School of Chemistry
Faculty of Maths and Physical Sciences

UNIVERSITY OF LEEDS

Aerosol and Ozone: Does Meteoric Smoke Control Nucleation in PSC?

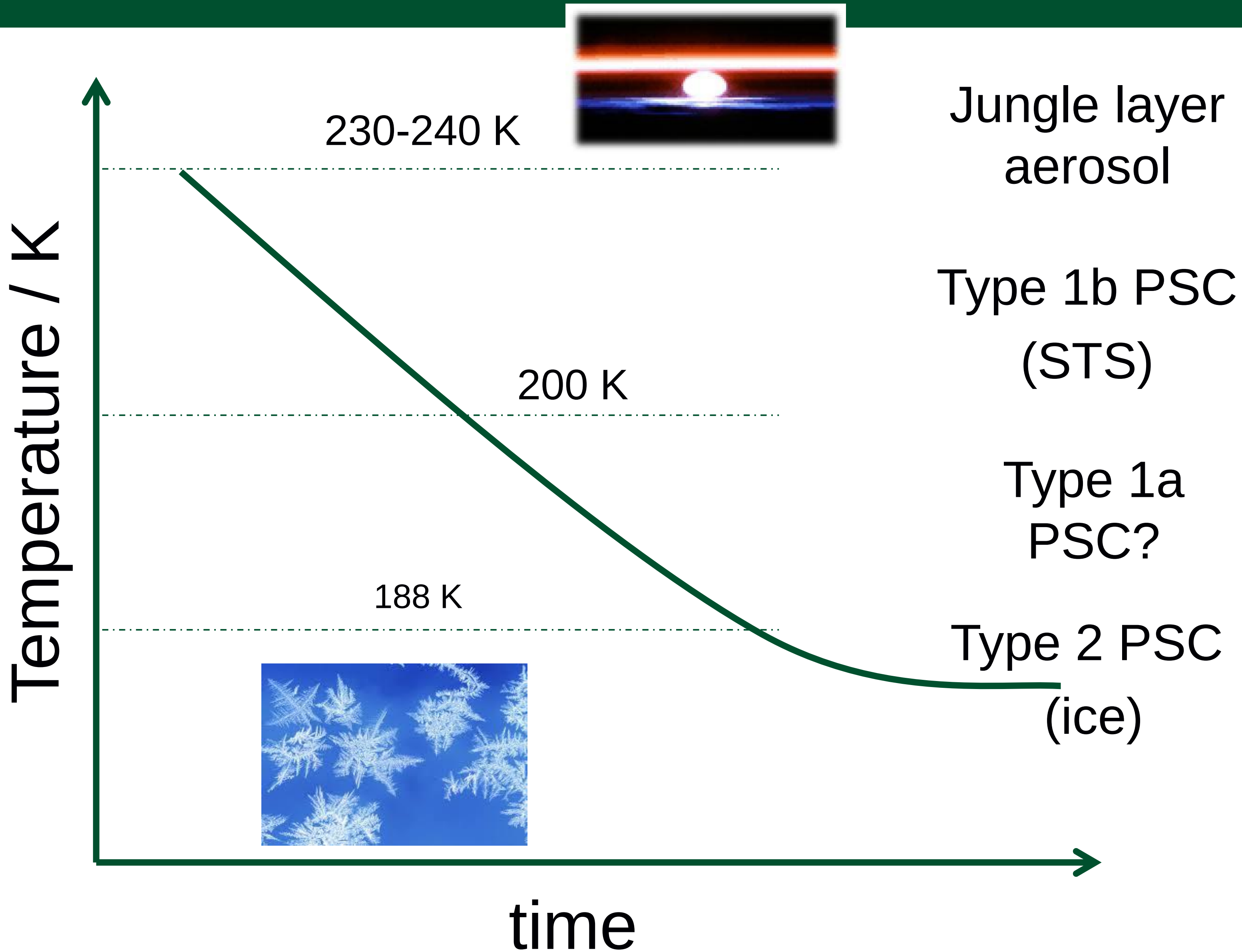
Alexander D. James *

Benjamin Murray	&	John Plane
Ice Nucleation	&	Mesosphere / MSP



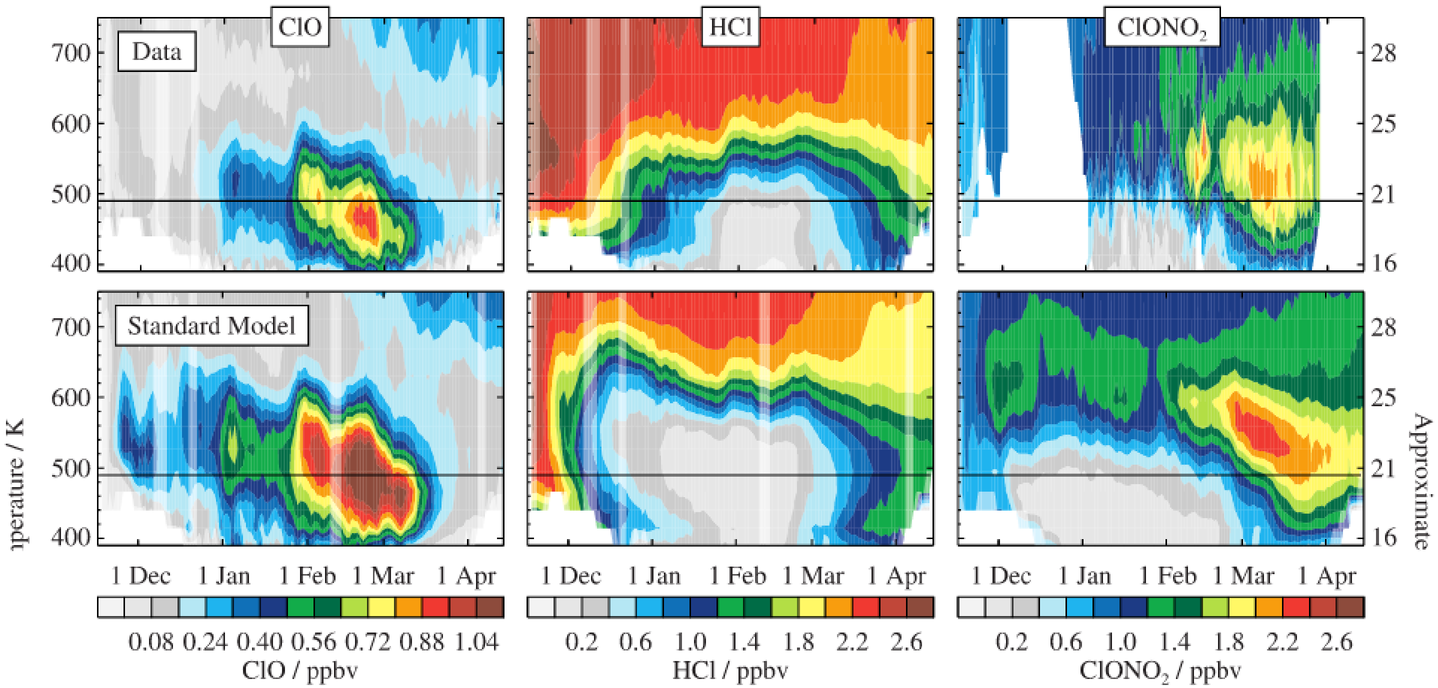
* cmaj@leeds.ac.uk

Background


230-240 K
Jungle layer aerosol
Type 1b PSC
(STS)
200 K
Type 1a PSC?
188 K
Type 2 PSC
(ice)
Temperature / K
time

Background

UNIVERSITY OF LEEDS



Heterogeneous chemistry on PSCs produces species which actively catalyse O_3 destruction

