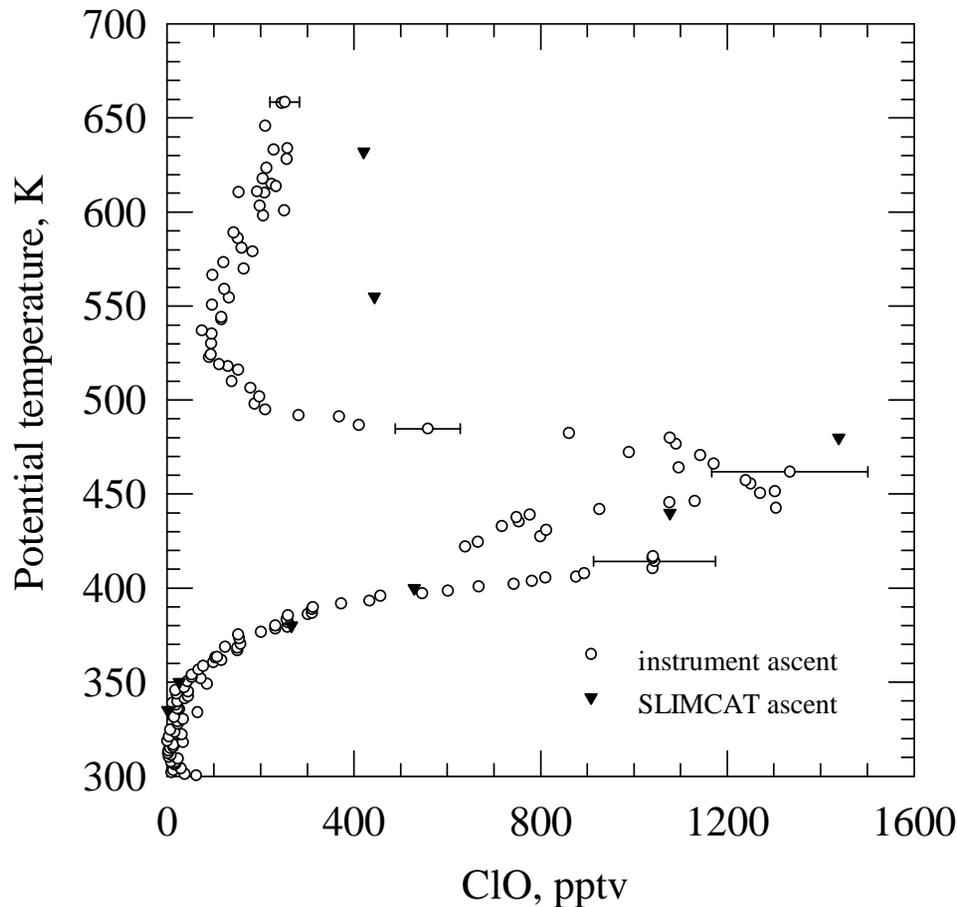


Fig. 5. Comparison between the vertical profile from the ClO instrument and from SLIMCAT on 8 March. The overall uncertainty of the ClO instrument data is shown by the error bars (1 standard deviation) at selected points in the profile.

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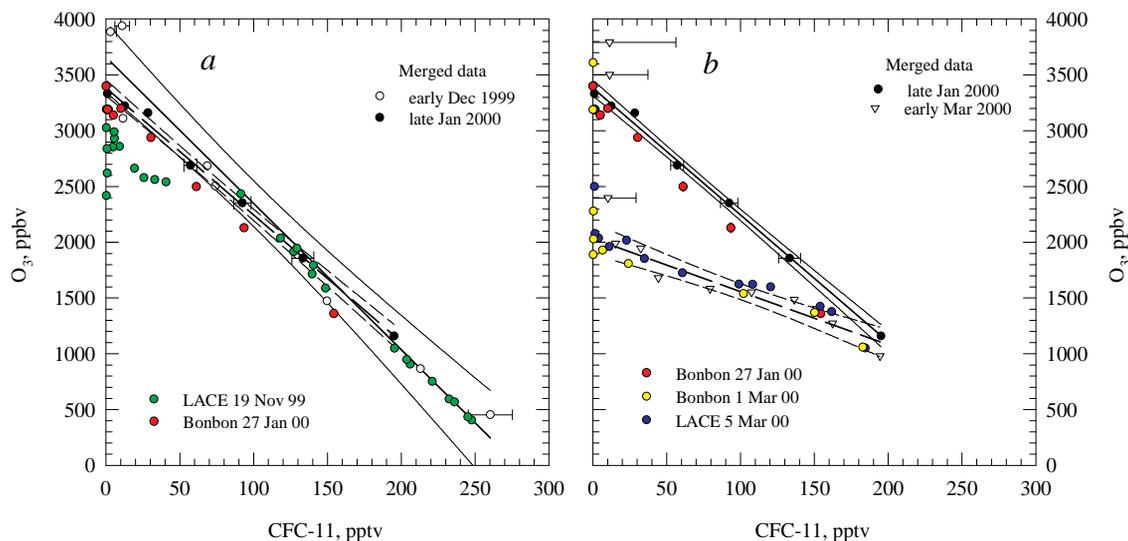


Fig. 6. The relationship between CFC-11 and ozone for the DIRAC/DESCARTES merged flight ensembles with data from Bonbon and LACE shown for comparison. Panel (a): early winter, the solid line is a 1st order regression fit to the early December ensemble, the dashed line is a 1st order regression fit to the late January ensemble (dotted lines are 95% confidence limits). Panel (b): late winter, the solid line is a 1st order regression fit to the late January ensemble, the dashed line is a 1st order regression fit to the early March ensemble.

