



Contribution of  
liquid, NAT and ice  
particles to chlorine  
activation

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# Contribution of liquid, NAT and ice particles to chlorine activation and ozone depletion during Antarctic winter and spring

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## Abstract

Heterogeneous reactions in the Antarctic stratosphere are the cause of chlorine activation and ozone depletion, but the relative roles of different types of PSCs in chlorine activation is an open question. We use multi-year simulations of the chemistry-climate model EMAC to investigate the impact that the various types of PSCs have on Antarctic chlorine activation and ozone loss.

One standard and three sensitivity EMAC simulations have been performed. The results of these simulations show that the significance of heterogeneous reactions on NAT and ice particles, in comparison to liquid particles, is subordinate regarding chlorine activation and ozone depletion in Antarctic winter and spring. The heterogeneous chemistry on liquid particles is sufficient to activate at least 90 % of the chlorine reservoir species. With the exception of the upper PSC regions between 10 and 30 hPa where temporarily the ice particles have a relevant contribution to the chlorine activation and during the initial PSC occurrence with short NAT contributions the liquid particles alone are sufficient to activate almost all of the available chlorine.

In the model simulations heterogeneous chemistry on liquid particles is responsible for more than 90 % of the ozone depletion in Antarctic spring. Only up to 5 DU of column ozone in high southern latitudes is depleted by chlorine activation due to additional heterogeneous chemistry on ice particles and less than 0.5 DU due to additional heterogeneous chemistry on NAT particles.

## 1 Introduction

Polar Stratospheric Clouds (PSCs) consist of supercooled ternary solution (STS, type 1b PSC), Nitric Acid Trihydrate (NAT, type 1a PSC) and ice particles (type 2 PSC). An accurate representation of PSCs and cold binary solutions of the sulphuric acid background/water aerosol is essential for chlorine activation and polar ozone depletion in chemistry-climate-models (CCMs). The liquid (cold binary aerosols and STS particles)

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Mauersberger (1993). For the ice nucleation 50 % H<sub>2</sub>O supersaturation is assumed to be necessary.

Besides the parameterisations for the formation of liquid and ice particles, there are two parameterisations for the formation of NAT particles included in the PSC submodel.

It is possible to choose either a “thermodynamical NAT parameterisation” assuming instantaneous thermodynamical equilibrium (Hanson and Mauersberger, 1988) or a new parameterisation for NAT particles based on the efficient growth and sedimentation algorithm of Carslaw et al. (2002) and van den Broek et al. (2004) implemented into the submodel by Kirner et al. (2011). In both cases NAT formation takes place below the NAT existence temperature ( $T_{\text{NAT}}$ , depending on the pressure and on the partial pressures of HNO<sub>3</sub> and H<sub>2</sub>O) with the assumption of a necessary super cooling of 3 K (Schlager and Arnold, 1990; Dye et al., 1992).

#### 4 Performed simulations

Four different EMAC simulations were performed using the new submodel PSC: a standard simulation (*Standard*) from 2000 to 2012 with heterogeneous chemistry on liquid, NAT and ice particles and three additional sensitivity simulations from 2005 to 2012. In the three sensitivity simulations we only changed the heterogeneous chemistry on PSC particles by switching on and off the chemistry on liquid, NAT and ice particles. We performed one sensitivity simulation without heterogeneous chemistry (*NoHet*), one sensitivity simulation with heterogeneous chemistry only on liquid particles (*Liquid*) and one sensitivity simulation with heterogeneous chemistry only on liquid and NAT particles, but excluding heterogeneous chemistry on ice particles (*LiquidNAT*) (see also Table 1).

The formation and distribution of the PSC particles is the same for the four simulations. Also the denitrification caused by the sedimentation of NAT particles is simulated in all simulations consistently. Only the assumptions regarding the heterogeneous chemistry on liquid, NAT and ice particles are different.

