# The prevalence of meteoric-sulphuric particles within the stratospheric aerosol layer

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**Abstract**

The widespread presence of meteoric smoke particles (MSPs) within a distinct class of stratospheric aerosol particles has become clear from in-situ measurements in the Arctic, Antarctic and at mid-latitudes. We apply an adapted version of the interactive stratosphere aerosol configuration of the composition-climate model UM-UKCA, to predict the global distribution of meteoric-sulphuric particles nucleated heterogeneously on MSP cores. We compare the UM-UKCA results to new MSP-sulphuric simulations with the European stratosphere-troposphere chemistry-aerosol modelling system IFS-CB05-BASCOE-GLOMAP. The simulations show a strong seasonal cycle in meteoric-sulphuric particle abundance results from the winter-time source of MSPs transported down into the stratosphere in the polar vortex. Coagulation during downward transport sees high latitude MSP concentrations reduce from ˜500 per cm3 at 40km to ˜20 per cm3 at 25km, the uppermost extent of the stratospheric aerosol particle layer (the Junge layer). Once within the Junge layer's supersaturated environment, meteoric-sulphuric particles form readily on the MSP cores, growing to 50-70nm dry-diameter (Dp) at 20-25km. Further inter-particle coagulation between these non-volatile particles reduces their number to 1-5 per cc at 15-20km, particle sizes there larger, at Dp ˜100nm. The model predicts meteoric-sulphurics in high-latitude winter comprise >90% of Dp > 10nm particles above 25km, reducing to ˜40% at 20km, and ˜10% at 15km. These non-volatile particle fractions are slightly less than measured from high-altitude aircraft in the lowermost Arctic stratosphere (Curtius et al., 2005; Weigel et al., 2014), and consistent with mid-latitude aircraft measurements of lower stratospheric aerosol composition (Murphy et al., 1998), total particle concentrations also matching in-situ balloon measurements from Wyoming (Campbell and Deshler, 2014). The MSP-sulphuric interactions also improve agreement with SAGE-II observed stratospheric aerosol extinction in the quiescent 1998-2002 period. Simulations with a factor-8-elevated MSP input form more Dp>10nm meteoric-sulphurics, but the increased number sees fewer growing to Dp ˜100nm, the increased MSPs reducing the stratospheric aerosol layer's light extinction.

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# 1. INTRODUCTION:
# METEORIC PARTICLES WITHIN THE STRAT-AEROSOL LAYER

The existence of the stratospheric aerosol layer was confirmed in the late 1950s by mid-latitude balloon-borne impactor measurements (Junge et al., 1961).

The role of extra-terrestrial particle influence on the stratospheric aerosol layer has remained a topic of continued debate since the first particle composition measurements of the stratospheric aerosol layer (Junge and Manson, 1961).

When publishing the first measurements of the stratospheric aerosol layer, Junge et al. (1961) explain that the high concentrations of particles in the 10 to 100nm size range, measured by Aitken particle counter, suggest a peak in the particle size distribution at ~30nm, similar to that observed earlier in the troposphere via ion mobility counter (Junge et al. 1955).

Hamill et al. (1976) presented an overview of particle formation mechanisms, with both homogeneous and heteorogenous nucleation effective pathways, and the earliest models of the stratospheric aerosol layer including both particle sources.

Rosen et al. (1978) applied a steady state 1D model of the stratospheric aerosol layer, noting many sulphuric particles contain solid inclusions and identified a "transition zone" at the uppermost Junge layer, with particles reaching above 35km evaporating down to their core size.

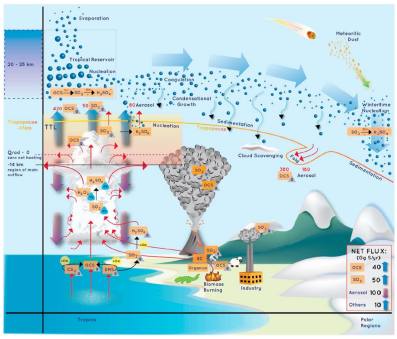
The first 1D time-dependent numerical model to assess the evolution of the Junge layer vertical distribution was developed by Turco et al. (1979). Their model included the sulphur chemistry and microphysics that combine to effect stratospheric aerosol properties, and specifically identify the size distribution and vertical distribution of the sub-population of particles with a refractory core inclusion, aligning with recent measurements from that time (Farlow et al., 1977) showing ~30% of stratospheric particles are of this type.

Hunten et al. (1980) applied a steady state dust microphysics model to assess the growth of meteoric smoke population, finding the particles are ~5nm diameter upon transport into the uppermost Junge layer.

Turco et al. (1981) progressed the Turco et al. (1979) 1D model to additionally resolve the nucleation of MSP-core sulphuric particles, these internally-mixed particles forming alongside pure sulphuric particles.

That 1981 paper discusses how the two particle types interact, MSP-sulphuric particles forming droplets around the MSP cores, the model experiments suggesting most meteoric material is found within particles larger than 100nm in the lowermost Junge layer.

# 2. MOSTLY HOMOGENEOUS NEW PARTICLE FORMATION IN LOWER STRAT-AEROSOL LAYER



Homogeneous nucleation of "pure sulphuric particles" occurs all year-round in the tropical upper troposphere and generates the major source of stratospheric particles (Brock et al., 1995).

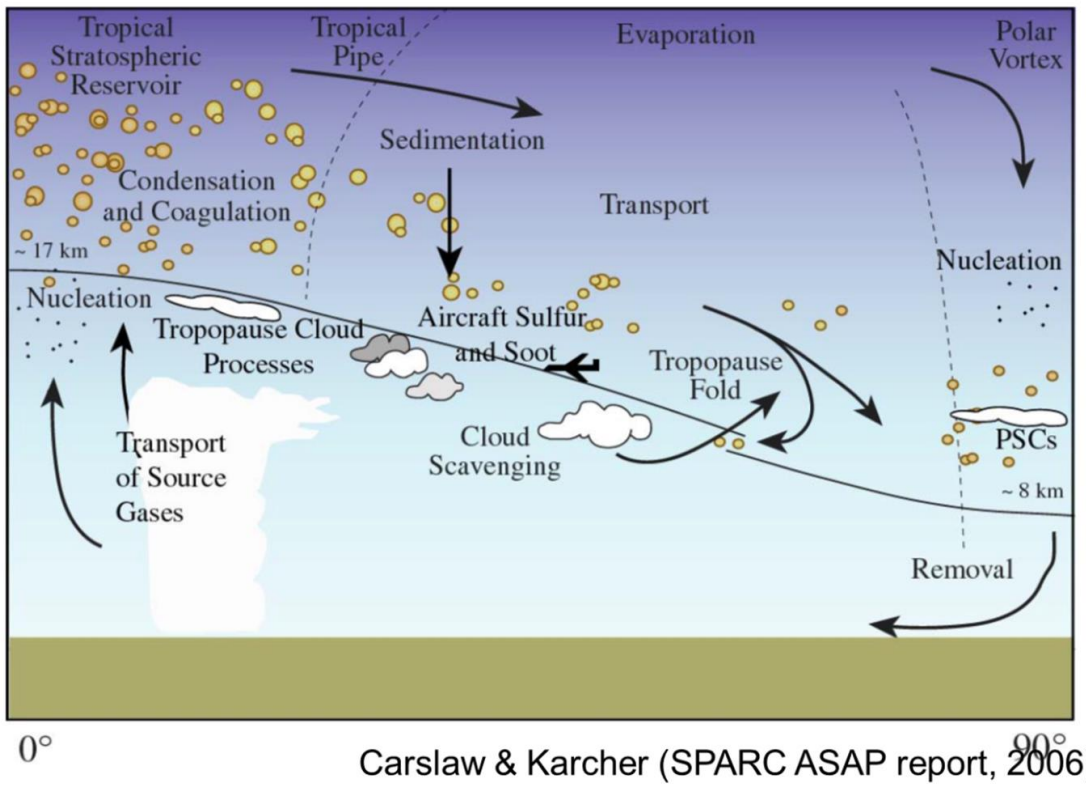
The newly formed particles are transported upwards into the tropical stratospheric reservoir and then meridionally within the prevailing Brewer-Dobson circulation, its seasonal cycle tending to preferentially transport towards the winter pole (e.g. Kremser et al., 2016).

Homogeneous nucleation also occurs in springtime in the polar stratosphere, for example causing the observed October CN maximum at 20km at McMurdo station (Hofmann et al, 1989).

Mills et al. (2005) explain the mechanism involving photolysis of $H_2SO_4$ vapour and $SO_3$ to cause a build-up of $SO_2$ during polar night and new particle formation in spring.

