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Sensitivity of Arctic ozone loss to stratospheric H_2O

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[1] Likely causes of a future increase in stratospheric H_2O are a rise in tropospheric CH_4 and H_2 leakages from an increased integration of hydrogen into the energy supply system. Here we evaluate the impact of potential future stratospheric H_2O increases on Arctic ozone loss by comparing ozone loss proxies based on two different mechanisms of chlorine activation. In particular, the H_2O dependence of the volume of air is analyzed where temperatures are low enough to form nitric acid trihydrate, denoted as V_{PSC}, and for Cl activation on liquid sulfate aerosols, denoted as V_{ACl}. We show that V_{ACl} increases faster than V_{PSC} with increasing H_2O mixing ratios in the altitude range of 400 K to 550 K potential temperature. As a consequence, the additional ozone column loss is expected to be most pronounced for cold winters and large H_2O increases and to be significantly higher when V_{ACl} is used as a proxy. **Citation:** Feck, T., J.-U. Grooß, and M. Riese (2008), Sensitivity of Arctic ozone loss to stratospheric H_2O, *Geophys. Res. Lett.*, *35*, L01803, doi:10.1029/2007GL031334.

1. Introduction

[2] Polar stratospheric ozone depletion is driven by temperature-dependent processes that lead to the release of active chlorine (Cl, ClO, Cl_2O_2, Cl_2) from its reservoir species (HCl, $ClONO_2$). Hence, the understanding of the relationship between temperature and ozone loss is one of the key parameters for the estimation of future ozone depletion. Today it is assumed that chlorine activation is connected with the existence of polar stratospheric clouds (PSCs) that consist either of nitric acid trihydrate (NAT) or supercooled ternary solutions (STS). PSCs are present in the winter atmosphere below the NAT threshold temperature T_{NAT} at about 195 K [*Hanson and Mauersberger*, 1988]. Because it is generally accepted that chlorine activation also occurs at about 195 K, the formation of NAT is commonly regarded as the dominant process in the activation of chlorine [e.g., *World Meteorological Organization* (*WMO*), 2003].

[3] *Rex et al.* [2004] found an empirical relationship between the Arctic ozone column loss and the total volume of air exposed to temperatures below T_{NAT}, denoted by them as V_{PSC}, which was confirmed by *Tilmes et al.* [2004] in an independent study. Interestingly, ozone column loss and V_{PSC} show a compact linear correlation. Therefore, V_{PSC} seems to be a convenient proxy for ozone loss. *Rex et al.* [2004] noted that their findings were quite surprising since other factors like the degree of denitrification in the Arctic vortex, the timing of the PSC periods during winter, the location of PSCs within the Arctic vortex and the stratospheric sulfate aerosol loading should also influence the ozone loss.

[4] A recent study (K. Drdla, Temperature thresholds for polar stratospheric ozone loss, submitted to *Science*, 2007, hereinafter referred to as Drdla, submitted manuscript, 2007; see auxiliary materials) investigated Cl activation on liquid sulfate aerosols and solid aerosols in a more comprehensive way. Drdla showed that Cl activation is sufficiently fast on liquid sulfate aerosols and that the threshold temperature for Cl activation T_{ACl} ($\approx$195 K) depends only on the potential temperature, the sulfate aerosol surface area density (SAD) and the H_2O mixing ratio. Therefore Drdla concluded that Cl activation is solely controlled by liquid sulfate aerosols. Drdla also demonstrated that T_{NAT} roughly coincides with T_{ACl} under current atmospheric conditions and concluded that T_{NAT} only fortuitously describes both PSC formation and Cl activation. Because atmospheric conditions are likely to change in future it is important to know whether either liquid sulfate or solid aerosols dominate Cl activation since this could lead to significant differences in the prediction of future polar ozone loss.

[5] Stratospheric H_2O, for instance, is predicted to increase as a result of increased tropospheric CH_4 levels [*Röckmann et al.*, 2004] and H_2 leakages from an increased integration of hydrogen into the energy supply system [*Schultz et al.*, 2003; *Tromp et al.*, 2003; *Warwick et al.*, 2004]. There are also variations in the stratospheric H_2O mixing ratio that might be due to changes in the tropopause temperature or the tropical upwelling circulation [*Randel et al.*, 2006]. Box model calculations by *Röckmann et al.* [2004] indicate that the projected increase of tropospheric methane by WMO scenarios will lead to an increase of the upper stratospheric H_2O mixing ratio of about 0.4 ppmv by 2050. A similar increase was proposed by *Riese et al.* [2006] as a realistic upper limit of the impact of a H_2 economy.

