

1 On the unexplained stratospheric ozone losses during cold 2 Arctic Januaries

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9 **Abstract.** Using a combination of data from Match, POAM II,
10 POAM III and MLS we show that the chemical loss rate of
11 Arctic O₃ during January of four cold winters (1992, 1995,
12 1996, and 2000) is consistently faster than can be accounted
13 for by assuming complete activation of reactive chlorine and
14 standard reaction kinetics. However, O₃ loss rates measured
15 during late February and early March 1996 are shown to be
16 consistent with observations of ClO. The faster than expected
17 O₃ loss rates during January are shown to occur when air
18 parcels are illuminated at high solar zenith angles (SZAs
19 between ~85 and 94°), and to result in cumulative O₃ loss of
20 ~0.5 ppmv. The cause of the rapid January O₃ loss is unclear,
21 but may be related to a photolytic process at high SZA that is
22 poorly represented by current photochemical models.

23 1. Introduction

24 Proper understanding of the timing and extent of chemical
25 depletion of Arctic O₃ during winter is a prerequisite for
26 developing reliable assessments of future ozone. Early studies
27 suggested consistency between *observed* rates of chemical O₃
28 loss (hereafter referred to as O₃ *loss_obs*) and *modeled* loss rates
29 (O₃ *loss_mdl*) based on measured concentrations of ClO and
30 BrO and relevant laboratory kinetics [e.g., Salawitch *et al.*,
31 1990]. These studies focused primarily on the February to
32 March time period and were limited by large (e.g., factor of
33 two) uncertainties in O₃ *loss_obs* [Schoeberl *et al.*, 1990].

34 Several recent studies suggest that observed rates of
35 chemical loss of Arctic O₃ are considerably faster than
36 expected. Becker *et al.* [1998, 2000] reported that O₃ *loss_obs*
37 for mid-January was more than a factor of two greater than
38 loss rates found using a parcel-trajectory photochemical model.
39 Hansen *et al.* [1997] reported that the accumulated O₃ loss
40 observed at 69.3°N in late March 1996 was ~50% larger than
41 values calculated using a chemical transport model (CTM). A
42 similar discrepancy has been reported based on analyses of O₃
43 from the POAM (Polar Ozone and Aerosol Measurement) II
44 satellite instrument using a different CTM [Deniel *et al.*,
45 1998].

46 Using a combination of data from the Match technique,
47 POAM II, POAM III and the Microwave Limb Sounder
48 (MLS), we show that Arctic ozone loss rates during cold

1 Arctic Januaries are consistently faster than is currently
2 understood. Our study focuses on O₃ loss rates measured by
3 the Match technique [e.g., *Rex et al.*, 1993, 1997, 2002; von
4 der Gathen, 1995] for four cold Arctic winters that underwent
5 significant chemical ozone depletion during January. We use a
6 simple theoretical framework for modeling chemical ozone
7 loss rates [*Salawitch et al.*, 1993] that is based on abundances
8 of ClO specified either from MLS satellite observations
9 [*Santee et al.*, 1996] or by assumptions regarding the level of
10 chlorine activation. We investigate the consistency between
11 O₃ *loss_obs* and O₃ *loss_mdl* for different time periods of Arctic
12 winter.

13 2. Chemical Loss of Arctic Ozone: January

14 Fig. 1 shows values of O₃ *loss_obs* on isentropic surfaces of
15 the lower stratosphere found by the Match technique for four
16 winters that underwent significant chemical loss. These
17 measurements are based on data collected by ozonesondes
18 from dozens of stations in a coordinated manner that allows air
19 masses to be sampled multiple times as they traverse the vortex
20 [e.g., *Rex et al.*, 1998, 1999]. The loss rates are expressed in
21 ppbv/sunlit hour, a convenient way to account for variations in
22 solar insolation. The sunlit times are defined as periods at solar
23 zenith angles (SZA) less than 95°. The discussion in this
24 section focuses on ozone loss rates for January of each winter.

