# How is chlorine activation affected by the composition of Polar Stratospheric Clouds (STS versus NAT) in the ATLAS CTM?


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## Motivation

- Explore impact of known uncertainties in heterogeneous processes on ozone depletion and chlorine activation
- Use ATLAS CTM and sensitivity runs for every uncertainty
- Focus on activation on liquid ternary solutions (STS) only versus activation on STS plus solid NAT particles (Wohltmann et al., 2013, Drdla and Müller, 2012)...
- ...and on reaction rate coefficients

## ATLAS Model

- Lagrangian model
- Stratospheric chemistry: 180+ reactions, 47 species
- Rate constants from JPL 2011
- $Cl_2O_2$ photolysis from Burkholder et al. (1990)
- Heterogenous chemistry: Reactions on NAT, ice, STS
- Particle-based denitrification model (DLAPSE): Nucleation, sedimentation, growth of "NAT rocks"

## Heterogeneous chemistry module

- STS: Carslaw et al. (1995), form up to ice frost point
- NAT, ice: Form instantly in equilibrium if given supersaturation is exceeded
- NAT, ice: Predefined number density, uniform particle radius (calculated)
- NAT forms from STS

| Reaction | STS | NAT | Ice |
|---|---|---|---|
| $ClONO_2 + H_2O$ | HR/Shi | HR/AM | 0.3 |
| $ClONO_2 + HCl$ | HR/Shi | HR/AM | 0.3 |
| $N_2O_5 + H2O$ | HR | 0.0006 | 0.02 |
| $N_2O_5 + HCl$ | - | 0.003 | 0.03 |
| $HOCl + HCl$ | HR/Shi | 0.1 | 0.2 |

## Model setup

- ERA Interim
- Resolution: 150 km
- Vertical: Potential temperature and heating rates
- December 2009–March 2010

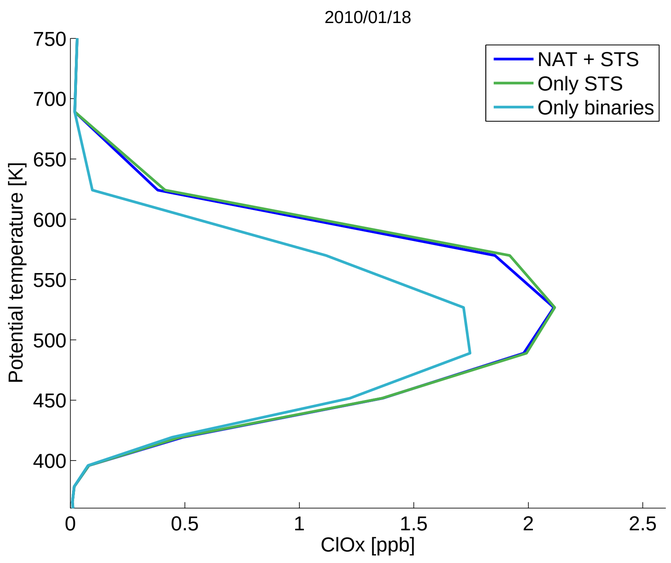
## Reference run

- NAT (Hanson+Ravishankara) and STS (Shi et al.)
- Supersaturation $HNO_3$ over NAT of 10 (3 K supercooling)
- Number density STS: 10 $cm^{-3}$
- Number density NAT: 0.1 $cm^{-3}$
- Number density ice: 0.01 $cm^{-3}$
- Nucleation rate NAT rocks: $7.8 \cdot 10^{-6}$ particles per h and $cm^3$

