

Severe ozone depletion in the cold Arctic winter 2004–05

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[1] During a flight of the M55 Geophysica into the Arctic polar vortex on 7 March 2005, ozone, halogen species, tracers and water vapor were measured. Up to 90% chlorine activation and up to 60% ozone loss were found above 14 km, reflecting the low temperatures and extensive PSC formation prevalent in the Arctic stratosphere over the 2004/05 winter. Observations are generally well reproduced by CLaMS model simulations. The observed levels of active chlorine can only be reproduced by assuming significant denitrification of about 70%. Moderate dehydration up to 0.5 ppm is observed in some locations. We deduce a partial column ozone loss of 62 (+8/−17) DU below 19 km on 7 March. **Citation:** von Hobe, M., et al. (2006), Severe ozone depletion in the cold Arctic winter 2004–05, *Geophys. Res. Lett.*, 33, L17815, doi:10.1029/2006GL026945.

1. Introduction

[2] Strong chemical polar ozone loss in winter is caused by chlorine compounds activated on polar stratospheric clouds (PSCs) that form at low temperatures [Solomon, 1999]. While in the Antarctic a stable and cold polar vortex has led to an ozone hole in virtually each winter since the early 1980s, the year-to-year variability in the Arctic is much greater [e.g., Tilmes et al., 2004]. Stratospheric cooling induced by climate change could intensify polar ozone depletion against the trend of declining chlorine levels [e.g., Rex et al., 2004]. In order to check on the model representation of key processes involved in polar stratospheric ozone loss we compare in-situ measurements and simulations of the Chemical Lagrangian Model of the Stratosphere (CLaMS) [e.g., Grooß et al., 2005; Konopka et al., 2004].

[3] The winter 2004/2005 was one of the coldest on record leading to extensive PSC formation [Manney et al., 2006] (see also S. Tilmes et al., Chemical ozone loss in the Arctic and Antarctic stratosphere between 1992 and 2005, submitted to *Geophysical Research Letters*, 2006) (hereinafter referred to as Tilmes et al., submitted manuscript, 2006), considerable chlorine activation [Dufour et al., 2006], denitrification [Kleinböhl et al., 2005] and ozone loss [Manney et al., 2006]. Here, we present in-situ measurements of O₃, ClO_x

($\cong \text{ClO} + 2 \text{Cl}_2\text{O}_2$), tracers and H₂O carried out during a flight of the high altitude research aircraft M55 Geophysica into the Arctic vortex on 7 March 2005. Total inorganic chlorine (Cl_y) and passive ozone are computed from tracer data and correlations in order to assess the extent of chlorine activation and chemical ozone loss. The consistency between observations and CLaMS 3-D CTM simulations is investigated. CLaMS is also used in a box model version in an attempt to constrain denitrification based on observed ClO_x and verify the denitrification predicted by the CTM version. Finally we integrate and extrapolate the observed vertical profile of ozone loss to total column ozone loss at the end of the winter that can be compared to estimates based on other methods and to ozone loss observed in previous Arctic winters.

2. Experiment

2.1. Observations

[4] The flight of the Geophysica on 7 March extended from 48.1° to 58.2°N, corresponding to 50° to 88° equivalent latitude, and spanned the altitude range 13 to 19 km (see auxiliary material¹ for a map and more detailed information on potential vorticity (PV) and equivalent latitude). O₃ was measured with a time resolution of 1 s and a precision of <10% by the FOZAN-II (Fast Ozone Analyzer) [Ulanovsky et al., 2001; Yushkov et al., 1999], a two channel chemiluminescent instrument calibrated in flight every 15 minutes. N₂O and CH₄ were measured by HAGAR (High Altitude Gas Analyzer, [Volk et al., 2000]) with a time resolution of 90 s and mean precisions of ~0.6% and ~2% of the respective tropospheric background values. Calibrations were performed in-flight with two standards linked to within 1% of the NOAA-CMDL scale. ClO, Cl₂O₂ and BrO were measured by the HALOX instrument [von Hobe et al., 2005] with time resolution of 10 s, 3 min and 10 min respectively. No ClO, Cl₂O₂ and BrO data could be recorded on the outbound flight leg. H₂O was measured with a frequency of 1 s^{−1} by FISH (Fast In situ Stratospheric Hygrometer) based on the Lyman-α photofragment fluorescence technique [Zöger et al., 1999]. Temperature was measured by Rosemount sensors, while static pressure and position are from the aircraft avionics system.

