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Modelling the progression of aerosol particles in the Arctic Stratosphere including the seasonal source of meteoric smoke particles

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We use the v8.4 UK Chemistry and Aerosol (UKCA) model, a stratosphere-troposphere composition-climate model, to simulate two types of stratospheric aerosol particles - pure sulphuric acid particles and sulphuric acid particles with a MSP-core.

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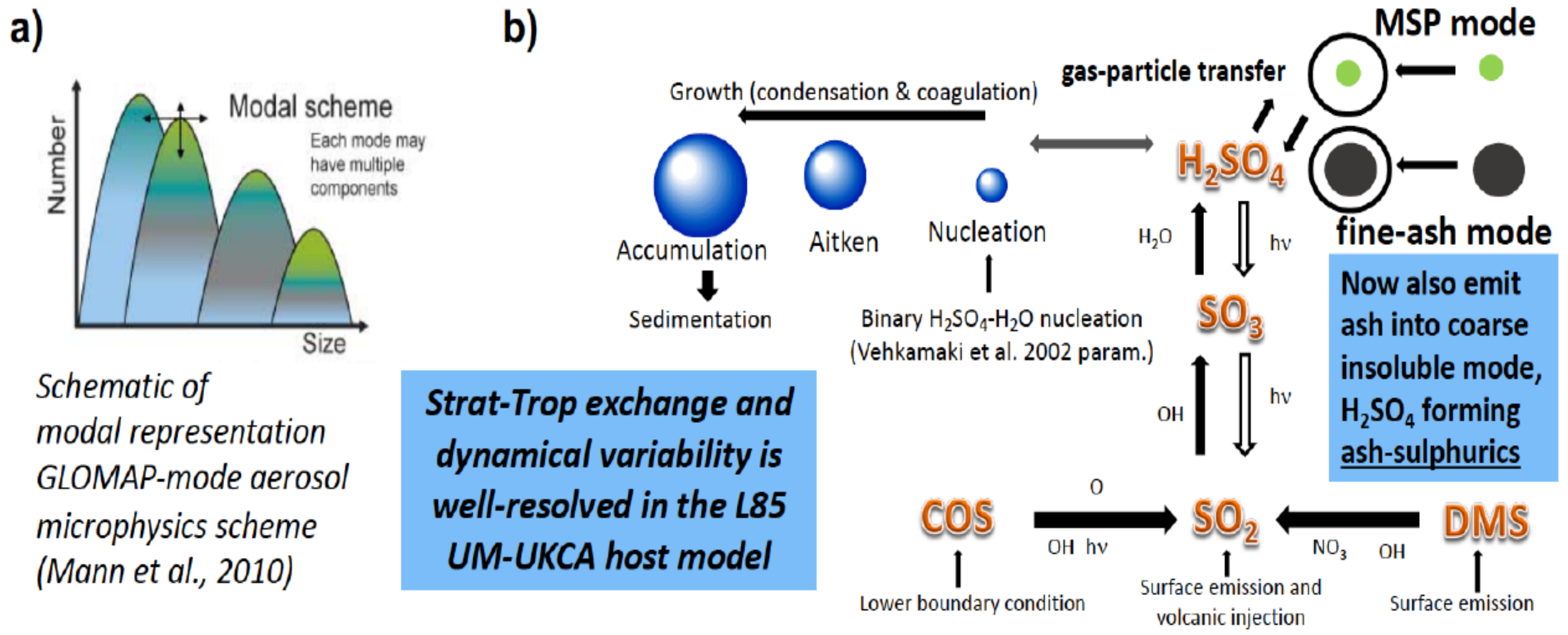
UK's interactive stratospheric aerosol model **UM-UKCA** (Dhomse et al., 2014; Mann et al., 2015) **adapted to resolve pure sulphurics & meteoric-sulphurics** (Brooke et al., 2017; Marshall et al., 2018) and now new configuration which co-emits volcanic SO2 and ash → **also ash-sulphuric particles**