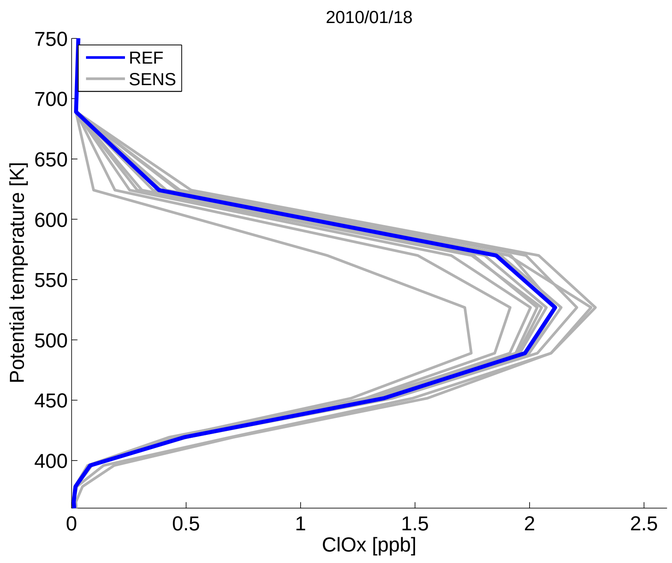
## Sensitivity runs

| | |
|---|---|
| REF | Reference run: NAT and STS |
| ONLY-LIQ-TER | Activation only on STS (no NAT) |
| ONLY-LIQ-TER-HR | As above, but with rates of Hanson and Ravishankara (1994) for STS |
| ONLY-LIQ-BIN | Activation only on binaries (no uptake of HNO3 allowed) |
| ABBATT | Rates of Abbatt and Molina (1992) for NAT |
| | ...and 7 more (not discussed here) |

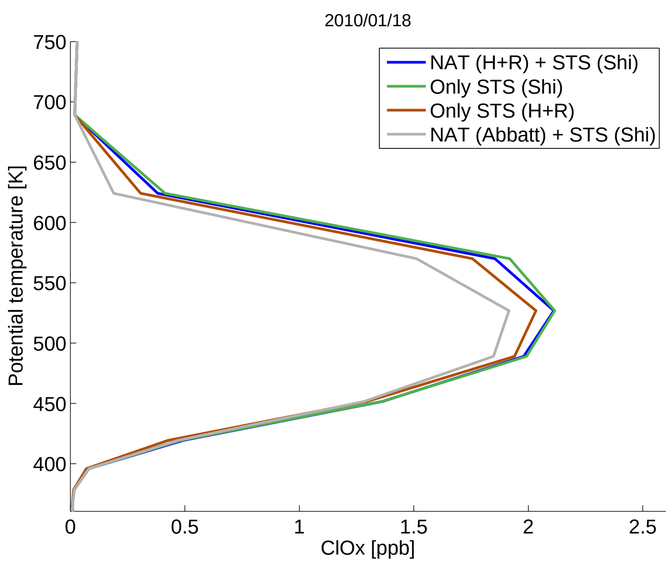
## NAT versus liquid clouds (Chlorine activation)