2.2. Calculation of Ozone Loss

[5] Passive ozone used for estimation of chemical ozone loss by the tracer-tracer correlation method [e.g., Tilmes et al., 2004] is calculated from HAGAR N₂O observations using correlations derived from MLS O₃ and N₂O observations

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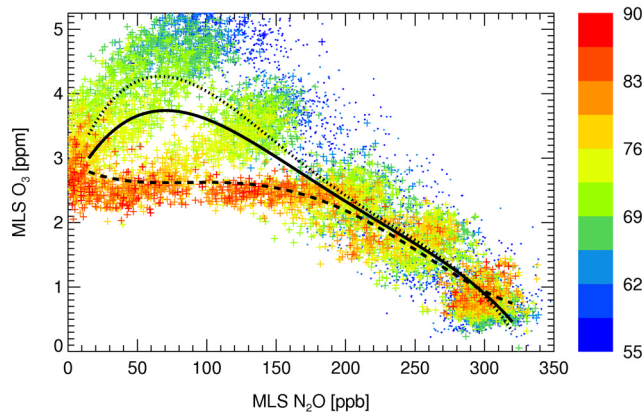


Figure 1. EOS MLS AURA (V1.51, <http://mls.jpl.nasa.gov/joe/>) O₃ vs. N₂O observations for the period 25.–31.12.2004 colored according to eq. latitude in °N. Lines show N₂O:O₃ correlations as described in the text (1: dotted, 2: dashed, 3: solid).

between 25 and 31 December 2004 (Figure 1). This procedure is complicated by the fact that early winter O₃ and N₂O were higher near the vortex edge than in the vortex core, and that considerable mixing between these two regions occurred from late February to early March [Manney *et al.*, 2006]. Different correlations also shown in Figure 2 (1: vortex edge, 65°–75°N eq. Lat.; 2: vortex core, 75°–90°; 3: vortex average, 65°–90°) were derived using equivalent latitude bands as defined for this time period by Manney *et al.* [2006]:

$$[O_3] = 2.64 + 5.76 \times 10^{-2}[N_2O] - 6.40 \times 10^{-4} \times [N_2O]^2 + 2.34 \times 10^{-6}[N_2O]^3 - 3.05 \times 10^{-9}[N_2O]^4 \quad (1)$$

$$[O_3] = 2.95 - 1.28 \times 10^{-2}[N_2O] + 1.73 \times 10^{-4} \times [N_2O]^2 - 9.04 \times 10^{-7}[N_2O]^3 + 1.31 \times 10^{-9}[N_2O]^4 \quad (2)$$

$$[O_3] = 2.45 + 4.29 \times 10^{-2}[N_2O] - 4.50 \times 10^{-4} \times [N_2O]^2 + 1.55 \times 10^{-6}[N_2O]^3 - 1.95 \times 10^{-9}[N_2O]^4 \quad (3)$$

with [O₃] in ppm and [N₂O] in ppb, valid for 15 ppb < [N₂O] < 320 ppb. Because of the mixing processes that took place inside the vortex over the winter, it is impossible to clearly identify the origin of the air masses investigated in this study. In the following, ozone loss is calculated using correlation (3) with upper and lower uncertainties determined from correlations (1) and (2).

2.3. Model Simulations

[6] The CLaMS model [e.g., Grooß *et al.*, 2005; Konopka *et al.*, 2004] is used in all simulations with trajectories and temperatures derived from analyses of the European Centre for Medium-Range Weather Forecasts (ECMWF).

[7] A hemispheric 3-dimensional CLaMS simulation for the winter, termed “CLaMS-H” in the following, is used for comparison. It was initialized based upon MLS-AURA satellite data and tracer-tracer correlations for 20 November 2004, when the polar vortex had already formed and temper-

atures in the Arctic stratosphere were still above the PSC formation threshold. Denitrification is simulated by following individual NAT particle trajectories along which the particles may grow and sediment due to their vertical settling velocity using the method described for the winter 2002/03 by Grooß *et al.* [2005]. The nucleation of NAT particles was assumed at all locations with $T < T_{\text{NAT}}$ using a nucleation rate of $7.8 \cdot 10^{-6} \text{ cm}^{-3} \text{ h}^{-1}$. The horizontal resolution of the simulation is 100 km.

