The presence of and effects from meteoric-sulphuric particles within the stratospheric aerosol layer

NATURAL ENVIRONMENT RESEARCH COUNCIL

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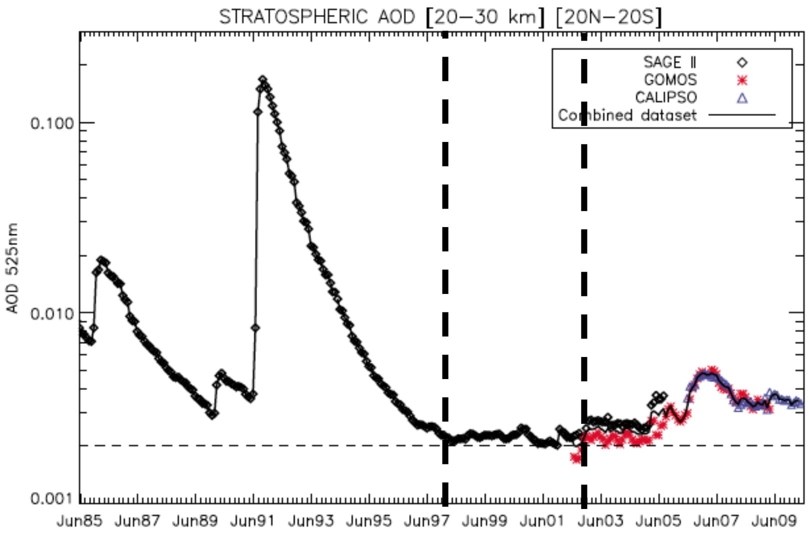
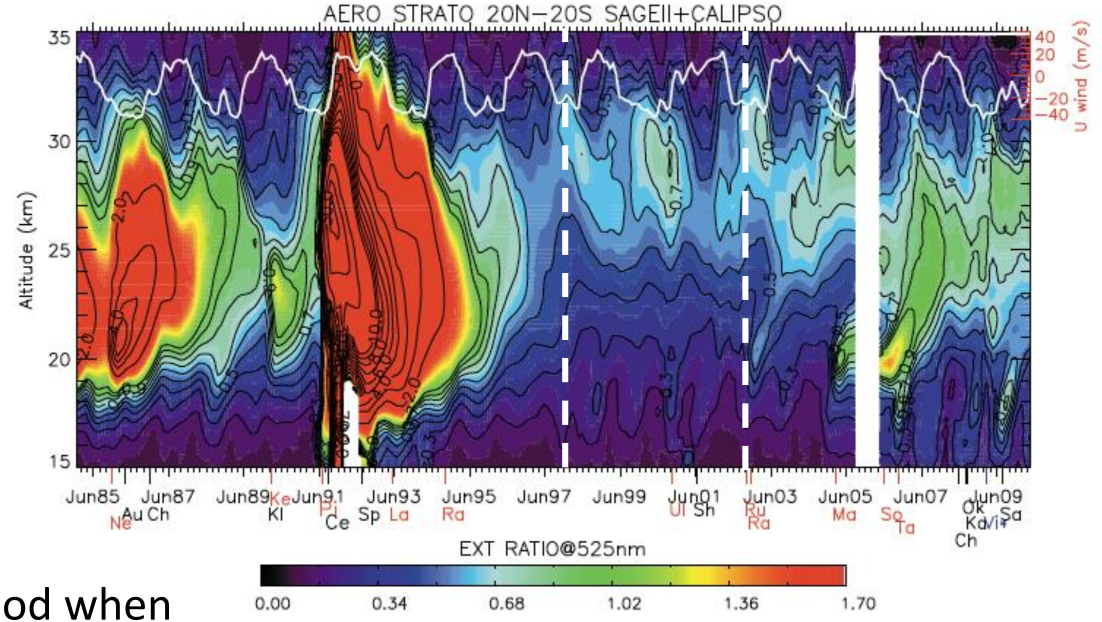
AGU Chapman conference: Stratospheric aerosol in post-Pinatubo era.

This talk re: the **quiescent stratospheric aerosol layer**, as observed 1998-2002.





1998-2002 was an unusual 5-year period when Junge layer remained in "quiescent conditions".

Vernier et al., (2011, Geophys. Res. Lett.)

PALMS stratospheric particle composition measurements show meteoric signature (iron) in 50% particles of quiescent mid-latitude Junge layer.

Murphy et al. (1998)

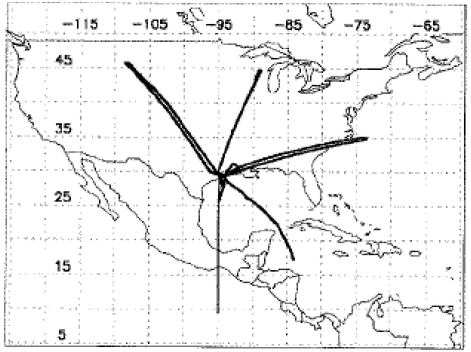
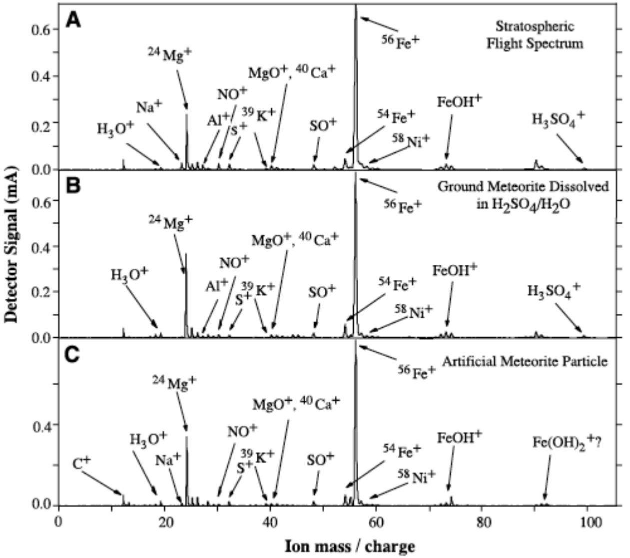


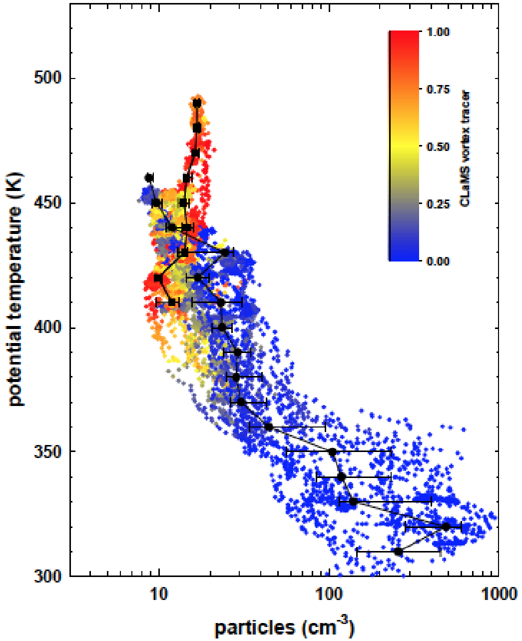
Fig. 1. Flight tracks of the WB-57F Aerosol Mission. Two flights were flown down the 95°W meridian.

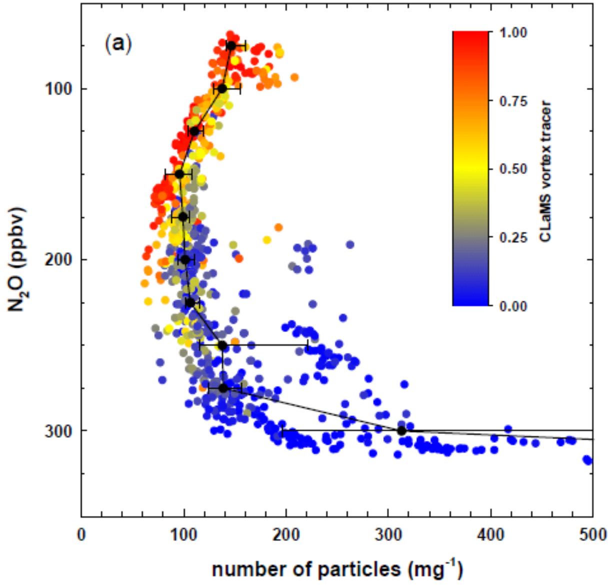
The concentration of meteoritic material in stratospheric aerosols is too low to change the refractive index significantly, but meteoritic material may be important for optical properties if it absorbs light at certain wavelengths. Also, meteoritic material could possibly affect the nucleation of stratospheric aerosols. Recent work has shown that a large fraction of air descending from the mesosphere into the stratosphere is concentrated in the winter polar regions (*29*), where large numbers of aerosols less than 0.1 μm in diameter have been observed (*30*). Our data suggest that many

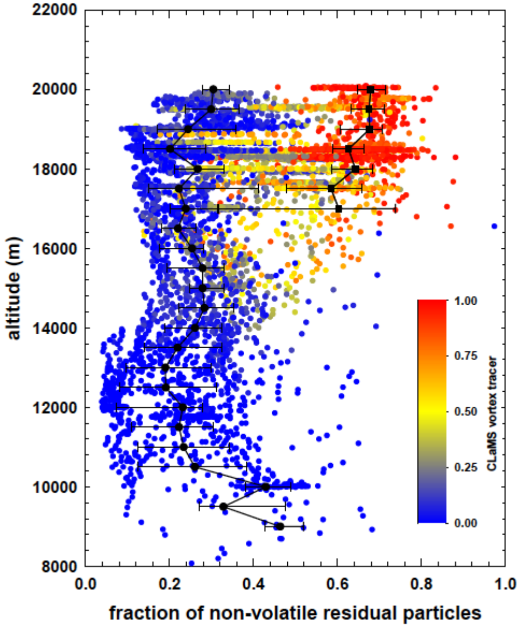
Cziczo et al. (2001)



(*16*). Particles produced from dissolved ground meteorite were sampled with the PALMS instrument, and the spectra exhibited good correlation with those obtained during flight (Fig. 1). This result demonstrated that it







Atmos. Chem. Phys., 5, 3053–3069, 2005
www.atmos-chem-phys.org/acp/5/3053/
SRef-ID: 1680-7324/acp/2005-5-3053
European Geosciences Union

Atmospheric Chemistry and Physics

Fig. 5. Fraction f of non-volatile residual particles (ratio of non-volatile residual particles n_{nv} to total particle concentration n_t) as altitude profile with vortex tracer indicated by color coding **(a)**. Data from flights as indicated in Fig. 1, except for entire flight of 12 March, and some parts of flights on 8, 9, and 11 February and 8 March when CN-Counter with heated aerosol channel was not operational.