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to 90° S and over the five years from 2005 to 2009. The illustrated results are valid for the standard and for all three sensitivity simulation, i.e. the formation and development of stratospheric aerosol and PSCs are the same for each simulation. In average NAT is in the EMAC simulations visible from mid-May until October in the area of lower temperatures. The relatively slow increase of the number density from 0 to the average maximum of over  $200 \text{ m}^{-3}$  by end of May is caused by different first NAT appearances in the individual years 2005 to 2009. The decrease between June to September is caused by the sedimentation of larger NAT particles and the related denitrification. Ice particles exist mainly from June to September with mean maxima of over  $40\,000 \text{ m}^{-3}$ . In comparison to liquid and NAT particles they have the largest surface densities with values up to  $38 \mu\text{m}^2 \text{ cm}^{-3}$ . The liquid particles have the second largest surface densities with values up to  $6.8 \mu\text{m}^2 \text{ cm}^{-3}$ , the NAT particles the smallest with maxima of  $0.11 \mu\text{m}^2 \text{ cm}^{-3}$ .

## 6 Contribution of NAT, ice and liquid particles to stratospheric chlorine activation and ozone depletion

By comparing the standard and the sensitivity simulations it is possible to determine the contribution of the heterogeneous reactivity of liquid, NAT and ice particles to polar chlorine activation and corresponding ozone depletion in polar spring. In this way it is possible to assess the impact of the liquid particles by subtraction the results of the simulation *NoHet* from the results of the simulation *Liquid*. The absolute contribution of additional NAT chemistry is calculated by subtraction the results of the simulation *Liquid* from the simulation *LiquidNAT*, and the contribution of additional heterogeneous chemistry on ice particles by subtraction the results of the simulation *LiquidNAT* from the simulation *Standard*. For a better statistics we averaged the  $\text{ClO}_x$  and  $\text{O}_3$  results over the southern high latitudes from 80° S to 90° S and over the five year time period from 2005 to 2009 (Figs. 3 and 4) and the ozone columns as zonal mean over the same five years (Fig. 5).



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The total effect on ozone chemistry of applying heterogeneous chemistry on different PSC particles is illustrated in form of the time development of ozone columns in the Southern Hemisphere in Fig. 5. Liquid particles are mainly responsible for the ozone depletion noticeable as a decrease in ozone columns by more than 130 DU in early October in high southern latitudes. During September to October in the southern high latitudes the contribution of liquid particles to the simulated ozone depletion is at least 95 %. The contribution of additional heterogeneous chemistry on ice particles only reaches up to 5 DU and is always smaller than 5 %, the contribution of additional heterogeneous chemistry on NAT particles is less than 0.5 DU and thus almost negligible. On the other hand the NAT particles are essential for the denitrification in polar spring and thus have an additional indirect influence on the formation of the ozone hole.

## 7 Conclusions

The significance of heterogeneous reactions on NAT and ice particles with respect to chlorine activation and ozone depletion in Antarctic winter and spring is small compared to reactions on liquid particles in standard and sensitivity simulations performed with the EMAC model. The simulated heterogeneous chemistry on liquid particles is sufficient to activate at least about 90 % of the chlorine. With the exception of the upper PSC regions between 10 and 30 hPa, where temporarily the ice particles have relevant contribution to chlorine activation and during the initial PSC occurrence with NAT contributions, the liquid particles are sufficient to activate almost all of the available chlorine reservoirs alone.

In the model simulations, heterogeneous chemistry on liquid particles is responsible for more than 90 % of the ozone depletion in polar spring. Only up to 5 DU of column ozone in high southern latitudes is depleted by chlorine activation due to additional heterogeneous chemistry on ice particles and less than 0.5 DU due to additional heterogeneous chemistry on NAT particles.



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**Table 1.** The four performed EMAC simulations with different assumptions for heterogeneous chemistry.