2. Impact of Future H_2O Levels

[6] Here we analyze the impact of stratospheric H_2O increases on two different mechanisms of chlorine activation and the consequences for the prediction of Arctic ozone loss on the basis of ECMWF operational analysis of the past two decades. Instead of performing simulations with a global 3-D model, which tends to have difficulty in reproducing polar ozone loss [e.g., *Austin et al.*, 2003], we use proxy-ozone column loss correlations as, for example, given by *Rex et al.* [2004]. Therefore we investigate the impact of H_2O increases on the evolution of the total volume of air exposed to temperatures below T_{NAT} (V_{PSC}) and T_{ACl} (V_{ACl}). Both V_{PSC} and V_{ACl} are used as proxies to estimate

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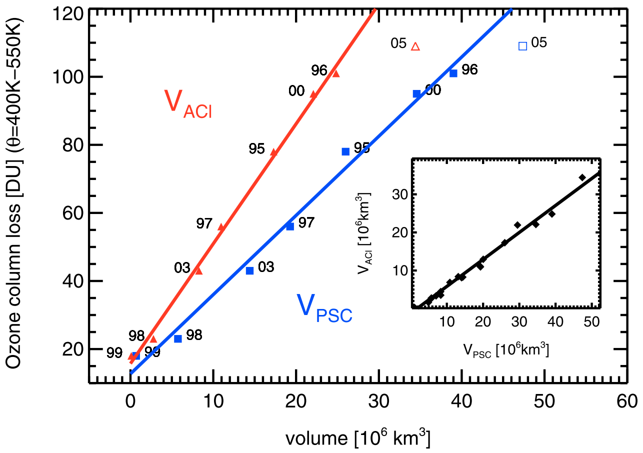

Figure 1. Relation of averaged ozone column loss in Dobson units (DU) versus V_{ACl} and V_{PSC} for the Arctic winters 1995 to 2000, 2003 and 2005. Blue and red solid lines represent the corresponding linear fits for all winters except 2005. According to *Rex et al.* [2006] this winter is characterized by an unusual meteorology, which caused significant ozone depletion below 400 K potential temperature. The small inset shows a linear and compact correlation for V_{PSC} and V_{ACl} for all Arctic winters between 1984 and 2005.

possible additional Arctic ozone column losses in the altitude range of 400 K to 550 K potential temperature due to a H_2O increase. Ozone column loss values are taken from *Rex et al.* [2006] and were derived by the so-called vortex average method. Hydrogen leakage scenarios of a future H_2 economy served as the basis for simulations of potential future stratospheric H_2O increases due to an increase of tropospheric H_2 [*Schultz et al.*, 2003; *Tromp et al.*, 2003; *Warwick et al.*, 2004].

[7] Because in this study we consciously concentrate on a single effect of a complex problem it should be pointed out that our study has certain limitations. It does not account for dynamical feedbacks of a potential H_2O increase. Moreover, neither the emission trade-offs that would be associated with the H_2 economy (e.g. decline of CO_2 and CH_4 emissions) nor the declining levels of anthropogenic halogens that would eventually reduce chemical ozone loss are considered. Furthermore, it should be noted that it is generally assumed that a global H_2 economy is unlikely to be put in place while stratospheric chlorine mixing ratios are high. This certainly has an impact on the proxy/O_3-loss correlations that are presented here.

2.1. V_{PSC} and V_{ACl} Under Current Conditions

[8] We computed V_{PSC} and V_{ACl} from ECMWF operational analysis for the period 1984 to 2005. Both V_{PSC} and V_{ACl} are calculated between 400 K and 550 K potential temperature and averaged over the period from mid-December to the end of March using the NAT equilibrium temperature T_{NAT} [*Hanson and Mauersberger*, 1988] and the Cl activation threshold T_{ACl} (Drdla, submitted manuscript, 2007), respectively. In both cases, we apply the same Arctic winter H_2O profiles to our calculations. These are based on the H_2O climatology compiled by *Grooß and Russell* [2005] from a data analysis of the Halogen Occultation Experiment (HALOE) and averaged over equivalent latitudes ≥65°N. According to the method used by *Rex et al.* [2004] for the calculation of T_{NAT}, we select an Arctic vortex average HNO_3 winter profile derived from a simulation without denitrification [*Grooß et al.*, 2005].

