

How is chlorine activation affected by the composition of Polar Stratospheric Clouds and background aerosol particles?

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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 solid NAT particles versus activation on liquid ternary or binary solutions (see Drdla and Müller, 2012)...
- ...and on reaction rate coefficients

ATLAS Model

- Lagrangian model
- Stratospheric chemistry: 180+ reactions, 47 species
- Rate constants from JPL 2011
- Cl₂O₂ photolysis from Burkholder et al. (1990)
- Heterogeneous 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 ₂ O	HR/Shi	HR/AM	0.3
ClONO ₂ + HCl	HR/Shi	HR/AM	0.3
N ₂ O ₅ + H ₂ O	HR	0.0006	0.02
N ₂ O 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

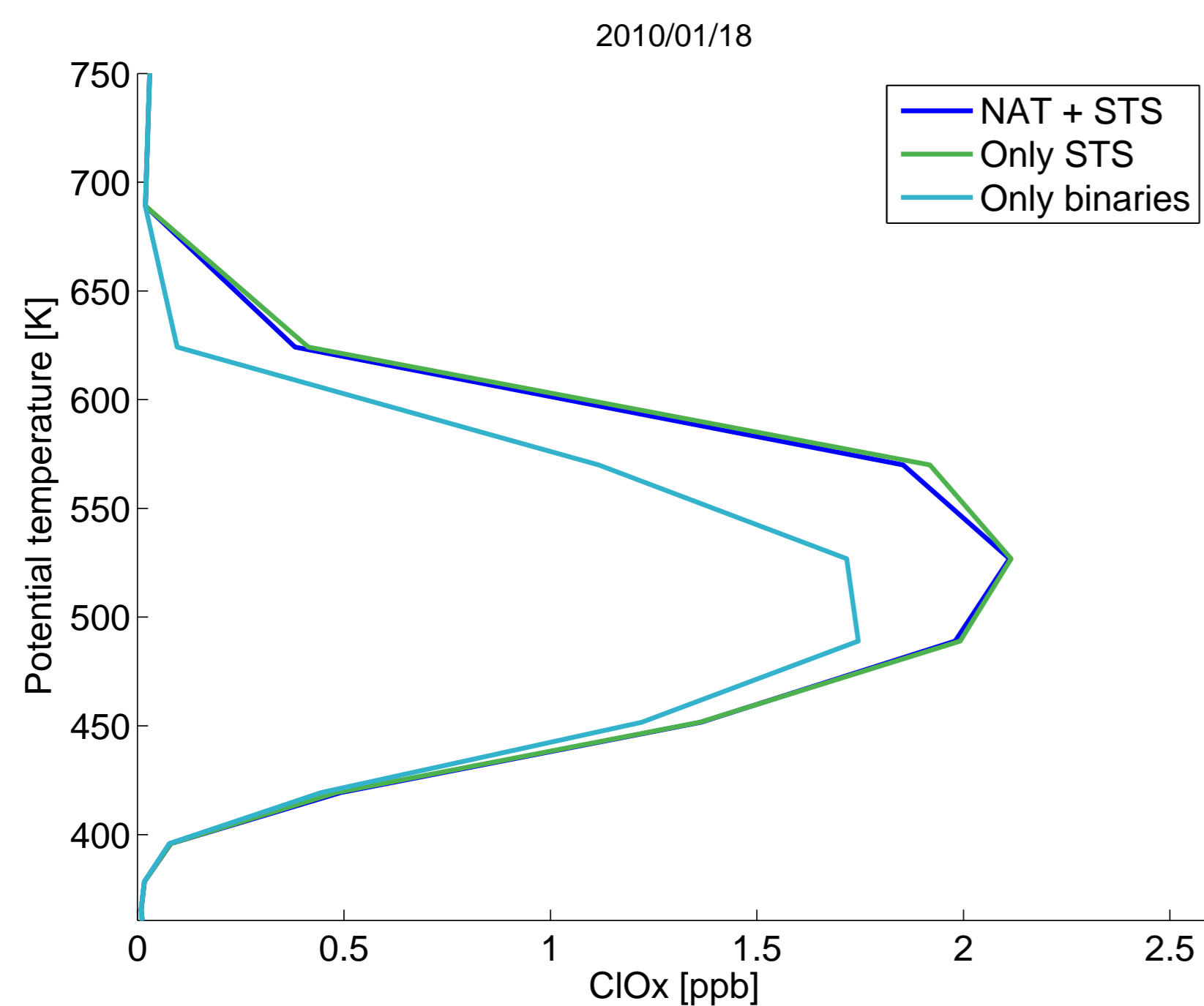
Reference run

- NAT (Hanson+Ravishankara) and STS (Shi et al.)
- Supersaturation HNO₃ 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 · 10⁻⁶ particles per h and cm³

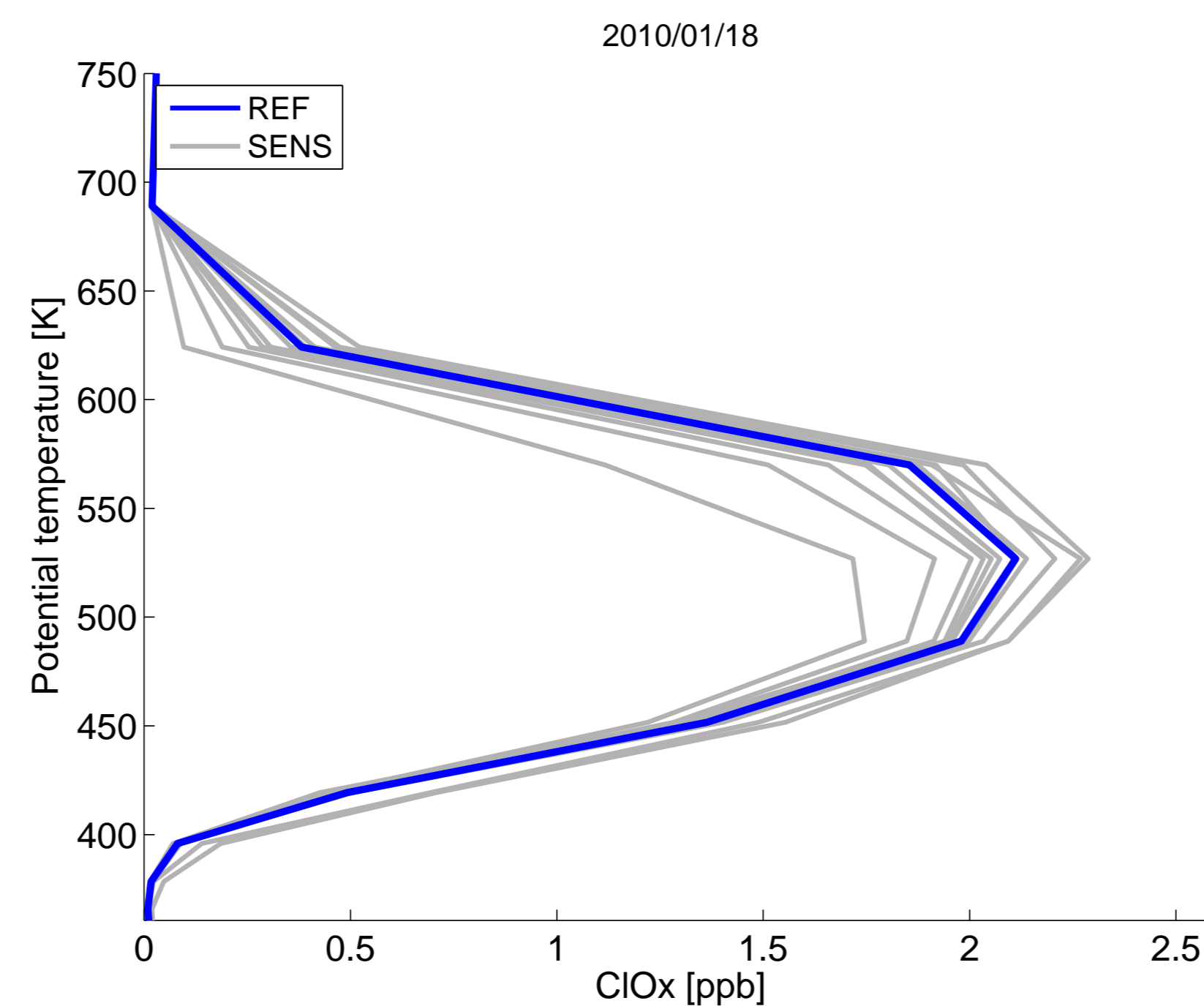
Sensitivity runs

- 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)