Long-range transport of particles nucleated in the Arctic stratosphere is known also to cause the observed spring-time particle concentration CN layer observed at Laramie (Campbell et al., 2014).

That the stratospheric aerosol layer can be explained principally based on the transport, growth and sedimentation of homogeneously nucleated particles (with additional particle formation in polar spring) remained the consensus presented in the SPARC 2006 Assessment of Stratospheric Aerosol Properties (Thomason and Peter, 2006).



# 3. IN-SITU OBSERVATIONS INDICATE ALSO SUBSTANTIAL METEORIC-ORIGIN PARTICLES

1. High-altitude stratospheric aerosol particle composition laser ablation mass specrometer (NOAA PALMS instrument on NASA WB-57)

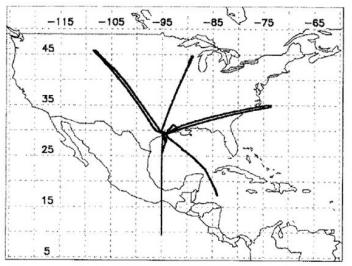
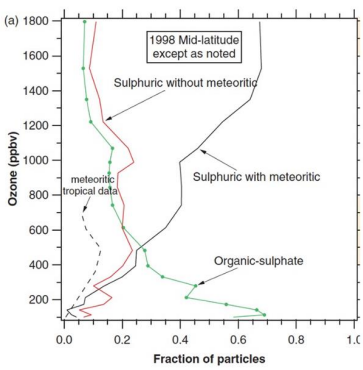
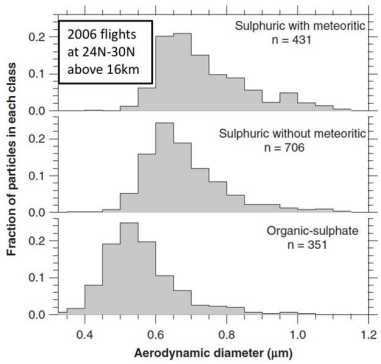



**Fig. 1.** Flight tracks of the WB-57F Aerosol Mission. Two flights were flown down the 95°W meridian.




Figures from Murphy et al. (1998, 2014)

2.High-altitude refractory particle concentration measurements from the M55 Geophysica aircraft

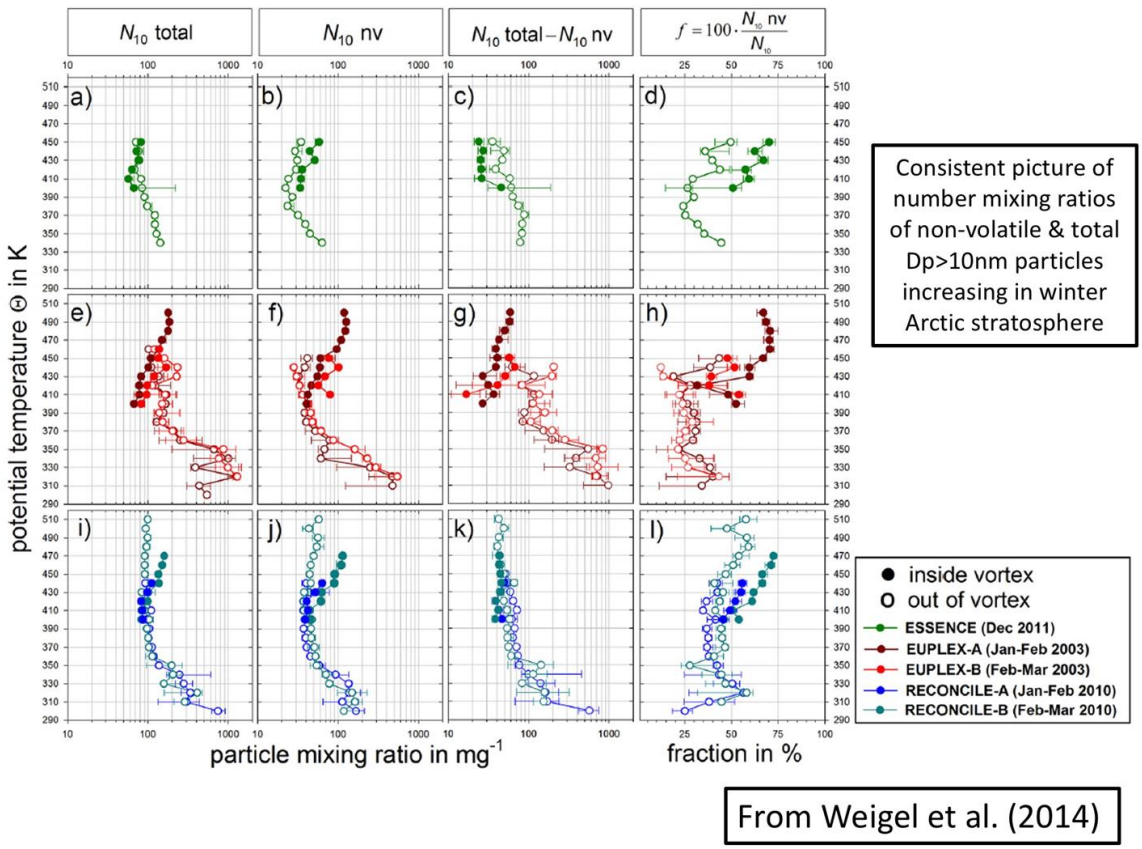



Figure from Weigel et al. (2014)

3. Balloon refractory particle concentration measurements from McMurdo and Laramie

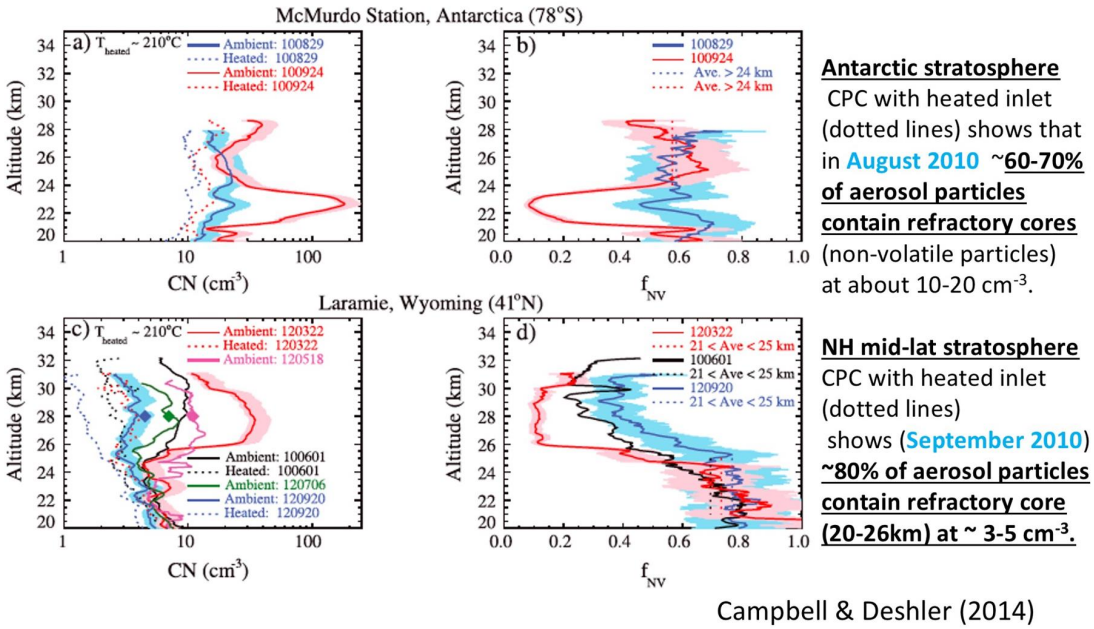



Figure from Campbell and Deshler (2014)

# UK CHEMISTRY & AEROSOL PROJECT (UKCA)

- Collaboration between UK National Centre for Atmospheric Science (Leeds, Cambridge, Oxford) & UK Met Office since 2005
- Has built aerosol-chemistry sub-model in the UK Met Office Unified Model, being applied for a range of applications (climate,air quality, Earth system science, weather)
- Chemistry schemes & aerosol configurations including for stratosphere-troposphere
- Multi-component aerosol microphysics scheme (GLOMAP)
- Global variations in aerosol particle size distribution --> sedimentation and SW & LW radiative effects
  |
- UKESM includes UKCA chemistry-aerosol sub-model so that it simulates strat-trop ozone and strat-trop aerosol interactively, each radiatively coupled for composition-dynamics interactions.