Santee et al., *J. Geophys. Res.: Atmos.*, **2008**, 113, D12307

Background

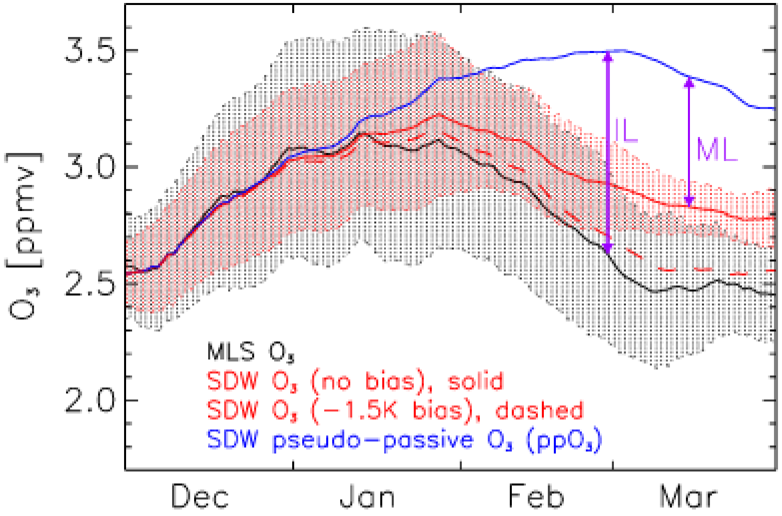


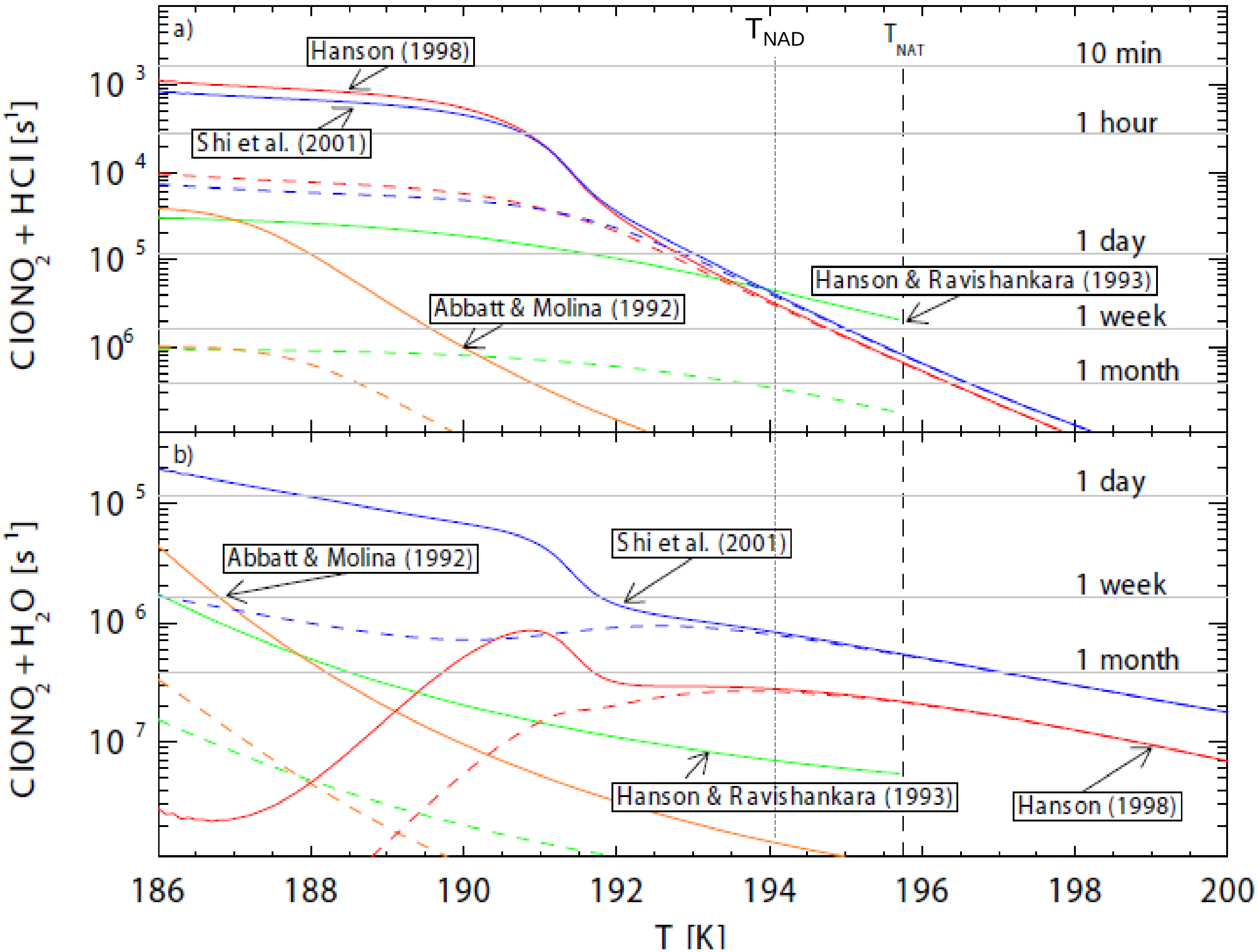
Figure 3. Evolution of vortex averaged MLS O_3 (black), SD-WACCM O_3 (red), and SD-WACCM pseudo-passive ozone (ppO_3, blue) at 475 K. Here, inferred loss (IL, purple) is the difference between MLS O_3 (black) and SD-WACCM ppO_3 (blue); modeled loss (ML, purple) is the difference between SD-WACCM O_3 (red) and SD-WACCM ppO_3 (blue). The black (red) shaded area is MLS (SD-WACCM) O_3 vortex average ± one standard deviation (σ) of the data used for the vortex average. Also shown is the evolution of vortex averaged SD-WACCM O_3 with a −1.5 K bias for heterogeneous chemistry (dashed red, see text).

Biasing all model processes which involve aerosol 1.5 K colder improved agreement between SD-WACCM and MLS O_3

Brakebusch et al., *J. Geophys. Res.: Atmos.*, **2013**, 118, 2673-2688

Background

UNIVERSITY OF LEEDS

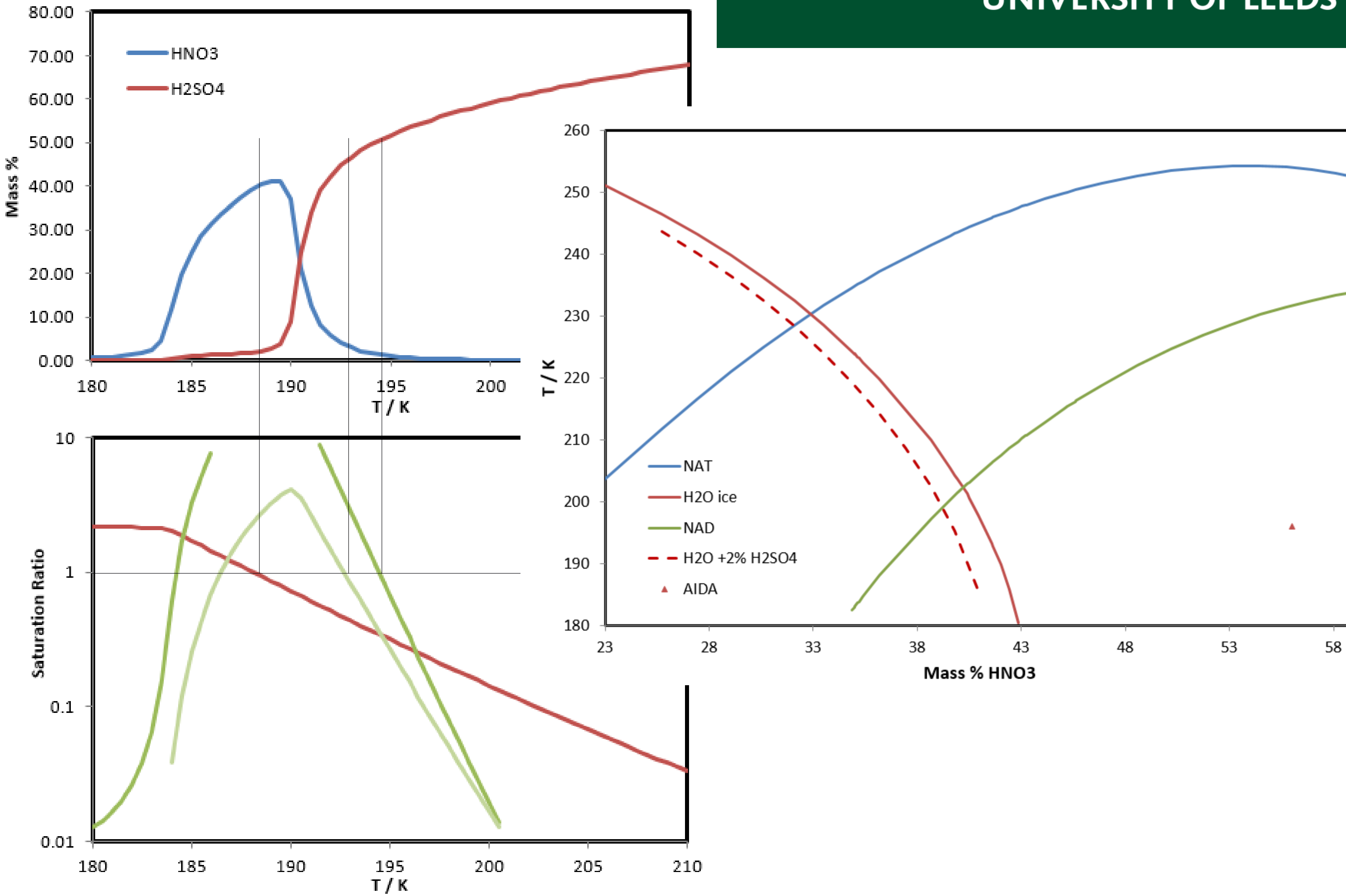


first-order loss rates (s^{-1}) for (a) $ClONO_2$+HCl and (b) $ClONO_2+H_2O$ for different parameterisations and aerosol types for typical stratospheric conditions (50 hPa, 5 ppmv H_2O, 1 ppbv HCl, 0.5 ppbv $ClONO_2$, 10 ppbv HNO_3, 0.15 ppbv H_2SO_4 and 10 background aerosol particles cm^{-3}).

