Arctic ozone loss and climate change

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[1] We report the first empirical quantification of the relation between winter-spring loss of Arctic ozone and changes in stratospheric climate. Our observations show that ~15 DU additional loss of column ozone can be expected per Kelvin cooling of the Arctic lower stratosphere, an impact nearly three times larger than current model simulations suggest. We show that stratospheric climate conditions became significantly more favorable for large Arctic ozone losses over the past four decades; i.e., the maximum potential for formation of polar stratospheric clouds increased steadily by a factor of three. Severe Arctic ozone loss during the past decade occurred as a result of the combined effect of this long-term climate change and the anthropogenic increase in stratospheric halogens. *INDEX TERMS:* 0340 Atmospheric Composition and Structure: Middle atmosphere—composition and chemistry; 0370 Atmospheric Composition and Structure: Volcanic effects (8409); 1620 Global Change: Climate dynamics (3309); 1610 Global Change: Atmosphere (0315, 0325); 3349 Meteorology and Atmospheric Dynamics: Polar meteorology. **Citation:** Rex, M., R. J. Salawitch, P. von der Gathen, N. R. P. Harris, M. P. Chipperfield, and B. Naujokat (2004), Arctic ozone loss and climate change, *Geophys. Res. Lett.*, *31*, L04116, doi:10.1029/2003GL018844.

1. Arctic Ozone Loss During the Past Decade

[2] We have analyzed about 2000 ozonesoundings within the Arctic polar vortex to quantify chemical loss of ozone for ten recent winters. Figure 1 shows the evolution of the vortex averaged ozone profile over the course of these winters.

[3] Air masses inside the polar vortex are relatively isolated from air at lower latitudes and, over the time scale of weeks, they cool radiatively and their potential temperature decreases, inducing downward transport of ozone rich air. To account for this effect, in Figure 1 the ozone mixing ratio has been plotted versus spring equivalent potential temperature ($e\Theta$). $e\Theta$ is the potential temperature a given air mass reached by the end of March. The calculation of $e\Theta$ is based on radiative transport calculations [*Shine*, 1987] as described in *Rex et al.* [2002]. Hence, the changes in ozone profiles shown in Figure 1 during the course of an Arctic winter provide a measure of chemical removal of ozone [*Rex et al.*, 2002]. Chemical ozone loss derived with this approach agrees well (within about 20%) with results from other techniques [*Harris et al.*, 2002].

[4] We have estimated the impact of chemical loss on total column ozone (ΔO_3) from the data in Figure 1. The vortex averaged profiles of ozone loss have been determined as the differences between the end of March (red) and the early January (blue) curves in Figure 1. These have been converted into concentration versus altitude profiles, using the vortex averaged temperature and pressure profiles from late March. The total column loss was calculated as the vertical integral of the loss profiles between 14 and 24 km altitude [see *Rex et al.*, 2002, for details]. The lower limit of this range ($\Theta \approx 380$ K) is close to the bottom of the well isolated part of the polar vortex. The results shown in Figure 1 indicate that for most winters ozone loss at this level is small. Also, the effect of any chemical loss in the vertical region below 14 km on the total ozone column in the Arctic would be limited because of rapid exchange with mid latitude air. Formation of PSCs above the vertical range considered here is unlikely and consequently significant chemical loss of ozone is not expected above 24 km. The estimated uncertainty of ΔO_3 is ~10–15 DU [*Rex et al.*, 2002], mainly due to uncertainties in the calculated cooling rates and potential impact of mixing across the vortex edge. Column ozone loss exhibited significant year-to-year variability during the past decade (Figure 2a, red bars), even though total halogen abundances hardly varied [*WMO*, 2003]. Total column ozone in the Arctic during late March typically ranges from 300–500 DU (average values for the last ten days in March calculated from all ozonesonde observations for the region covered by the polar vortex). Our results indicate that chemical loss of column ozone, which has varied between 0 and about 100 DU during the past decade (Figure 2), has contributed about half of the observed year to year variability of total column ozone.

2. Relation Between Ozone Loss and PSC Formation Potential

[5] We calculated the stratospheric volume where conditions were cold enough for the existence of PSCs from meteorological analyses provided by the European Centre for Medium-Range Weather Forecasts (ECMWF). Figure 2a shows the average value of the potential PSC volume over the period mid-December to end of March (V_{PSC}). V_{PSC} was calculated between $\Theta = 400$ and 550 K using the NAT equilibrium temperature and standard profiles for HNO_3 and H_2O (see *Rex et al.* [2002] for details). Observations of the location of PSCs agree well with this definition for V_{PSC} [*Rex et al.*, 2002]. The degree of Arctic ozone loss and V_{PSC} are closely related (Figure 2b). The slope of a linear fit is