# INTERACTIVE STRATOSPHERIC AEROSOL MODELLING IN BOTH UM-UKCA & ECMWF

"Strat-enabled GLOMAP" within UM-UKCA

- GLOMAP adapted to simulate aerosol across the stratosphere & troposphere (Dhomse et al., 2014)
- Absorptive heating within volcanic aerosol clouds added (feedback on dynamics) in Mann et al. (2015)
- Through 2016-7, strat-enabled GLOMAP developed to simulate meteoric-sulphuric particles alongside homogeneously nucleated particles as in classification in Murphy et al. (2014).
- Adaptations described in Brooke et al. (2017, JGR) and applied for VolMIP Tambora-ISA integrations (Zanchettin et al., 2016; Marshall et al., 2018, Clyne et al., 2020).
- Applied for pre-industrial, 1960s, 1980s, 1990s and 2000s strat-aerosol layer and new major volcanic aerosol datasets produced (Agung, El Chichon & Pinatubo), Dhomse et al., (2020, ACP).

"Strat-enabled GLOMAP" in ECMWF model

- GLOMAP integrated into ECMWF Integrated Forecast System (IFS) during MACC project (2009-2015).
- Applied for global tropospheric aerosol forecasts (Woodhouse et al., 2011; Mann et al., 2015) as "IFS-GLOMAP" with several improvements in source and sink processes during phase 1 of Copernicus Atmospheric Monitoring Service (CAMS).
- Has achieved parity of skill in forecasting AOD reached by operational aerosol scheme IFS-AER (e.g. Remy et al., 2019).
- Volcanic configuration of IFS-GLOMAP validated during 2018 for case studies of Calbuco and Holhuraun eruptions (Remy et al., 2018).
- Latest interactive stratospheric aerosol capability from UM-UKCA (including meteoric-sulphuric interactions) added.
- New tropospheric-stratospheric aerosol and chemistry forecasting system IFS-CB05-BASCOE-GLOMAP (ICBG) that combines IFS-GLOMAP with strat-trop chemistry system IFS-CB05-BASCOE (see Huijnen et al., 2016, GMD).
- Case studies of 2019 Raikoke and 1991 Pinatubo aerosol clouds with ICBG confirm the capability of the new volcanic aerosol forecasting system.

# SUMMARY & FUTURE WORK

- Simulations show that, consistent with observations, in volcanically quiescent conditions, large proportion of particles in upper mid-lat & high-latitude strat-aerosol layer are of meteoric origin.

- UM-UKCA interactive stratospheric aerosol model adapted to simulate also MSP-sulphuric particles within GLOMAP aerosol microphysics scheme.

- MSP-sulphuric particles are now a core component of "strat-enabled GLOMAP", e.g. as applied for UM-UKCA simulations for ISA-MIP HErSEA simulations (Timmreck et al., 2018; Dhomse et al. 2020) & VolMIP Tambora-ISA experiment (Zanchettin et al., 2016, Marshall et al., 2018; Clyne et al., 2020)

- Model experiments show the heterogeneously nucleated MSP-sulphuric particles are strongly influential year-round in mix of particles in the stratospheric aerosol layer

- The presence of meteoric-sulphuric particles changes the growth and residence times of pure sulphuric particles & weakens new particle formation in polar winter/spring

- UK Natural Environment Research Council research project investigating meteoric influence on stratospheric aerosol and PSCs with range of observational project partners

- Dr. Kamalika Sengupta (Leeds global modelling PDRA) writing paper on the progression of the particle mix through Arctic winter stratospheric aerosol layer, comparing to refractory particle concentrations from M55 Geophysica flights in the Arctic, and to balloon particle concentration measurements in NH mid-latitudes.

# ABSTRACT

The widespread presence of meteoric smoke particles (MSPs) within a distinct class of stratospheric aerosol particles has become clear from in-situ measurements in the Arctic, Antarctic and at mid-latitudes.

We apply an adapted version of the interactive stratosphere aerosol configuration of the composition-climate model UM-UKCA, to predict the global distribution of meteoric-sulphuric particles nucleated heterogeneously on MSP cores. We compare the UM-UKCA results to new MSP-sulphuric simulations with the European stratosphere-troposphere chemistry-aerosol modelling system IFS-CB05-BASCOE-GLOMAP.

The simulations show a strong seasonal cycle in meteoric-sulphuric particle abundance results from the winter-time source of MSPs transported down into the stratosphere in the polar vortex. Coagulation during downward transport sees high latitude MSP concentrations reduce from ~500 per cm3 at 40km to ~20 per cm3 at 25km, the uppermost extent of the stratospheric aerosol particle layer (the Junge layer).

Once within the Junge layer's supersaturated environment, meteoric-sulphuric particles form readily on the MSP cores, growing to 50-70nm dry-diameter (Dp) at 20-25km. Further inter-particle coagulation between these non-volatile particles reduces their number to 1-5 per cc at 15-20km, particle sizes there larger, at Dp ~100nm.

The model predicts meteoric-sulphurics in high-latitude winter comprise >90% of Dp>10nm particles above 25km, reducing to ~40% at 20km, and ~10% at 15km.

These non-volatile particle fractions are slightly less than measured from high-altitude aircraft in the lowermost Arctic stratosphere (Curtius et al., 2005; Weigel et al., 2014), and consistent with mid-latitude aircraft measurements of lower stratospheric aerosol composition (Murphy et al., 1998), total particle concentrations also matching in-situ balloon measurements from Wyoming (Campbell and Deshler, 2014).

The MSP-sulphuric interactions also improve agreement with SAGE-II observed stratospheric aerosol extinction in the quiescent 1998-2002 period.

Simulations with a factor-8-elevated MSP input form more Dp>10nm meteoric-sulphurics, but the increased number sees fewer growing to Dp ~100nm, the increased MSPs reducing the stratospheric aerosol layer’s light extinction.

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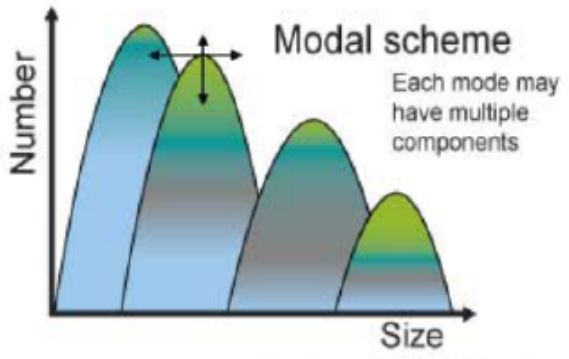
# Interactive stratospheric aerosol configuration of UK's composition-climate model UM-UKCA

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**

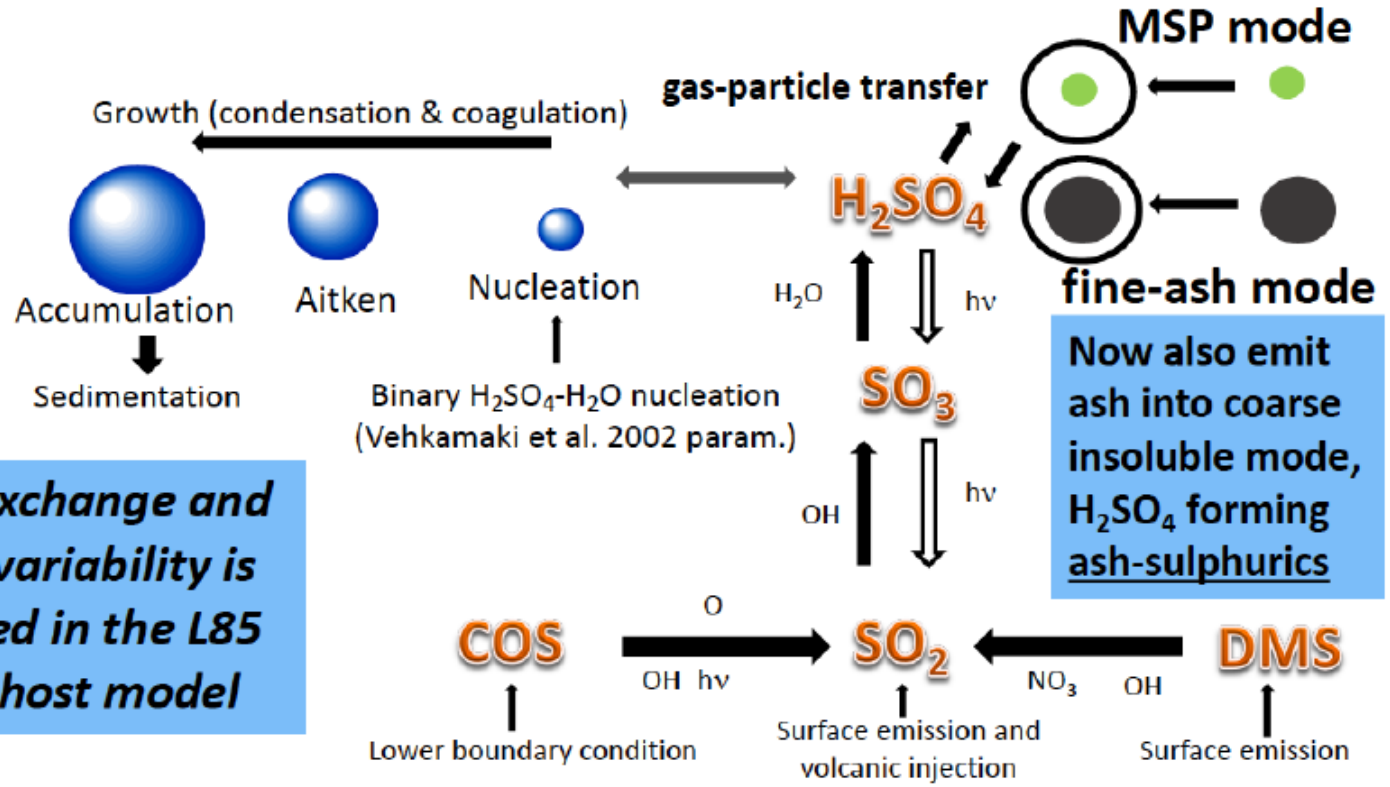
## UKCA sulphur chemistry & aerosol microphysics