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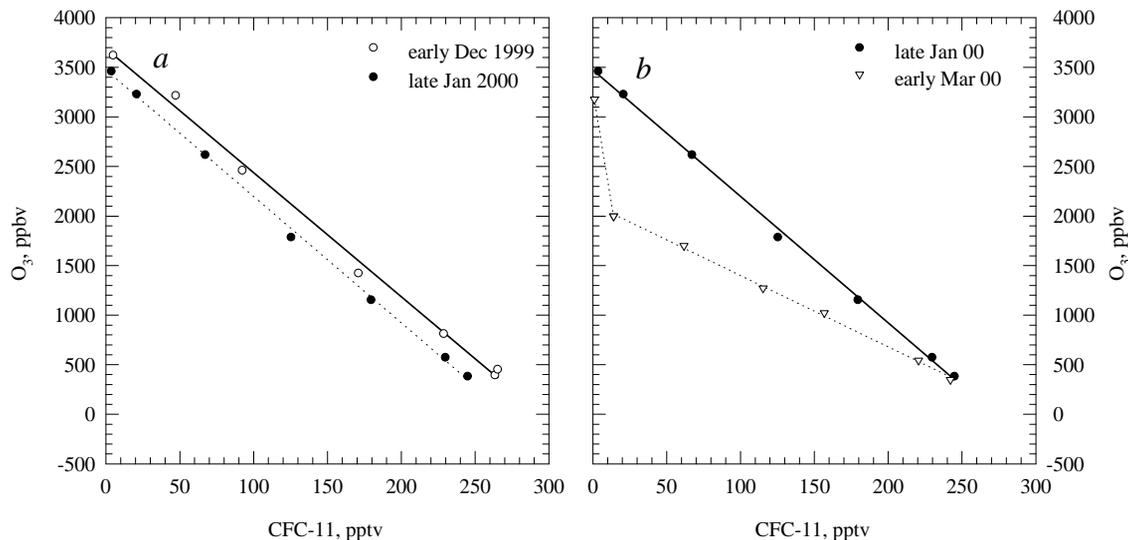


Fig. 7. The relationship between SLIMCAT CFC-11 and ozone for the DIRAC/DESCARTES merged flight dates (panel **a**: early December and late January; panel **b**: late January and early March). Solid lines are 1st order regression fits to the earlier ensemble in each panel, dashed lines are 1st order regression fits to the later ensemble in each panel.

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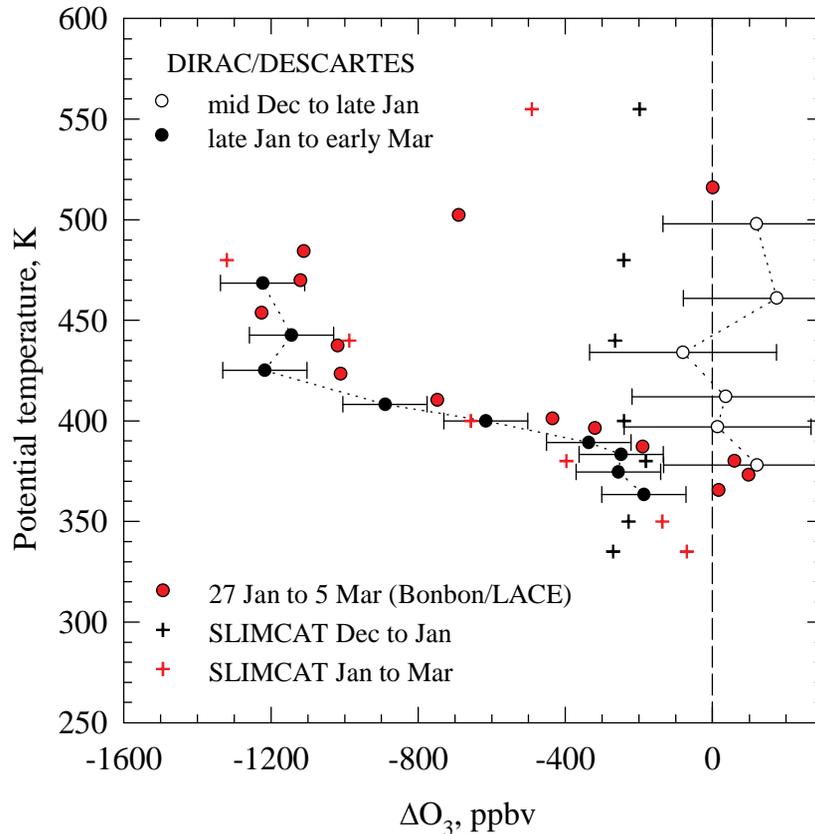


Fig. 8. The vertical distribution of ozone change from early December to late January and from late January to early March derived from the relationships between CFC-11 and ozone for the merged DIRAC/DESCARTES flights. Also shown is the profile of ozone change between 27 January and 5 March based on the Bonbon and LACE data. The SLIMCAT vertical ozone change is shown for the same periods as the merged DIRAC/DESCARTES ozone change plots.

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