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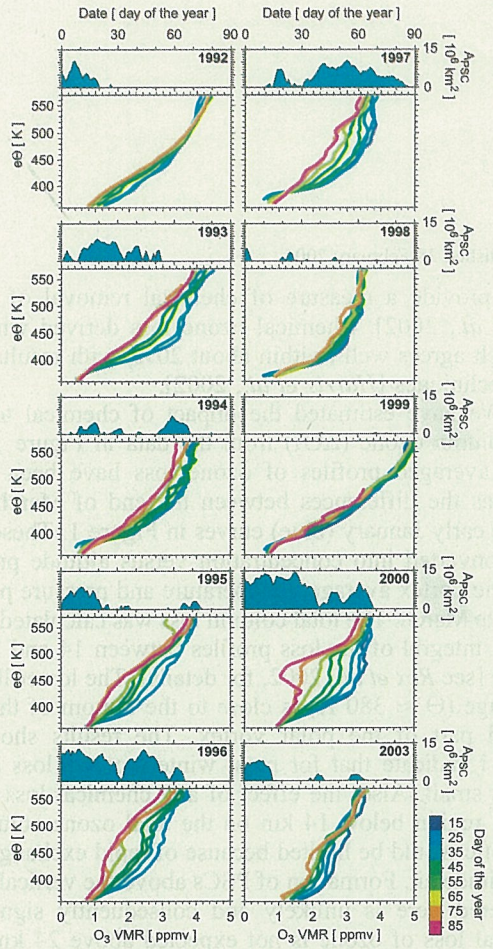

Figure 1. Blue shaded areas: Geographical area covered by temperatures below T_{NAT} (A_{PSC}) on the surface $e\Theta$ = 460 K for ten winters between 1992 and 2003 (for winter 2001 our approach to derive ozone loss is not applicable, because the vortex break up overlapped a period of significant ozone loss; in 2002 the number of ozone sondes launched was not sufficient for our analysis). Colored curves: Evolution of the vortex averaged ozone mixing ratio versus $e\Theta$ through the same winters.

2.1 ± 0.2 DU per 10^6 km^3, and the correlation coefficient is 0.96.

[6] The compactness of the empirical relation between ΔO_3 and V_{PSC} is quite striking. One could expect that ΔO_3 may also be influenced by a variety of other factors, such as the presence of volcanic aerosols [e.g., *Deshler et al.*, 1996], the degree of denitrification [e.g., *Rex et al.*, 1997; *Waibel et al.*, 1999], the timing of the PSC periods during winter, and the location of PSCs within the Arctic vortex.

[7] Large concentrations of volcanic aerosol from the eruption of Mt. Pinatubo were present in the Arctic stratosphere during the first half of the 1990s [*Thomason et al.*, 1997]. We find that the deviations of ΔO_3 from the linear fit tend to relate to the abundance of aerosols, in a manner consistent with expectation and observations from the Antarctic [*Deshler et al.*, 1996]. However the deviations are hardly larger than the estimated uncertainty of the measured ozone loss (inset in Figure 2b). The slope of the fit through the data points is not significantly impacted by the aerosol effect. It becomes 2.2 ± 0.1 DU per 10^6 km^3 if the measured ozone loss is corrected by 14 DU $\times$ log (ρ_a μm^{-2}cm^3), where ρ_a denotes the surface area density of the stratospheric aerosol. The correlation coefficient between the corrected ΔO_3 and V_{PSC} is 0.99.

[8] The observations show that variability in the other conditions mentioned above has not introduced much scatter into the relation between ΔO_3 and V_{PSC}, probably because these factors correlate with V_{PSC} or cancel to a certain degree. For example, the degree of denitrification may correlate with V_{PSC}, because extensive and persistent exposure of air to PSCs is required for Arctic denitrification to occur [*Fahey et al.*, 2001]. The location of the low temperature regions may have a limited impact on ΔO_3. When the low temperatures are near the edge of the polar vortex (baroclinic situation), the lifecycle of individual PSC particles is short and denitrification is less likely [e.g., *Santee et al.*, 1998], reducing ΔO_3. But, in the baroclinic situation, larger fractions of air inside the Arctic vortex circulate through the low temperature region and are processed by PSCs, thus increasing levels of reactive chlorine (ClO_x) and ΔO_3. The timing of the PSC events may have a limited impact because faster ozone loss later during late winter (due to more sunlight) is offset by the more rapid deactivation of ClO_x due to faster photolysis of HNO_3. Quantitative photochemical model studies are required to study these effects and to