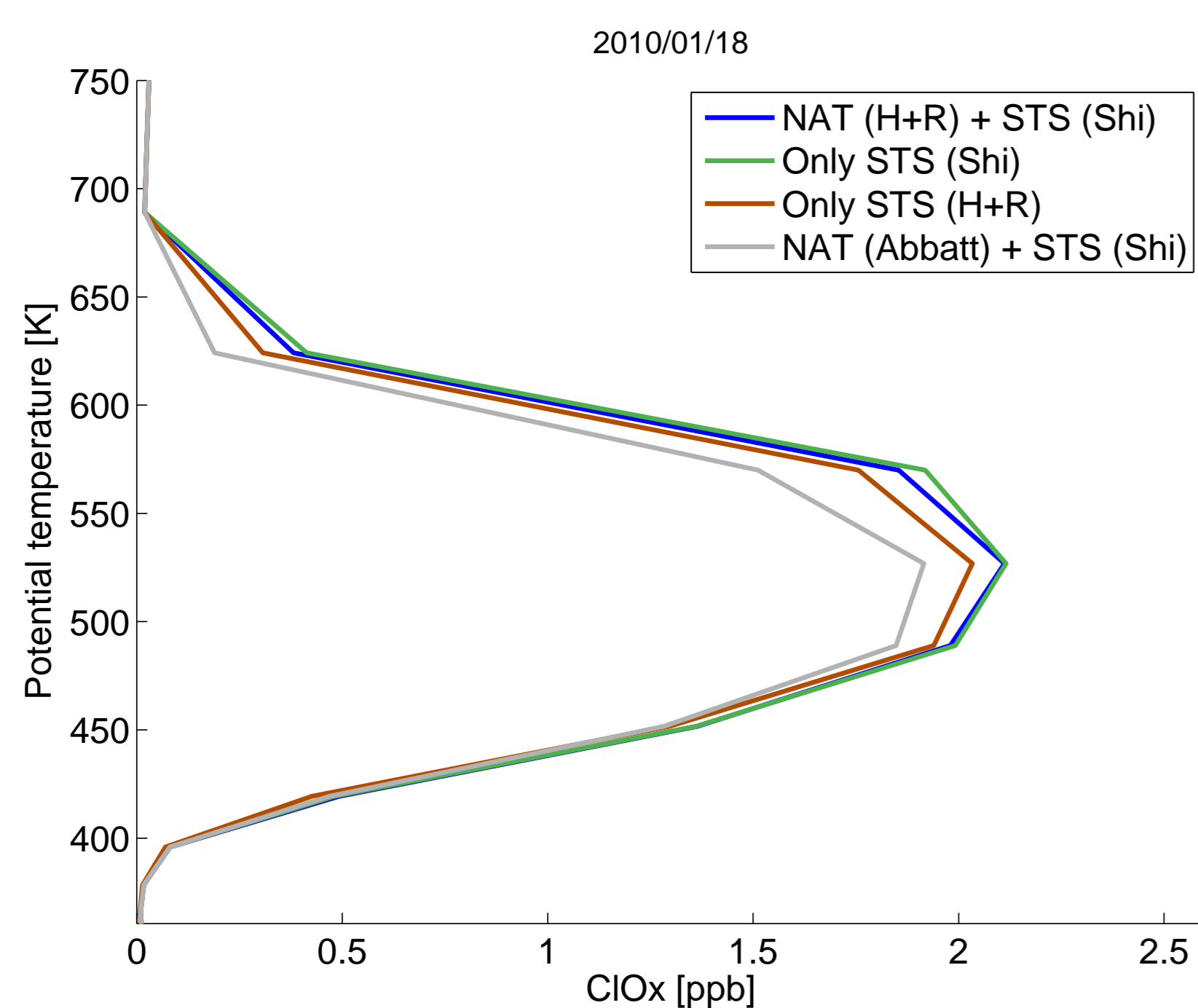
NAT versus liquid clouds (Chlorine activation)