UKCA sulphur chemistry & aerosol microphysics

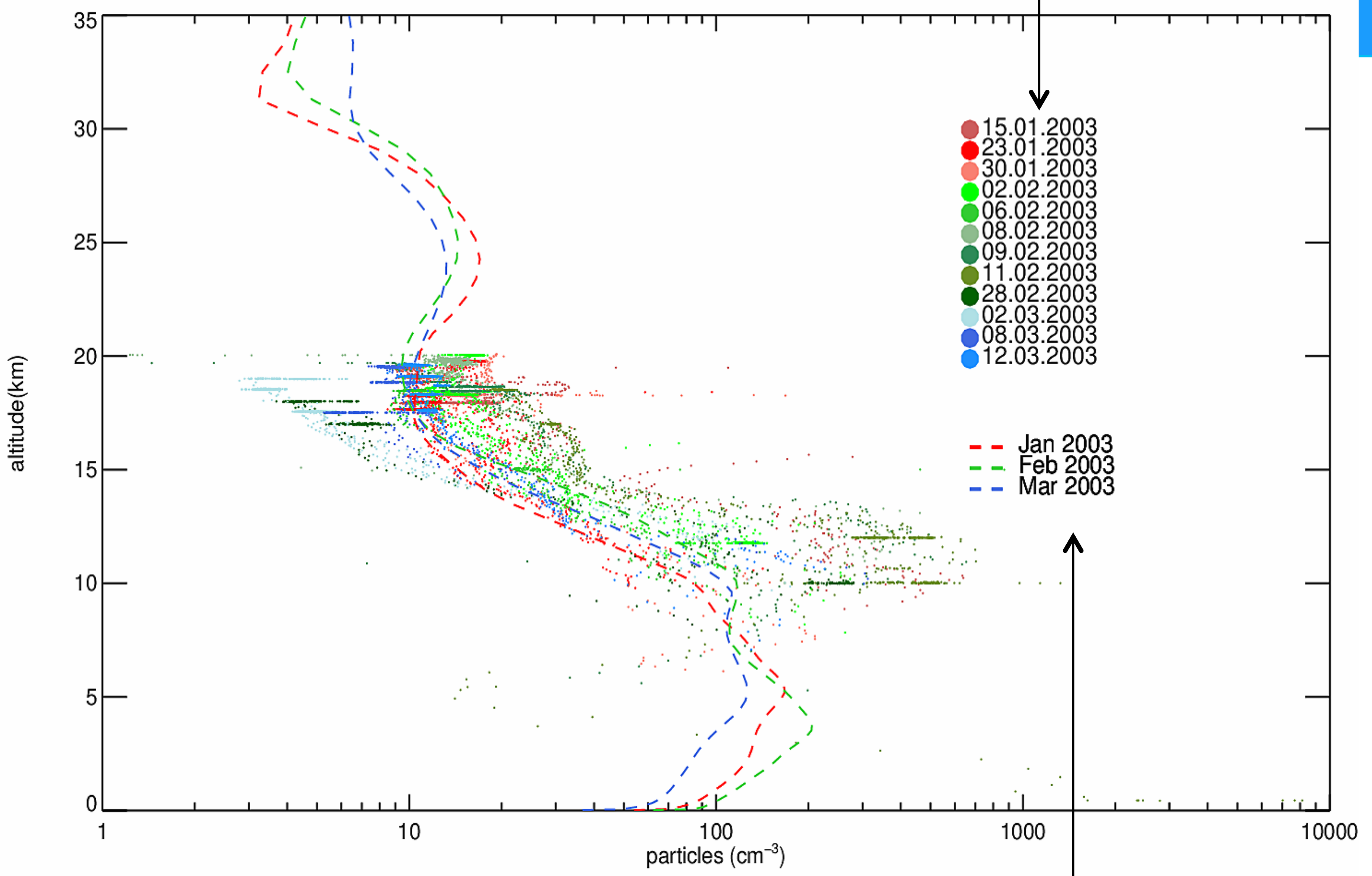


Schematic of UM-UKCA stratospheric sulphur chemistry (see Dhomse et al., 2014) and coupling to GLOMAP-mode aerosol microphysics: adapted to resolve both pure sulphuric & meteoric-sulphuric particles (Murphy et al., 2014)

Model configuration includes:

- Nudging to reanalysis data for 2001-2003
- Daily influx of 7.9 tons of meteoric smoke particles (MSP)
- photolysis of sulphuric acid to SO_2 at high altitudes

Observations of particles above 10nm diameter (Weigel, 2014)



Mean monthly modelled number concentration of particles over 10nm in diameter, CN10

Our model is able to reproduce the vertical profile of particles over 10nm diameter, as observed in in-situ measurements.

We used the nudged configuration of the UM-UKCA stratosphere-troposphere composition-climate model to reproduce the vertical profile of stratospheric particles measured in-situ during the EUPLEX 2003 campaign.

Mean monthly CN10 number concentration between 1998 and 2003 averaged over 30-45N, 45-60N and 60-90N