25 Chemical loss of O₃ per sunlit hour peaks in January of all
26 winters due to greater abundances of ClO [*Rex et al.*, 1997,
27 2002]. Data for January 1995 and 2000 are shown for the
28 isentropic surfaces that experienced the largest ozone loss rates
29 (490 and 500 K respectively). For 1992 and 1996, sufficient
30 numbers of ozonesonde observations are not available to
31 precisely define loss rates above 475 K. Therefore, for those
32 years, ozone loss rates at the 475 K level are given in Fig. 1.

33 As noted above, *Becker et al.* [1998, 2000] could not
34 account for the rapidity of ozone loss during January 1992 (at
35 475 K) and January 1995 (at 490 K). They used a Lagrangian
36 photochemical box model with a comprehensive description of
37 gas phase and heterogeneous reactions.

38 To our knowledge, during the cold Arctic Januaries
39 discussed here, measurements of ClO at the required altitudes
40 are not available, or not sufficiently frequent, to constrain
41 model runs along the trajectories used in Match. Therefore we
42 have chosen a different approach. To illustrate the extent of the
43 discrepancy, we have used a photochemical box model to
44 calculate the level of ClO_x (ClO+2×ClOOCl) that would be
45 required to account for the observed O₃ loss rates along Match
46 trajectories. In the model we use a simple theoretical
47 framework for the representation of the diurnal variation of
48 ClO, ClOOCl, OCIO, BrO, BrCl, and atomic O [*Salawitch et*
49 *al.*, 1993] (note 1). The calculations assume constant ClO_x
50 along each Match trajectory, account for the effects of
51 variations in temperature and solar insolation on O₃ *loss_mdl*,
52 and use kinetic parameters from JPL00-3 [*Sander et al.*, 2000]
53 (note 2).

54 The level of ClO_x necessary to account for the observed
55 ozone loss rates in January exceeds 5 ppbv for each winter
56 analyzed. This is larger than 3.7 ppbv, the total amount of
57 inorganic chlorine present in the stratosphere [*WMO*, 1998].

1 Ozone loss rates for January found by assuming ClO_x equals
2 3.7 ppbv are also shown in Fig. 1. The failure to fully account
3 for O_3 *loss_obs*, even assuming complete activation of ClO_x , is
4 robust for reasonable uncertainties in the reaction coefficients
5 of the primary ozone loss cycles ($\text{ClO}+\text{ClO}$ and $\text{BrO}+\text{ClO}$).
6 These analyses suggest that loss of O_3 in January occurs by a
7 process that is not well represented by current photochemical
8 models.

9 The discrepancy between O_3 *loss_obs* and O_3 *loss_mdl* is
10 significant considering the uncertainty in the measured loss
11 rates. The error bars for O_3 *loss_obs* in Fig. 1 represent 1σ
12 statistical uncertainties assuming a Gaussian distribution of the
13 error of individual Match events (note 3). Observed ozone loss
14 rates exceed the maximum possible modeled loss rate
15 (assuming $\text{ClO}_x=3.7$ ppbv) by 2σ to 3σ for late January 1992
16 and by 1σ to 2σ for parts of mid to late January of other years.
17 Our assessment that this discrepancy is significant is based
18 also on the consistent observation of faster than expected
19 ozone loss rates for all cold Januaries during the past decade.
20 Finally, the Match observation of essentially zero rates of
21 chemical O_3 loss for January of warm winters (e.g., 1998 and
22 1999) [Schulz *et al.*, 2001], when higher levels of planetary
23 wave activity pose greater challenges to the Match approach
24 than for cold winters, increases our confidence in the validity
25 of the observed January loss rates shown here.

26 A statistical analysis of the ozonesonde data has been
27 performed to determine whether sunlight exposure is
28 associated with chemical ozone loss. A bivariate linear
29 regression has been applied to the data, allowing for different
30 rates of ozone change for the sunlit and dark (defined as $\text{SZA} >$
31 95°) segments of the trajectories [Rex *et al.*, 1999]. Ozone
32 depletion, expressed in units of ppbv/hr, is found only for the
33 sunlit segments (Fig. 2). No significant change in O_3 is found
34 for the times the airmasses are in complete darkness. The
35 consistency of these results for four winters suggests that the
36 unaccounted for ozone loss process is photolytic.