Simulations	Time period	Heterogeneous chemistry
<i>Standard</i>	2000–2012	on liquid, NAT and ice particles
<i>NoHet</i>	2005–2012	none
<i>Liquid</i>	2005–2012	on liquid particles
<i>LiquidNAT</i>	2005–2012	on liquid and NAT particles

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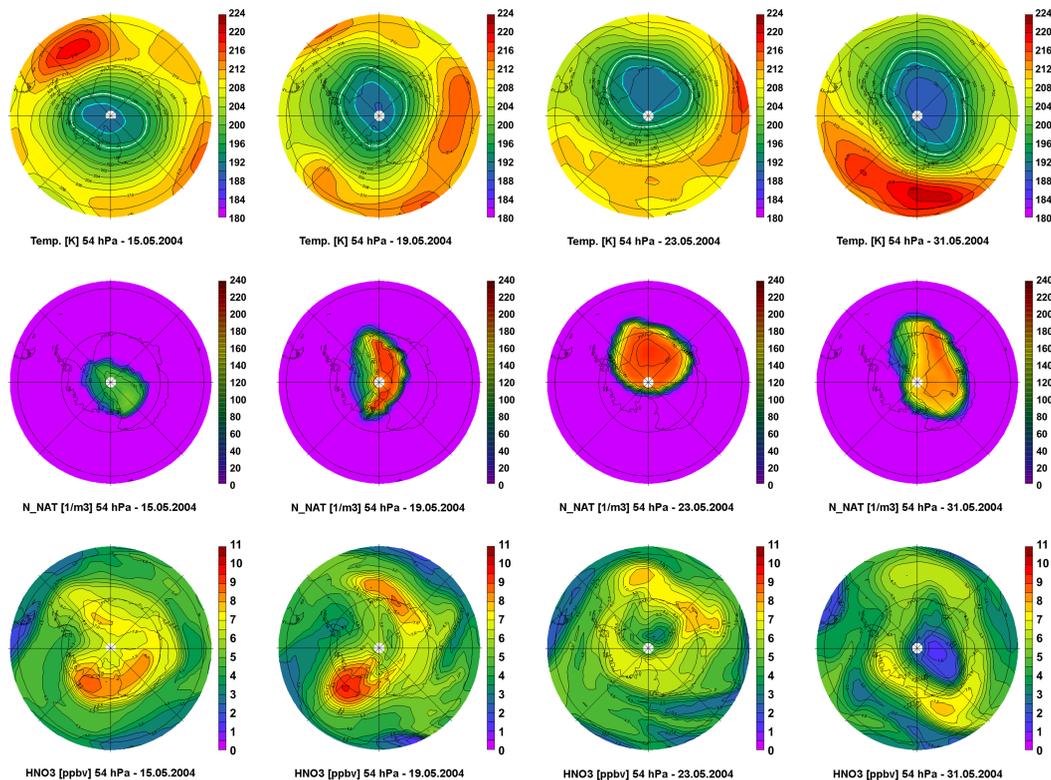
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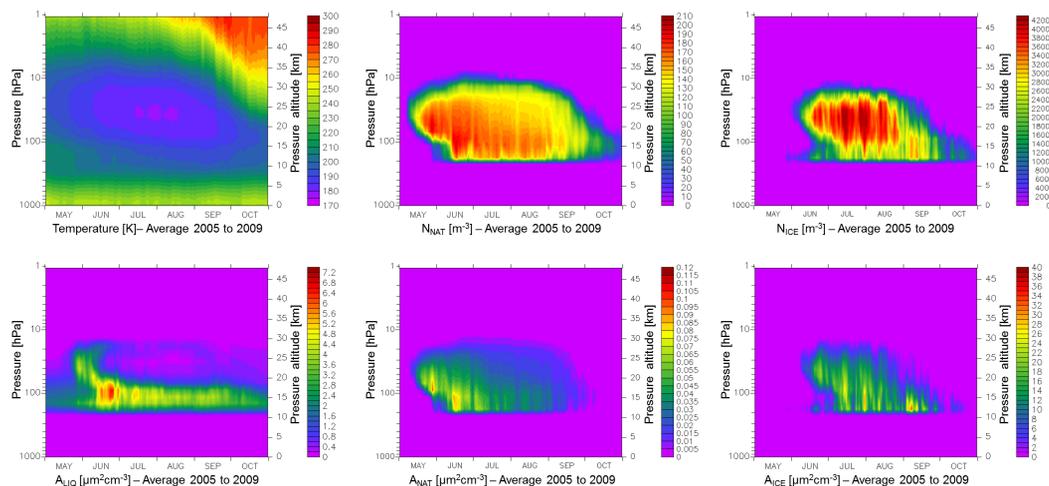
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**Figure 1.** Denitrification in the standard simulation – time series of the temperature in K (top row) and the distribution of  $N_{\text{NAT}}$  in  $\text{m}^{-3}$  (second row) and  $\text{HNO}_3$  in ppbv (bottom row) for southern polar latitudes on 54 hPa (approx. 20.5 km) for the times 15 May 2004, 19 May 2004, 23 May 2004, and 31 May 2004. In the temperature plots the isothermes of 195 K (white) and 192 K (light blue) are marked.