Observations of meteoric material and implications for aerosol nucleation in the winter Arctic lower stratosphere derived from in situ particle measurements

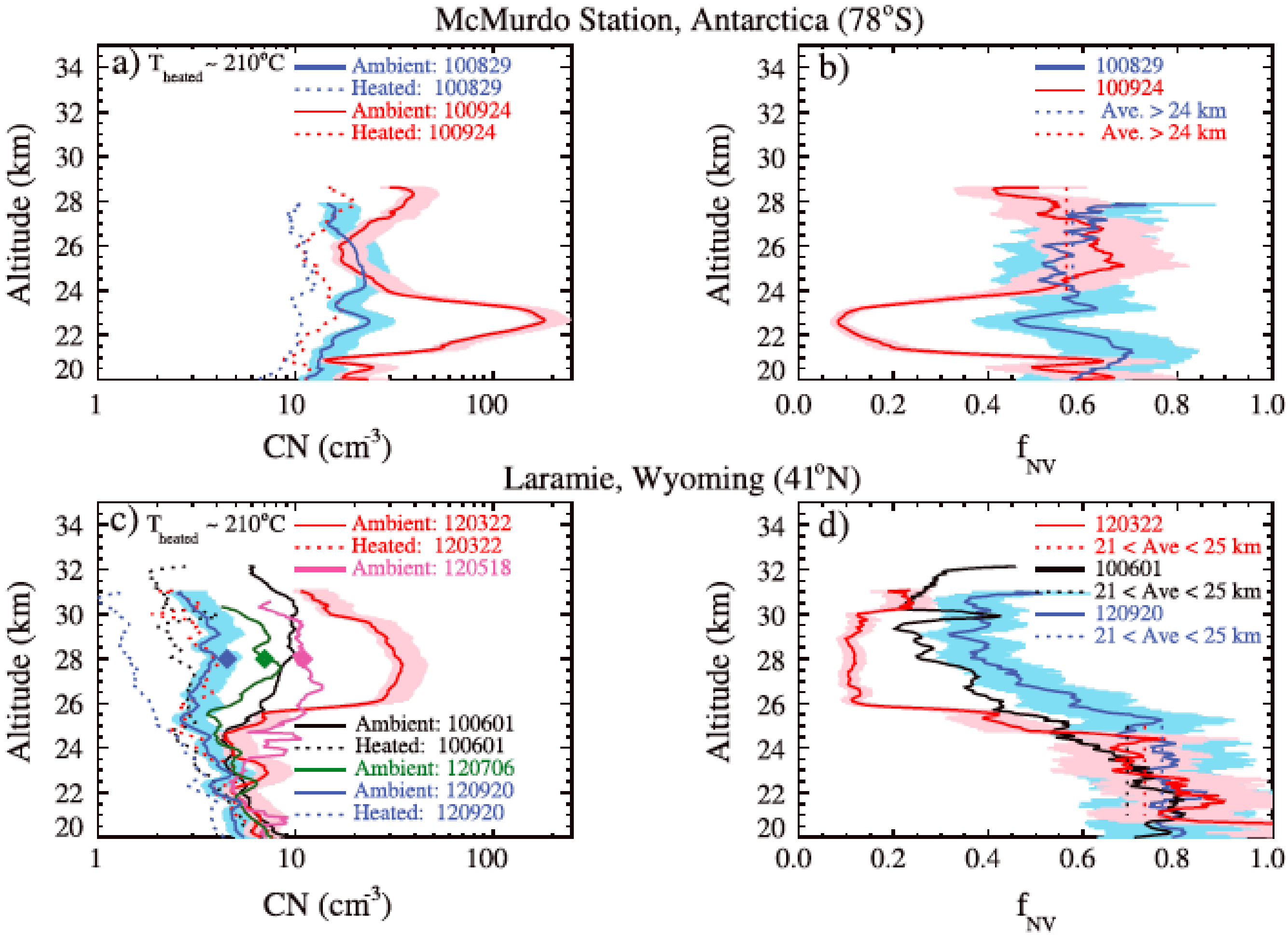
J. Curtius[1], R. Weigel[2], H.-J. Vössing[1], H. Wernli[1], A. Werner[3], C.-M. Volk[3], P. Konopka[4], M. Krebsbach[4], C. Schiller[4], A. Roiger[5], H. Schlager[5], V. Dreiling[6], and S. Borrmann[1,2]

Curtius et al. (ACP, 2005)

COPAS measurements of **Arctic stratospheric aerosol** during EUPLEX campaign (Jan-Mar 2003).

Two COPAS instruments on Geophysica high-altitude aircraft (with different size cut-offs).

Each is a two-channel CPC instrument: with two inlets, **one heated to evaporate volatile material**



Campbell & Deshler (2014)

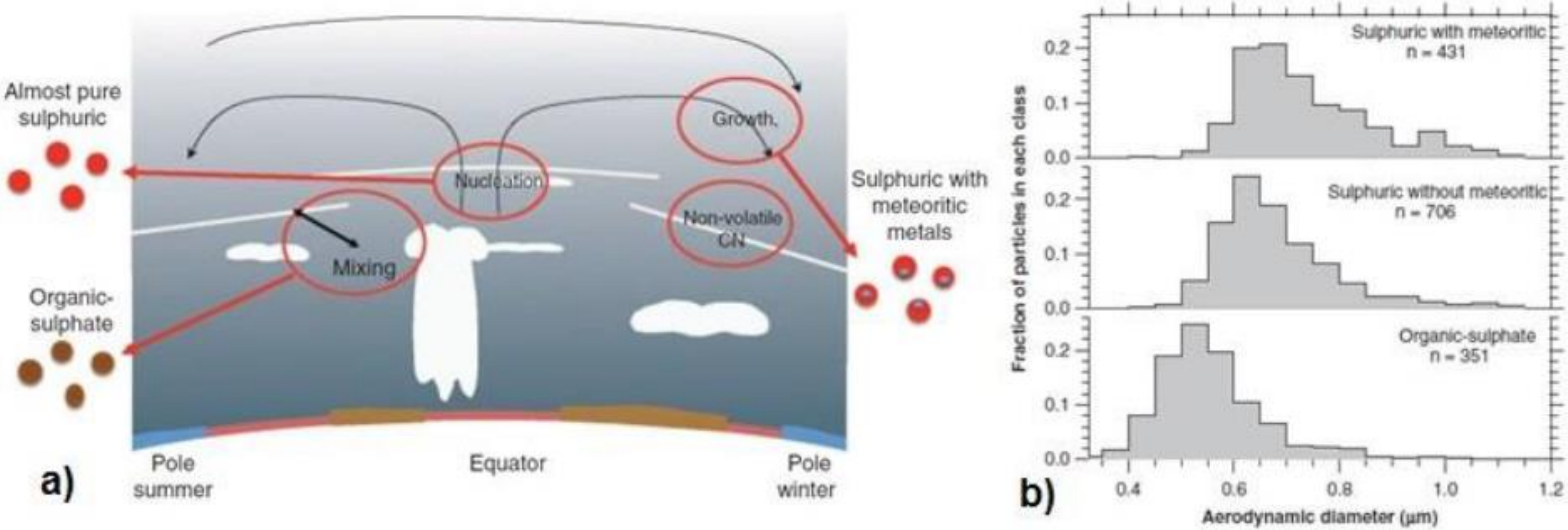


***Figure 1** a) New understanding of stratospheric sulphuric particle types (Murphy et al., 2014) b) their observed distinct size distribution, and therefore residence time in the Junge layer.*

Particles collected at an altitude of 20 km over Australia by exposing electron-microscope grids covered with a nitrocellulose film in the airstream from a *U*-2 aircraft have been examined by the present author. These consist almost exclusively of hygroscopic particles of the size and concentration reported by Junge *et al.* A typical collection is illustrated in Fig. 1. It has been confirmed that ammonium sulphate is a major constituent of these particles. Within the aggregations of water-soluble material are small, dense, insoluble particles many of which are undoubtedly of extra-terrestrial origin. These are of such a size that most of them would escape collection if they were not encased in the much greater mass of sulphate.