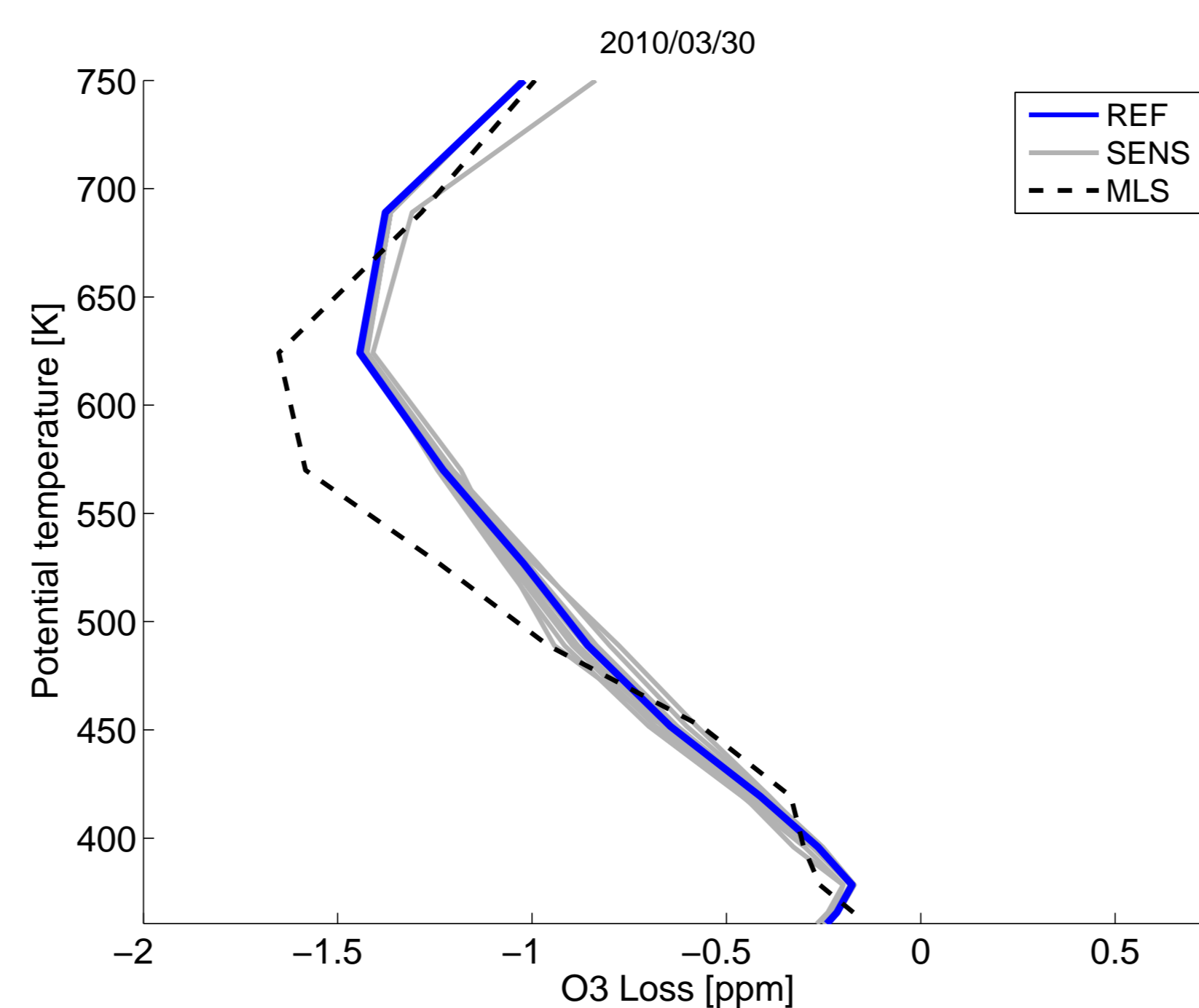
All sensitivity runs (Chlorine activation)



