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Supporting Online Material for  
**The Sensitivity of Polar Ozone Depletion  
to Proposed Geoengineering Schemes**

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## Online Supporting Material:

### The sensitivity of polar ozone depletion to proposed Geo-engineering schemes

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#### **The surface area density (SAD) of liquid-phase sulfate aerosols in polar regions:**

The volcanic eruption of Mt Pinatubo in 1991 resulted in a large injection of sulfur in the tropical stratosphere and strongly increased the surface area density (SAD) of liquid-phase sulfate aerosols. Stratospheric transport processes distributed these aerosols towards the polar regions in the following months. Strongly enhanced SAD values were observed in the years immediately following the eruption in the entire stratosphere (see the Assessment of Stratospheric Aerosol Properties (ASAP) (*S1*)). Fig. S1 shows SAD values averaged over high northern and southern latitudes and between 375 and 600 K potential temperature for the years 1984-2001. Large SAD values of more than 20  $\mu\text{m}^2/\text{cm}^3$  were observed in 1992 and 1993 at high latitude regions of both hemispheres and enhanced SAD values were present up to 1995, which increased chlorine activation and therefore chemical ozone destruction.

#### **Relationship between column ozone loss (DU) and PACl:**

The relationship between chemical ozone depletion and the potential for chlorine activation (PACl, as defined in the main paper) for Arctic and Antarctica for the years between 1992 and 2005 is shown in Fig. 2S.

Chemical ozone loss for the Arctic and Antarctica was derived for winters between 1992 (*S2*) and 2003 (*S3*) based on satellite and aircraft observations and for the Arctic winter of 2005 (*S4*, *S5*), for altitudes between 380 and 550 K potential temperature, using tracer-tracer correlations (Fig S2, grey triangles). PACl was derived using MetO meteorological analyses (*S6*). As in earlier studies (*S7*, *S8*), the relation for the Arctic does not correspond to zero ozone loss when PACl reaches zero. In the Arctic, Lee-wave induced chlorine activation occurs even when synoptic temperatures, on which PACl is based, do not reach the PSC threshold (*S9*).

For Antarctica, chemical ozone loss is saturated during the last 10-15 years (grey solid squares). To derive a linear relationship between ozone loss and PACl for the years between 1960 and 1990 we use results from the Whole Atmosphere Chemistry Climate Model (WACCM3) that allowed for changing halogen content of the stratosphere

between 1960 and present. Tilmes et al. (S10) have shown that WACCM3 Antarctic ozone loss in the center of the polar vortex is realistically simulated compared to recent observations. The slope of this relationship is mainly affected by the temporal development of EESC in the stratosphere.

The chemical loss of column ozone for the Antarctic is significantly less than for the Arctic, for equal values of PACI. For Antarctica, we expect less strong Lee wave activity compared to the Arctic. Further, the Arctic vortex is dynamically more active, which results in a larger fraction of the vortex exposed solar radiation than in the Antarctic vortex. Three-dimensional chemistry and transport models simulate well the near linear relation between chemical loss of column ozone in the *Arctic* and  $V_{\text{PSC}}$ , the volume of air below PSC threshold temperatures over the course of winter (S11). The quantity  $V_{\text{PSC}}$  is closely related to PACI. Also, models simulate reasonably well the temporal evolution of the *Antarctic* ozone hole (S12). Hence, the different relations between chemical ozone loss and PACI evident in the two hemispheres appear to reflect differences in ozone availability and the timing and locations of chlorine activation.

It would be desirable to include another volcanic period to corroborate the validity of the relation between ozone loss and PACI for the Arctic. El Chichón, which erupted in 1982, imparted a considerable impact on the stratospheric aerosol loading. Satellite observations provide a record of the perturbation to total column ozone of enhanced aerosols associated with El Chichón. However, observations needed to quantify chemical loss of column ozone (i.e., to separate dynamical and chemical effects on total column ozone) did not become available until 1991, when the HALOE instrument was launched onboard the NASA UARS platform. Hence, we are not able to conduct an analysis of  $\text{O}_3$  loss vs. PACI for the El Chichón time period.

The linear relation between PACI and chemical ozone loss breaks down when the ozone loss is saturated and all the ozone in the partial column that is exposed to cold conditions is destroyed. Saturation of ozone loss occurs in the Antarctic at about 150 DU (Fig. S2). For the Arctic, more efficient poleward transport of ozone results in about 200 and 250 DU of ozone being present before the onset of chemical loss (3). Hence, for the Arctic, considerably more ozone has to be destroyed before saturation of ozone loss occurs (open triangles).