[9] Because no observation dataset is available that provides a detailed SAD climatology for the Arctic polar vortex, we use an average stratospheric SAD background value of 0.5 $\mu m^2\ cm^{-3}$ for the calculation of T_{ACl}. This value is extracted from a dataset provided by D. B. Considine [*Stratospheric Process and Their Role in Climate*, 2006]. Higher SAD values are not considered here due to the unpredictability of future volcanic eruptions. For current H_2O mixing ratios and background SAD, the quantities of V_{PSC} and V_{ACl} derived in this way show a linear and compact correlation (see Figure 1, inset). The slope of a linear fit is 0.71 ± 0.02 · 10^6 km^3 V_{ACl} per 10^6 km^3 V_{PSC} with a correlation coefficient (r) of 0.994.

2.2. Arctic O_3 Loss Versus V_{PSC} and V_{ACl}

[10] In accordance with the empirical method used by *Rex et al.* [2004], we plotted Arctic ozone column loss data derived by *Rex et al.* [2006] on the basis of the vortex average method against V_{PSC} and V_{ACl}. Figure 1 shows the corresponding correlation for the years 1995 to 2000, 2003 and 2005. Data points for 1992 to 1994 are not shown in Figure 1 as the Mount Pinatubo eruption in 1991 caused significantly higher stratospheric SAD values. Data points for 2001 and 2002 are omitted since these winters were characterized by a weak vortex and high temperatures and therefore the ozone loss was low. The winter 2005 is shown in Figure 1, but has not been considered for the linear interpolation since this winter was characterized by an unusual meteorology, which caused significant ozone depletion below 400 K potential temperature [*Rex et al.*, 2006]. For all other winters ozone column loss shows a good linear correlation with both proxies V_{PSC} and V_{ACl}, but with different slopes due to the differently defined temperature thresholds (see Figure S1 of the auxiliary materials).[1] The slope of a linear fit in the first case (V_{ACl}, red solid line) is 3.52 ± 0.10 DU per 10^6 km^3 V_{ACl} (r = 0.998), whereas the linear fit in the second case (V_{PSC}, blue solid line) is 2.33 ± 0.11 DU per 10^6 km^3 V_{PSC} (r = 0.995). This difference would become important if a stratospheric H_2O increase were to lead to a decoupled evolution of V_{PSC} and V_{ACl} and, consequently, to a divergence of the compact correlation described in subsection 2.1.

2.3. Stratospheric H_2 and H_2O Scenarios

[11] We selected four H_2 scenarios that reflect the ongoing discussion about the potential increase of atmospheric H_2 due to leakages caused by a future hydrogen economy [*Schultz et al.*, 2003; *Tromp et al.*, 2003; *Warwick et al.*, 2004]. These scenarios are listed in Table 1, whereby S1–S3 are assumed to be the realistic lower (S1) and upper limits (S2 and S3) of a H_2 economy, respectively. It has been asserted that S4 is unrealistic due to the high leakage rate assumption [*Prather*, 2003]. However, as an extreme assumption we also take the latter scenario into account.

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[1] Auxiliary materials are available in the HTML. doi:10.1029/2007GL031334.

Table 1. Box Model Initial Values for Hydrogen at 15°N and 100 hPa for All H_2 Scenarios and the Corresponding Simulation Results of the Absolute and Relative Arctic H_2O Increase Averaged Over Equivalent Latitudes $\geq$65°N, From 400 K to 550 K Potential Temperature and From December to March[a]

			ΔH_2O	
Scenario	Reference	H_2, ppmv	ppmv	%
Reference case		0.56	0.	0.
S1	*Schultz et al.* [2003]	0.65	0.03	0.6
S2	*Schultz et al.* [2003]	1.10	0.18	3.6
S3	*Warwick et al.* [2004]	1.40	0.27	5.6
S4	*Tromp et al.* [2003]	2.30	0.58	11.8

[a] Hydrogen, H_2; Arctic H_2O increase, ΔH_2O.