37 Since Match results are available for a range of potential
38 temperature surfaces, the accumulated loss of ozone can be
39 calculated on surfaces that follow the diabatic descend of air
40 [Rex *et al.*, 1997]. For 1994/1995 and 1999/2000 the range of
41 theta levels for which Match results are available is sufficiently
42 broad to do the accumulation on various descending surfaces,
43 resulting in a vertical profile of the overall loss at the end of
44 January (Fig. 3).

45 Accumulated O_3 loss measured by POAM II and POAM III
46 for January 1995, 1996, and 2000, found by allowing vortex
47 averaged ozone to descend using calculated cooling rates
48 [Hoppel *et al.*, 2002], compares well with Match observations
49 considering the respective uncertainties (Fig. 3).

50 Significant chemical removal of O_3 during January has been
51 reported by other techniques. Accumulated chemical loss of
52 0.5 ppmv of ozone at 465 K was observed by MLS during
53 January 1995 (Fig. 3), in excellent quantitative agreement with
54 ozone reductions found by Match [Harris *et al.*, 2002].

55 In January 2000 ozone loss derived by Match peaks at ~ 0.54
56 ± 0.2 ppmv between potential temperature levels of 480 and
57 520 K. Accumulated loss of O_3 was moderate ($\sim 0.22 \pm 0.13$
58 ppmv) at the 444 K surface, close to the cruise altitude of the

1 NASA ER-2 aircraft during January. Therefore, the finding of
2 little or no chemical loss of ozone (0.0 ± 0.15 ppmv) from ER-
3 2 observations during January 2000 [Richard *et al.*, 2001] is
4 not inconsistent with the analyses presented here.

5 **3. Chemical Loss of Arctic Ozone: February and** 6 **March**

7 Changes in ozone per sunlit hour are smaller in February
8 and March compared to January because of partial recovery of
9 ClO_x to the ClNO_3 reservoir [Rex *et al.*, 1997, 2002]. In this
10 section, we use MLS observations of ClO to calculate loss
11 rates along the Match trajectories, and compare them to Match
12 estimates of O_3 *loss_obs*.

13 1996 is the only year for which Match observations of rapid
14 ozone loss overlapped with sufficiently dense MLS
15 observations of ClO to allow the reconstruction of ClO_x along
16 the match trajectories. MLS observations during rapid ozone
17 loss events in other years were not available due to the
18 monthly yaw of the Upper Atmospheric Research Satellite
19 (UARS) or were not sufficiently dense due to difficulties with
20 the MLS scan mechanism in later years.

21 We have reconstructed the abundance of active chlorine
22 along the Match trajectories by interpolating between mixing
23 ratios of ClO_x that have been inferred from MLS
24 measurements of ClO close to the respective trajectories. We
25 use Version 5 MLS retrievals, which provide a better definition
26 of the vertical distribution of ClO than previous MLS retrievals
27 [Livesey *et al.*, 2002]. O_3 *loss_mdl* was calculated along each
28 Match trajectory at 475 K for this time period. All other
29 assumptions (i.e., BrO_x , O_3) are as previously described.¹

30 Excellent agreement is found between decreases in O_3 along
31 the Match trajectories and modeled loss based on MLS
32 observations of ClO for late February/early March 1996 (Fig.
33 4). The ozone loss rates (e.g., change per sunlit hour) are
34 considerably slower than observed during January.
35 Abundances of ClO_x inferred from MLS ClO along Match
36 trajectories range from ~ 1.2 to 2.6 ppbv. Most importantly, the
37 Match trajectories spend a considerably smaller portion of their
38 overall sunlit time at high SZA (e.g., between ~ 85 and 94°)
39 than is found for the January trajectories.

40 **4. Discussion**

41 We turn our attention to speculation regarding the cause of
42 the rapid ozone loss during January. Standard photochemical
43 models predict relatively slow rates of polar O_3 loss at high
44 SZA (e.g., between 85 and 94°) because strong attenuation of
45 UV light, due to the high O_3 slant columns, limits the
46 photolysis rate of ClOOCl and hence the overall rate of O_3 loss
47 by the $\text{ClO}+\text{ClO}$ and $\text{BrO}+\text{ClO}$ cycles.