In order to reveal these insoluble particles for examination it is first necessary to remove most of the soluble material by floating the specimen grids on purified water, collecting face upward. Dialysis for a suitable length of time leaves only the insolubles surrounded by a residual ring which establishes the position of the original material, as shown in Fig. 2. Occasional rings are devoid of insoluble particles; but as particles of the expected appearance are found on the film nearby, it is believed that each aggregate of soluble material contains at least one insoluble particle. This has been confirmed by exposing several collections of stratospheric particles to an intense electron beam: the sulphate evaporates, leaving behind at least one stable particle for each 'parent' particle.

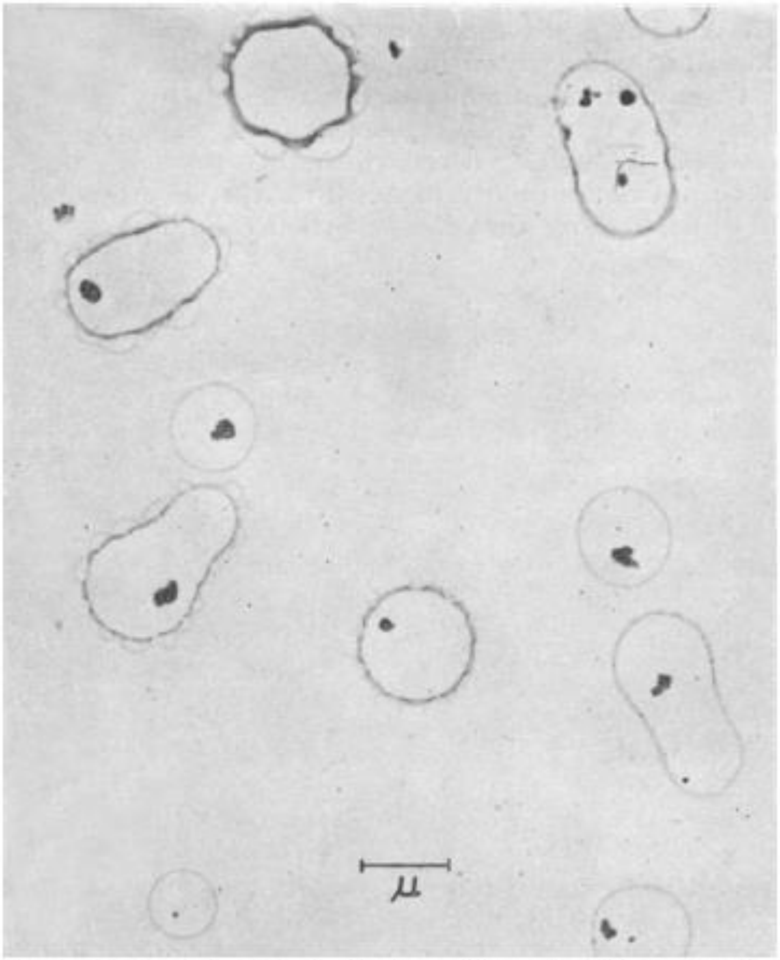


Fig. 2. Residue after dialysis of water-soluble material from collection of January 31, 1963

Mossop (1963, Nature) --- (CSIRO, Sydney, Australia)

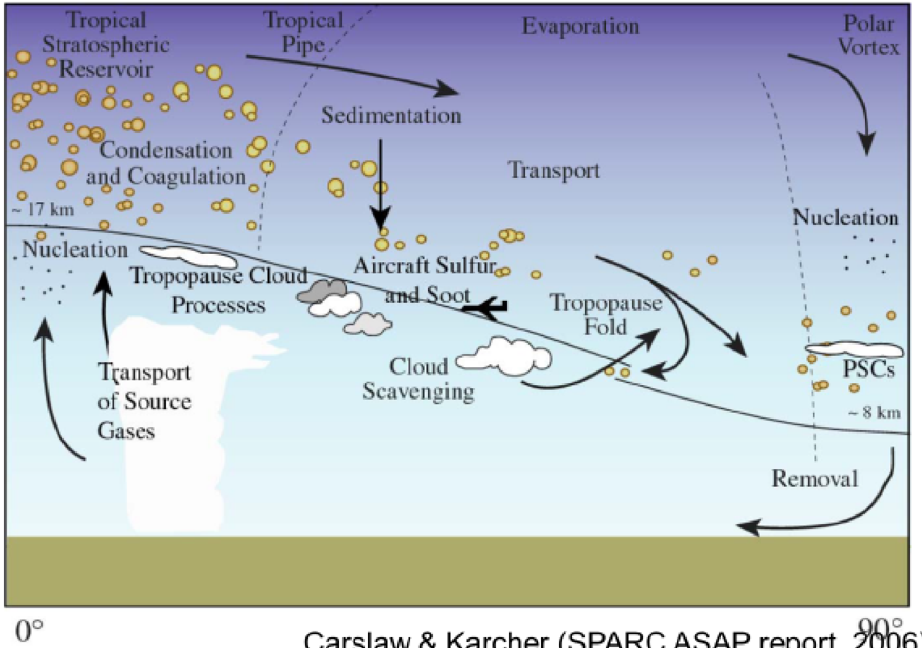
U-2 particle collectors at 20km in latitudes 20-45^{o} South.

Jan to Mar 1963 – quiescent conditions (prior to Agung plume)
Apr to May 1963 – ash-sulphuric particles in Agung plume.

MSPs & stratospheric aerosol

- Meteoric smoke particles form in mesosphere & transported into the polar stratosphere but interactive stratospheric aerosol models include only limited representation of their effects (if any)

Q: How does the presence of the MSPs alongside the homogeously nucleated particles affect the Junge layer?



Carslaw & Karcher (SPARC ASAP report, 2006)

3. Interactive stratospheric aerosol module with MSP

Include MSPs within existing accumulation insoluble mode (but size restrictions removed to allow to cover terrestrial and extra-terrestrial dust.

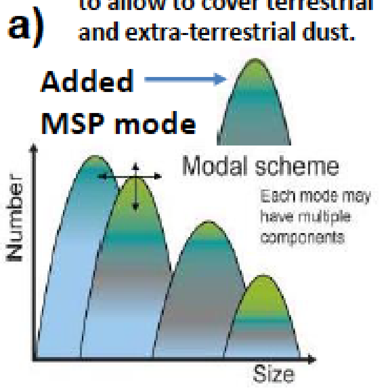


Fig 2a. Schematic of modal representation GLOMAP-mode aerosol microphysics scheme (Mann et al., 2010)

(*) We prescribe number- and mass- mixing ratios of MSP p<0.2hPa based on WACCM-CARMA sectional runs (Bardeen et al. , 2008).