Solid red and blue lines depict ternary aerosol (STS), dashed lines binary aerosol. Solid green and orange lines represent NAT particles with density 10^{-1} cm^{-3}, dashed lines 10^{-4} cm^{-3}. The NAT and NAD formation temperature (T_{NAT}) under these conditions is shown by the vertical dashed line.

Background

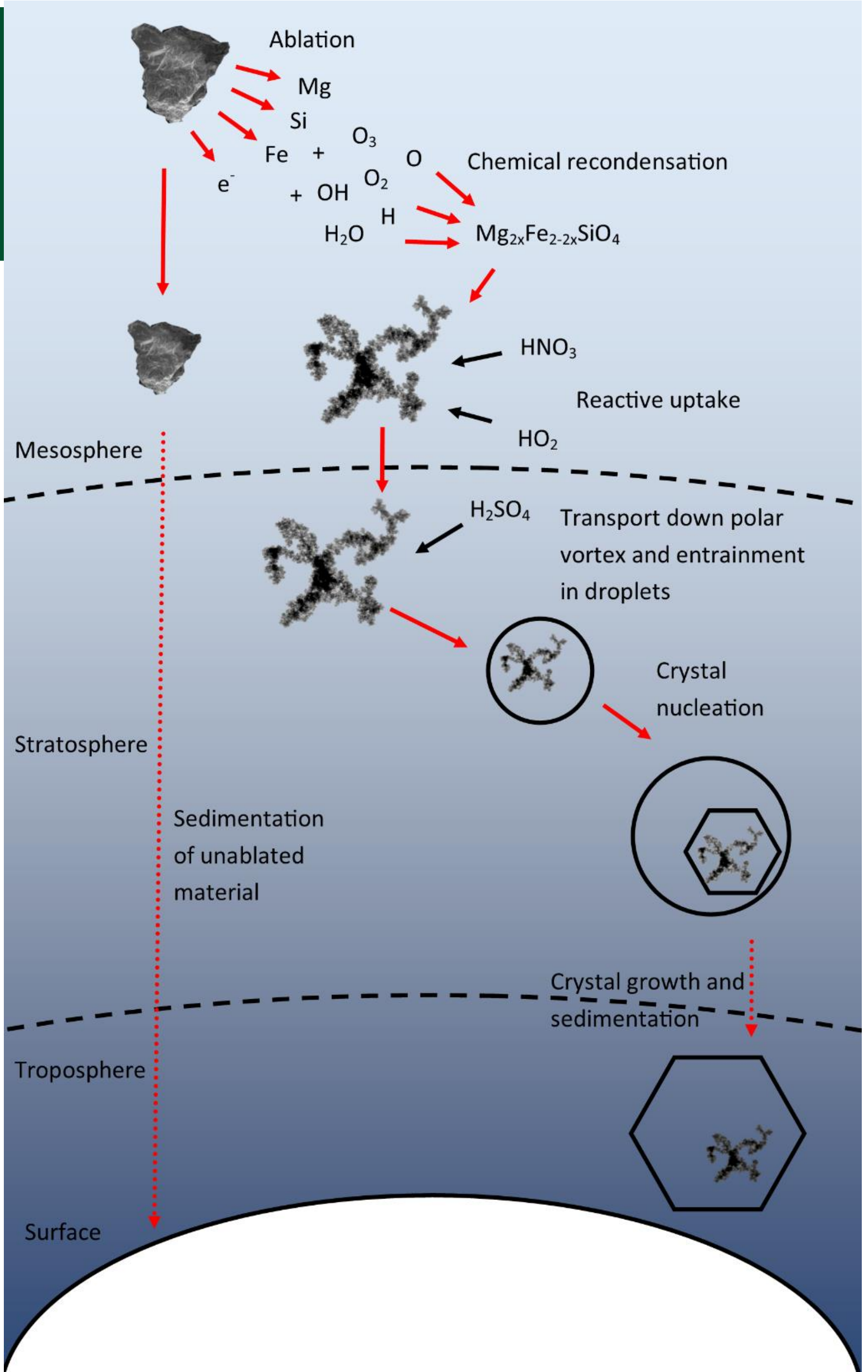
UNIVERSITY OF LEEDS



Background

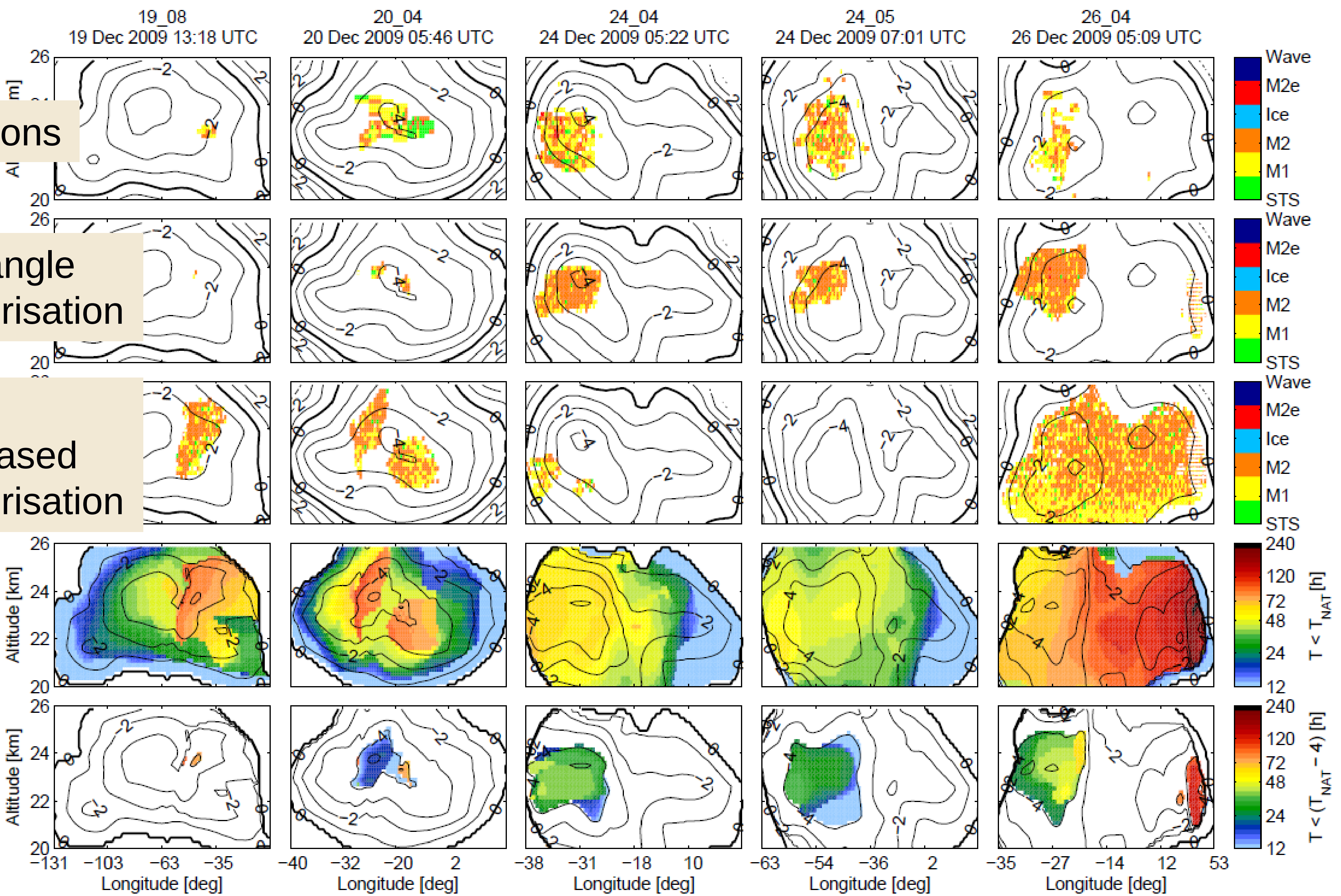
Meteoric smoke particles:

Formed from re-condensation of metal vapours released on ablation of meteoroids.