48 Prior to the SOLVE/THESEO 2000 field campaign, we had
49 speculated that photolysis of ClOOCl by an unknown state in
50 the near IR (wavelengths > 800 nm, which is optically thin
51 even at high SZA) might provide a strong enhancement to the
52 abundance of Cl and ClO during twilight. Such photolysis
53 could lead to significant increases in chemical ozone loss rates
54 at high SZA. Little change would occur for calculated loss

1 along trajectories in late February and early March because air
2 parcels spend a much smaller fraction of overall sunlit time at
3 high SZA. However, in-situ observations of ClO and ClOOCl,
4 obtained from the NASA ER-2 during SOLVE, provide
5 evidence that ClOOCl does not photolyze at an appreciable
6 rate in the near IR [R. M. Stimpfle, private communication,
7 2002]. Hence, we are left to ponder other possible causes of
8 the observed rapid loss of ozone during January.

9 It is possible that O₃ is lost by reactions on the surface of
10 PSCs (polar stratospheric clouds). The upper limit for the
11 reaction probability of this process, 2.5×10^{-4} on the surface of
12 nitric acid trihydrate [Sander *et al.*, 2000], suggests that this
13 process could contribute significantly to ozone loss in January.
14 For each January considered here, air was exposed to
15 considerable amounts of PSCs during both day and night. Our
16 finding that loss of ozone occurs only during sunlit periods
17 suggests that, if direct loss on PSC surfaces is responsible,
18 such loss must be driven by photons.

19 Observations indicate that BrO does not fall off with
20 increasing SZA near sunset as rapidly as expected [Wahner
21 and Schiller, 1992; Avallone and Toohey, 2001]. It is difficult
22 to reconcile these observations with existing photochemical
23 theory. Nonetheless, it is not clear how enhanced BrO in
24 twilight could lead to appreciable increases in chemical loss
25 rates since ClO is observed to decline with increasing SZA
26 essentially as expected [Kreher *et al.*, 2002; R. M. Stimpfle,
27 private communication, 2002]. A reactive partner is required
28 for the enhanced levels of BrO, since the self-reaction of BrO
29 is too slow to contribute appreciable amounts of ozone loss.
30 Perhaps loosely bound higher oxides of ClOOCl contribute to
31 the rapid ozone loss found in January, either by reaction with
32 BrO or in other yet unidentified ozone loss cycles [Sander *et al.*,
33 1989]. Better understanding of the photochemistry of this
34 time period requires observations at high SZA, and appropriate
35 potential temperature levels (e.g., 480 to 520 K), of BrO, other
36 radicals, and a variety of chlorine species to test the budget and
37 partitioning of halogens in the stratosphere.

38 5. Concluding Remarks

39 We have analyzed data from Match, POAM II, POAM III
40 and MLS to assess our understanding of Arctic ozone loss
41 rates. The consistent inability to fully account for observed
42 ozone loss rates during cold Arctic Januaries suggests the
43 existence of a currently unknown ozone loss mechanism.
44 Detailed data analyses indicate that this loss process involves a
45 photolytic step. Observed ozone loss later during winter (e.g.,
46 mid February to early March) is in good quantitative
47 agreement with model results based on observed ClO,
48 suggesting that the unknown ozone loss mechanism is most
49 important at high SZA and low temperatures typical of January
50 conditions.

51 During cold Arctic Januaries we find cumulative ozone loss
52 of about 0.5 ppmv. Although our and other analyses point
53 toward the existence of a currently unknown ozone loss
54 mechanism, its relative impact is modest for winters with
55 massive ozone depletion. For such winters, loss of ozone
56 predominantly occurs during February and March by known
57 catalytic processes (ClO+ClO and ClO+BrO) that operate

1 efficiently under conditions of high solar illumination.
 2 Nonetheless, the January discrepancy demands further
 3 investigation because reliable assessments of future Arctic
 4 ozone depletion require a full understanding of all significant
 5 processes that affect ozone.

6
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16 Notes

17 1. Values of BrO are found by specifying the sum, BrO+BrCl, as a
 18 function of potential temperature such that observed mixing ratios of
 19 BrO in the Arctic vortex [Harder *et al.*, 1998] are reproduced.
 20 Measurements of O₃ from Match are also specified along each
 21 trajectory.