a)




*Schematic of modal representation GLOMAP-mode aerosol microphysics scheme (Mann et al., 2010)*

b)




***Strat-Trop exchange and dynamical variability is well-resolved in the L85 UM-UKCA host model***

*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)*

**Fig 1: Panels a & b = monthly-varying MSP climatology from WACCM-CARMA c) to f) = MSP-sulfuric particle N and mean size**

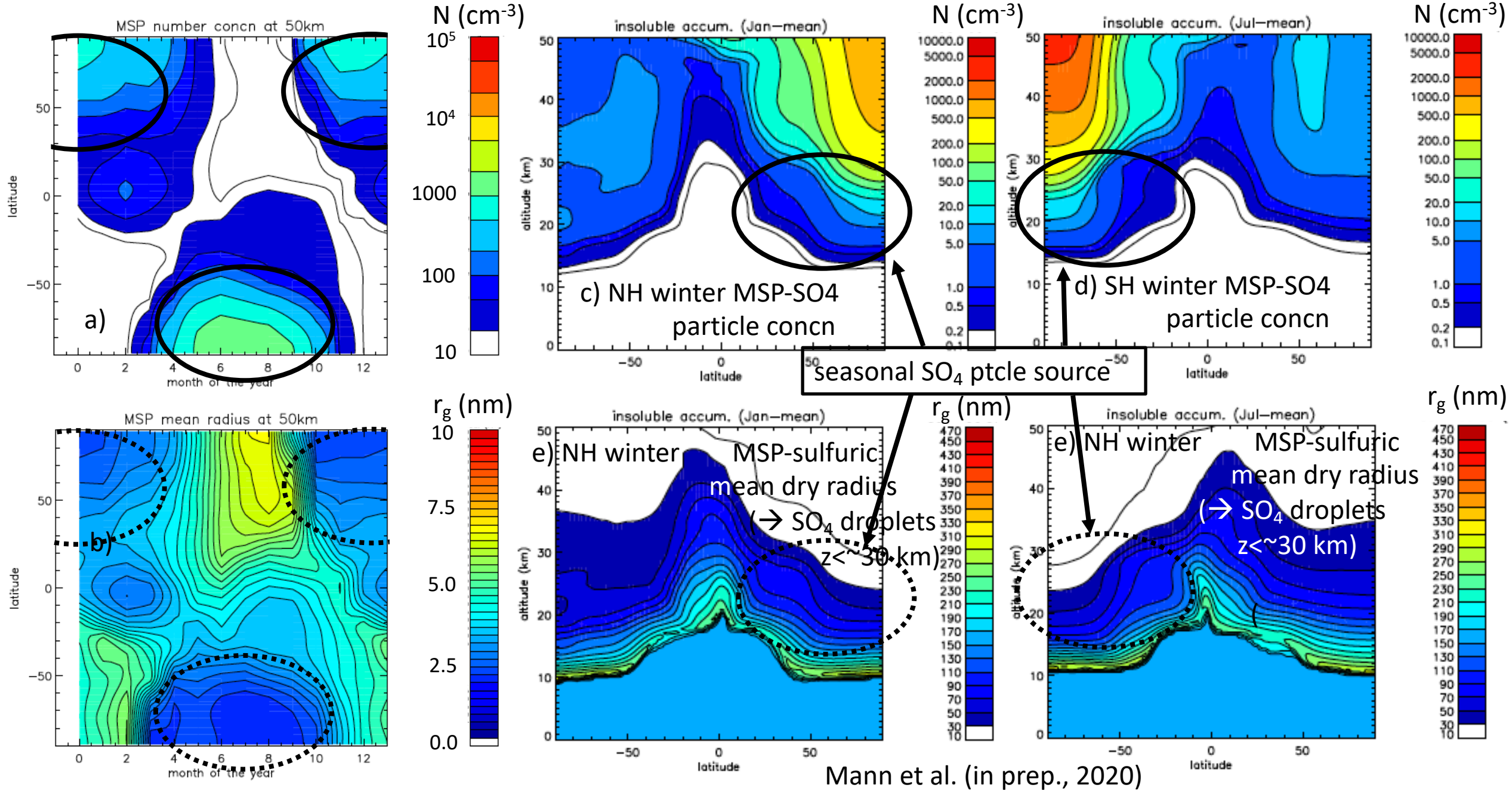
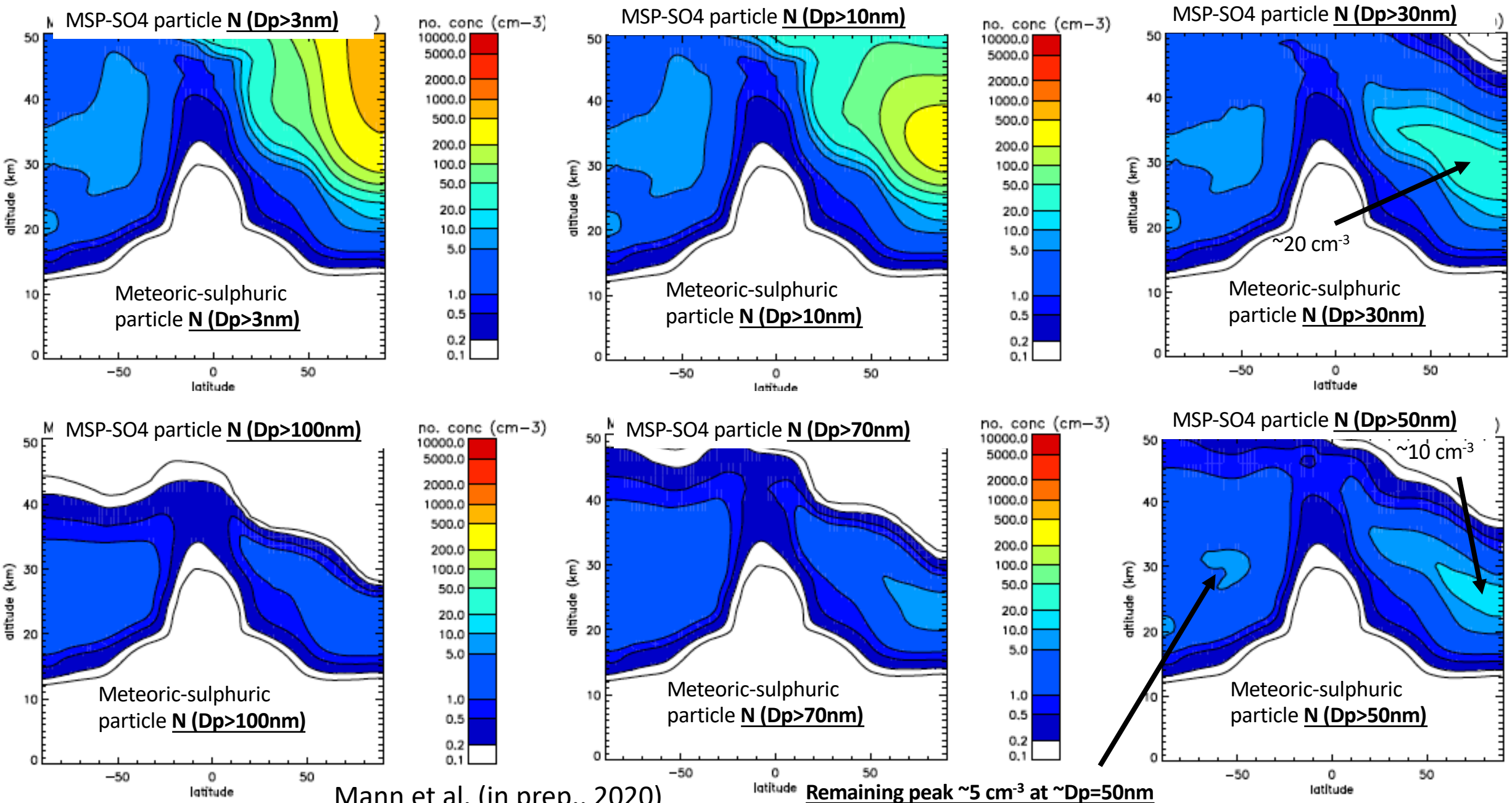