Motivation

UNIVERSITY OF LEEDS



Background

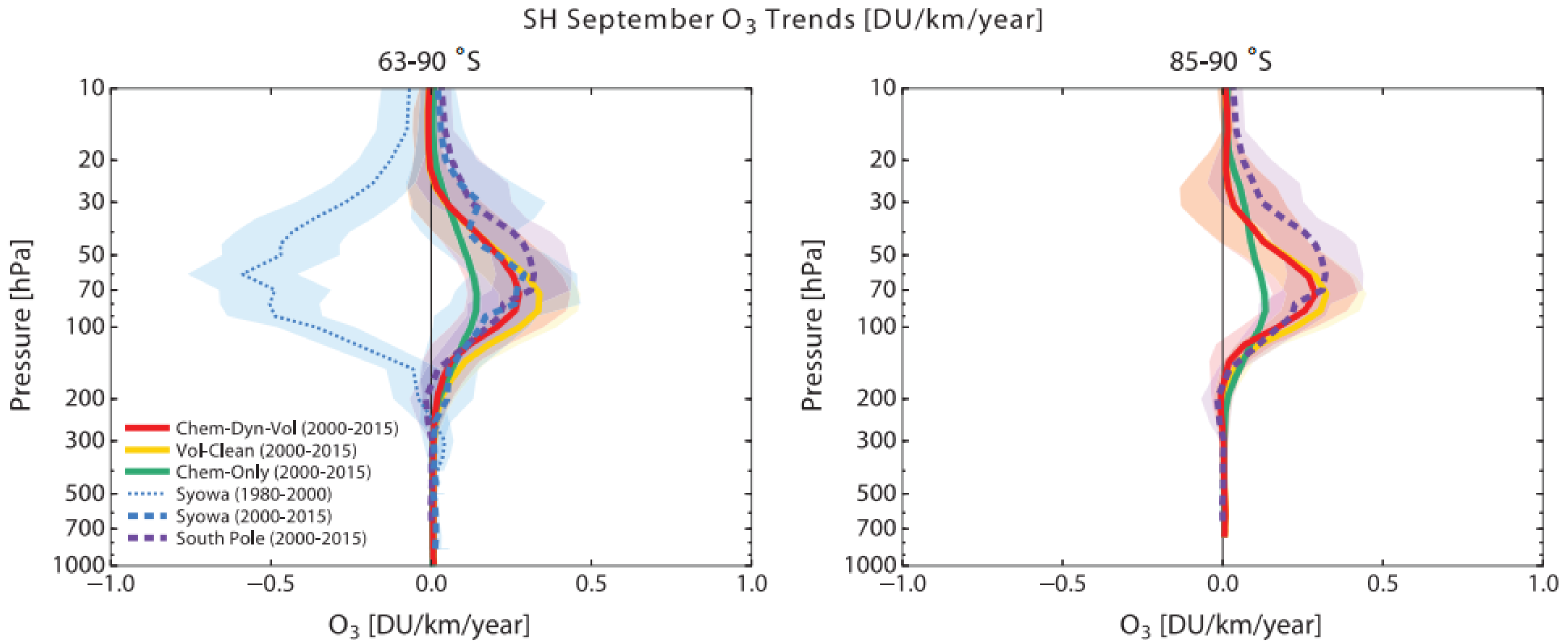
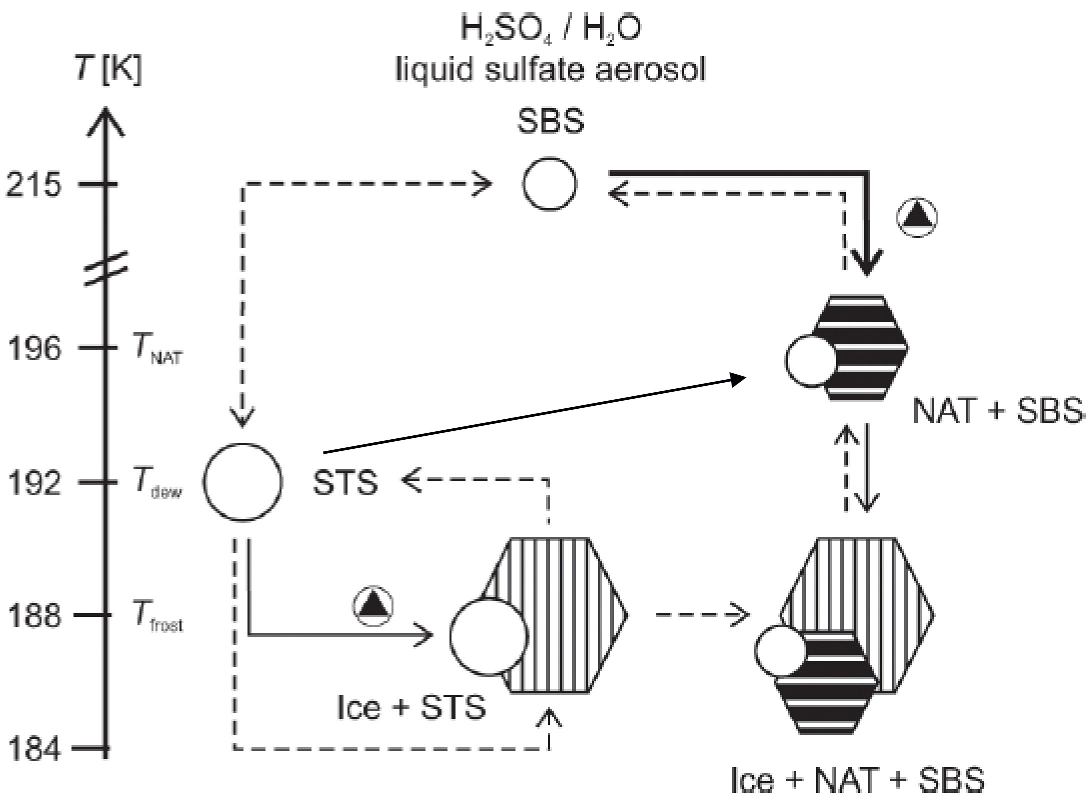


Fig. 2. Trends in Southern Hemisphere (SH) polar cap ozone profiles in September. Ozone data from balloons at the Syowa (69°S, 39.58°E) (**Left**) and South Pole (**Right**) stations, along with model simulations averaged over the polar cap and over 85°S to 90°S, respectively, are shown versus pressure. The shading represents the uncertainties on the trends at the 90% statistical confidence interval.

South polar ozone shows signs of recovery but predictive capacity in the Arctic requires understanding of aerosol processes and climate change.

Solomon et al., *Science*, **2015**, 353, 269-273

Background



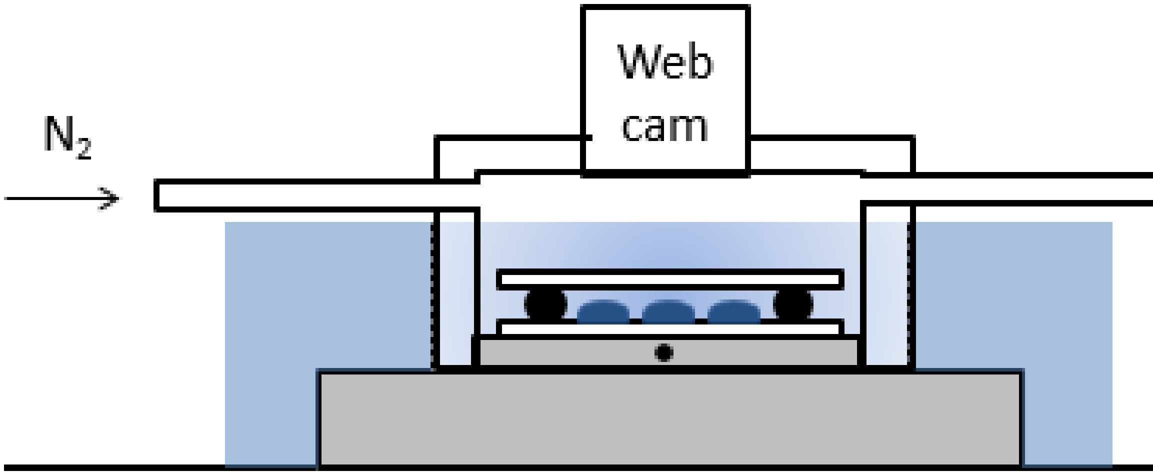
'It is estimated that denitrification due to this mechanism [het. nucleation on H_2O ice] could potentially be responsible for as much as 80% of that observed.'