22
 23 2. Use of the Bloss *et al.* (2001) rate for ClO+ClO+M rather than the
 24 JPL00-3 [Sander *et al.*, 2000] rate has essentially no effect on our
 25 model calculations because a faster rate titrates ClO into ClOOCl.
 26 Hence, the increase in O₃ loss due to the ClO+ClO cycle is nearly
 27 balanced by the calculated decrease due to the BrO+ClO cycle.

28
 29 3. An analysis of the individual errors of the Match events shows that
 30 the distribution of errors is Gaussian [Rex, 1993]. However, based
 31 on the sample size, it is hard to exclude a small, non-Gaussian
 32 component at the far edge (e.g., beyond 2 σ) of the distribution. Since
 33 faster than expected ozone loss rates are observed for all cold
 34 Januaries, it is unlikely that the discrepancy discussed in this paper is
 35 due to a statistical fluctuation of the data.

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41
 42
 43 Figure 1. Chemical loss rate of O₃ (O₃ *loss_obs*) in the Arctic
 44 vortex for indicated years and isentropic surfaces based on the
 45 Match method (red boxes; error bars represent 1σ uncertainty).
 46 The abundance of ClO_x necessary to account for O₃ *loss_obs*
 47 along each trajectory (green dots; see text) for JPL00-3
 48 kinetics is also shown. Also shown is an estimate of O₃ *loss_mdl*
 49 for January of each year assuming ClO_x=3.7 ppbv (blue dashed
 50 line).

51
 52 Figure 2. Rate of change of ozone along Match trajectories for
 53 data collected during sunlit conditions (defined as SZA < 95°)
 54 and during dark periods (SZA > 95°) based on bivariate
 55 regressions for data collected between 5 and 31 January of
 56 each year for the set of matches used in Fig. 1. Error bars are
 57 1σ estimates of the statistical uncertainty. During January 1996
 58 a much smaller number of ozone soundings have been
 59 performed compared to the other years shown here and the
 60 uncertainty of the bivariate analysis is much larger.

61
 62 Figure 3. Accumulated chemical loss of O₃ versus potential
 63 temperature for Januaries of 1992 (black), 1995 (red), 1996
 64 (green), and 2000 (blue). Results from Match analyses are
 65 shown by solid lines with solid markers. The single open
 66 marker represents a result from MLS for 1 to 31 January 1995
 67 [Harris *et al.*, 2002]. The dotted lines represent ozone losses
 68 derived from POAM II and III measurements. For these an
 69 ozone versus PV relation was derived from POAM

1 measurements made during day 32 ± 2 days. The vortex
2 average ozone profile was calculated based on these relations
3 at various heights. The ozone loss was then estimated by
4 comparing this profile with subsided vortex average profiles
5 calculated with the same approach for day 5 ± 2 days. Error
6 bars represent 1σ uncertainties.

7
8 Fig. 4. The chemical loss of O_3 measured by Match between
9 20 February 1996 and 3 March 1996 at 475 K versus the
10 amount of sunlight exposure along each Match trajectory (red
11 squares) and the computed reduction in O_3 along the same
12 Match trajectory based on MLS Version 5 measurements of
13 ClO (green crosses). Error bars represent the 1σ sigma
14 standard deviation of the measured and modeled changes in
15 O_3 .
16