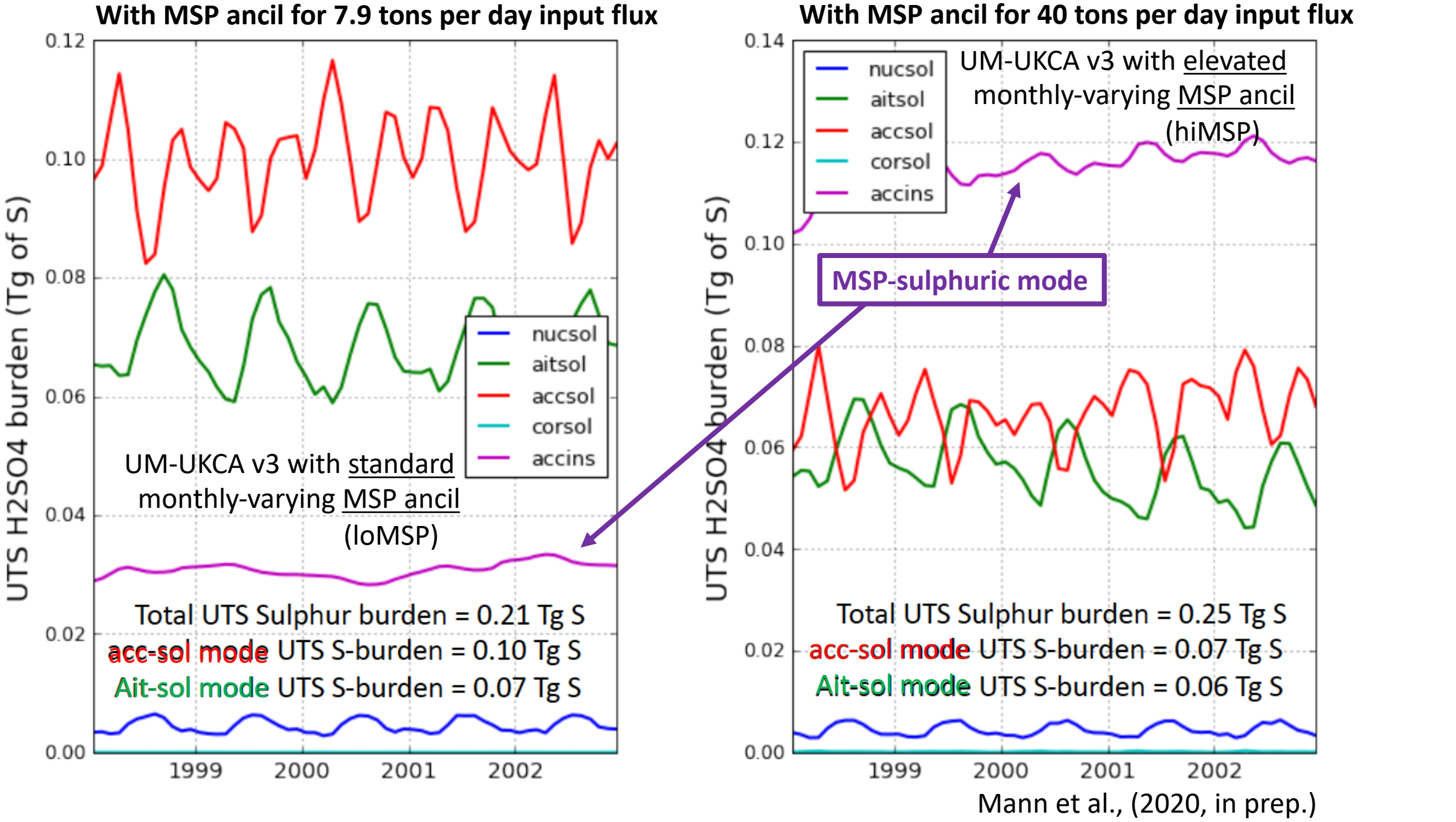
**Figure 6**: UM-UKCA simulated size-resolved **meteoric-sulphuric particle concentrations** (5-year January-mean)




Mann et al. (in prep., 2020)

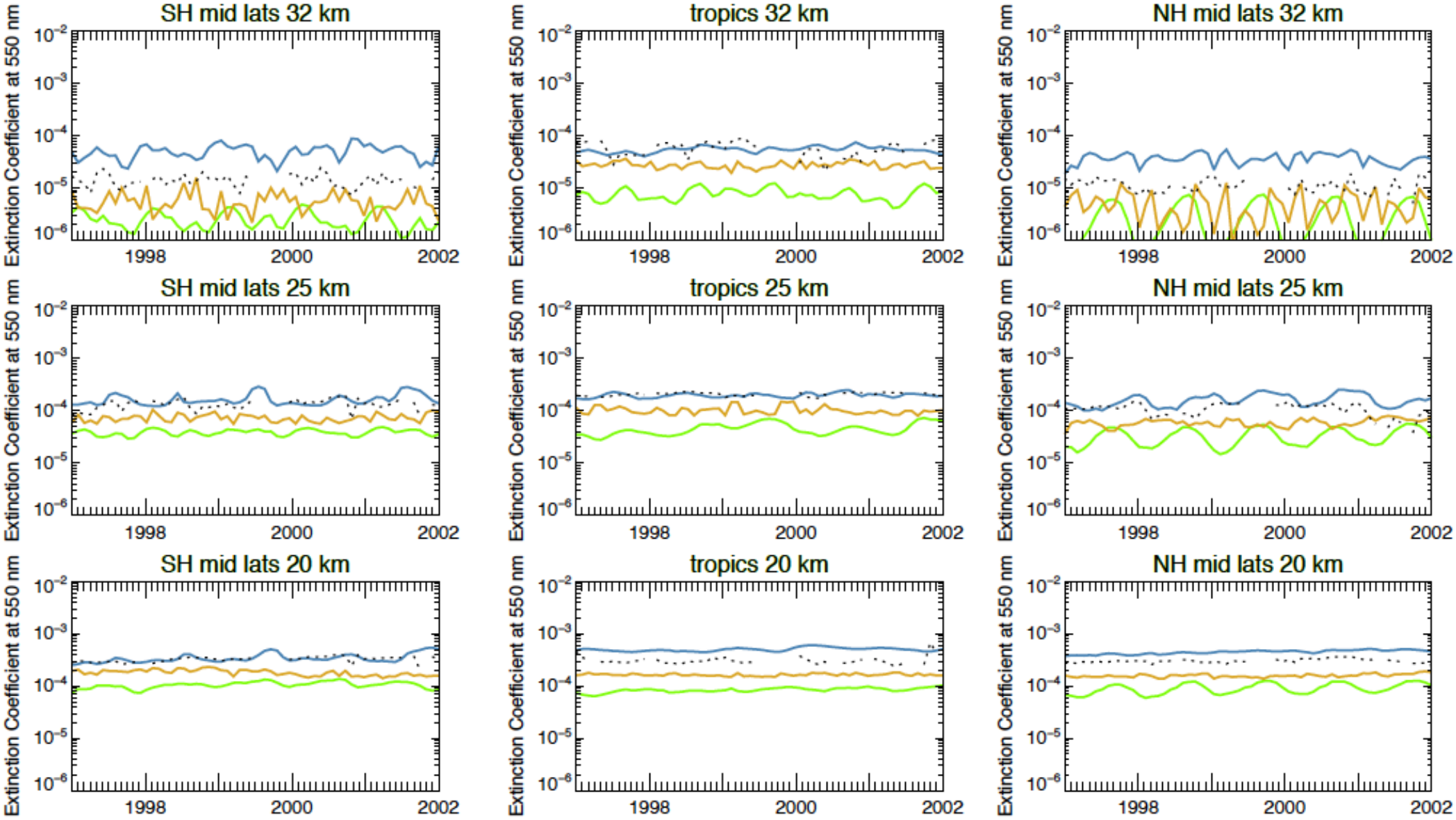
**Remaining peak ~5 cm$^{-3}$ at ~Dp=50nm**