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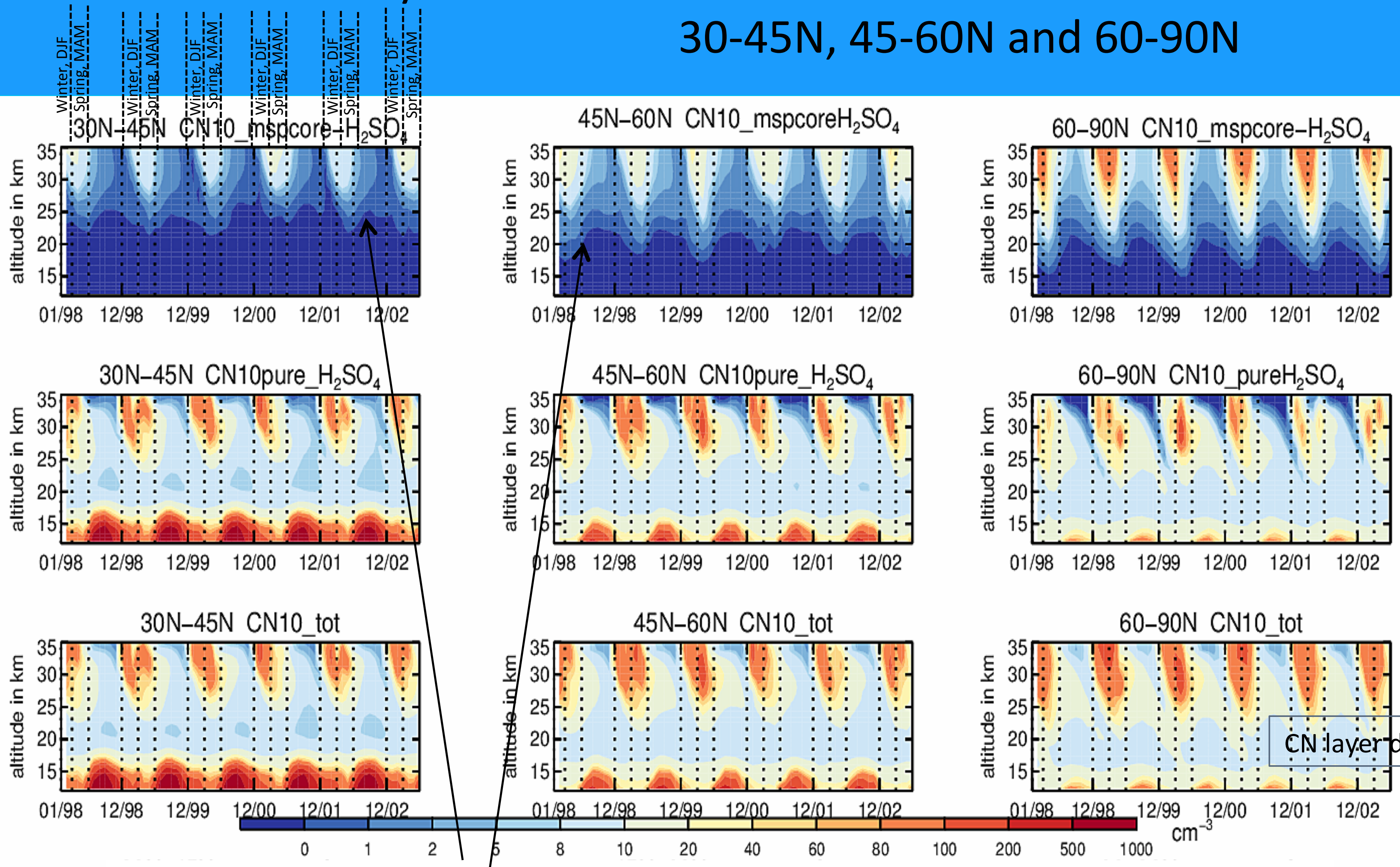

Influx of MSP from higher altitudes occurs upto about 20km in the winter time Arctic

Number concentrations of particles with a msp core reaches 80-100 per cc between 28-30km and funnels down to the lower latitudes with the polar vortex subsidence.

Number concentrations of sulphuric acid particles increases in Spring to 20-40 particles per cc around 20km to 100-200 particles per cc between 25-30km. After the polar night the rapid formation of sulphuric acid and the presence of meteoric course facilitate nucleation.

CN layer descends from 30 to 25km in the Spring

The MSPs funnel down the polar vortex reaching the tropics and mid-latitudes

% of sulphuric acid particles with meteoric core simulated in the model

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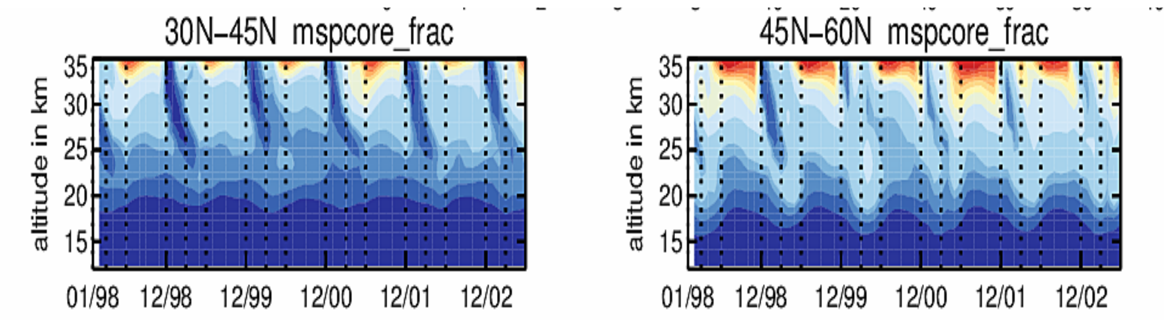
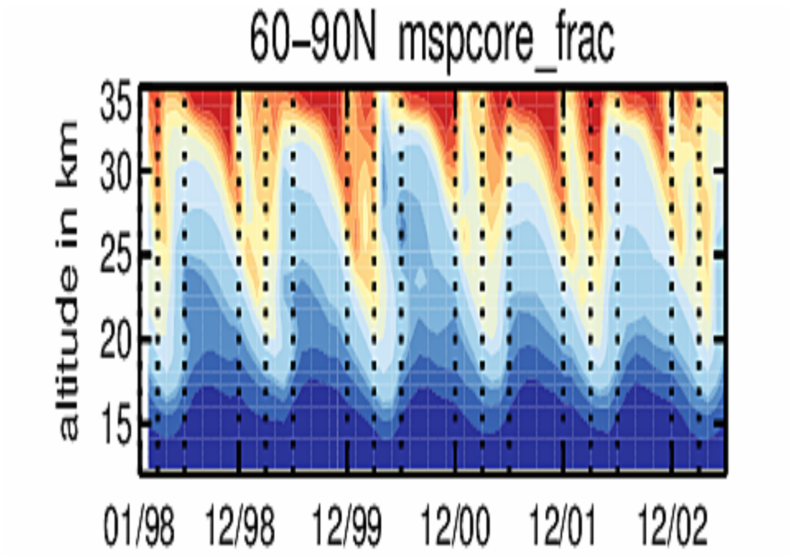
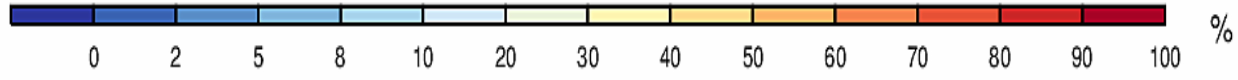