Mann et al., *J. G. R.: Atmos.*, **2005**, 110, D08202

Hoyle et al., *Atmos. Chem. Phys.*, **2013**, 13, 9577–9595

EF 600 Setup

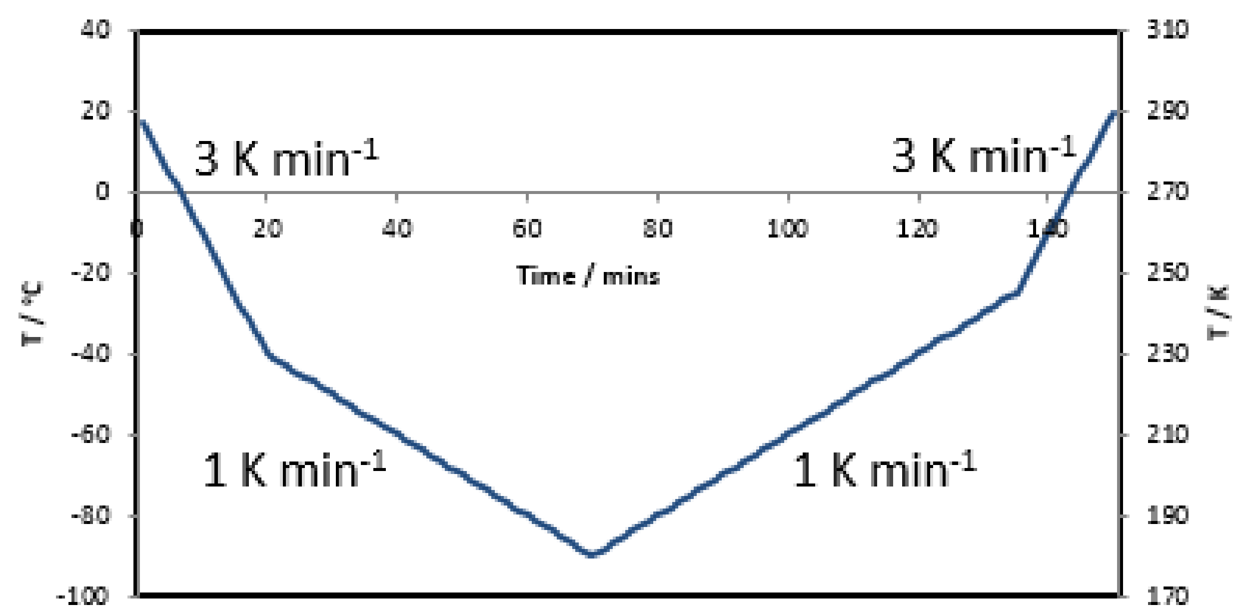
ERSITY OF LEEDS



Heterogeneous particles

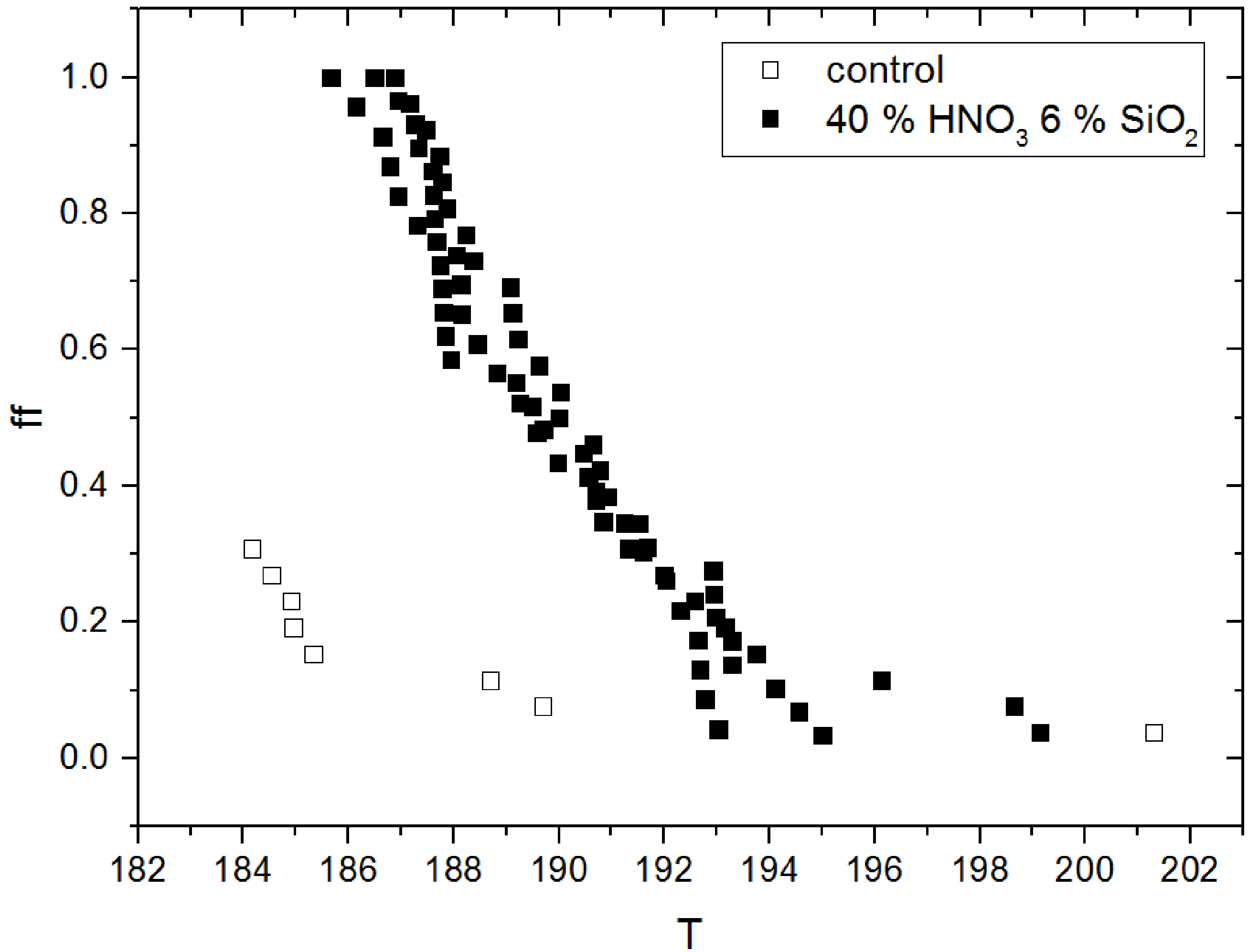
Sonicated in H_2O for 1 hour

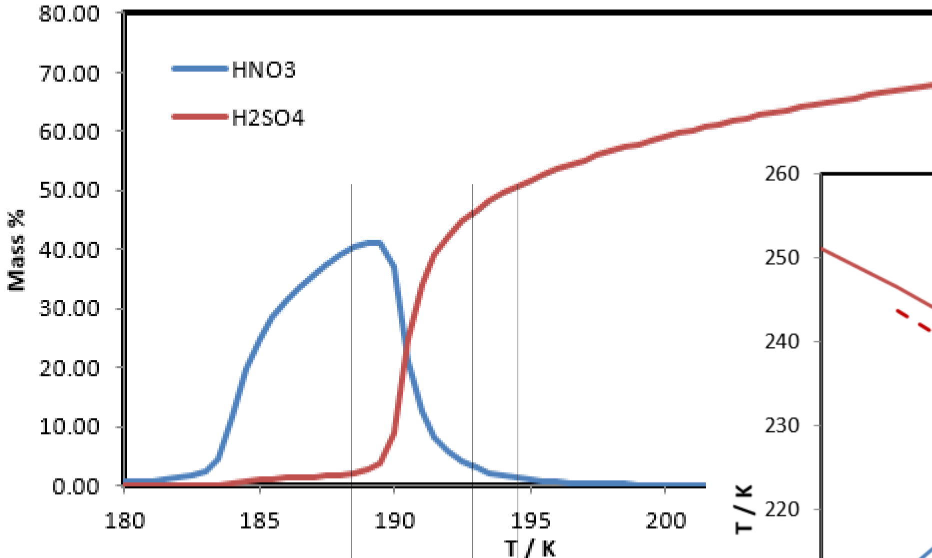
Used to dilute HNO_3



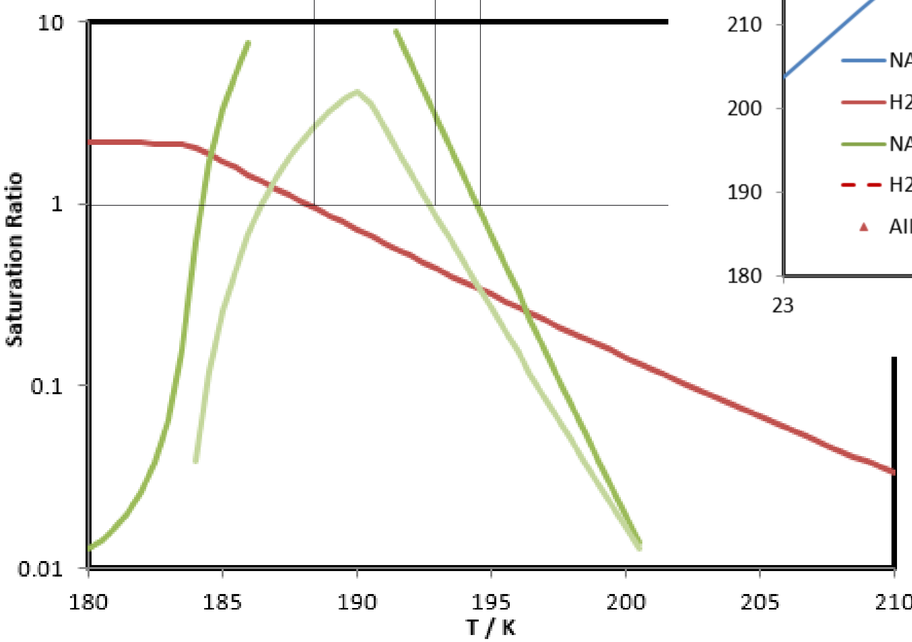