**Fig 3.5: Stratospheric aerosol particle sulphur burden – impact of MSPs on size & residence time of aerosol particles in stratosphere**



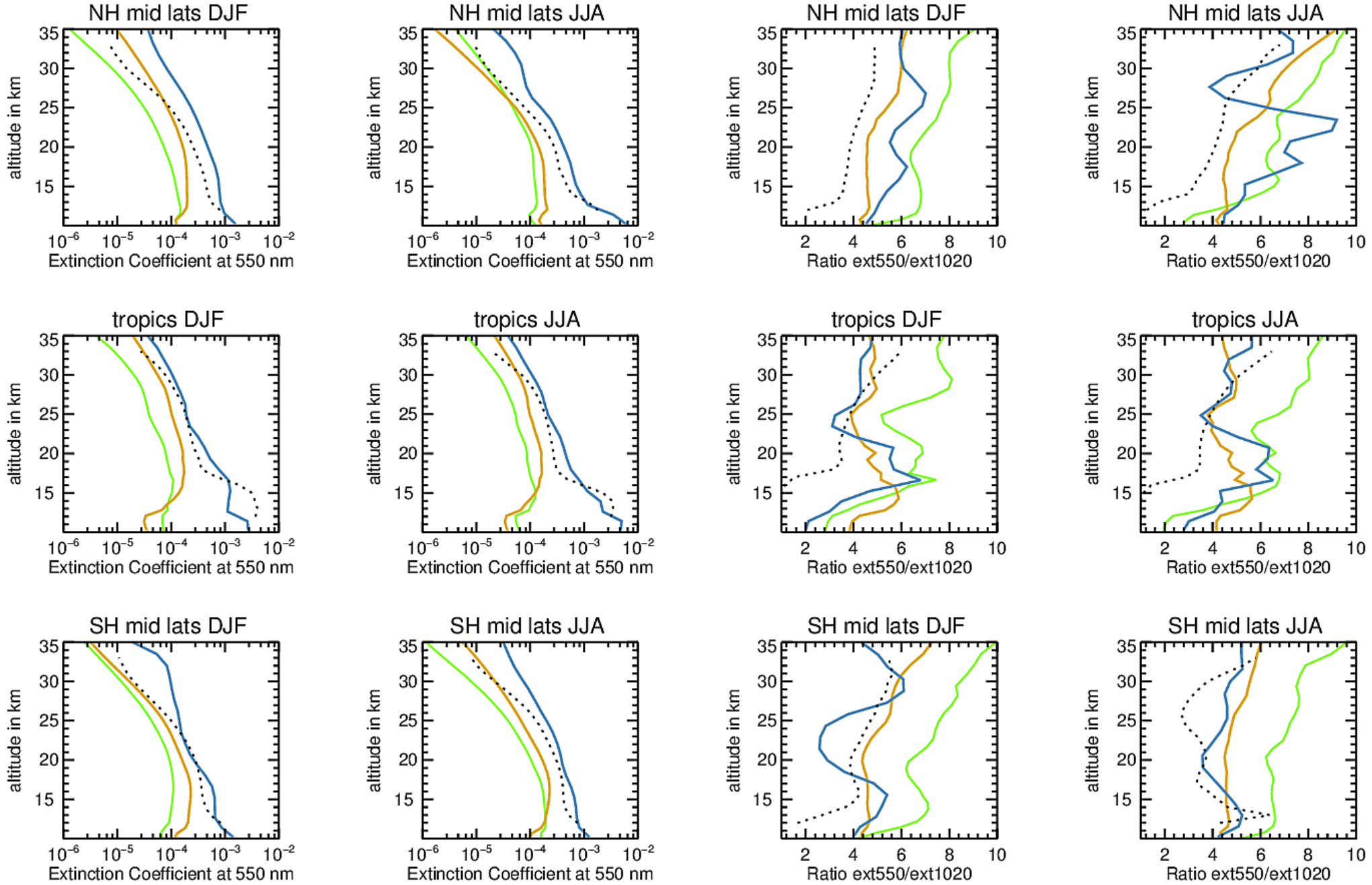

Mann et al., (2020, in prep.)

**Fig 4: 550nm extinction timeseries at 20, 25, 32km in tropics and NH, SH mid-latitude (5-yrs of monthly-means [xnelf]**



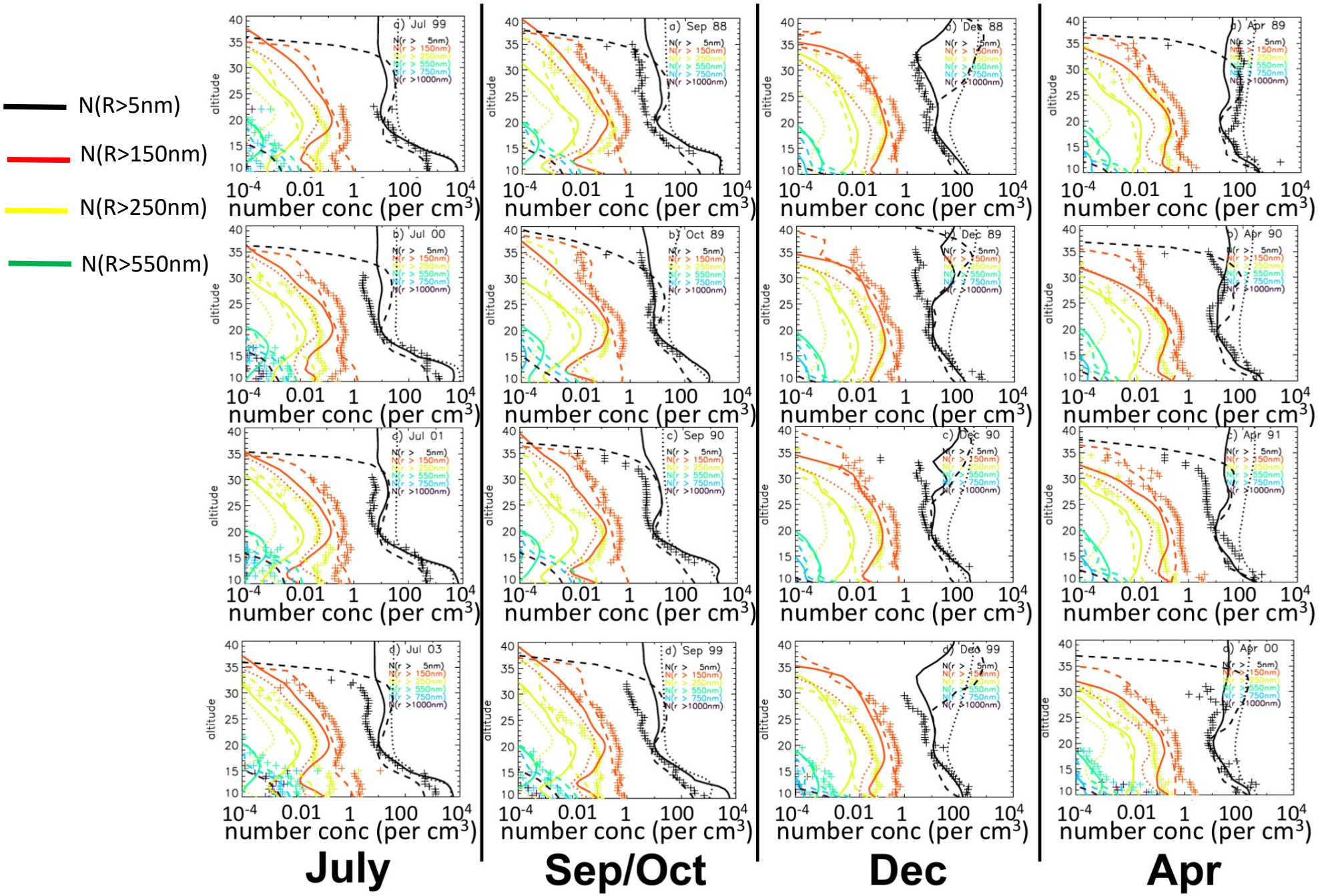

Mann et al.
(in prep., 2020)

# Fig 5: 550nm and 1020nm extinction tropics and NH mid-latitude [same as ASAP2006 profiles] (5-yr mean Jan & July [xnelf]