Particles with a msp-core are maximum during winter at the pole:
Over 80% of particles in the stratospheric aerosols around 35km contain a msp-core whereas at 25km, over 50% of particles contain a msp-core.

The fraction of msp-core particles decreases from winter to spring in 60-90N but increases in lower latitudes, showing the gradual downward descent of the meteoric particles.



Seasonal variation in aerosol concentrations over 2-km altitude bins between 21-35km

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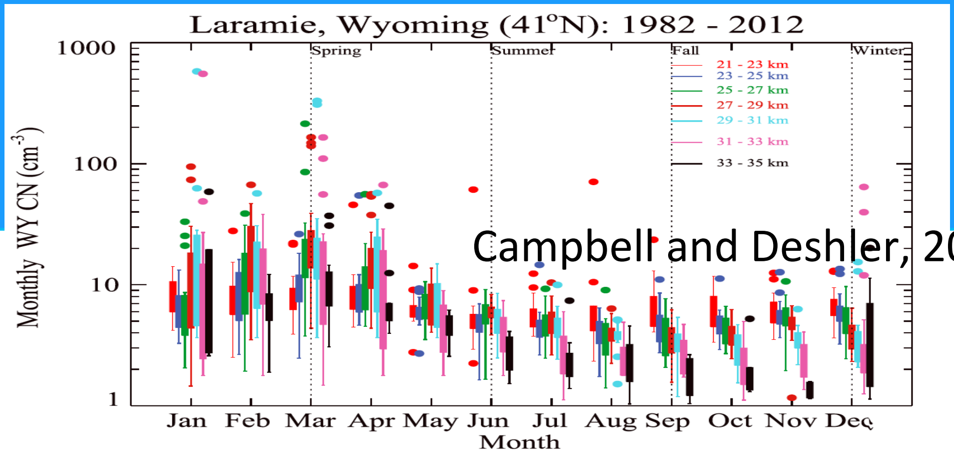
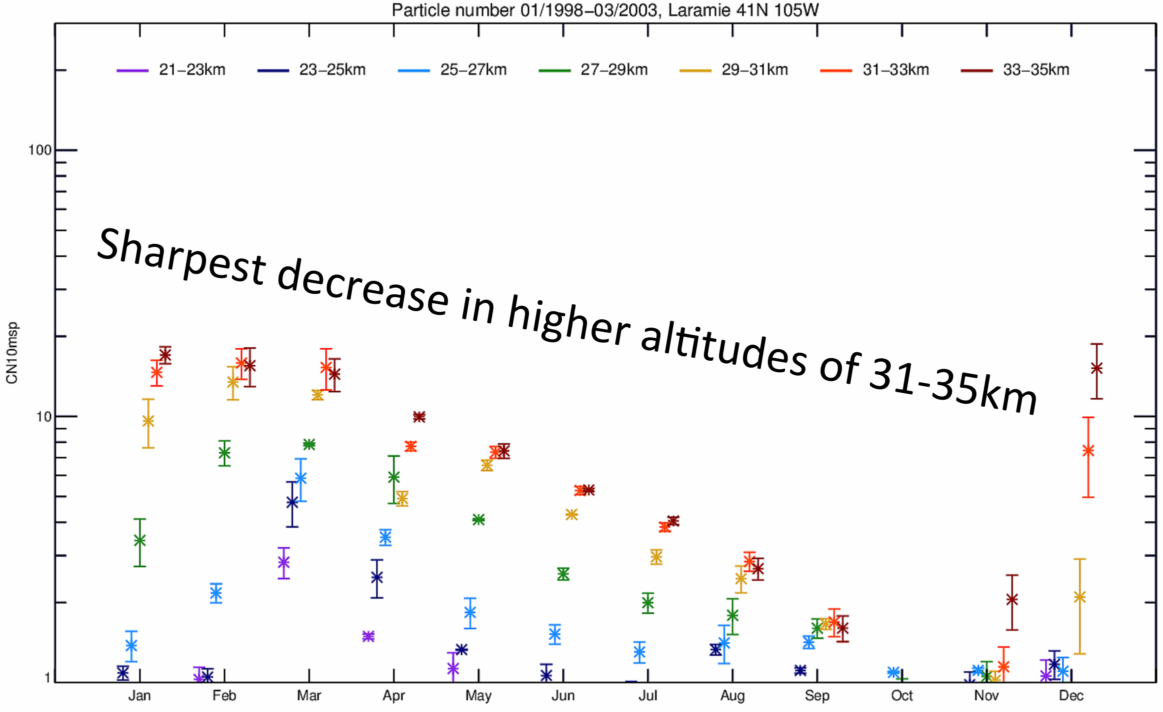