[12] The relative importance of increased H_2 levels for a potential increase in stratospheric H_2O is analyzed using a box model, which consists of the chemistry module of the Chemical Lagrangian Model of the Stratosphere (CLaMS) [*McKenna et al.*, 2002]. In our analysis we follow the procedure suggested by *Röckmann et al.* [2004]. According to this procedure, we carried out CLaMS box simulations with a single air parcel that ascends from the tropical stratosphere (15°N, 100 hPa) to 1 hPa along an idealized trajectory over a period of 4 years, in accordance with the so-called tape recorder effect noted by *Mote et al.* [1996].

[13] The air parcel was initialized by applying a typical chemical composition of air masses near the tropical tropopause based on HALOE data for H_2O and CH_4 [*Grooß and Russell*, 2005] and simulations of the Mainz 2-D model [*Grooß*, 1996]. Table 1 shows the averaged (Θ = 400–550 K, Dec.–Mar.) Arctic H_2O increase (ΔH_2O) for the different hydrogen scenarios. We translate the results of this tropical box model simulation to polar latitudes by the following assumptions. Air masses with equal CH_4 mixing ratios encounter a comparable state of hydrogen oxidation regardless of whether they were transported poleward by the global-scale mean circulation or whether they stayed in the tropics. At Arctic altitudes where chemical ozone depletion occurs ($\Theta \approx 400-550$ K) we find typical CH_4 mixing ratios from 0.67 to 1.26 ppmv in the HALOE climatology. This corresponds to an altitude range from 2.7 to 5.8 hPa in the tropical simulation. The simulations with different H_2 mixing ratios indicate that in this altitude range 21% to 52% of H_2 has been converted to H_2O. The applicability of this method was confirmed by comparing CH_4-H_2-H_2O correlations for different trajectories. The results show that the pathway of an air parcel and the H_2 initial value have only a minor impact ($\leq$1%) on the correlations.

2.4. Impact of H_2O Increase on V_{PSC} and V_{ACl}

[14] Based on these assumptions, we modify Arctic HALOE H_2O profiles to account for the effects of air masses with increased tropical H_2 mixing ratios that enter the stratosphere through the tropical tropopause. These profiles are used to recalculate the corresponding average V_{PSC} and V_{ACl} values for all hydrogen scenarios as listed in Table 1. Figure 2 shows the corresponding absolute (Figure 2, left) and relative increase (Figure 2, right) of V_{PSC} and V_{ACl} versus the average ΔH_2O increase for three winters that reflect the full spectrum of Arctic stratospheric winter conditions during the past two decades (1999: warm, 1997: medium, 1996: cold). The dotted lines in Figure 2 indicate a polynomial fit to the data points. The absolute increase of V_{PSC} and V_{ACl} with increasing H_2O is very similar except for the warm winter. However, in all cases the relative increase of V_{ACl} with increasing H_2O is larger than V_{PSC}. For a H_2O increase of 0.58 ppmv (S4) and medium to cold winters, the relative volume increase is almost a factor of 2 larger for V_{ACl} than for V_{PSC}.

[15] We also estimated the radiative feedback of a H_2O increase on V_{PSC} and V_{ACl} using the relation between stratospheric H_2O increase and temperature response as given by *Forster and Shine* [2002]. Accordingly, a H_2O increase of 3.6% or 0.18 ppmv (Table 1, S2) will cool the Arctic vortex by $\approx$0.13 K. Based on this assumption, we recalculated the V_{PSC} and V_{ACl} values for the lower temperatures. Our results show that in the case of V_{ACl} (V_{PSC}) radiative forcing would increase ΔV_{ACl} (ΔV_{PSC}) further by 4% (3%) during a cold winter like 1996. Thus, the radiative feedback due to a H_2O increase is somewhat smaller than the direct impact of H_2O on V_{ACl} and V_{PSC} (Figure 2, S2), but is of a similar order of magnitude. However, the indirect impact due to H_2O radiative forcing has a stronger effect on V_{ACl}. Both direct and indirect effects of a H_2O increase indicate a larger impact on ozone loss when V_{ACl} is used as a proxy.