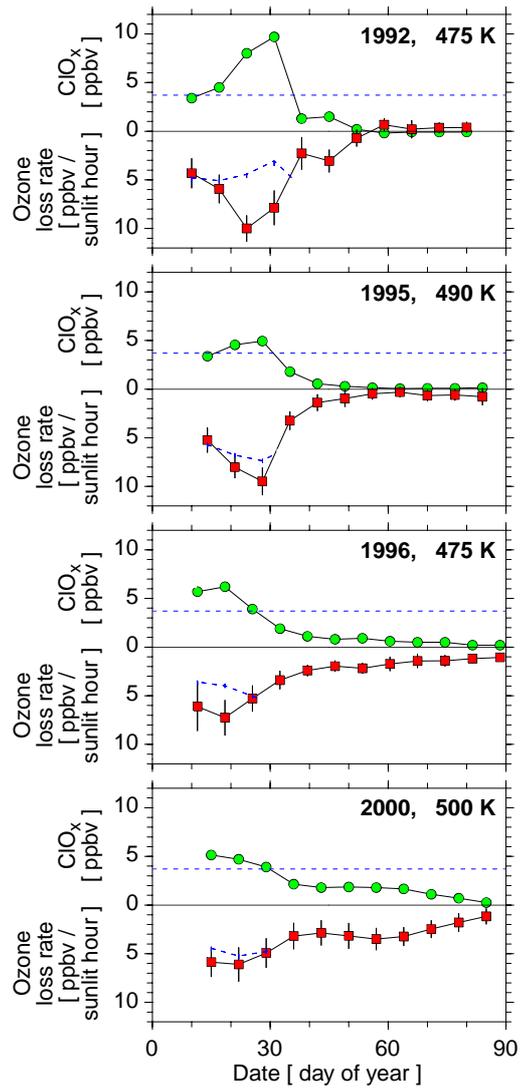


Figure 1, Rex et al.

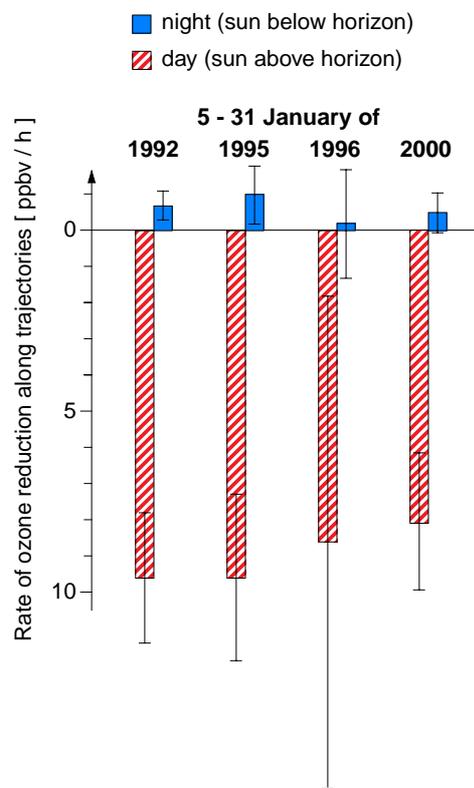


Figure 2, Rex et al.

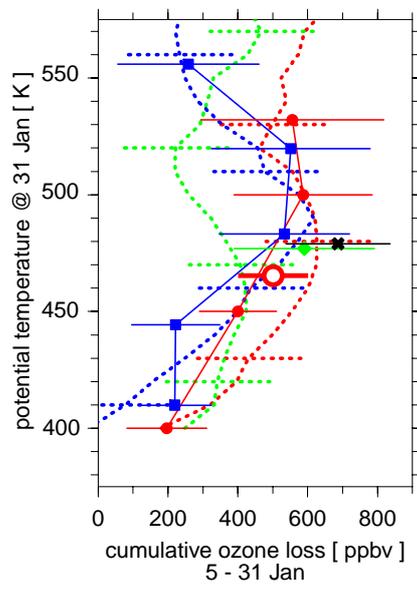


Figure 3, Rex et al.

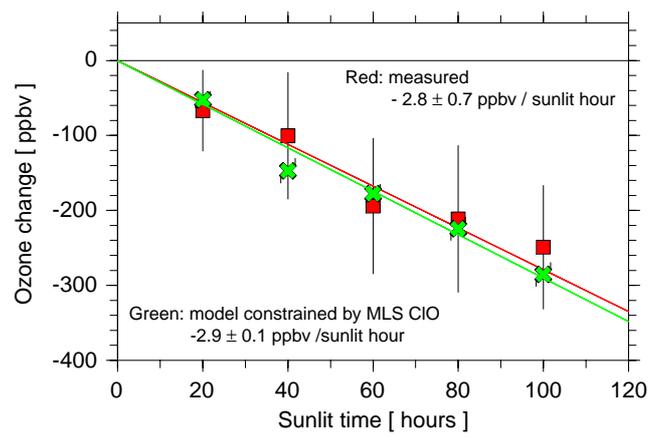


Figure 4, Rex et al.