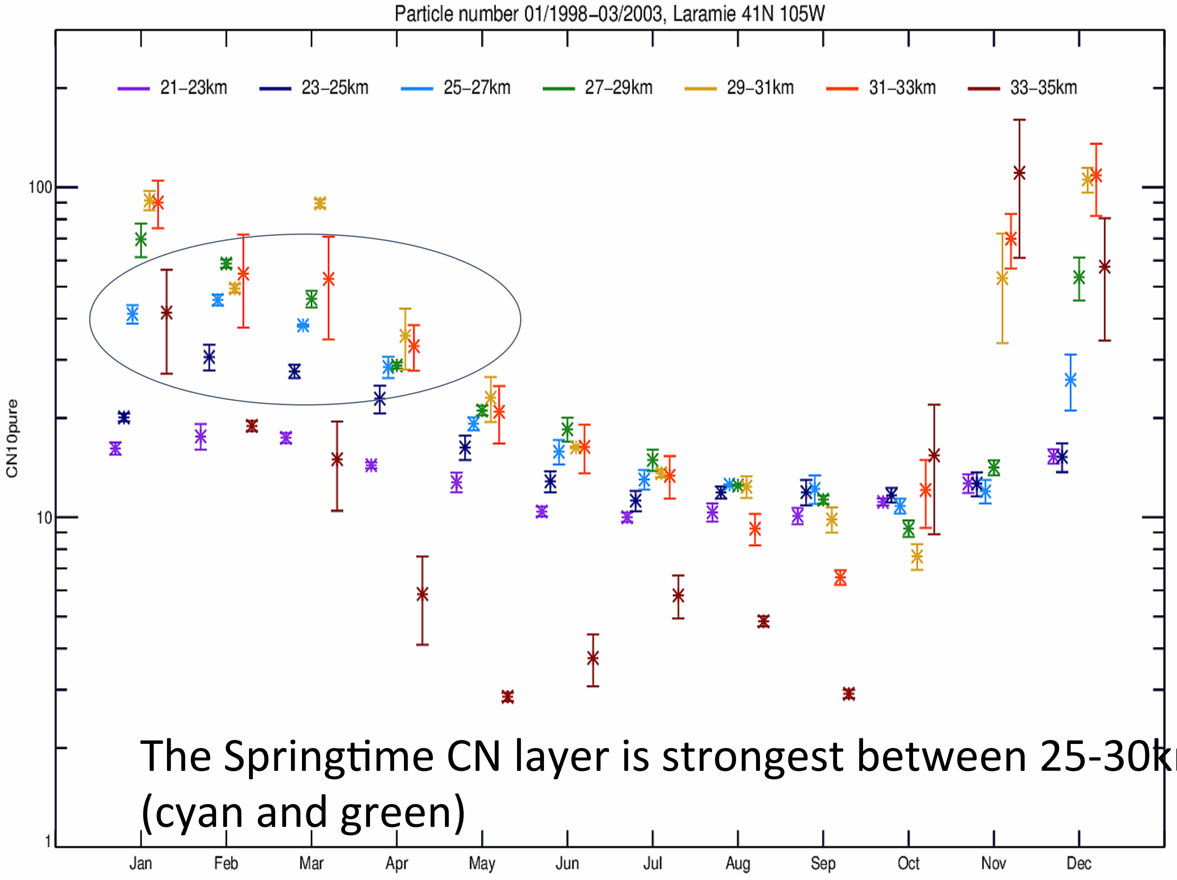
Campbell and Deshler, 2014

Sulphuric acid particles with msp core



Sharpest decrease in higher altitudes of 31-35km

Pure sulphuric acid particles



The Springtime CN layer is strongest between 25-30km (cyan and green)

Monthly mean simulated size distribution of aerosol particles at 90N, 30E from December 2002 to May 2003