The impact on ozone loss of a hypothetical geo-engineering in past Arctic winters for `GeoEng_Large_Aerosol` and `GeoEng_Small_Aerosol` (as defined in the main text) is illustrated in Fig S2. The abscissa of the solid black triangles is the value of PACI for contemporary levels of chlorine and for a very cold Arctic winter. The ordinate is based on an extrapolation of the linear relationship towards higher PACI values. We assume that the relation between Arctic ozone loss and PACI will remain linear until saturation is reached, which is borne out by our model calculations for the Antarctic (small solid squares, Fig. S2). The figure shows that, for a small aerosol approach to geo-engineering, the entire available column of Arctic ozone in the lower polar stratosphere might be depleted during a very cold winter (see discussion below). A large aerosol

approach would result in slightly less depletion, but the estimated loss, about 180 DU, approaches the entire available column abundance of ozone.

### **The impact of geo-engineering of observed meteorological conditions and past stratospheric halogen content:**

Enhanced SAD values in the stratosphere caused by man-made sulfur injection can strongly increase PACI compared to background conditions, depending on the prevailing meteorological conditions. Here, we calculate PACI time series for the Arctic and Antarctica, between 1992 and 2006 (Fig. S3a), for the following three hypothetical SAD scenarios: the background case, the GeoEng\_Large\_Aerosol case and the GeoEng\_Small\_Aerosol case. Further, we show PACI and chemical ozone loss based on the observed aerosol record from the Assessment of Stratospheric Aerosol Properties (ASAP).

Figure S3b shows chemical loss of Arctic ozone derived using the empirical relationship illustrated in Fig. 1b, extrapolated to higher level of PACI (Fig. S2). Antarctic ozone losses are not shown because, for contemporary levels of EESC, Antarctic ozone loss has “saturated” at near its maximum level. For all aerosol cases, PACI and chemical ozone loss for the Arctic vary strongly due to inter-annual temperature differences. The impact of the eruption of Mt. Pinatubo in 1991 is most pronounced for the moderately cold Arctic winter of 1993 and for the Antarctic winter of 1992 (Fig. S3a, dotted lines). Values of PACI for background SAD ( $PACI_{BACKGROUND}$ ) do not exceed 20%, even for cold Arctic winters (e.g., 2005), but values of PACI for the enhanced SAD cases are significantly larger, except for very warm Arctic winters. For the cold Arctic winters 1995, 1996, 2000, and 2005, PACI is more than three times larger than background values for the small aerosol geo-engineering scenario and about twice background for the large aerosol geo-engineering case.

The enhanced values of PACI associated with the small aerosol geo-engineering scenario would have resulted in ozone loss of 210-240 DU in 4 out of the past 15 Arctic winters (Fig. S3b, dashed dotted lines). This is about the entire amount of ozone in the lower Arctic stratosphere. Had the sulfate layer been enhanced by geo-engineering during these winters, ozone losses far in excess of the observed loss would have occurred. Moderately cold winters (e.g., 1993, 2003) would have 3 to 4 times larger values of PACI for geo-engineering cases compared to  $PACI_{BACKGROUND}$ . In this case, chemical ozone loss would be comparable to the loss observed for the very cold Arctic winter 2005.

For Antarctica, PACI would increase by 25% for large aerosol geo-engineering and by about 40% for the small aerosol geo-engineering (Fig. S3a). The much smaller variability of Antarctic PACI, compared to the Arctic, is due to the low values and small interannual variability of temperature in the Southern Hemisphere stratosphere. Antarctic chemical ozone loss is saturated in the lower stratosphere for the period under consideration and, therefore, ozone loss cannot exceed the observed value of ~150 DU (Fig. S2) unless geo-engineering were to result in an increase in the vertical extent of the ozone depletion region (see main text).