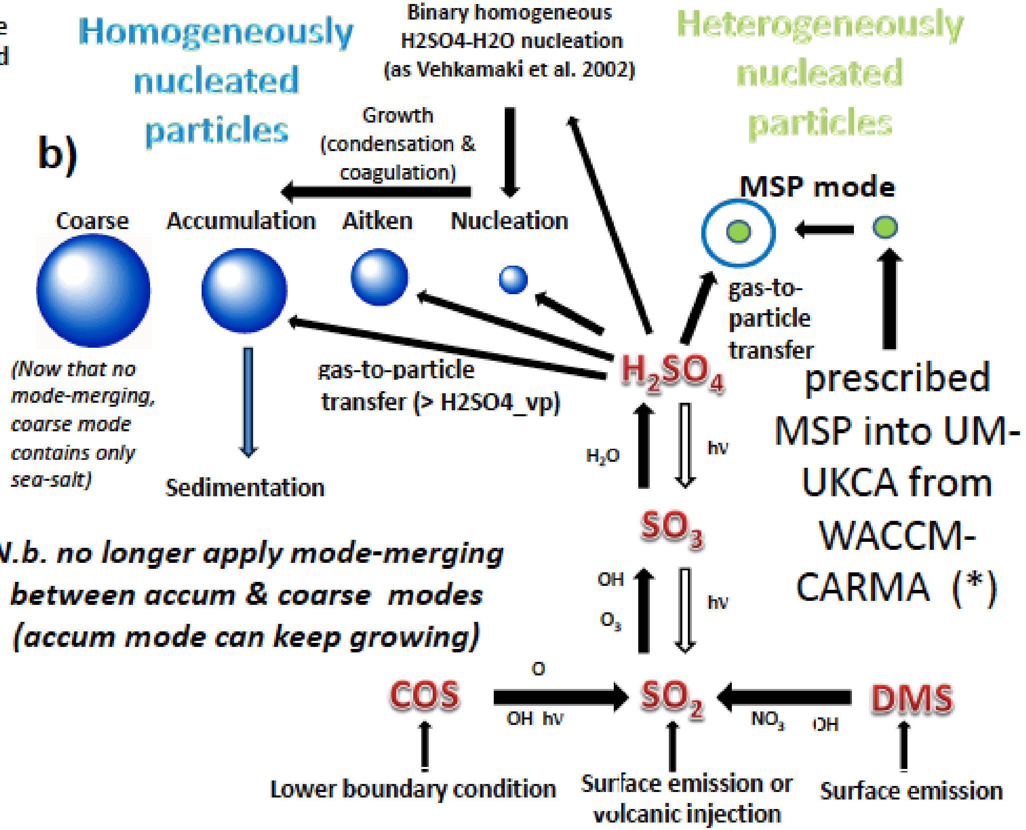


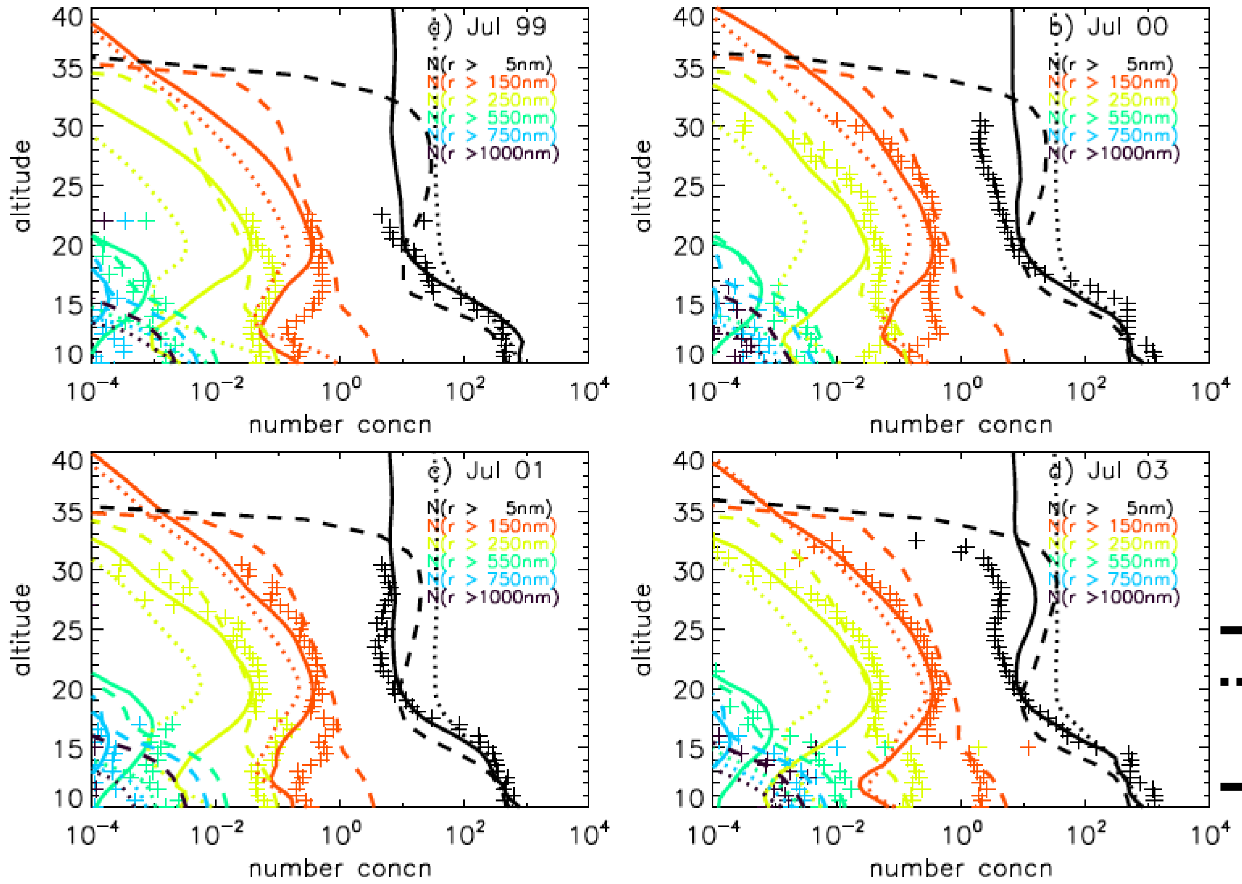
Fig 2b. Schematic of UM-UKCA stratospheric sulphur chemistry and coupling to GLOMAP-mode aerosol microphysics (see Dhomse et al., 2014) with whole-atmosphere "Chemistry of Stratosphere & Trosphere" (CheST).

UM-UKCA interactive stratospheric aerosol model experiments to How do meteoric-sulphuric & pure sulphuric particles co-exist?

- Introduced MSP into GLOMAP accumulation-insoluble mode, those particles competing for H_2SO_4 alongside the homogeneously nucleated particles in the soluble modes.
- N96 horizontal resolution with 85 vertical levels up to 80km 20+-year spin-up in present-day setting (GHG, ODS), as applied for MSP deposition to ice core (Brooke et al., 2017)
- Assess stratospheric aerosol-dynamcal quasi-equilibium reached as the MSP-interaction reduces the growth and shifts the vertical distribution of the sulphuric particles
- Map out the morphology of the Junge layer, quantifying the proportion of particles that have meteoritic inclusion.

Four seasons of quiescent particle concentrations at Laramie, Wyoming

Summertime (July) quiescent Junge layer balloon soundings (CPC and OPC)



Particle concentrations at Laramie measured by Condensation Nuclei Counter (CNC) clearly show this same transition.

See **decreasing particle concentrations** from the tropopause **up to ~20km**.

But then **above 20km** only slight decrease or **profile approximately constant**.

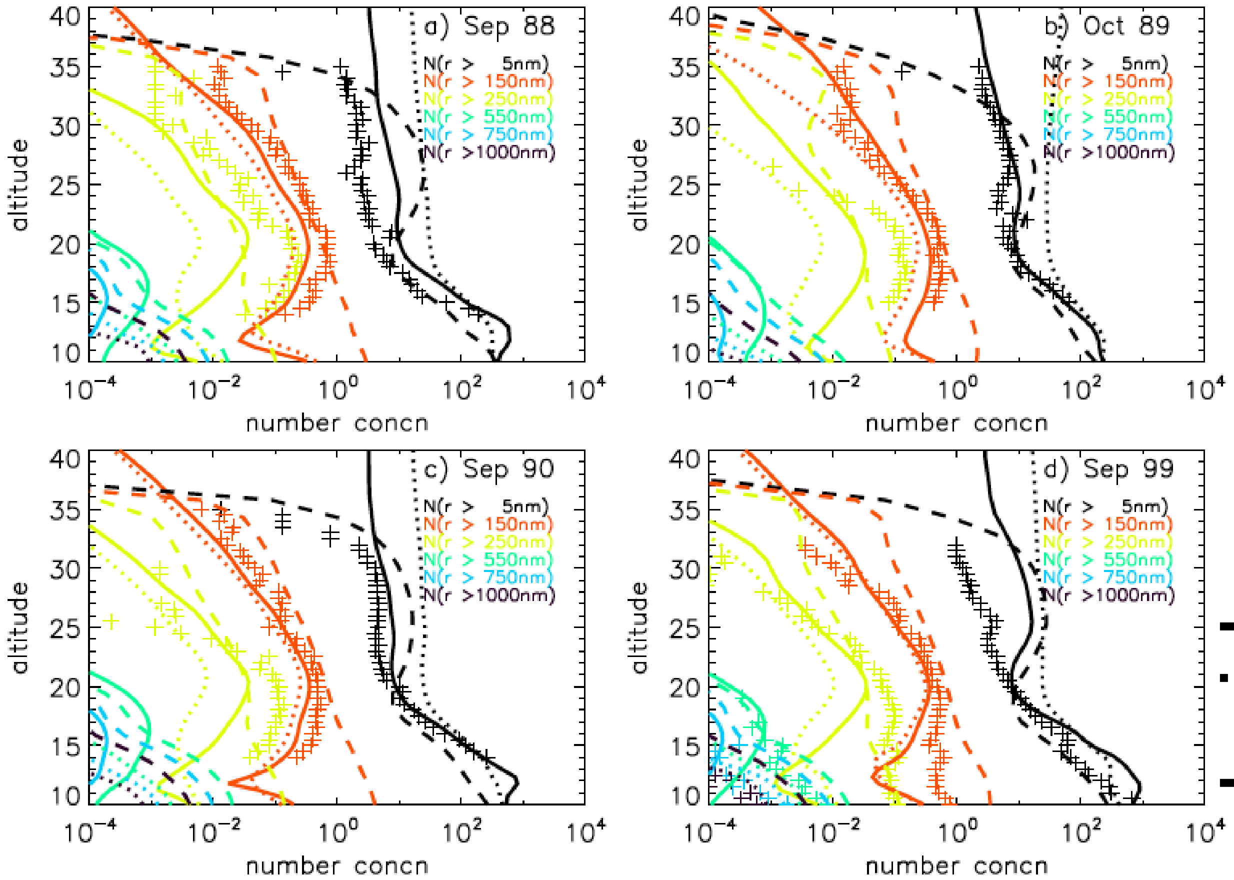
UM-UKCA (5 tons/day MSP interacting)

UM-UKCA (40 tons/day MSP interacting) (simulation from Brooke et al., 2017)

UM-UKCA (no MSP interaction) (only homogeneously nucleated particles) (simulation from Dhomse et al., 2014)

Four seasons of quiescent particle concentrations at Laramie, Wyoming

Autumn (September) quiescent Junge layer balloon soundings (CPC and OPC)