Fraction Frozen

UNIVERSITY OF LEEDS

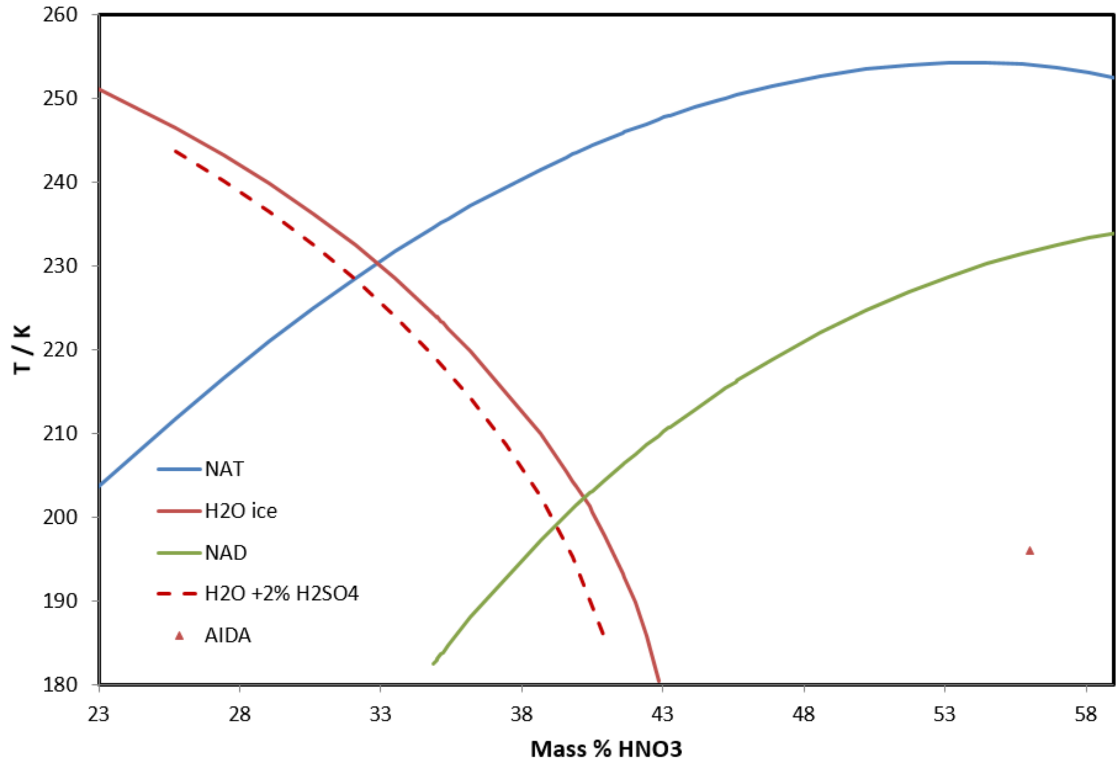




80.00
70.00
60.00
50.00
40.00
30.00
20.00
10.00
0.00
Mass %
HNO3
H2SO4
180
185
190
195
200
T / K



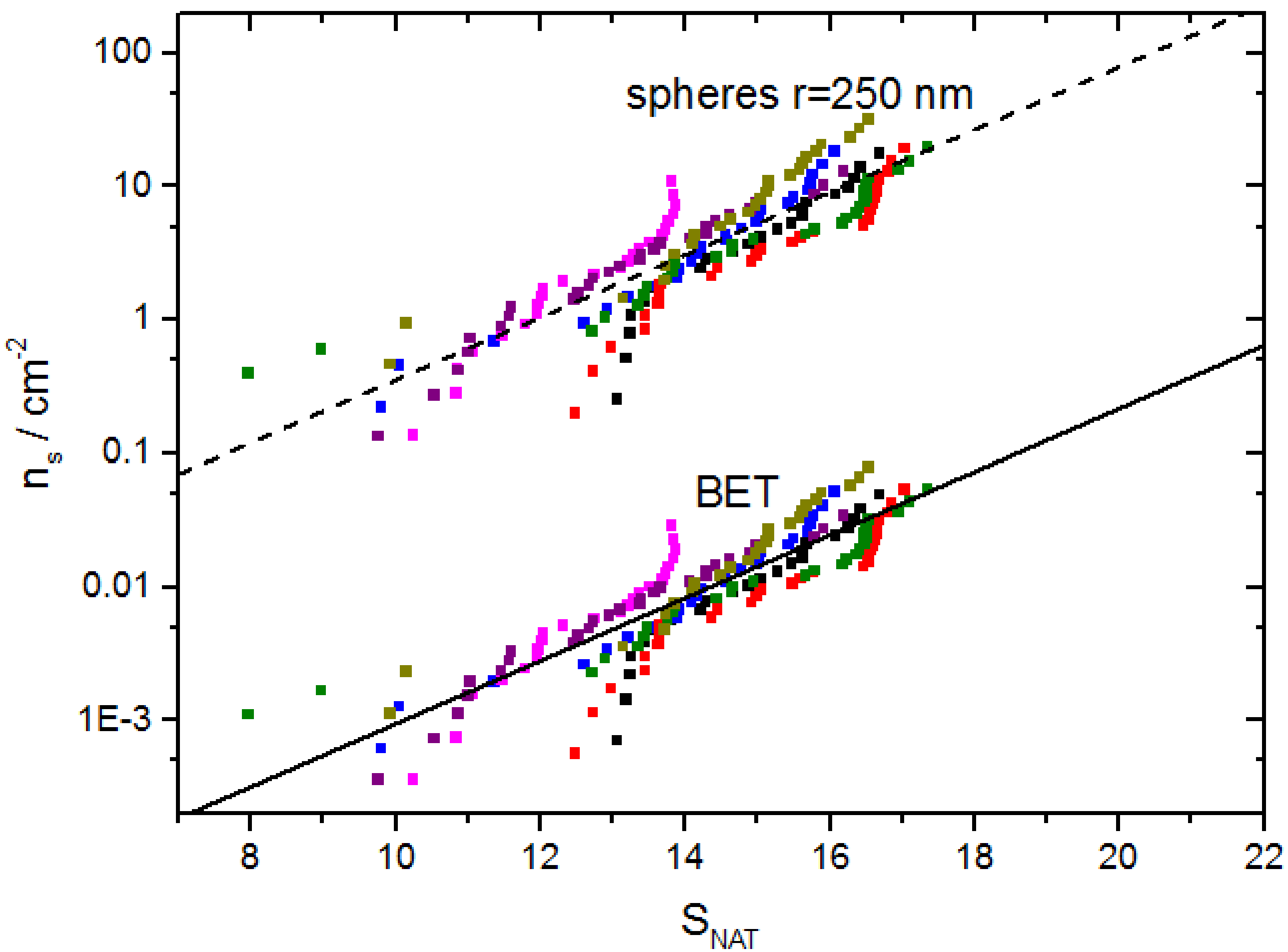
10
1
0.1
0.01
Saturation Ratio
180
185
190
195
200
205
210
T / K



260
250
240
230
220
210
200
190
180
T / K
NAT
H2O ice
NAD
H2O +2% H2SO4
AIDA
23
28
33
38
43
48
53
58
Mass % HNO3