### **SOM References and Notes**

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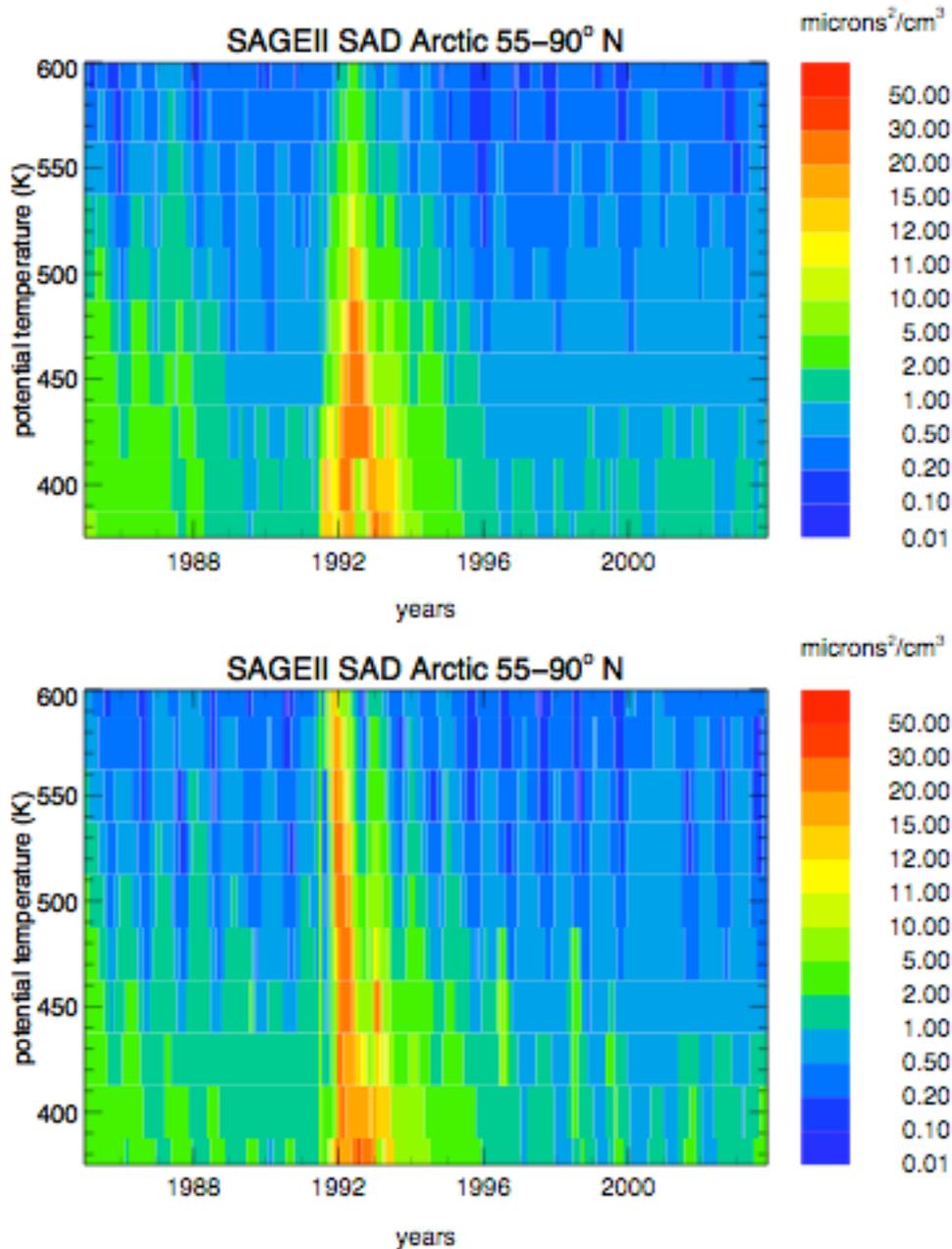


Fig. S1:  
 The surface area densities of liquid-phase aerosols (SAD) in  $\text{microns}^2/\text{cm}^3$  for high northern (top panel) and southern (bottom panel) latitudes between 1984 and 2001 plotted versus potential temperature, using the gap-filled SAD data set of monthly averaged SAD values produced by the Assessment of Stratospheric Aerosol Properties (ASAP) (1).