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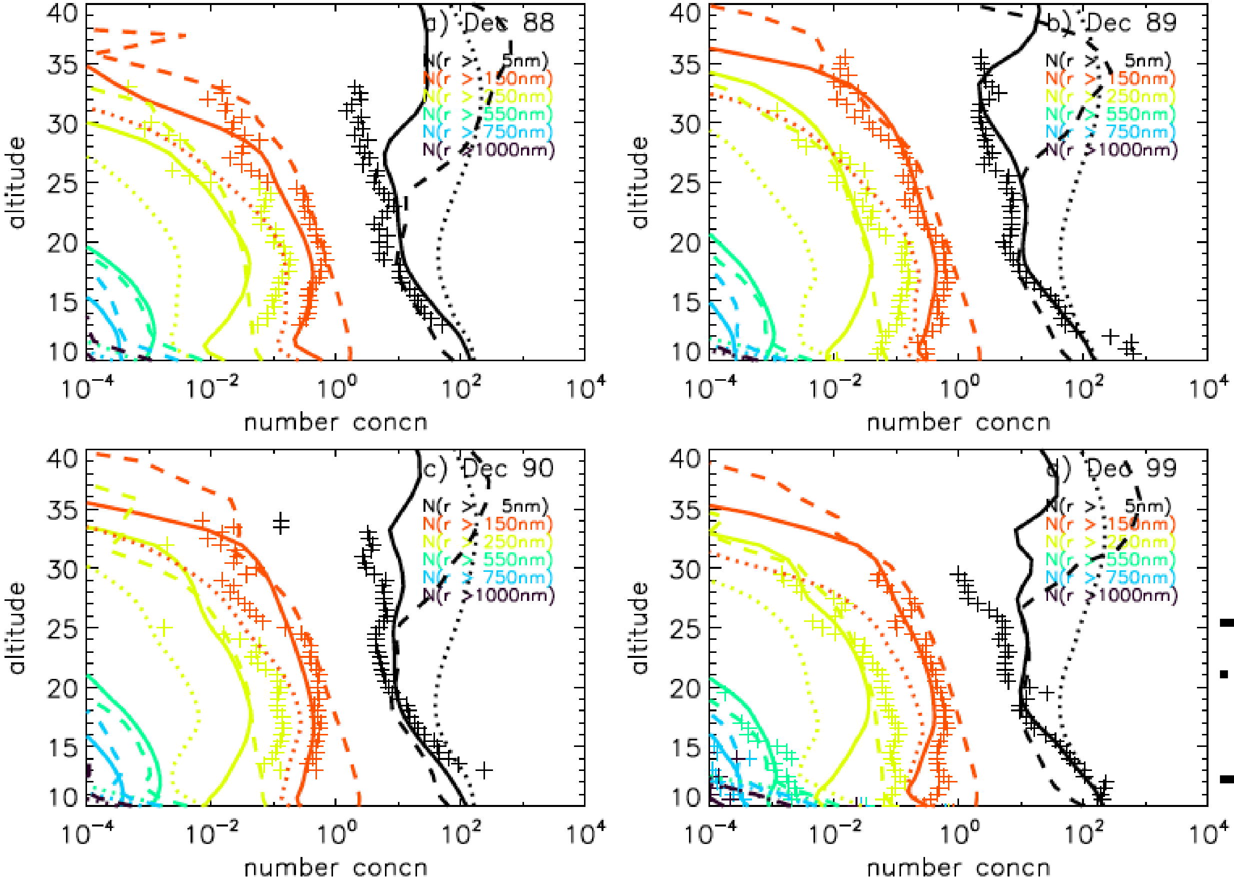
(simulation from Brooke et al., 2017)

UM-UKCA (no MSP interaction)
(only homogeneously nucleated particles)
(simulation from Dhomse et al., 2014)

Four seasons of quiescent particle concentrations at Laramie, Wyoming

Early winter (December) quiescent Junge layer balloon soundings (CPC and OPC)



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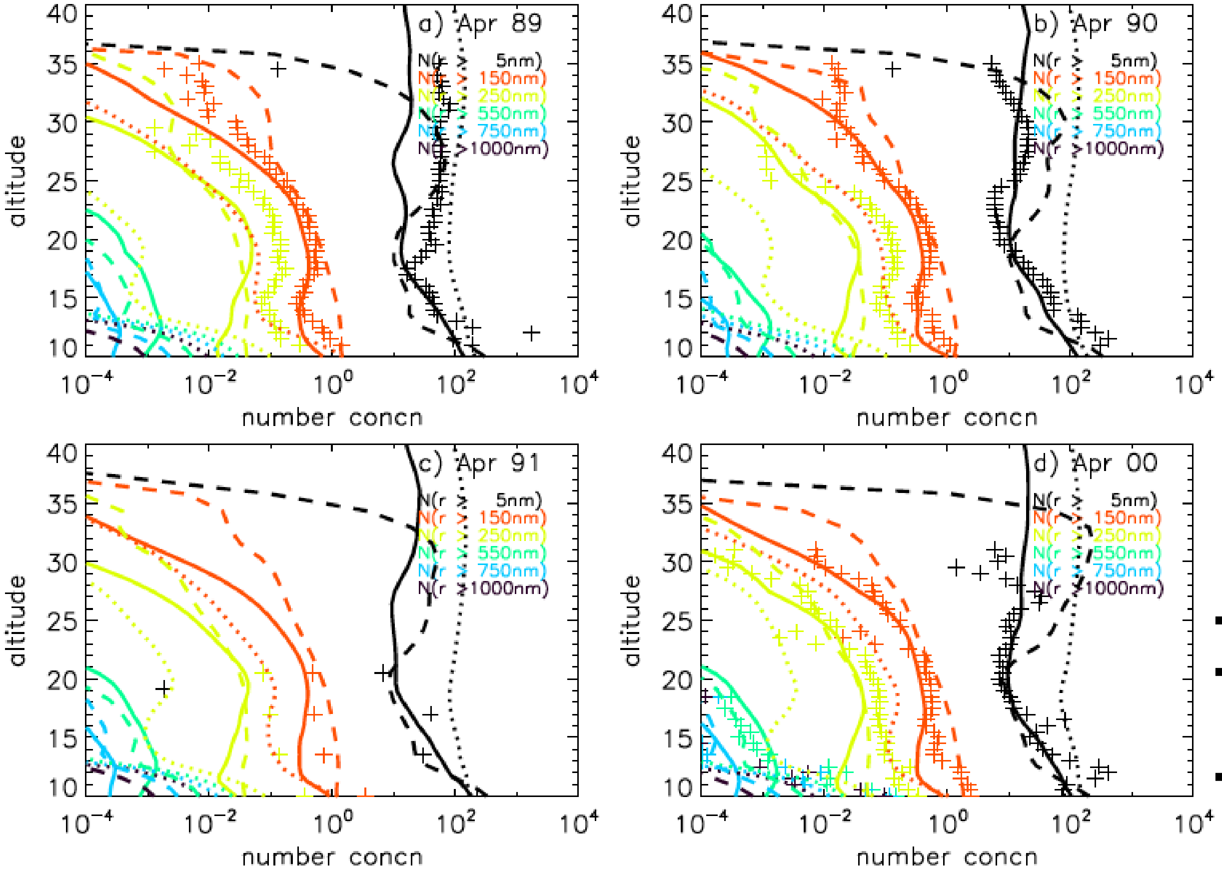
(simulation from Brooke et al., 2017)

UM-UKCA (no MSP interaction)
(only homogeneously nucleated particles)
(simulation from Dhomse et al., 2014)

Four seasons of quiescent particle concentrations at Laramie, Wyoming

Mid-spring (April) quiescent Junge layer balloon soundings (CPC and OPC)