2.5. Estimation of Future Arctic Ozone Losses

[16] Since *Rex et al.* [2006] showed that the linear correlation between V_{PSC} and ozone column loss for the coldest winter so far (2004/2005), which is characterized by a 37% larger V_{PSC} compared to the winter 1995/1996, still seems to be valid we assume that a reasonable extrapolation of the correlations shown in Figure 1 by 9% in the case of V_{PSC} and 16% in the case of V_{ACl} is justified. Hence, we estimate the direct effect of a H_2O increase (without considering radiative feedbacks) on additional Arctic ozone losses by applying the linear fits shown in Figure 1 to ΔV_{PSC} and ΔV_{ACl} for all H_2O scenarios (Table 1) and for all Arctic winters between 1984 and 2005.

[17] A comparison of the diagrams in Figure 3 shows that in both cases the estimated additional ozone loss is small for small H_2O increases ($\leq$0.15 ppmv) even during cold winters. Similar conclusions apply for warm winter conditions (e.g. 1999) where a H_2O increase of as much as 1 ppmv is expected to have no significant impact on the additional ozone loss (ΔO_3). However, for cold Arctic winter conditions and for a ΔH_2O between 0.18 and 0.27 ppmv (S2 and S3, Table 1) we estimated an additional ozone loss of up to 5 DU in the case of V_{PSC} compared to 8 DU in the case of V_{ACl}. For higher H_2 mixing ratios (S4, Table 1), a maximum additional ozone column loss of 11 DU in the case of V_{PSC} and 20 DU using V_{ACl} as a proxy is calculated. It should be noted that such high leakage rates are very unlikely to occur in a future global H_2 economy simply for economic reasons. It is more likely that significantly lower H_2 emissions rates (3% – 10%) will be achieved, which are comparable to the assumptions used here in scenarios S1 and S2 [*Schultz et al.*, 2003]. Assuming that liquid sulfate aerosols dominate the process of chlorine activation as suggested by Drdla (submitted manuscript, 2007) and an average undisturbed Arctic ozone column of 400 DU, a H_2 economy would increase the ozone column loss in this region by about 0.4% (S1) to 2% (S2). In the