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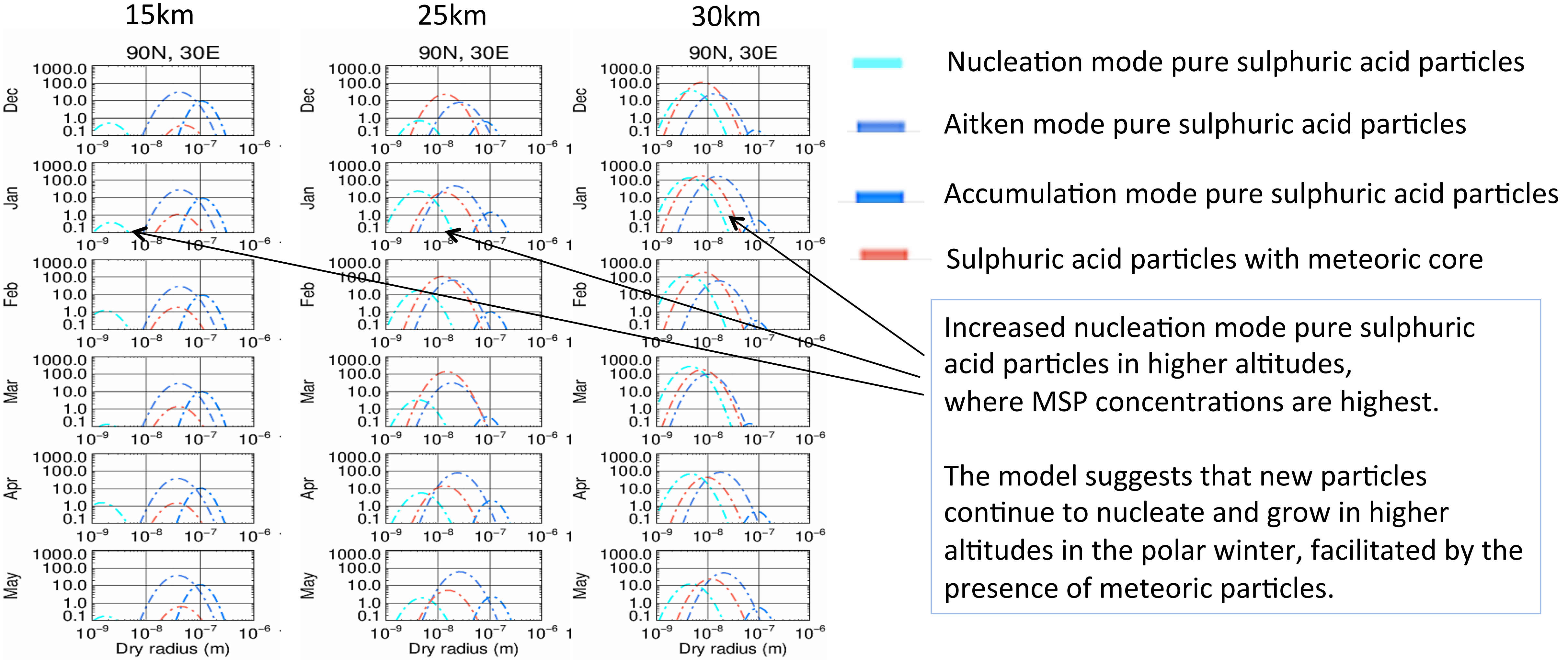

Increased nucleation mode pure sulphuric acid particles in higher altitudes, where MSP concentrations are highest.

The model suggests that new particles continue to nucleate and grow in higher altitudes in the polar winter, facilitated by the presence of meteoric particles.

At 25km in the CN layer, as Spring starts with sunlight and OH, sulphuric acid vapour concentrations recover and particles grow by condensation. The number of small particles reduce and the size distribution shifts towards larger particles towards the end of Spring.

5-day mean polar stereographic projection – Arctic, March 2003

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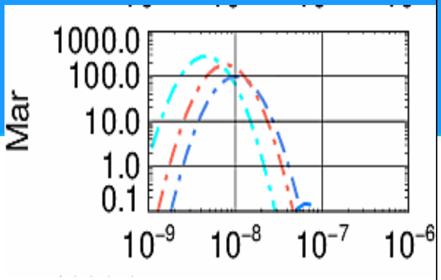
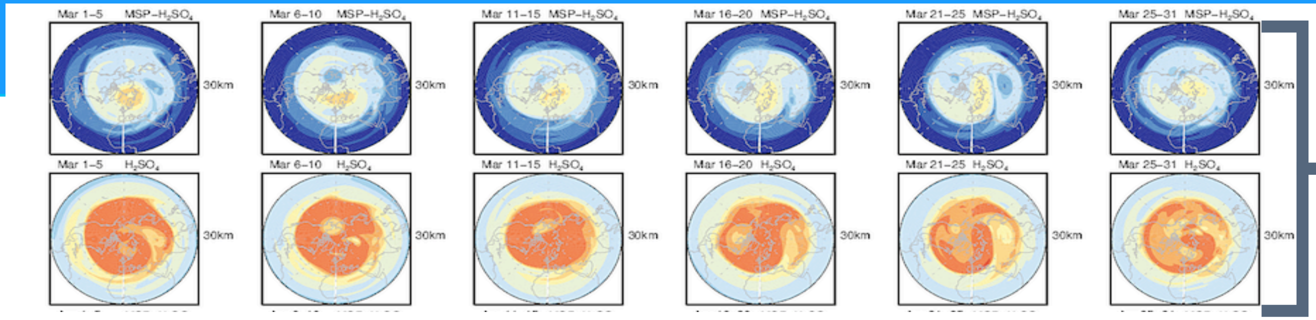
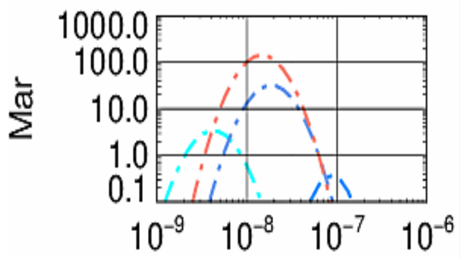