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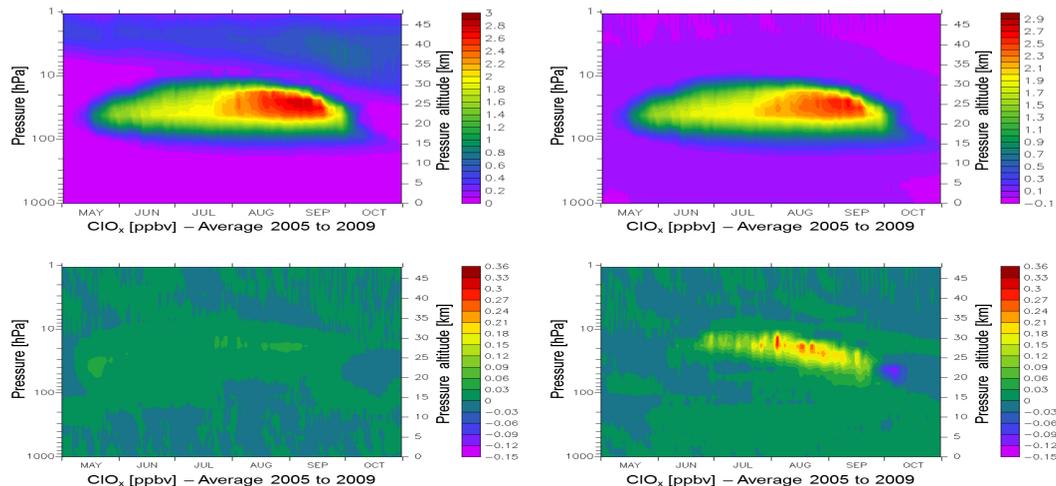
**Figure 2.** Stratospheric temperature, liquid and solid particles – time series averaged from 80° to 90° S and over the years 2005 to 2009 for temperature in K (top left), number densities of NAT ( $N_{\text{NAT}}$ ) and ice particles ( $N_{\text{ICE}}$ ) in  $\text{m}^{-3}$  (top center and right), as well as surface densities of liquid particles ( $A_{\text{LIQ}}$ ), NAT ( $A_{\text{NAT}}$ ) and ice particles ( $A_{\text{ICE}}$ ) in  $\mu\text{m}^2 \text{cm}^{-3}$  (bottom left to right).