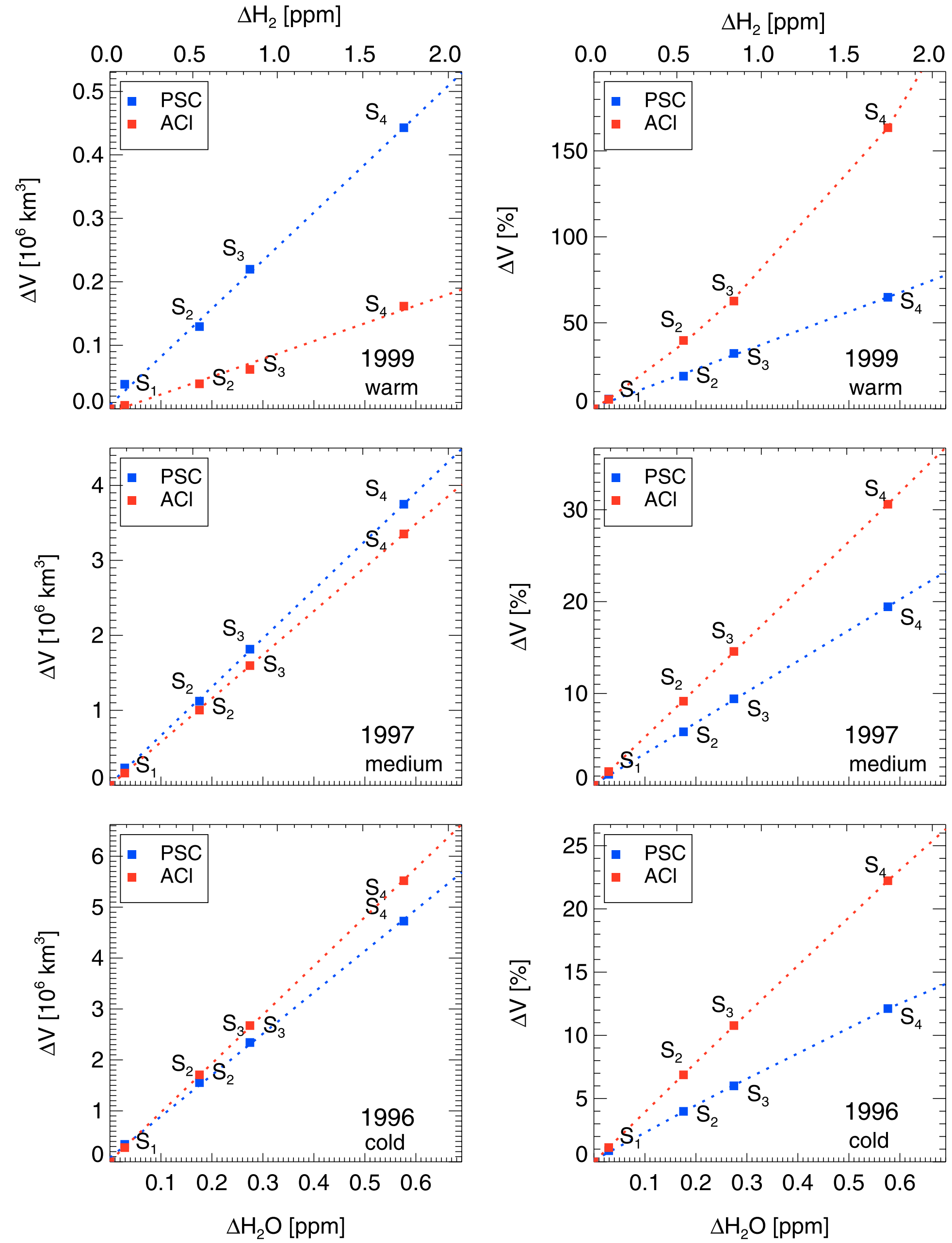

Figure 2. Absolute and relative change of V_{PSC} (blue dashed lines) and V_{ACl} (red dashed lines) due to an increase of stratospheric H_2O for a selection of Arctic winters (1999: warm, 1997: medium, and 1996: cold). Data points are fitted with a third-order polynomial. In addition, the corresponding hydrogen increase (ΔH_2) is displayed on the upper abscissa.

very unlikely event that stratospheric H_2 levels were to increase as much as assumed in scenario S4 [*Tromp et al.*, 2003] and an increase of the tropospheric methane level were to occur as predicted by WMO scenarios [*WMO*, 2003], a monotonic H_2O increase of as much as 1 ppmv would be possible. In this case, our calculations show an additional Arctic ozone loss during cold Arctic winters of up to 34 DU (9%) using V_{ACl} and 19 DU (5%) using V_{PSC} as a proxy.

3. Summary and Conclusions

[18] According to the methodology applied here, we calculated V_{PSC} and V_{ACl} utilizing ECMWF operational analysis for 22 different Arctic winters and different Arctic H_2O profiles motivated by a potential stratospheric H_2O increase due to a future H_2 economy. The comparison of the H_2O sensitivity of the two proxies indicates a stronger relative increase of V_{ACl} by almost a factor of 2 for a H_2O increase of 0.58 ppmv in medium to cold winters. This effect is increased further if radiative feedbacks due to a stratospheric H_2O increase are taken into account. The linear correlation of the proxies with ozone loss was used to provide estimates of the H_2O dependence of polar ozone loss. Under current stratospheric conditions, V_{ACl} and V_{PSC} are equally valid proxies for ozone depletion. However, the impact of a stratospheric H_2O increase on Arctic ozone loss for both proxies is different. If liquid sulfate aerosols