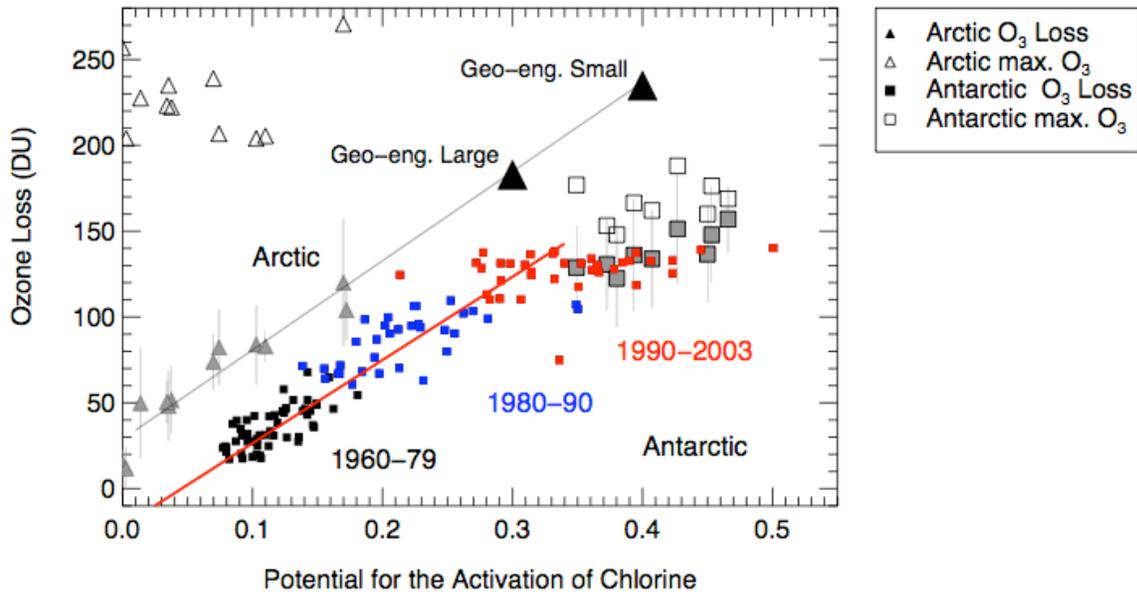


Fig. S2:

Relationship between column ozone loss (DU) and PACI in the Arctic and Antarctic polar vortex core for the years 1992 to 2005 derived from measurements. Monthly averaged ozone loss inside the vortex core in March in the northern hemisphere (gray solid triangles) and October in the southern hemisphere (gray solid squares) between 350 and 550 potential temperature, using the tracer-tracer method; and ozone loss in the Southern Hemisphere from WACCM3 simulations since 1990 (red filled circles), between 1980-90 (blue filled circles) and between 1960-80 (black filled circles). Correspondingly, the column in proxy ozone (ozone without chemical change) is shown for each winter from 1992 to 2005 using open symbols (triangles for northern hemisphere; squares for southern hemisphere) (adapted from Tilmes et al. 2007 (10), Figure 17). The grey line describes the linear fit for the Arctic based on observations (main Paper, Fig. 1b). The red line describes the linear fit for Antarctica based on WACCM3 model results. Maximum values of past Arctic ozone loss for the two different geo-engineering schemes are illustrated as large black triangles.

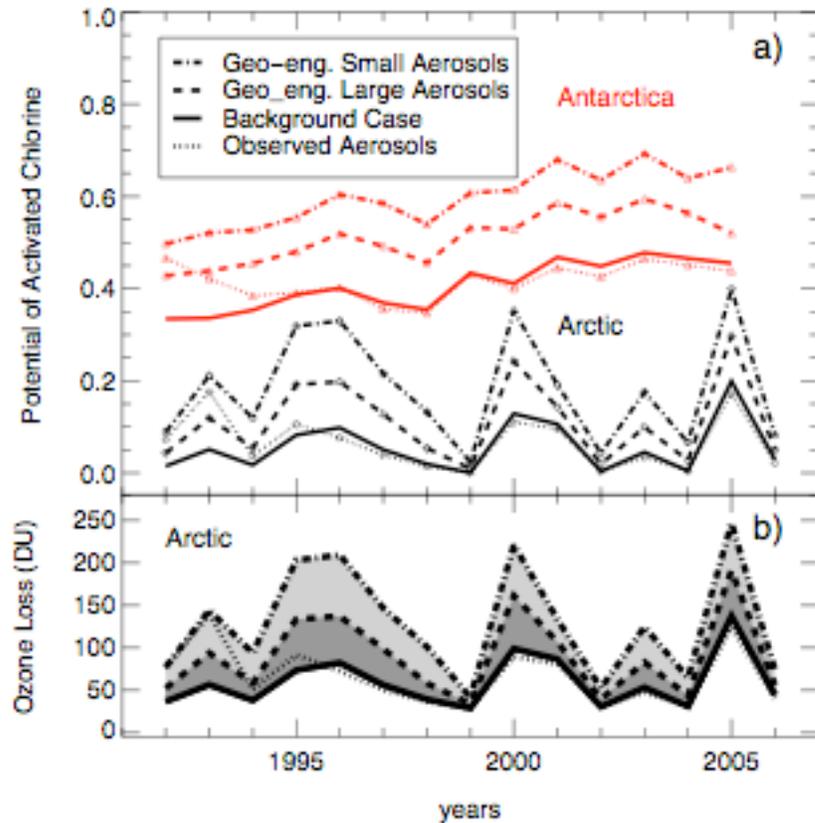


Fig.S3

Sensitivity of PACI for the Arctic and Antarctic (panel a) and Arctic ozone loss (panel b) to aerosol loading, for meteorological conditions between 1992 and 2006 from United Kingdom Meteorological Office analyses (6). Results for four aerosol cases are shown: observed aerosols, background aerosols (i.e., no volcanic or geo-engineering perturbation), and two geo-engineering schemes (small and large aerosols), applied hypothetically to past atmospheric conditions. The Arctic ozone loss estimates are found using the relation between chemical ozone loss and PACI shown in Fig. S2.