[8] Chemistry simulations were also performed along 10-day backward trajectories from the flight path. These simulations, referred to as “CLaMS-B10” below, were used to constrain NO_y ($\approx \text{HNO}_3 + \text{ClONO}_2$), for which no data are available. As the HNO₃ concentration determines the rate of chlorine deactivation to ClONO₂, initial HNO₃ can be optimized for agreement between simulated and observed ClO_x. Initial ClO_x was set equal to Cl_y (estimated from HAGAR CH₄ as in work by Grooß *et al.* [2002]), thus requiring the model to deactivate the maximum possible amount of active chlorine in the given amount of time. Initial HNO₃ was varied in small increments over a number of simulations. The amount of NO_y present in the simulation that best reproduced the observed ClO_x then represents an upper limit for NO_y. Parameters other than ClO_x and NO_y were initialized with Geophysica observations from 7 March. Results of this optimization are meaningful only above $\sim 390 \text{ K}$ because uncertainties in ClO_x and Cl_y are too large below this altitude. A 10-day period was chosen so that the main PSC period had ended before the start of the back trajectories. Minor PSC events were simulated on some trajectories. Heterogeneous processes potentially leading to renewed chlorine activation and further removal of HNO₃ are

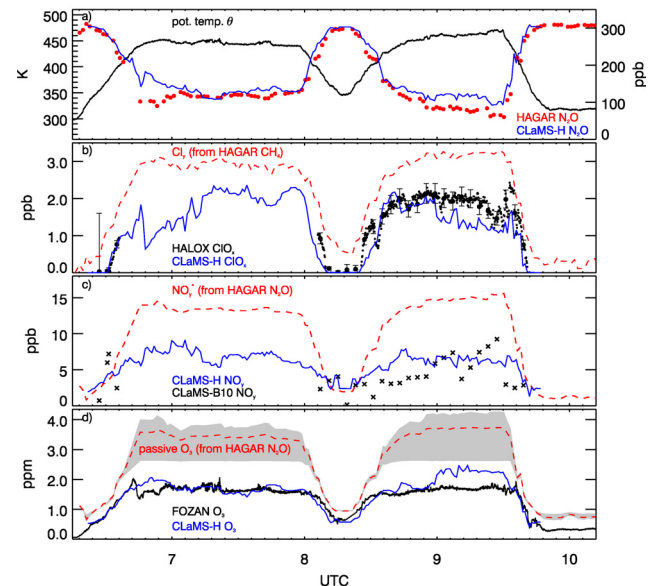


Figure 2. Comparison of observations to CLaMS simulations. In Figure 2b, large symbols with error bars correspond to measured [ClO_x], whereas the small symbols represent data where only [ClO] was measured and [Cl₂O₂] was interpolated (cf. auxiliary material); in Figure 2d, the dashed red line shows passive ozone inferred from N₂O using correlation 3 (see text) with the gray area denoting the range between correlations 1 and 2.