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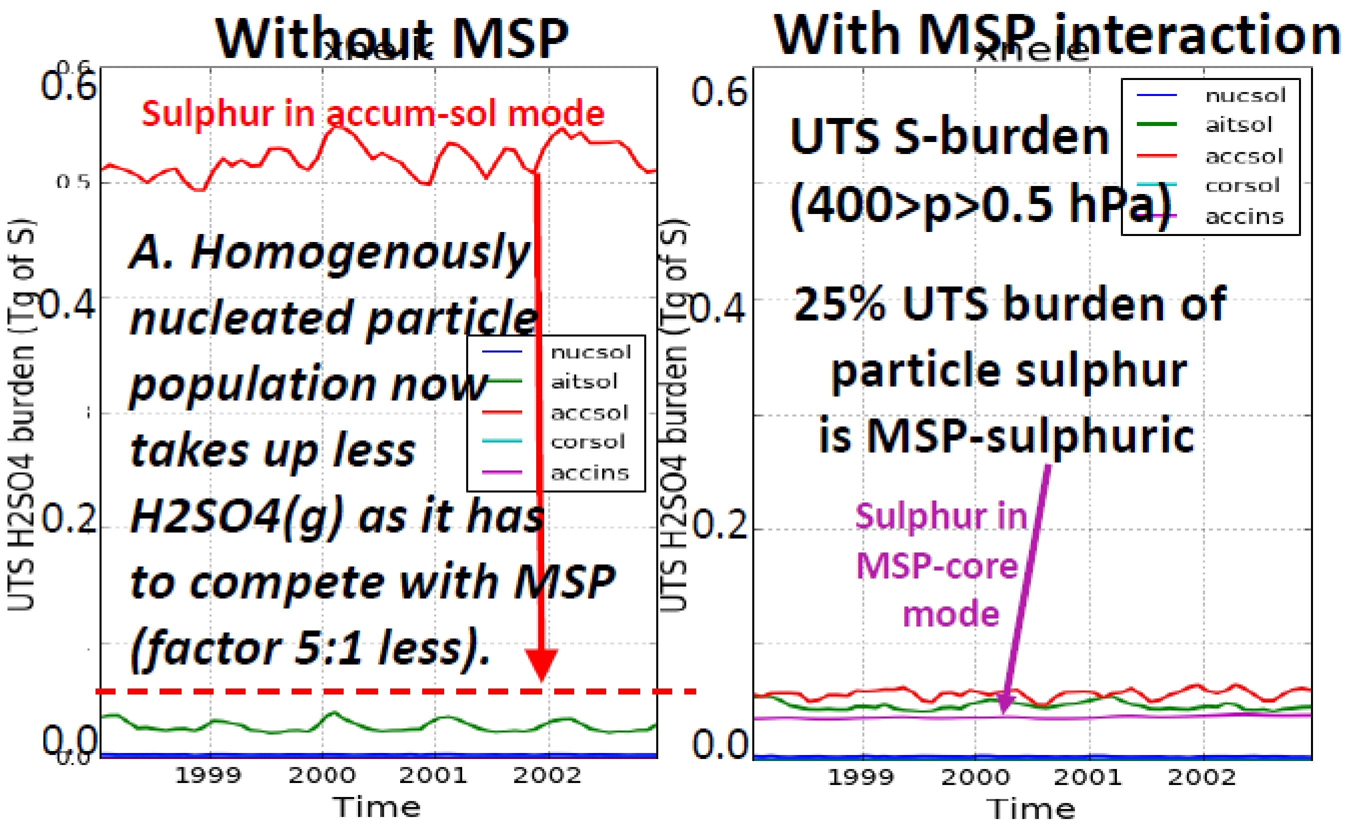
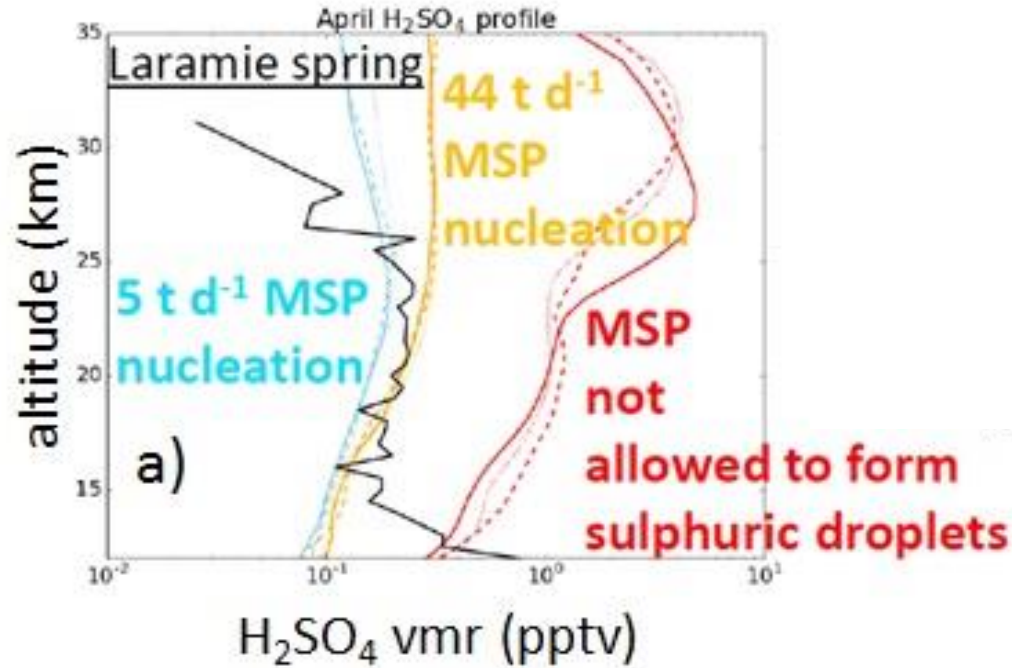
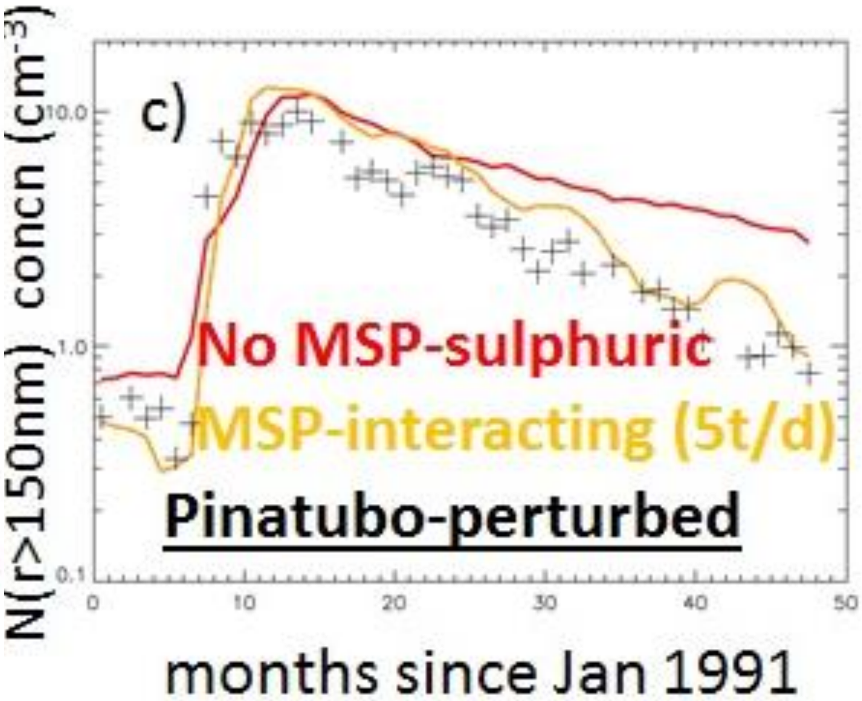


B. UTS particle S-burden also decreases as the particles are larger & shift towards tropopause



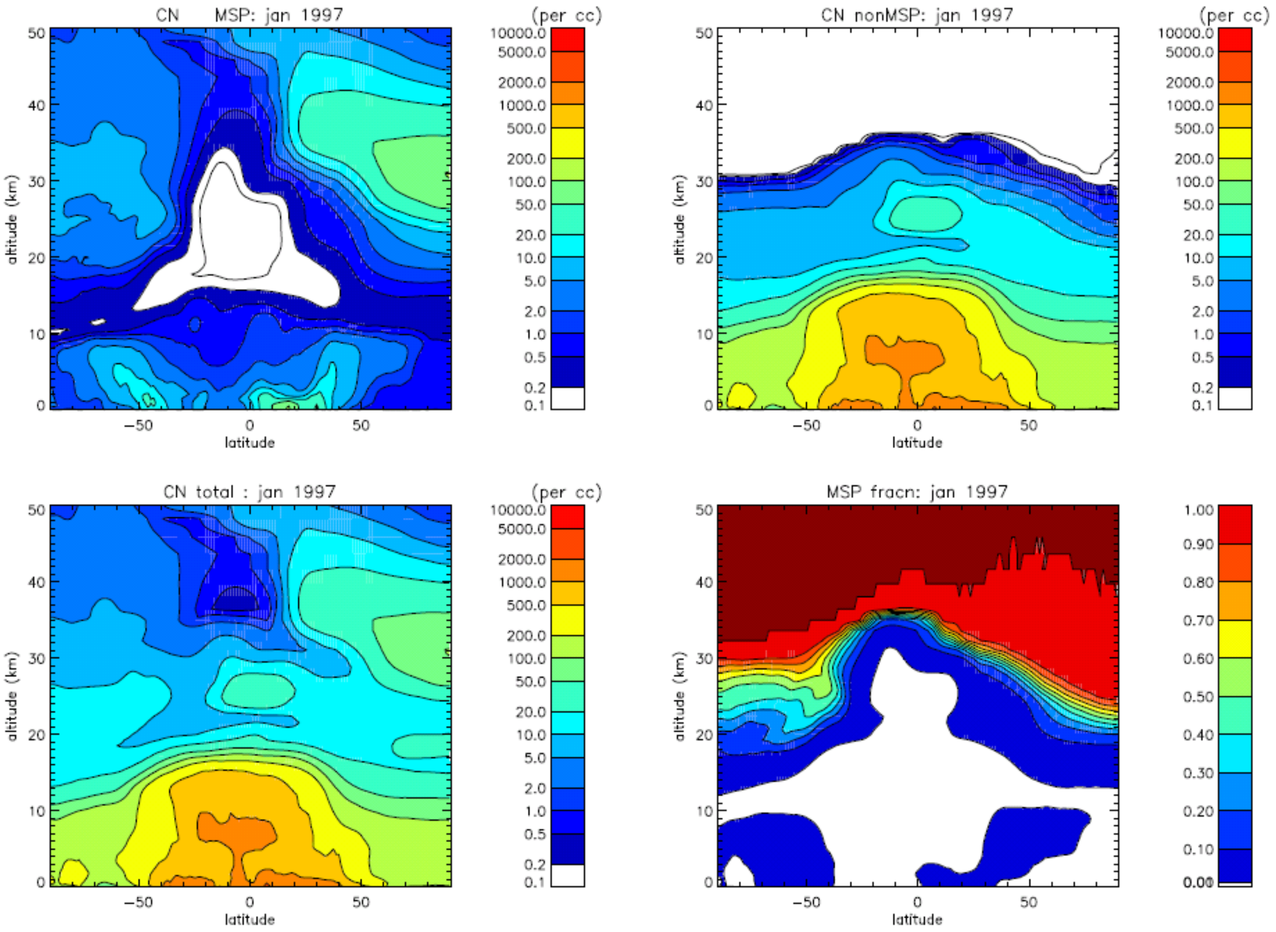


Particle phase sulphuric acid from size-distribution fits to CPC+OPC (Terry Deshler, U. Wyoming)