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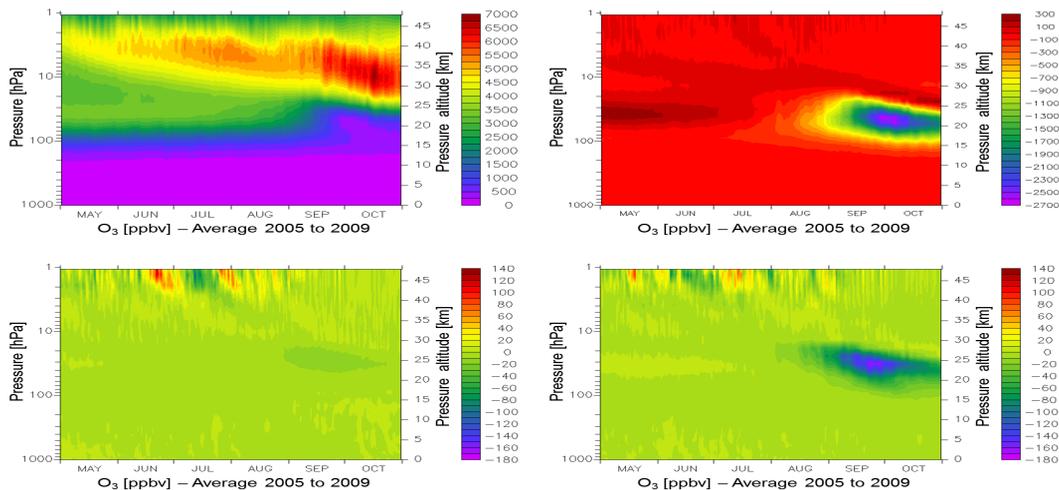
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**Figure 3.** Chlorine activation – time series of  $\text{ClO}_x$  in ppbv in the standard simulation during Antarctic winter and spring in  $80^\circ$  to  $90^\circ$  S averaged over the years 2005 to 2009 (top left) and contribution of liquid particles (top right), additional contribution by activation on NAT particles (bottom left) and additional contribution by activation on ice particles (bottom right).  $\text{ClO}_x$  is defined as the sum of  $\text{ClO} + \text{OCIO} + \text{HOCl} + 2\text{Cl}_2 + 2\text{Cl}_2\text{O}_2$ .

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**Figure 4.** Ozone depletion – time series of  $O_3$  in ppbv in the standard simulation during Antarctic winter and spring in  $80^\circ$  to  $90^\circ$  S averaged over the years 2005 to 2009 (top left) and contribution of liquid particles (top right), additional NAT particles (bottom left) and additional ice particles (bottom right). Note that the color bars have different units.

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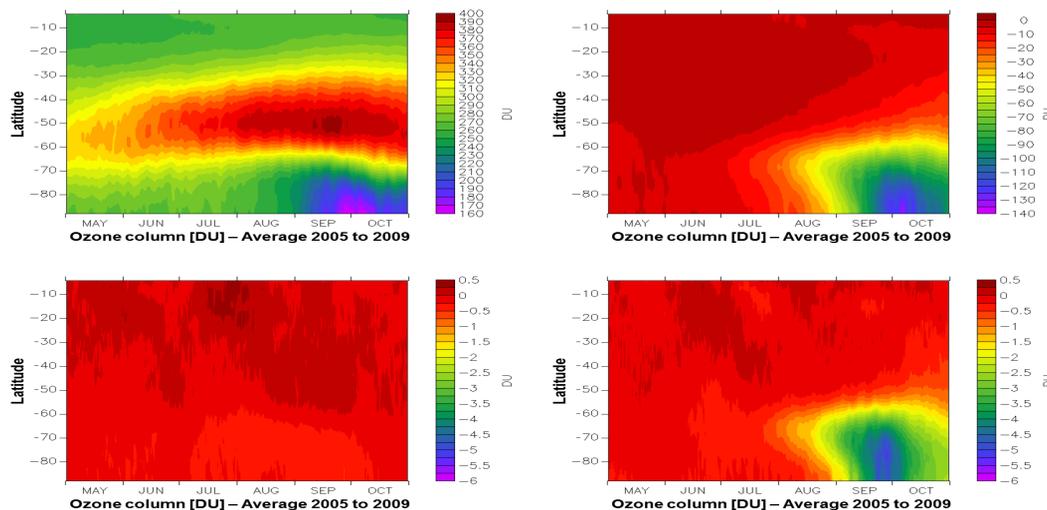
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**Figure 5.** Ozone columns – development of ozone column in DU in the standard simulation during Antarctic winter and spring in the Southern Hemisphere averaged over the years 2005 to 2009 (top left) and contribution of liquid particles (top right), additional NAT particles (bottom left) and additional ice particles (bottom right) to ozone depletion. Note that the color bars have different units.

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