n_s activity parameterisation

UNIVERSITY OF LEEDS



Atmospheric observations

UNIVERSITY OF LEEDS

C. Voigt et al.: NAT formation at low NAT supersaturation 1373

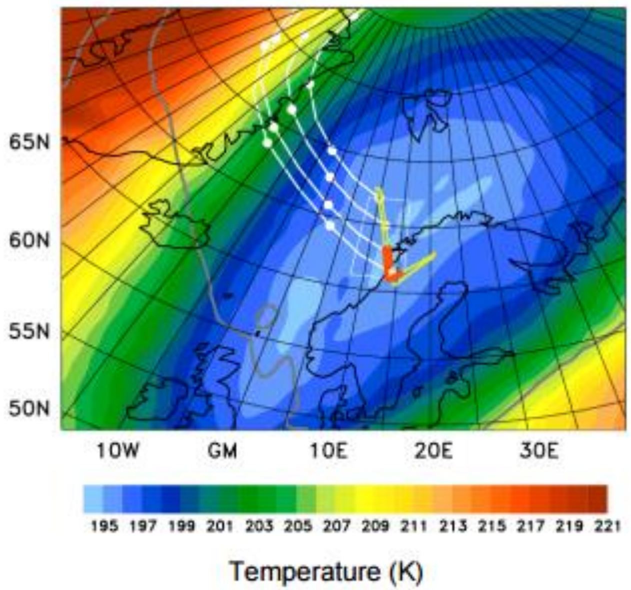
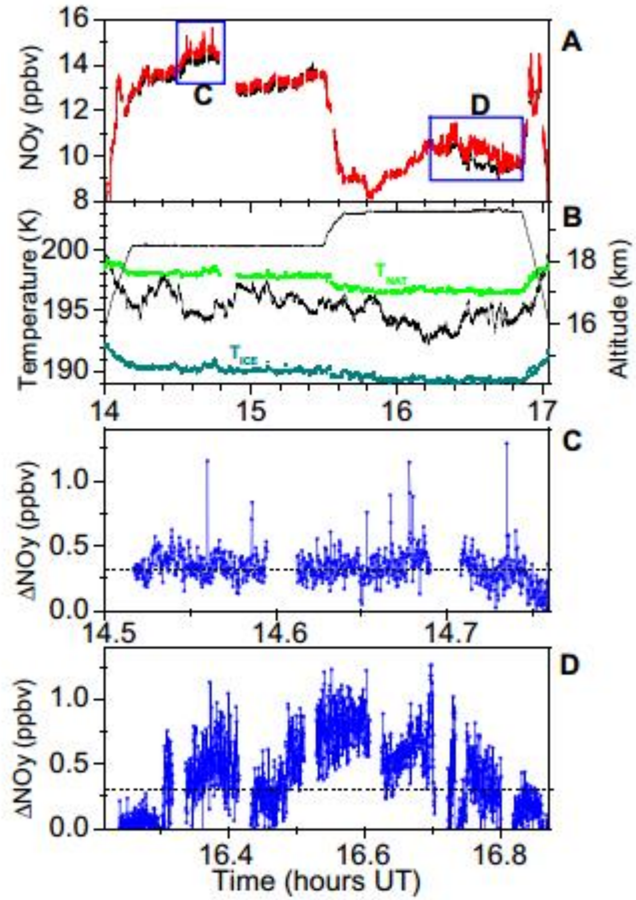
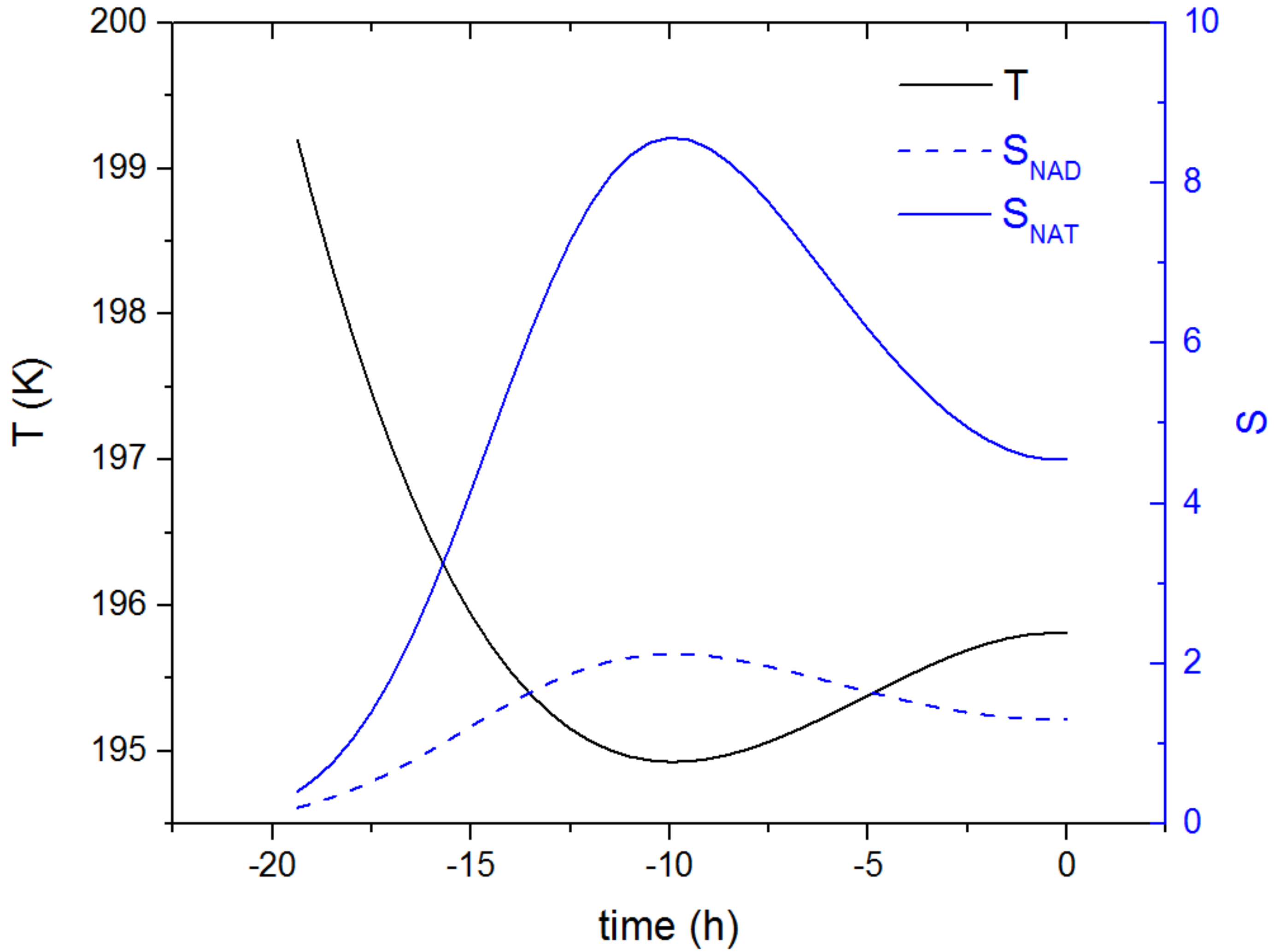


Fig. 1. ECMWF analysed temperature (color coded) at 70 hPa on 6 February 2003 at 12:00 UT. The flight path is marked in yellow and the part of the flight with particle observations is marked by the thick red line. White lines indicate selected air parcel backward trajectories. Each white dot marks a duration of 12 h. The white square indicates the region, in which the trajectories are released. Regions with $T < T_{NAT}$ are inside the blue 197 K contour line. The thick grey line is the vortex edge, defined by a potential vorticity of 3×10^{-6} km^2 kg^{-1} s^{-1}.



Voigt et al., *Atmos. Chem. Phys.*, **2005**, 5, 1371–1380

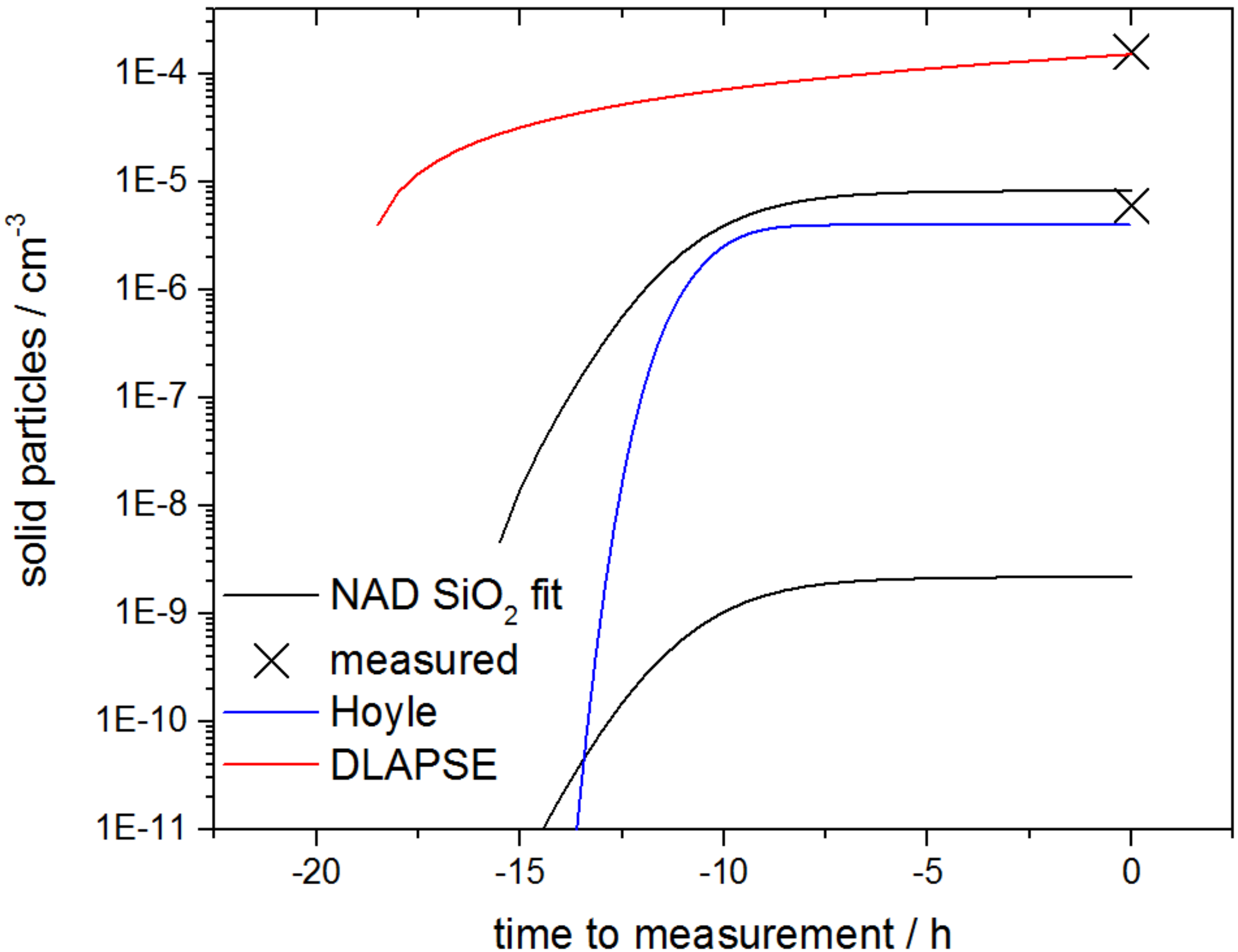
ECMWF => T and S



200
199
198
197
196
195
T (K)
10
8
6
4
2
0
S
-20
-15
-10
-5
0
time (h)
T
S_NAD
S_NAT