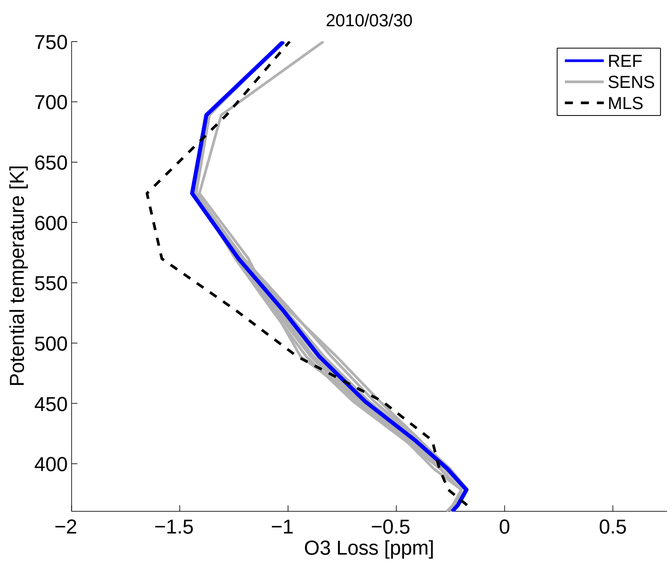

## All sensitivity runs (Chlorine activation)




## Different reaction rate coefficients (Chlorine activation)

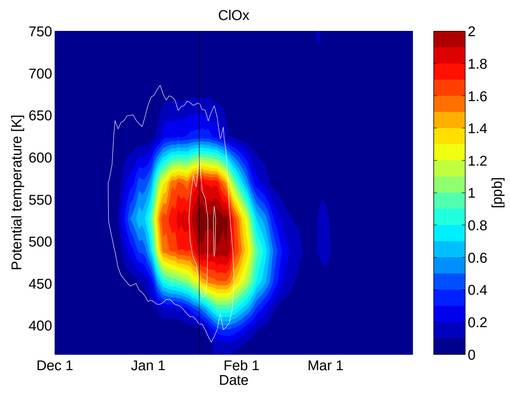



## All sensitivity runs (Ozone loss)



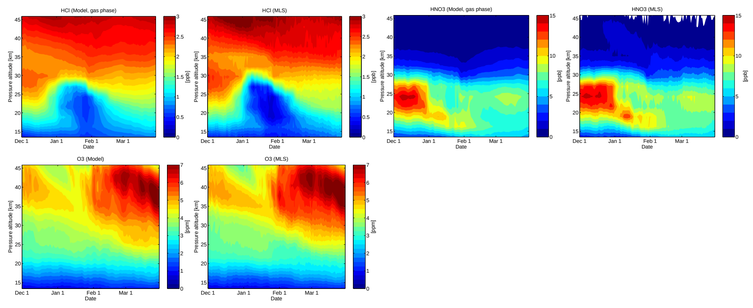

Vortex means of reference run and sensitvity runs for 18 January (ClOx) and 30 March (ozone loss)

## Chlorine activation (reference run)



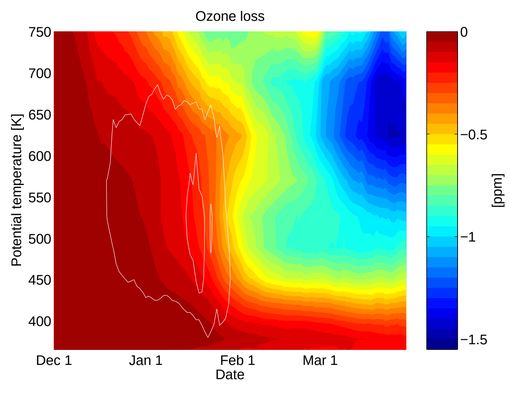

- $ClO_x = ClO + 2Cl_2O_2$
- Vortex average (Nash criterion)
- White lines: NAT/ice occurence

## Comparison to MLS (reference run)




Vortex means of $HCl$ (REF), $HNO_3$ (REF), $O_3$ (REF)
Model = Left, MLS = right

## Ozone loss (Reference run)




- Difference between passive ozone tracer and ozone
- Two maxima: $NO_x$ and $ClO_x$
- White lines: NAT/ice occurence
- Moderate loss compared to other winters

## Conclusions (NAT versus liquid)

- Activation on liquid aerosol alone (STS plus binaries) sufficient to explain observed magnitude and morphology of ozone depletion and chlorine activation
- Current estimates of NAT number density and supersaturation imply small role of NAT
- No final decision possible from our model runs which percentage of activation occurs on STS or NAT (relatively similar results, model bias to observations)
- No sufficient constraint from observations (e.g. Calipso) on the fractions that STS and NAT contribute to chlorine activation

## Conclusions (Reaction rates)

- Change between rates of Shi et al. or Hanson and Ravishankara for liquid aerosols has only minor impact
- Same is true for change between rates of Abbatt and Molina or Hanson and Ravishankara for NAT...
- ...but that is caused by the small role of NAT in the model run. More NAT clouds would cause large differences.

## Conclusions (general)

- Even (unrealistically) large changes in the underlying assumptions have only a small impact on the modeled ozone loss (≈10%)
- General morphology of all species is reproduced well
- Runs slightly overestimate HCl and underestimate ClOx and ozone depletion compared to MLS, Geophysica and ozone sondes

## References


- Poster based on Wohltmann et al. (2013), Uncertainties in modelling heterogeneous chemistry and Arctic ozone depletion in the winter 2009/2010, Atmos. Chem. Phys., 13, 3909-3929.
- Drdla and Müller (2012), Temperature thresholds for chlorine activation and ozone loss in the polar stratosphere, Ann. Geophys., 30, 1055-1073.



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