30km

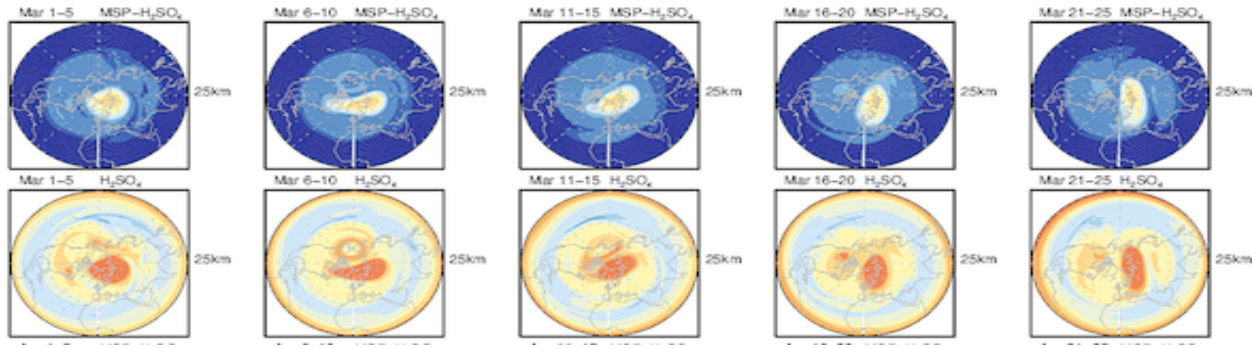


1st row: MSP-Sulphuric acid particles

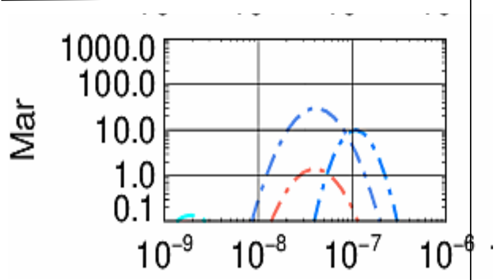
2nd row: Pure Sulphuric acid particles



25km

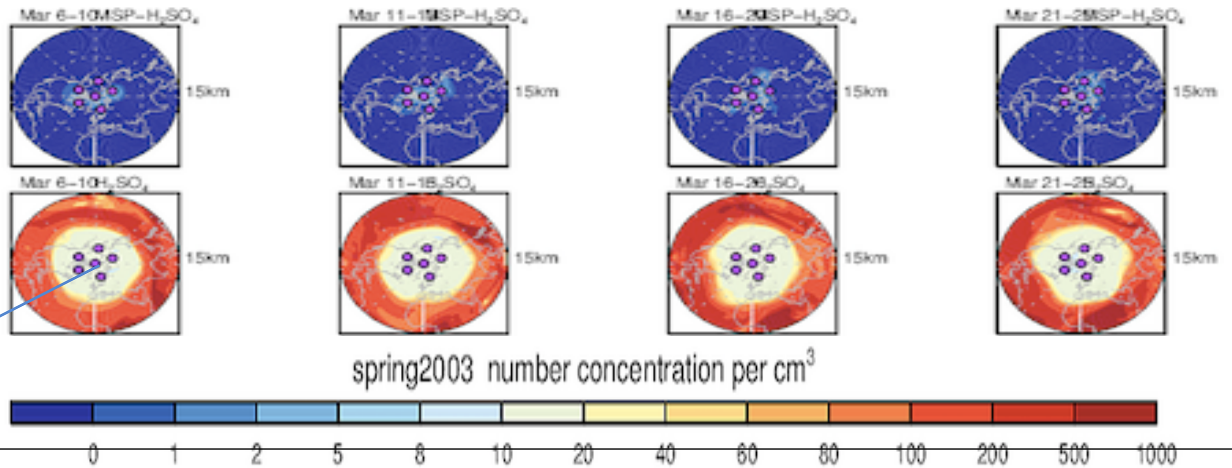


Increased concentrations of pure sulphuric acid particles in regions of high msp-core sulphuric particles.



15km

Size distribution at 90N 30E



The presence of meteoric particles has implications for homogeneous nucleation leading to the formation of pure sulphuric acid particles

Summary

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- A 3-D global stratospheric model is used to simulate the stratospheric aerosol layer between 1998 and 2003 with two types of particles: pure sulphuric acid and sulphuric acid with meteoric core
- The model configuration includes a daily influx of 7.9 tons of meteoric smoke particles and photolysis of sulphuric acid particles.
- Our model is able to simulate the vertical aerosol profiles observed during the EUPLEX 2003 campaign
- Our study shows over 50% of particles at the altitude of 25km contain a msp-core.
- The presence of meteoric cores produces particles in addition to homogeneous nucleation in the polar winter while in Spring, the size distribution shifts to larger particle sizes.

Weigel, R., Volk, C. M., Kandler, K., Hösen, E., Günther, G., Vogel, B., ... & Borrmann, S. (2014). Enhancements of the refractory submicron aerosol fraction in the Arctic polar vortex: feature or exception?. Atmospheric Chemistry and Physics, 14(22), 12319-12342.

Campbell, P., & Deshler, T. (2014). Condensation nuclei measurements in the midlatitude (1982–2012) and Antarctic (1986–2010) stratosphere between 20 and 35 km. Journal of Geophysical Research: Atmospheres, 119(1), 137-152.