Different reaction rate coefficients (Chlorine activation)

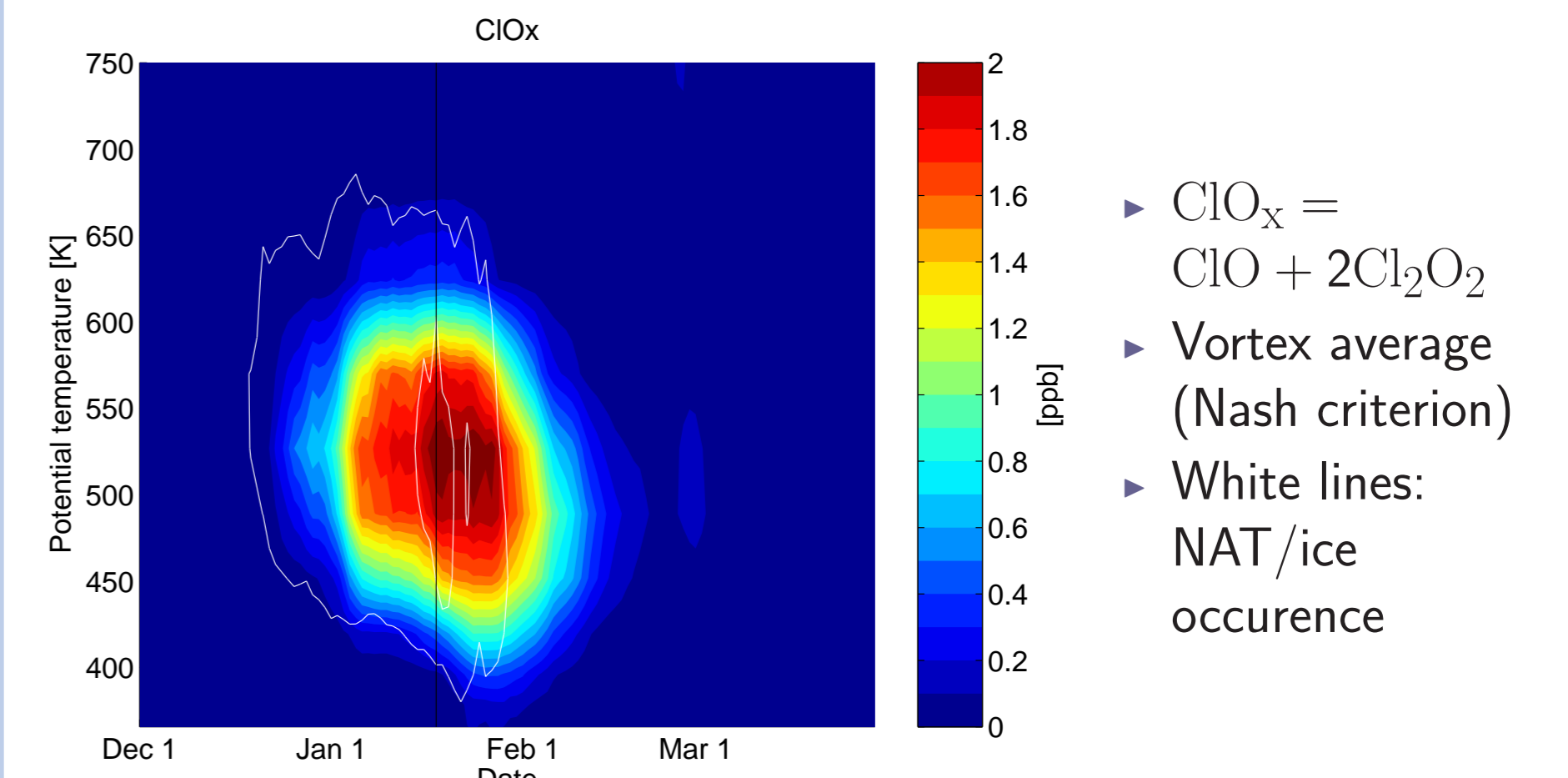


All sensitivity runs (Ozone loss)