Mann et al.
(in prep., 2020)

**Fig 3: NH mid-lat (Laramie) profiles of N(Dp>10nm, black), N(Dp>300nm,red), N(Dp>500nm, yellow) (single-yr mean profiles [xnelf]**




+++++ CPC & OPC observations

– – – No-MSP run (Dhomse14)

——— v3 loMSP run (Dhomse20)

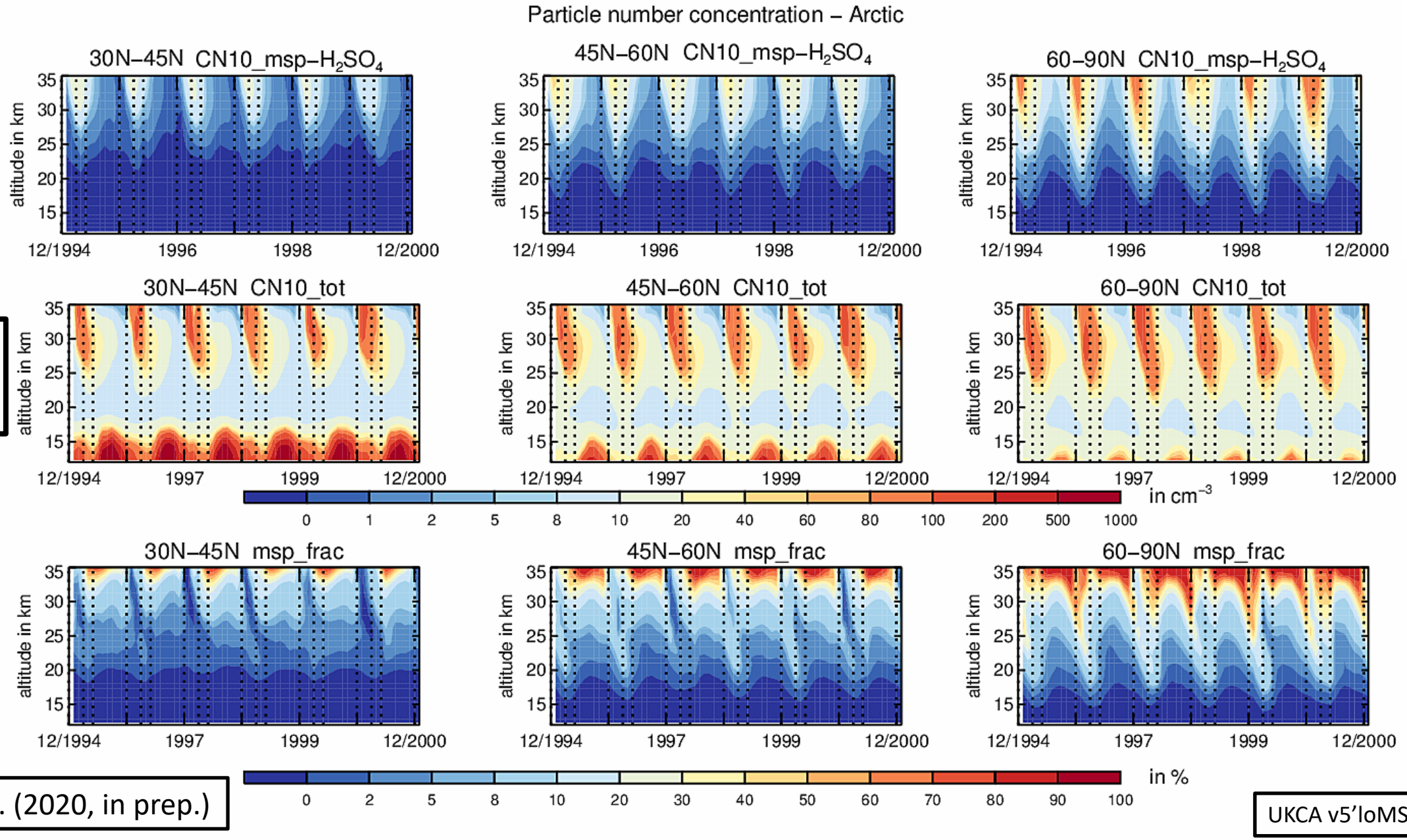
····· v3 hiMSP run (sensitivity simulation)

For this comparison, picked 4 flights during non-volcanic periods in same month

n.b. Updated Figure with re-calibrated OPC data from Deshler et al. (2019)

Mann et al. (in prep., 2020

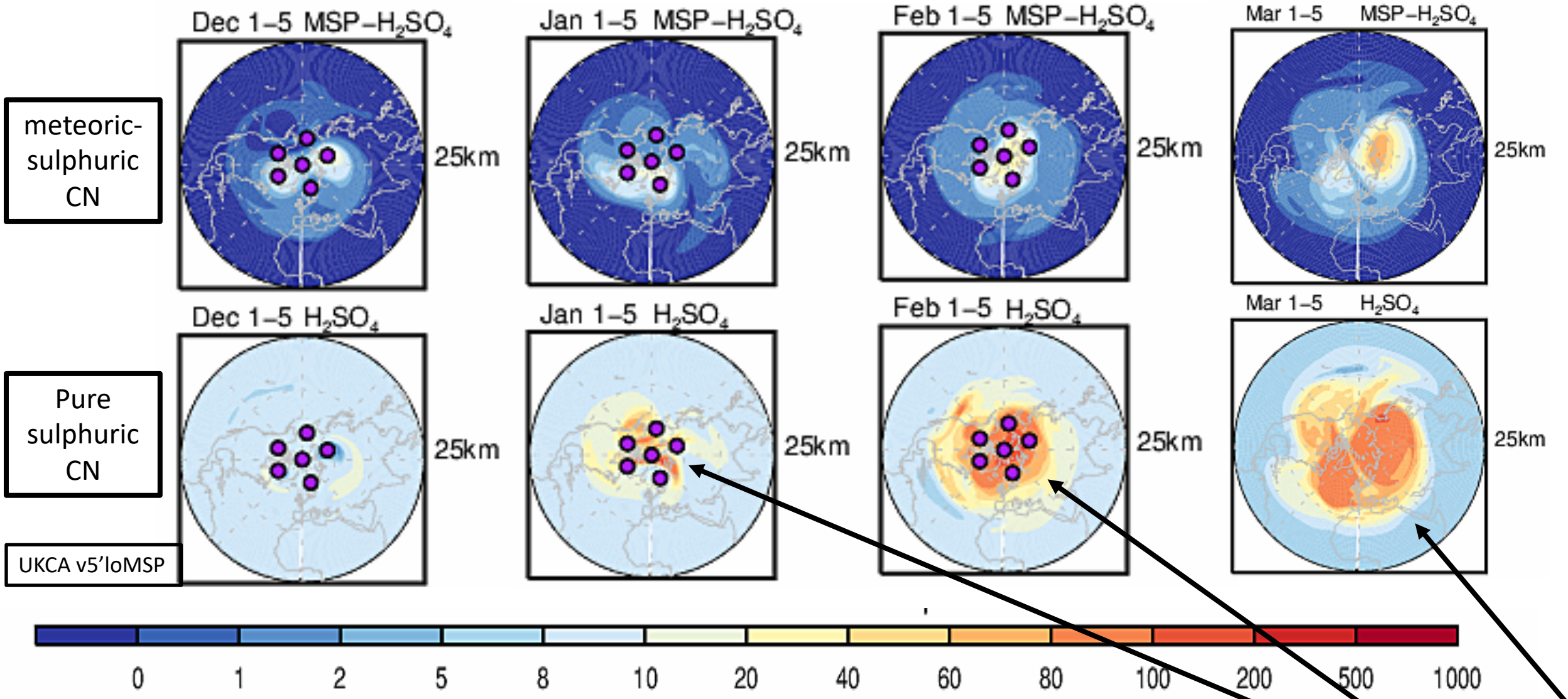
# UM-UKCA simulated Arctic winter strat-aerosol progression (volc. quiescent conditions)