Mann, Graham, James Brooke, Kamalika Sengupta, Lauren Marshall, Sandip Dhomse, Wuhu Feng, Ken Carslaw et al. The prevalence of meteoric-sulphuric particles within the stratospheric aerosol layer. No. EGU21-15390. Copernicus Meetings, 2021.

Modelling the progression of aerosol particles in the Arctic Stratosphere including the seasonal source of meteoric smoke particles

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- Meteoric Smoke Particles are considered to play a role in nucleation of solid nitric acid PSCs, strongly influencing heterogeneous chemistry in the Arctic.
- Here we use the UM-UKCA climate composition model to simulate the stratospheric aerosol layer in the Arctic with two types of particles: pure sulphuric acid and sulphuric acid with meteoric core

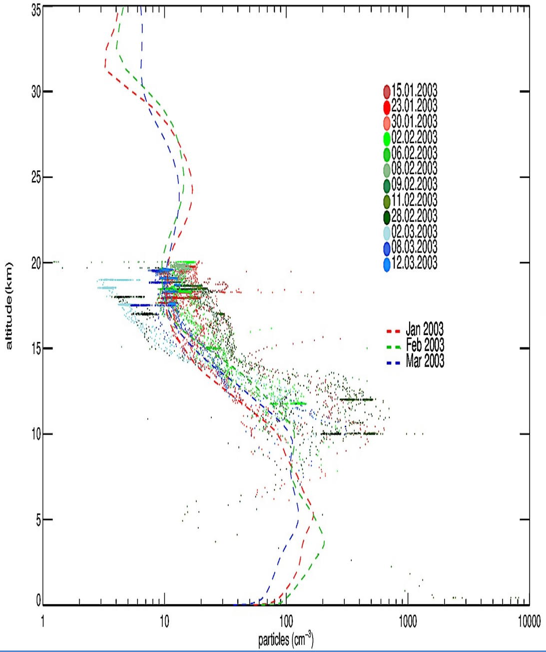


Figure 1: Comparison of simulated vertical profile with EUPLEX 2003 data. The dotted lines show modelled CN10 (per cc) for Jan, Feb and March 2003.

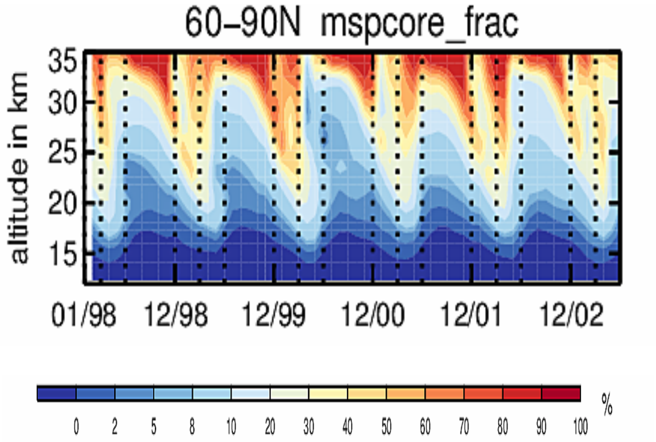
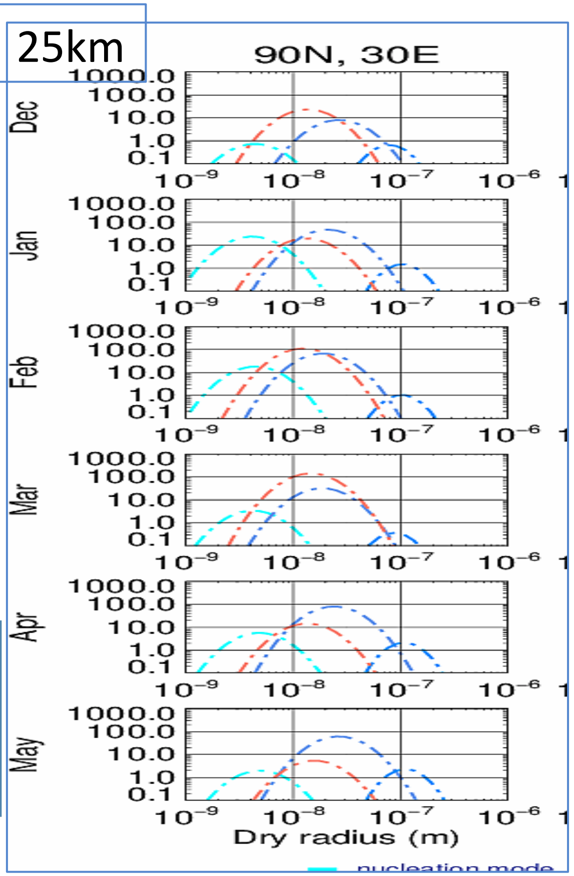


Figure 2: Fraction of particles above 10nm diameter with meteoric core

Over 50% of particles at the altitude of 25km contain a msp-core.



Our model shows that new particles continue to nucleate and grow in higher altitudes in the polar winter, dominated by particles with meteoric cores.

In the polar spring, when sulphuric acid concentrations are recovered, the size distribution shifts towards larger particles of pure sulphuric acid..

Figure 3: Winter to Spring evolution of the size distribution of aerosol particles in Arctic