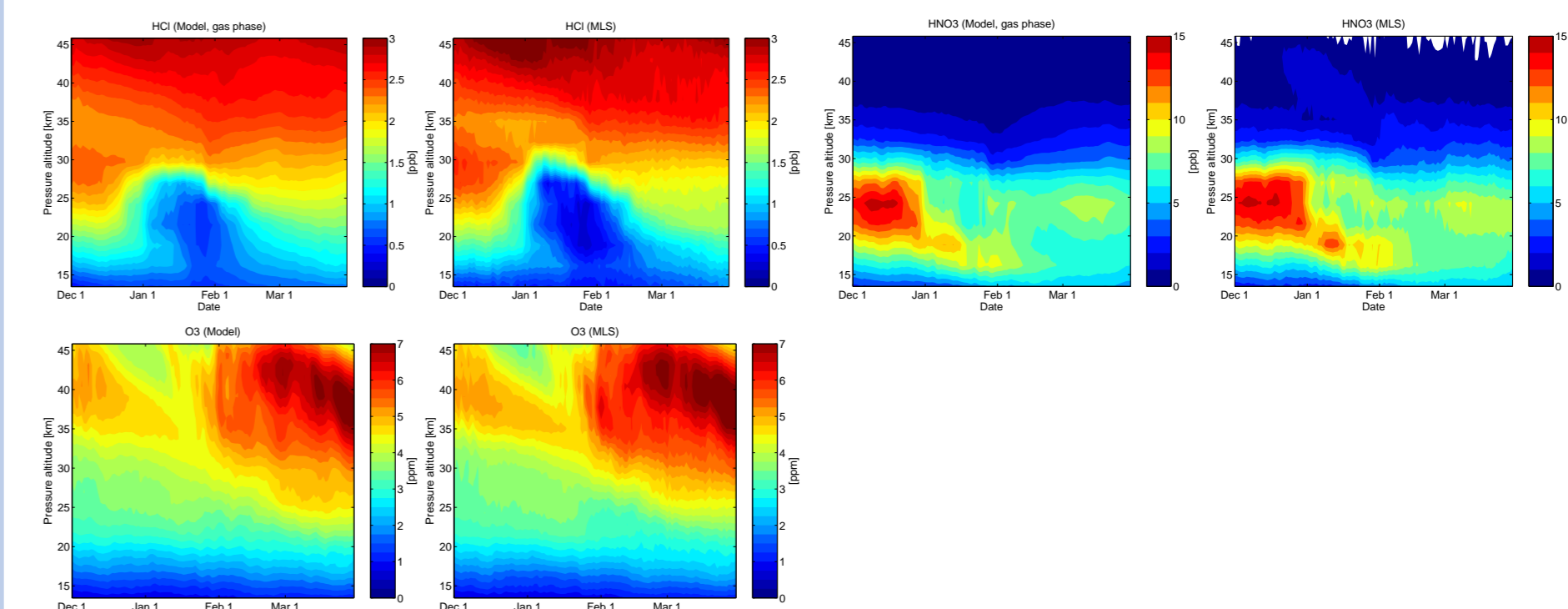


Vortex means of reference run and sensitivity runs for 18 January (ClO_x) and 30 March (ozone loss)

Chlorine activation (reference run)