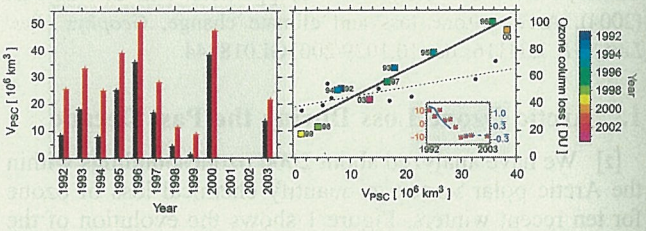

Figure 2. (a) Variation of V_{PSC} (black columns) and ΔO_3 (red columns) through the ten winters shown in Figure 1. ΔO_3 was estimated from the data shown in Figure 1 between day 15 and day 85 of each year. Days 25 and 75 were chosen as start or end date of the integration for years when the well isolated vortex established late (1994, 1999) or broke up early (1992, 1998, 2003). In these cases ozone loss is not expected during the omitted ten days. (b) Scatter plot of ΔO_3 versus V_{PSC}. Measurements are shown by colored squares. Black points are results from the SLIMCAT model (ΔO_3 as calculated from SLIMCAT vs. V_{PSC} based on UK Met Office data that is used in SLIMCAT. The small inset shows the deviations of measured ΔO_3 from the linear fit (in DU; red points, left hand scale). The solid blue line in the inset shows the logarithm of the aerosol surface area density (in μm^{2}cm^{-3}) at 15.5 km estimated from SAGE II data for 60 to 70°N [*Thomason et al.*, 1997]. *Deshler et al.* [1996] showed that the additional polar ozone loss due to the Pinatubo aerosol correlated linearly with the logarithm of the aerosol surface area density. The dashed blue line represents typical background values of surface area estimated from data measured before 1991.

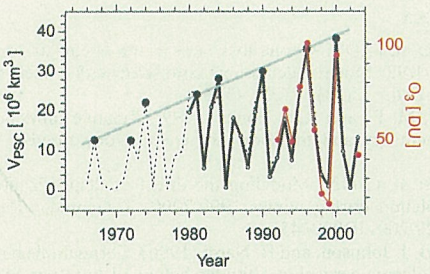

Figure 3. Evolution of V_{PSC} over the past 37 years composed from ECMWF data (solid black line) and data from the FU-Berlin (dashed line). The latter is available only on the 50 and 30 hPa pressure surfaces which are close to the 470 and 540 K potential temperature surfaces. To estimate V_{PSC} from this data, we have used $A_{PSC} = 0.8 \times A_{PSC}(50\ hPa) + 0.2 \times A_{PSC}(30\ hPa)$ to estimate the average PSC extent between between 400 and 550 K, where A_{PSC} denotes the average horizontal extent of temperatures low enough for PSCs to exist. During the overlapping time period, we find that 5.06 km $\times$ A_{PSC} is a very good approximation for V_{PSC} from ECMWF data. The gray line represents a linear fit through the maximum values of V_{PSC} during five year intervals. Also shown is ΔO_3 for the years 1992 to 2003 (red line), scaled such that V_{PSC} represents the best fit through ΔO_3.

fully understand the compactness of the observed relation shown in Figure 2.

3. Impact of Climate Change on Arctic Ozone Loss

[9] The linear quantitative empirical relation between ΔO_3 and V_{PSC} shown in Figure 2b allows us to assess the impact of changes in stratospheric temperature on Arctic ozone loss. For the years shown in Figure 2 we have applied a wide range of temperature offsets to the meteorological data and found that V_{PSC} would have increased by 7.7 $\pm$ 2.6 $\times$ 10^6 km^3 per Kelvin of uniform cooling throughout the vortex. In combination with the slope of ΔO_3 versus V_{PSC} from Figure 2b, this implies that about 15 DU additional ozone loss can be expected per Kelvin average cooling of the Arctic stratosphere. This is the first empirical measure of the relation between enhanced polar ozone loss and stratospheric cooling induced by climate change, and is only valid for the range of V_{PSC} shown in Figure 2.

[10] The empirical relation between ΔO_3 and V_{PSC} represents a principal diagnostic for assessing the ability of models to predict the evolution of polar ozone under different climate scenarios. This diagnostic allows the validation of both full chemistry chemical transport models (CTMs) and coupled chemistry-climate general circulation models (CCMs). The diagnostic can also be used to calibrate parametrizations of polar ozone loss used in CCMs with simplified chemistry.

[11] Calculations of ΔO_3 versus V_{PSC} from the SLIMCAT CTM [*Chipperfield and Jones*, 1999] for the winters 1992 to 2000 are compared to the observed relation in Figure 2b. Currently, many models tend to underestimate the chemical loss of Arctic ozone during cold winters [*WMO*, 2003], an effect most likely related to problems with the current schemes used to parametrize denitrification [*Davies et al.*, 2002] and to unexplained ozone losses during cold Arctic Januaries [*Becker et al.*, 1998; *Rex et al.*, 2003]. Conversely, the SLIMCAT model overestimates ozone loss during warm winters, suggesting that the effect of small, localized areas of temperatures below the thermodynamic PSC existence threshold is overestimated in the model, because of the equilibrium treatment of PSC occurrence (i.e., PSCs are instantaneously formed in the model once temperatures fall below T_{NAT}). The slope of a fit through the SLIMCAT model results for ΔO_3 vs. V_{PSC} is 0.71 $\pm$ 0.23 DU per 10^6 km^{-3}, a value nearly a factor of three smaller than observed. The model suggests only about 5.5 DU additional ozone loss would occur per Kelvin cooling of the Arctic stratosphere. Our observations indicate that the impact of potential, future stratospheric cooling on Arctic ozone loss is about three times larger than this model value.