Simulated decay of N(r>150nm) at Laramie post-Pinatubo peak is faster with MSP-interaction. Larger reff → faster sedimentation

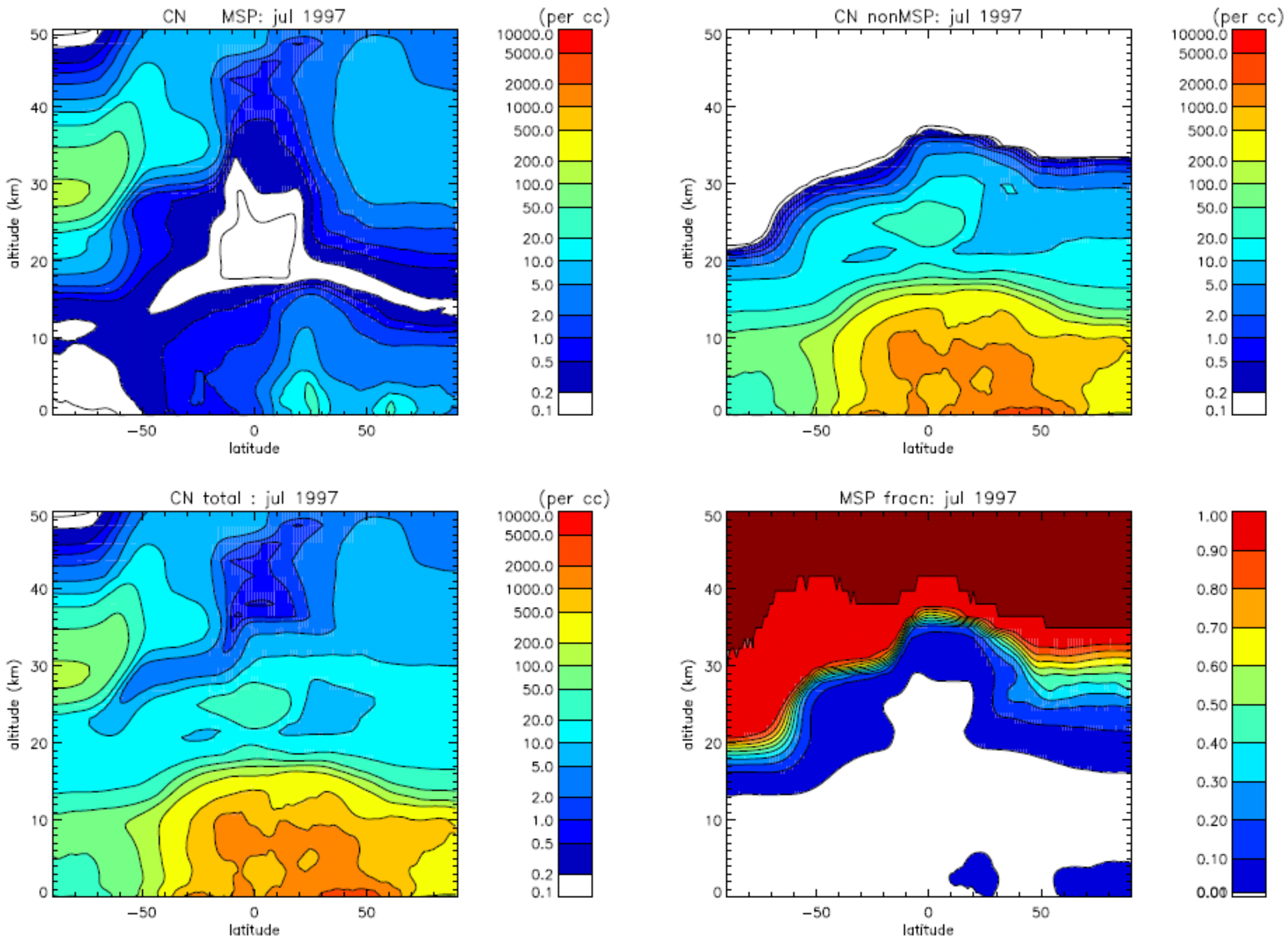
Morphology of quiescent Junge layer (pure-sulphuric and meteoric-sulphuric)

January mean



Morphology of quiescent Junge layer (pure-sulphuric and meteoric-sulphuric)

July mean



Summary and Future Work

-- Evidence from observations is clear that, in volcanically quiescent conditions, a substantial proportion of particles in the mid-latitude & high-latitude Junge layer are of meteoric origin

-- UM-UKCA interactive stratospheric aerosol model adapted to track meteoric-sulphuric particles within modal aerosol scheme. Now a core component of "strat-enabled GLOMAP" e.g. as applied for UM-UKCA simulations for VolMIP Tambora intecomparison experiment (Zanchettin et al., 2016, Marshall et al., 2018)

-- Model experiments show the heterogeneously nucleated "meteoric-sulphuric particles" strongly affect the vertical profile of stratospheric sulphur in the Junge layer.

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

-- UK Natural Environment Research Council has funded blue-skies research project to investigate meteoric influence on stratospheric aerosol and PSCs (to begin April 2018)

-- Leeds PDRA will compare "strat-enabled UM-UKCA" to the observations that indicate the meteoric origin (WB-57 PALMS & FCAS; refractory particles from Geophysica and balloons)

Meteoric particles under-represented in stratospheric particle lifecycle.

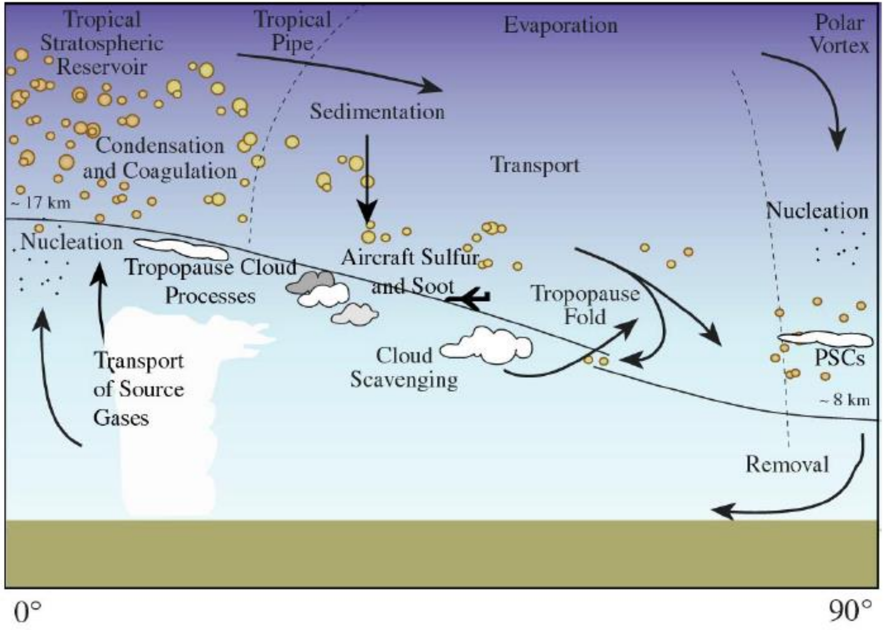
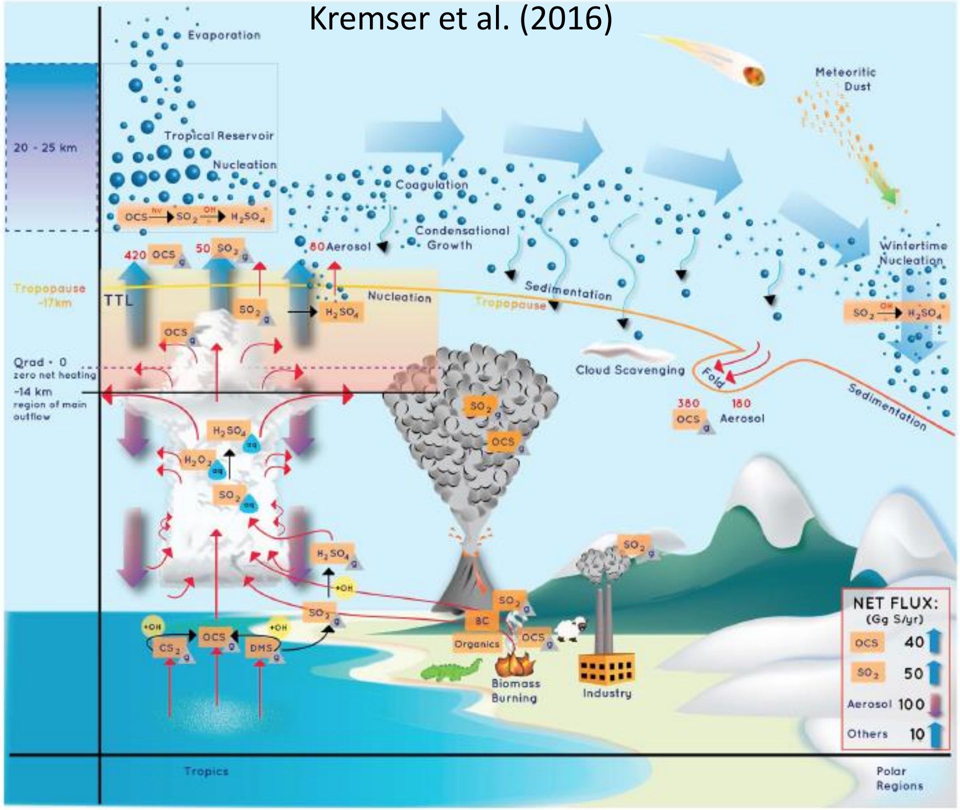