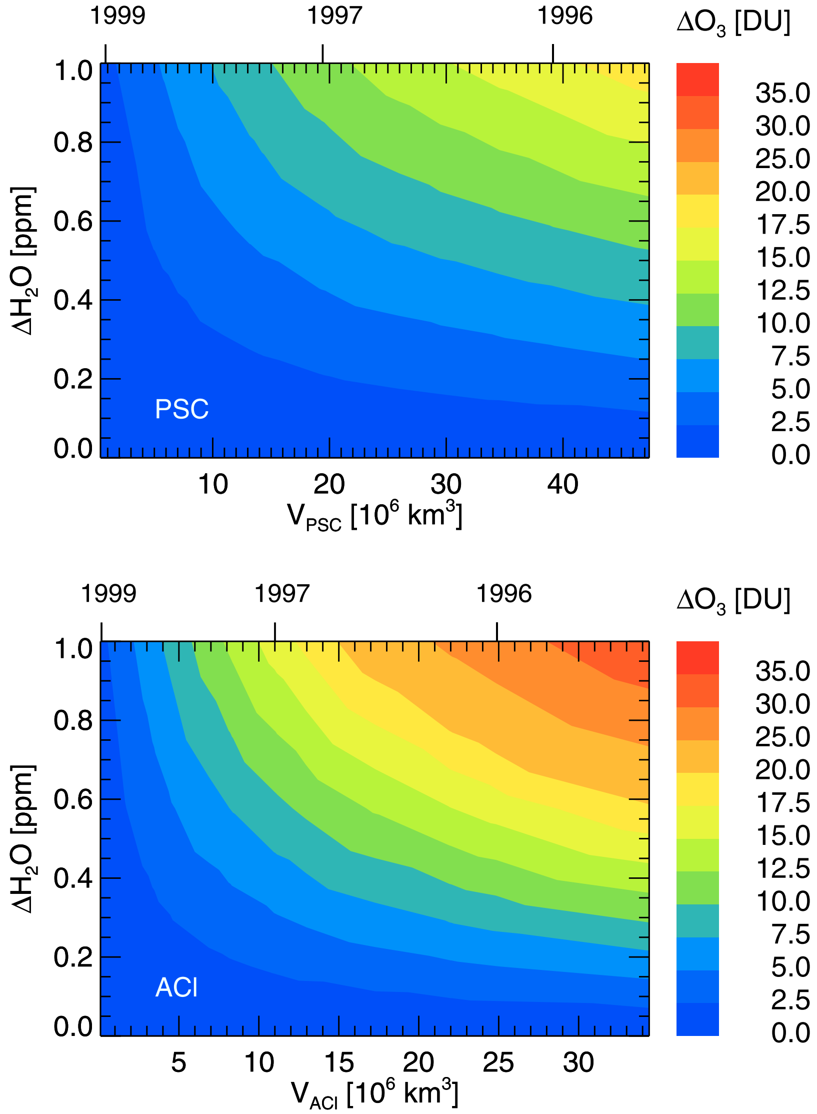

Figure 3. Contour plots of the additional Arctic ozone losses (ΔO_3) in Dobson units (DU) due to an increase of the stratospheric H_2O mixing ratio (ΔH_2O) as calculated for all Arctic winters between 1984 and 2005. (top) The impact of increased H_2O on solid aerosols, and (bottom) that on liquid aerosols for different Arctic winter conditions represented here by the proxies V_{PSC} and V_{ACl}.

dominate the process as suggested by Drdla, it is likely that V_{ACl} better describes chlorine activation and polar ozone loss under changing stratospheric H_2O conditions. This also becomes important for predicted increases of stratospheric H_2O mixing ratios due to increased stratospheric methane levels [*Röckmann et al.*, 2004] and decreases of stratospheric temperatures as expected due to future increases in greenhouse gasses [*Rex et al.*, 2006].

[19] Using V_{ACl} as a proxy indicates that a realistic upper limit of 0.54 ppmv (S2) for a tropospheric H_2 increase is expected to have no significant impact ($\leq$2%) on Arctic ozone loss. Higher H_2 emission scenarios (S4) combined with a projected increase of tropospheric methane levels [*Röckmann et al.*, 2004] suggest a possible stratospheric H_2O increase of 1 ppmv. This is estimated to cause an additional Arctic ozone loss of up to 9%. However, it is generally assumed that scenario S4 is far from being realistic [e.g., *Prather*, 2003]. Consequently, on the basis of our results and the predicted future decline of stratospheric chlorine loading by 2050 we do not expect any substantial impact of a future H_2 economy on Arctic ozone loss, nor is the H_2 economy projected to reach a global dimension before the middle of this century.

[20] **Acknowledgment.** The authors would like to thank Katja Drdla for providing the formula to calculate the temperature threshold for liquid sulfate aerosols prior to its publication.

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