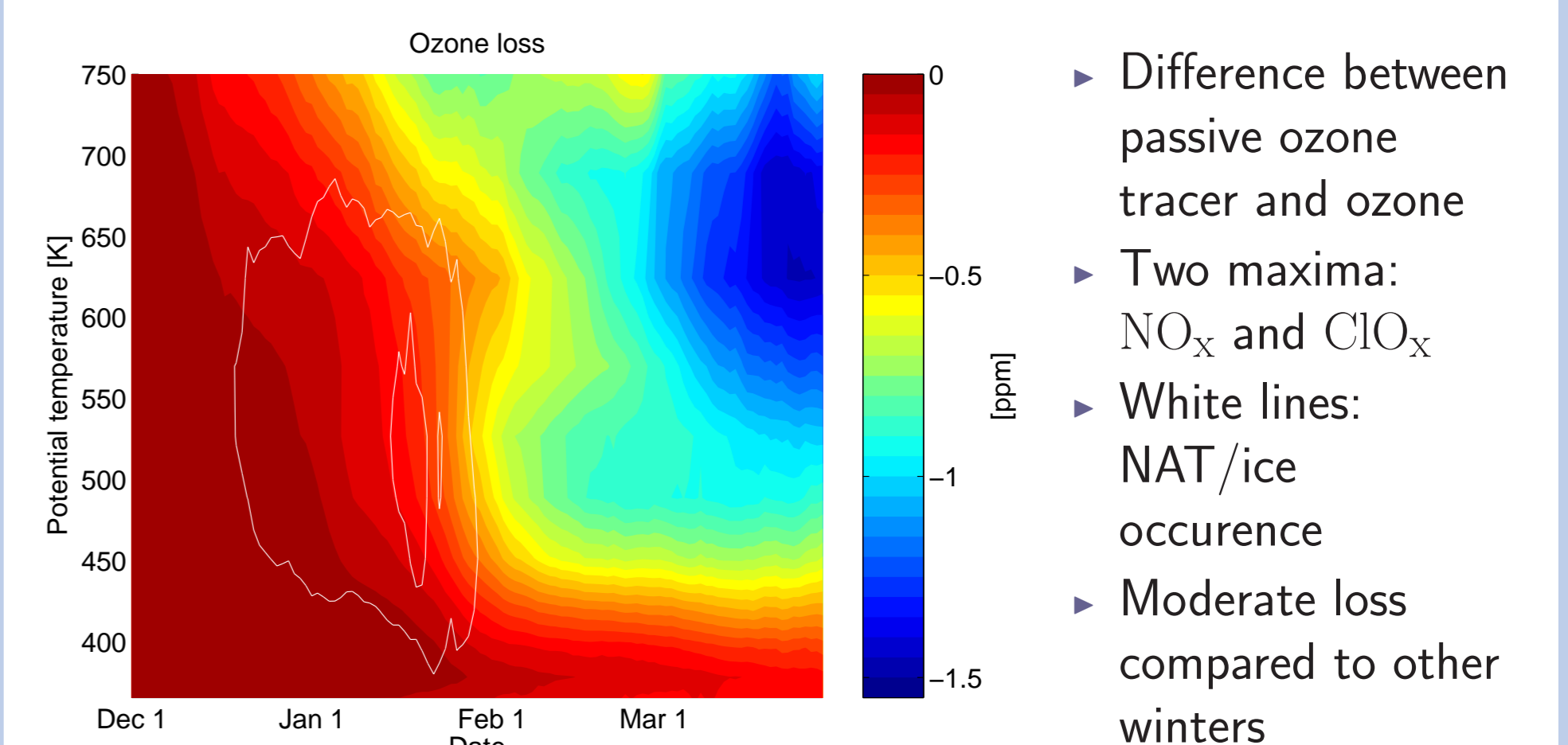


Comparison to MLS (reference run)



Vortex means of HCl (REF), HNO₃ (REF), O₃ (REF)
Model = Left, MLS = right

Ozone loss (Reference run)



Conclusions (NAT versus liquid)

- Activation on liquid aerosol alone sufficient to explain observed magnitude and morphology of ozone depletion and chlorine activation
- Even true for binary aerosols (no uptake of HNO₃ from gas-phase allowed)
- Current estimates of NAT number density and supersaturation imply small role of NAT, at least in 2009/2010 winter
- No final decision possible from our model runs which percentage of activation occurs on STS or NAT (relatively similar results, model bias to observations)

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.

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.

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 ClO_x and ozone depletion compared to MLS, Geophysica and ozone sondes