[12] The observations also indicate that chemical loss is a significant cause of the observed year to year variability of the Arctic ozone column. Based on SLIMCAT results *Chipperfield and Jones* [1999] concluded that, compared to dynamical variability, year-to-year changes in Arctic chemical ozone loss does not play a major role in the interannual variability of late winter column ozone. In contrast, the observations presented here indicate that chemistry contributed about half of the interannual variability. Figure 2 shows that the conclusion of Chipperfield and Jones is affected by the fact that SLIMCAT largely underestimates the year to year variability in the chemical loss term.

[13] The long term evolution of the climate in the Arctic stratosphere has been the subject of active recent research [e.g., *Ramaswamy et al.*, 2001]. These studies generally focused on minimum or average temperatures and concluded that observations indicate a slight cooling over the past decades, which is barely significant. *Pawson and Naujokat* [1999] found that the average area extent of PSC conditions has been increasing over the past decades. Our results show that V_{PSC} is the relevant climate parameter that drives the evolution of polar ozone loss.

[14] Figure 3 shows a time series for V_{PSC} over the past 38 years, based on meteorological data from the ECMWF and from the FU-Berlin stratospheric analyses, together with the evolution of ΔO_3 over the past decade. The severity of extreme ozone loss events in the Arctic is controlled by the cold Arctic winters; i.e., those that define the upper limit of the year-to-year variability of V_{PSC}. These winters reveal significant change in Arctic stratospheric climate over the past four decades. The value of maximum V_{PSC} during five year intervals has increased steadily, by about a factor of three, since the late 1960s. A linear fit through the solid points in Figure 3 has a slope of 7.86 $\pm$ 0.86 $\times$ 10^6 km^3 per decade. The basic conclusion of a large, steady rise in the maximum value of V_{PSC} does not depend on the length of the time interval. The slope is 6.16 $\pm$ 1.9, 7.88 $\pm$ 0.96 and 7.38 $\pm$ 1.30 $\times$ 10^6 km^3 per decade if maximum values of V_{PSC} during 4, 6, and 10 year intervals are chosen instead. Also, the slope does not significantly depend on the end points of the analyzed periods or potential differences of the two meteorological data sets; e.g., the slopes are 9.3 $\pm$ 2.8 and 6.6 $\pm$ 1.3 $\times$ 10^6 km^3 per

decade for the first half (based on FU-Berlin data alone) and the second half (based on ECMWF data alone) of the period respectively. These results demonstrate that the severe Arctic ozone losses reported for some winters during the 1990s [*WMO*, 2003] were not only the result of increased stratospheric halogen loading compared to earlier decades, but also resulted from a long term change in the climate of the Arctic stratosphere.

[15] It is currently not clear what caused the change in climatic conditions in the Arctic polar vortex. Cooling of the polar stratosphere is qualitatively consistent with the direct radiative impact from increased levels of greenhouse gases (GHG) in the atmosphere [e.g., *WMO*, 2003]. However, the indirect effect of changes in the dynamical structure of the stratosphere related to increased GHG forcing also may impact the wintertime polar temperatures [e.g., *Shindell et al.*, 1998; *Langematz*, 2000]. The range of results from current climate models used to study the effect of increasing GHG forcing on the temperatures in the polar stratosphere is broad, ranging from a slight increase in temperatures to severe cooling [*WMO*, 2003]. Hence, it is currently not possibly to unambiguously attribute the climate change observed in Figure 3 to increased GHG forcing. Another factor that could play a role is long term internal variability of the climate system. Model calculations suggest, however, that changes in radiative forcing due to decreased ozone alone are not sufficient to cause the observed decline in the temperature of the Arctic lower stratosphere [*Rosier and Shine*, 2000; *Langematz*, 2000].

[16] The empirical relation between ΔO_3 and V_{PSC} presented here does not appear to be well represented by current chemistry models. The relation between ΔO_3 and V_{PSC} may not be valid for larger values of V_{PSC} than have been observed. It is therefore imperative to develop a theoretical understanding of this relation as a prerequisite for reliably predicting the future evolution of Arctic ozone in a changing stratosphere characterized by rising abundances of GHGs and declining levels of chlorine.

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