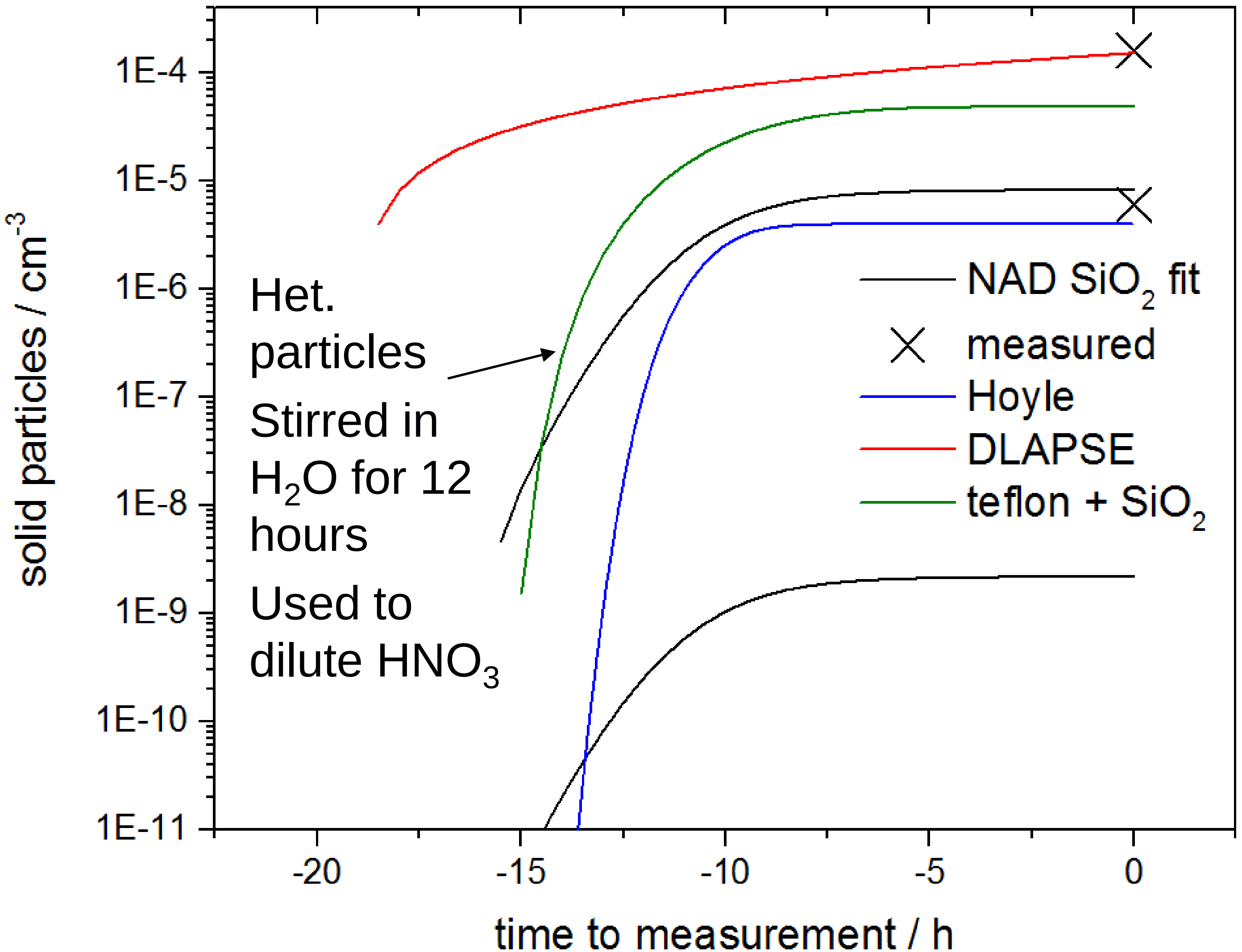
Comparison of nucleation schemes

UNIVERSITY OF LEEDS



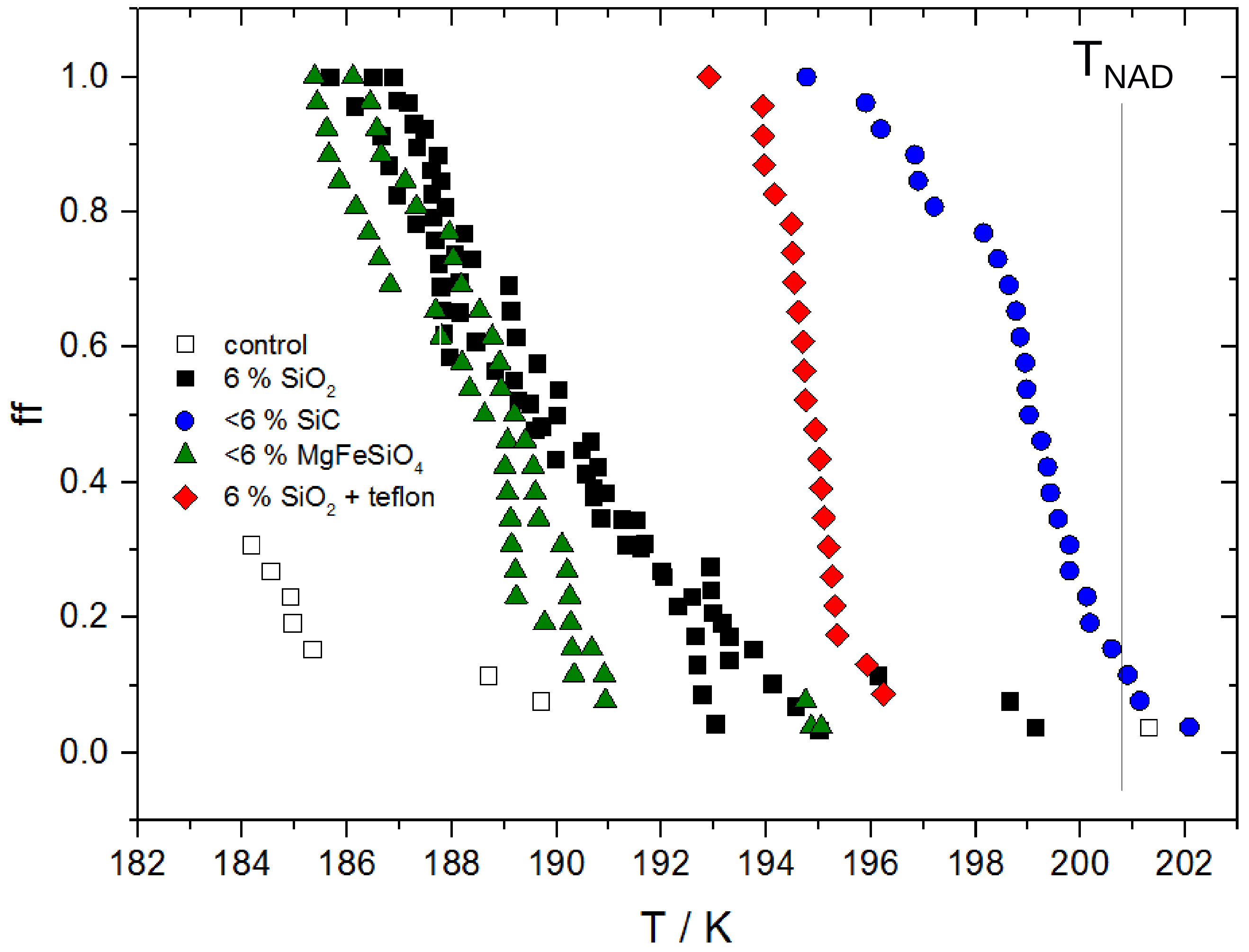
Interesting contamination effects

UNIVERSITY OF LEEDS



Other nuclei?

UNIVERSITY OF LEEDS



Conclusion / Further work

UNIVERSITY OF LEEDS

- Can measure nucleation kinetics of nitric acid hydrates in the lab.
- SiO_2 does not give sufficient activity to explain observed PSC crystal number densities
- Other materials can nucleate more efficiently, why?
- What other materials are atmospherically relevant?

Conclusion / Further work

UNIVERSITY OF LEEDS

Funding: ERC - CODITA
Levehulme - PETALS

Plane Group

Murray Group

Dr. Tamsin Malkin