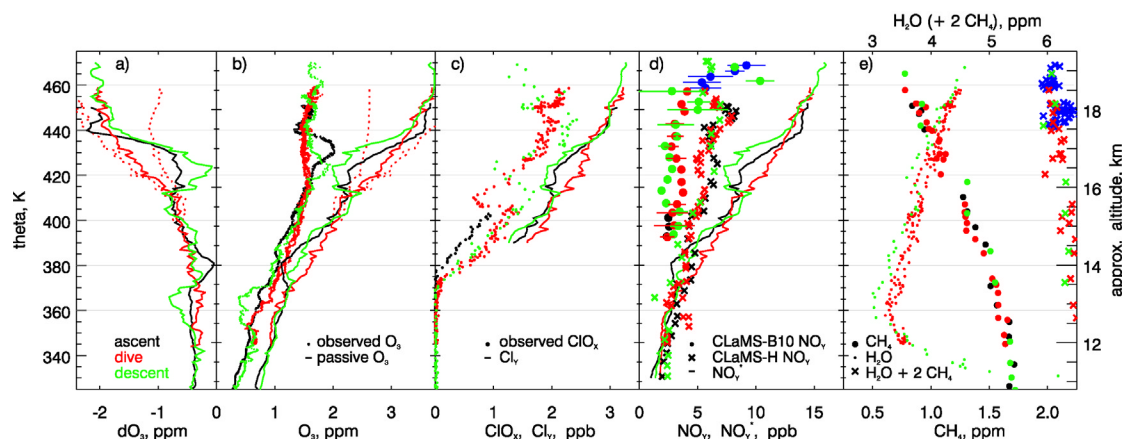


Figure 3. Vertical profiles of (a) chemical ozone loss dO_3 , (b) observed and passive O_3 (see text), (c) ClO_x and Cl_y (see text), (d) NO_y from CLaMS simulations and NO^* , and (e) observed CH_4 and H_2O and $H_2O + 2 CH_4$ (see text). Passive O_3 , Cl_y and NO^* are plotted using HAGAR tracer values interpolated with the aid of the CLaMS-H simulation. Dotted lines in Figures 3a and 3b give uncertainty estimates of passive ozone based on correlations (1) and (2) for the dive. In Figures 3d and 3e, data for parts of the flight between profiles are added in blue. In Figure 3d, mean and 1σ standard deviation from CLaMS-B10 for 5 K theta bins (2.5 K for the blue symbols) are given.

incorporated in the CLaMS-B10 simulations, and uncertainties in the estimated NO_y are limited to uncertainties in the model representation of these processes.

[9] To estimate the amount of ozone loss that occurred after 7 March, a CLaMS simulation along 30-day forward trajectories from the Geophysica flight path was employed (“CLaMS-F30”). All species relevant to ozone loss were initialized using observations, except HNO_3 and $ClONO_2$, for which the output of the CLaMS-B10 simulation was used. As CLaMS-B10 yields a lower limit for denitrification, CLaMS-F30 is expected to give a lower limit for ozone loss, but influence of denitrification within the constraints given here (cf. below) is expected to be small [Groß et al., 2005].

3. Results and Discussion

3.1. Chlorine Activation

[10] ClO_x mixing ratios observed by HALOX (Figure 2b; see auxiliary material for individual ClO , Cl_2O_2 and BrO measurements) reveal significant chlorine activation at altitudes between 14 and 20 km (Figure 3c), with ClO_x constituting up to 90% of Cl_y . However, the high degree of activation observed in the test flight is probably not representative for the entire vortex. The CLaMS-H simulation shows that the flight track extended into the region with the highest ClO_x levels in early March, whereas deactivation into $ClONO_2$ and HCl had progressed considerably further in other parts of the vortex [Dufour et al., 2006].

[11] Lower ClO_x mixing ratios observed during descent between 415 and 435 K and around 390 K are likely caused by intrusions of mid latitude air (see auxiliary material), while the dips in ClO_x between 405 and 420 K during the dive and around 450 K during dive and descent are more likely to be caused by higher NO_y levels (cf. below). During ascent and the first half of the return flight leg, ClO_x concentrations predicted by CLaMS-H compare extremely well to the observations (Figure 2b). However, CLaMS-H cannot resolve the small scale structure observed during dive and

descent. The underestimation of ClO_x by the model during the second half of the return flight leg (9:00–9:30 utc) may be caused by either an underestimation of the diabatic descent or an overestimation of in-mixing of non-vortex air in the hemispheric simulation, indicated by the overestimation of the passive tracer N_2O over the same time period (Figure 2a).

3.2. Denitrification and Dehydration

[12] Within the vortex a lower limit to denitrification of about 70% (compared to the NO_y^* reference state estimated using a polynomial fit to N_2O given by Popp et al. [2001]) above 400 K is predicted by CLaMS-B10 (Figures 2c and 3d), with more severe denitrification on some trajectories above 450 K, which is supported by observations of water vapor. Although the H_2O measurements themselves do not show any obvious dehydration (Figure 3e), the quantity $H_2O + 2 CH_4$ commonly used as a measure of dehydration [Schiller et al., 2002] is reduced by up to 0.5 ppm between 440 and 470 K and for some individual data points below (Figure 3e), suggesting permanent removal of H_2O by sedimenting ice particles in the Arctic vortex 2004/5, confirming an earlier study by Kivi et al. [2005].

[13] CLaMS-H mostly predicts less severe denitrification (on the order of 50%) than CLaMS-B10 (Figures 2c and 3d), but the shapes of the NO_y profiles generally agree quite well. It should be noted that NO_y is kept constant during the CLaMS-B10 runs (except where new particles form and sediment out) and mixing with less denitrified air masses is not accounted for, unlike in CLaMS-H. However, some overestimation of NO_y mixing ratios by CLaMS-H does not seem unreasonable given the uncertainties in the parameterizations of particle formation and sedimentation [Groß et al., 2005] and the uncertainties in vortex descent and mixing (cf. above).

[14] Both observed chlorine activation and predicted denitrification down to 370 K during dive and descent point to PSCs generally extending to lower altitudes in 2005 than in earlier cold winters, in good agreement with the notion that

ozone loss also extended to lower altitude [Manney *et al.*, 2006].