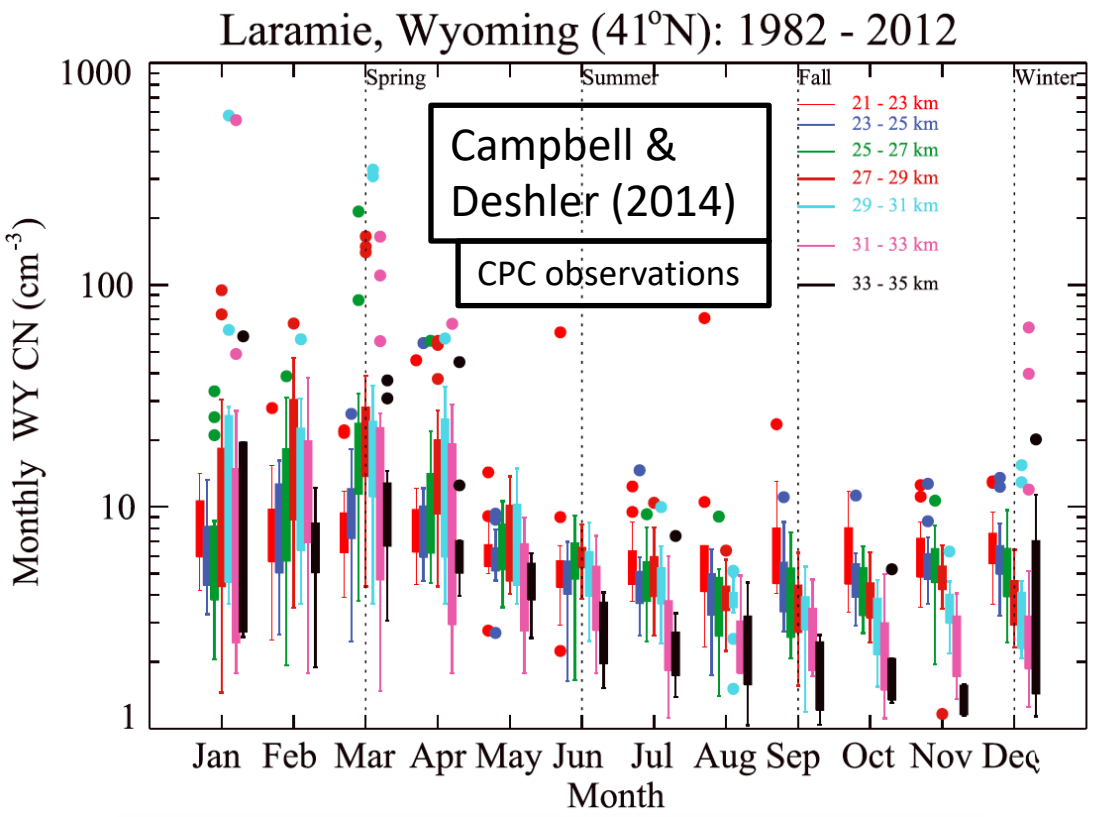

Sengupta et al. (2020, in prep.)

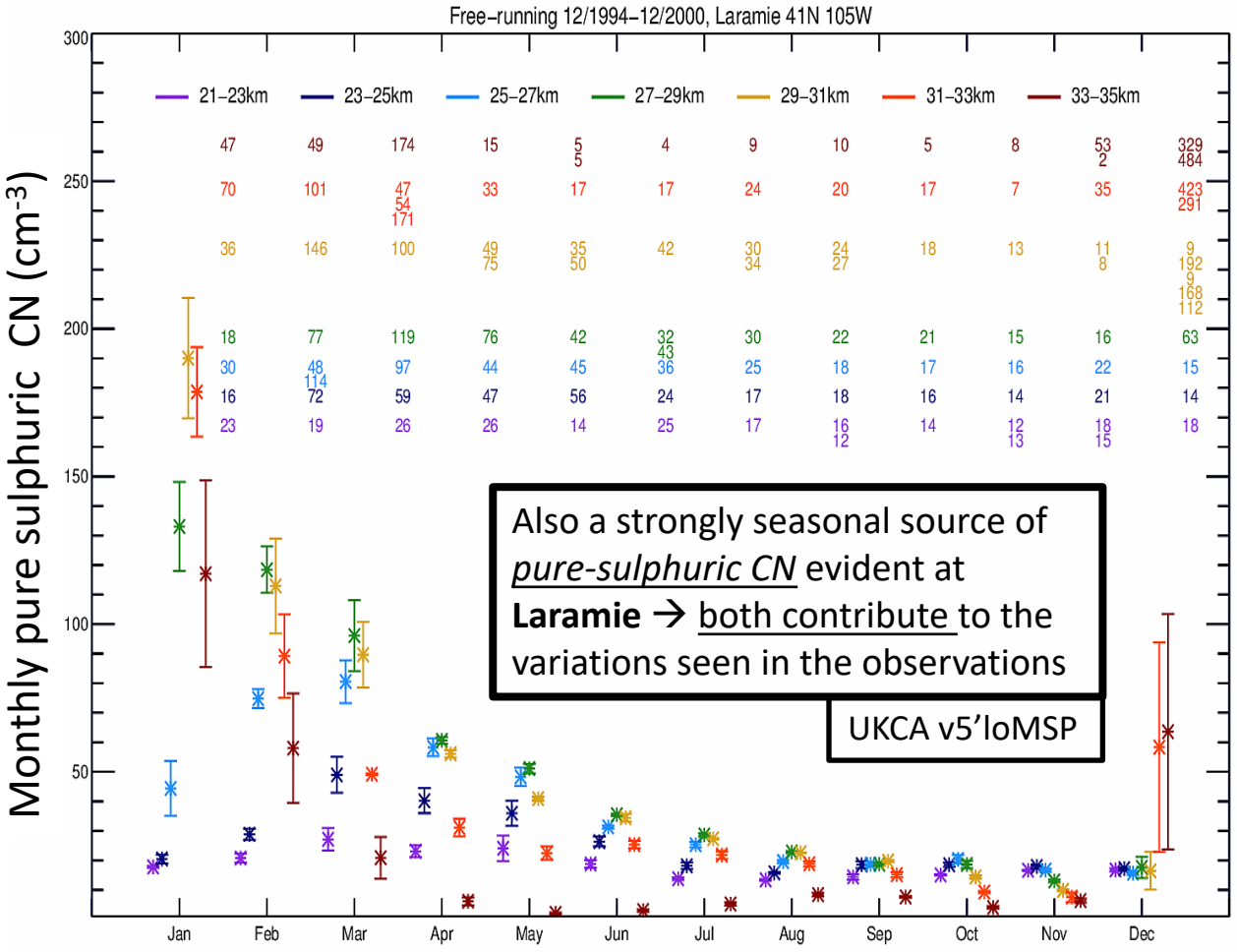
UKCA v5'loMSP

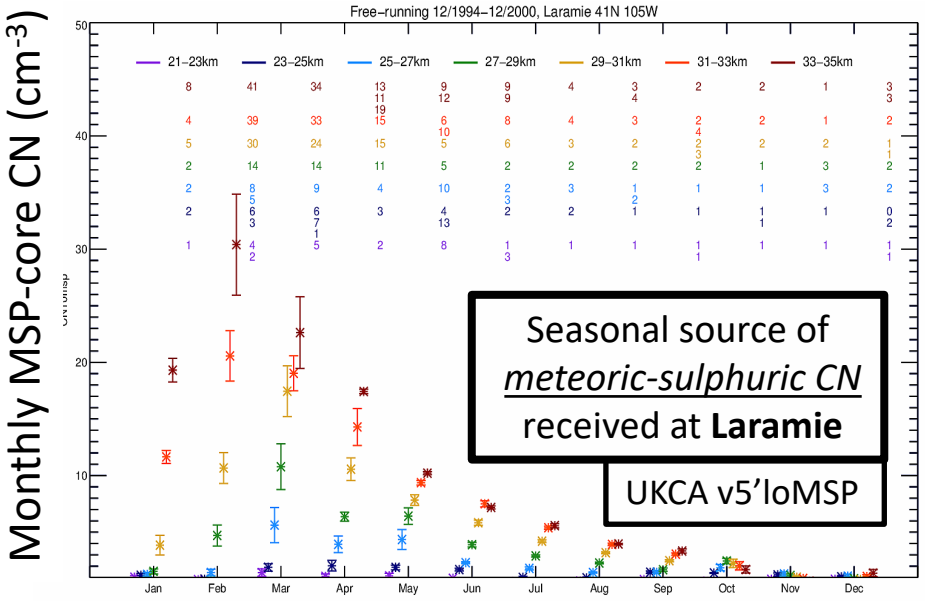
# UM-UKCA simulated Arctic winter strat-aerosol progression (volc. quiescent conditions)




Sengupta et al. (2020, in prep.)




Model represents the high variability in CN concentrations seen in Laramie CPC in Dec & Jan in the upper-most Junge layer (31-35km).

Each "seasonal dose" of meteoric-sulphuric particles co-incides with a stronger seasonal new particle formation of the pure sulphuric CN

Then transition to steady peak in pure sulphuric CN at 27-29km (green)

Sengupta et al. (2020, in prep.)