3.3. Ozone Loss

[15] A comparison of observed O_3 with passive O_3 deduced from correlation 3 suggests ozone depletion (dO_3) as high as 60% between 14 and 19 km (380 to 470 K theta), consistent with the high degree of chlorine activation and denitrification in this altitude range (Figure 3). The vertical structure in the ascent and descent profiles of O_3 and dO_3 probably results from mixing with non-vortex air (see auxiliary material). Ozone predicted by CLaMS-H follows the observations well over large parts of the flight (Figure 2d). Significant discrepancies are observed between 9:00 and 9:30 (coincident with the underestimation of ClO_x) and between 6:40 and 7:00 after the descent. In both cases, CLaMS-H also overestimates N_2O pointing to deficiencies in the simulation of the dynamics (descent and mixing, cf. ClO_x above) rather than the chemistry.

[16] Integration of the ozone loss (Figure 3a) over altitude yields partial column ozone loss of 57^{+7}_{-14} DU for the ascent profile (altitude range 11.6–18.0 km), 62^{+8}_{-17} DU for the dive (12.8–18.8 km), and 77^{+10}_{-22} DU for descent (11.7–19.2 km). This partial column ozone loss is comparable to values observed in the coldest Arctic winters over the past 15 years inferred from HALOE O_3 /HF correlations [Tilmes *et al.*, 2004]. Only the ozone loss for the dive is used in the following calculations, because this is least influenced by mixing processes.

[17] The shape of the ozone loss profiles at the highest altitudes reached by the Geophysica (Figure 3a) suggests significant ozone loss above 460 K (ozone loss between 460 and 470 K for the descent profile makes up $5.8^{+1.5}_{-3.1}$ DU). Evidence from the 5 coldest Arctic winters before 2004/5 (1992, 93, 95, 96 and 2000: average and standard deviation of 23 ± 6 DU above 460 K inferred from HALOE O_3 /HF correlations) suggests that variability of the partial column ozone loss at high altitudes in cold winters is small [Tilmes *et al.*, 2004]. Under the assumption that this is true also for the 2004/5 winter, and that ozone loss above 460 K falls into the same range as for the winters named above, the total column ozone loss on 7 March 2005 would be expected to fall between 75 and 103 DU, which would represent rather dramatic ozone depletion for this stage of the winter, especially with the amount of ClO_x still present and the high degree of denitrification. However, as mentioned above, chlorine deactivation had probably progressed much further in other regions of the vortex.

[18] Ozone loss following 7 March was estimated by the CLaMS-F30 simulation. It was limited to 19 DU due to rapidly increasing temperatures and slowly progressing chlorine deactivation in the investigated air masses. To aid the comparison with other ozone loss estimates for the 2004/5 winter, we add up the partial column ozone loss below 460 K observed during the dive on 7 March, the average partial column ozone loss above 460 K from previous years and the late winter ozone loss based on the CLaMS-F30 model calculations arriving at 104^{+10}_{-18} DU. We do want to stress that this value is only partially based on observations, and that one profile may not be representative of the entire vortex, especially considering the inhomogeneities present in this particular winter. The value given falls

into the lower uncertainty range of other studies of this winter: Rex *et al.* [2006] report a chemical ozone loss of 121 ± 20 DU between 380–550 K deduced using measurements from ozone sondes and the vortex average approach. Jin *et al.* [2006] derived 119 DU in mid-March from tracer correlations of ACE satellite measurements. A more dramatic decline in ozone levels was prevented by the early break-up of the vortex (the temperature increase following 7th March represents the final warming of the vortex [Manney *et al.*, 2006]) and associated in-mixing of ozone from unperturbed air masses.

[19] Nevertheless, the cold 2004/05 Arctic winter represents one of the strongest northern winters in terms of ozone loss (Tilmes *et al.*, submitted manuscript, 2006; Rex *et al.*, 2006), which both the Geophysica measurements and the CLaMS-H simulation show. The results from this winter are particularly interesting in the context of future climate change, if long periods with $T < T_{NAT}$ become a regular feature and aggravate ozone depletion in the Arctic [Rex *et al.*, 2004]. The generally good representation of the observations by the hemispheric CTM CLaMS initialized near the formation of the polar vortex more than 3.5 month prior to the flight demonstrates our ability to simulate the dynamic and chemical processes affecting the ozone layer in the polar vortex, which should add to our confidence in predicting future polar ozone loss.

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