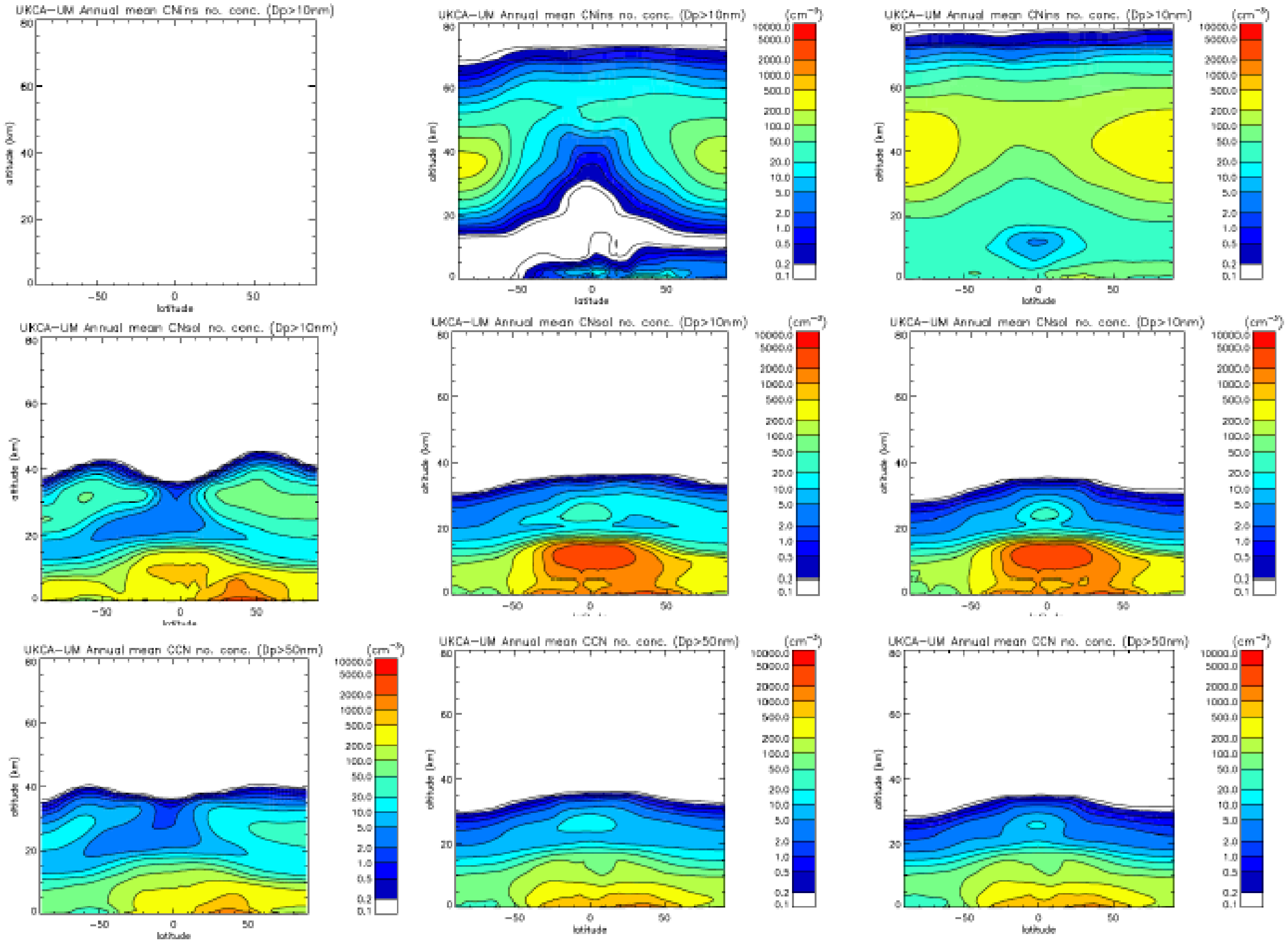
Figure 1.1. Schematic of the stratospheric aerosol life cycle [from Hamill et al., 1997].

SPARC Assessment of Stratospheric Aerosol Properties (2006)
Ch 1:"Stratospheric Aerosol Processes. (Carslaw and Karcher)

Kremser et al. (2016)



Updated stratospheric particle lifecycle schematic now includes Meteoric Dust but particle formation mechanism not recognised.

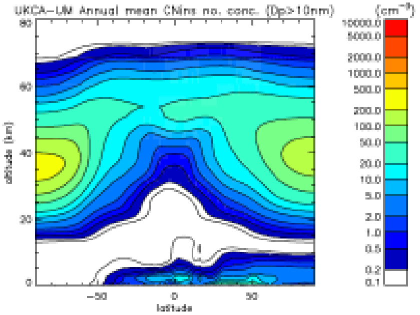


meteoric-sulphuric particle number N(Dp>10nm)

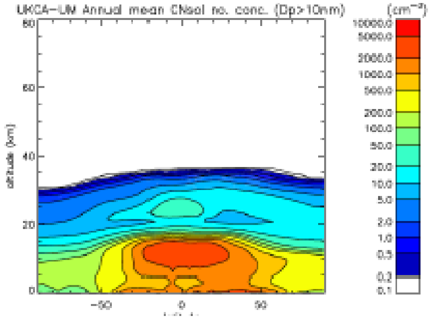
pure sulphuric particle number N(Dp>10nm)

pure sulphuric particle number N(Dp>50nm)

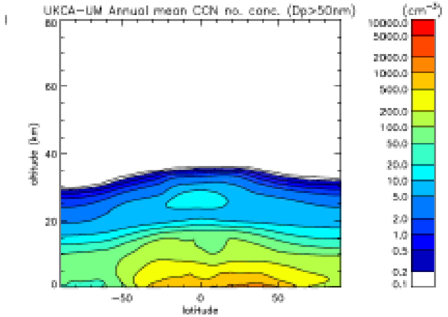
noMSP (Dhomse14) | loMSP, interacting with H_2SO_4 | hiMSP, interacting with H_2SO_4



meteoric-sulphuric particle number N(Dp>10nm)



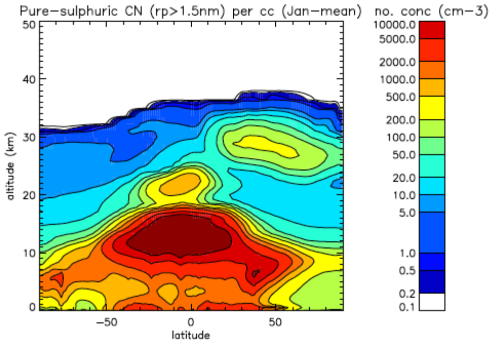
pure sulphuric particle number N(Dp>10nm)



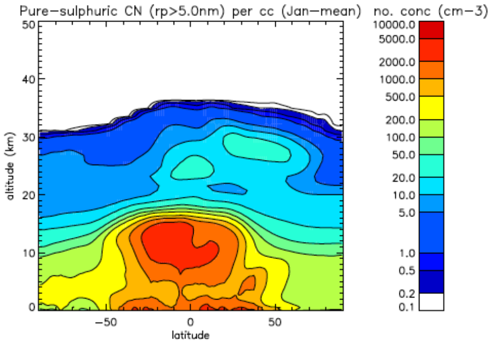
pure sulphuric particle number N(Dp>50nm)

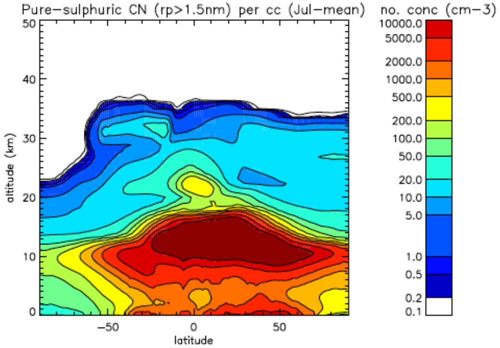
IoMSP, interacting with H_2SO_4

January-mean Pure sulphuric N(Dp>3nm)

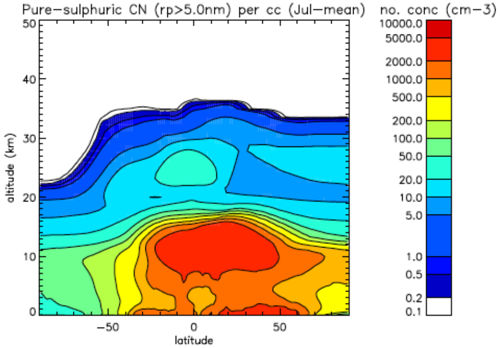


January-mean Pure sulphuric N(Dp>10nm)





July-mean Pure sulphuric N(Dp>3nm)



July-mean Pure sulphuric N(Dp>10nm)

Stratospheric Aitken particle concentration profiles

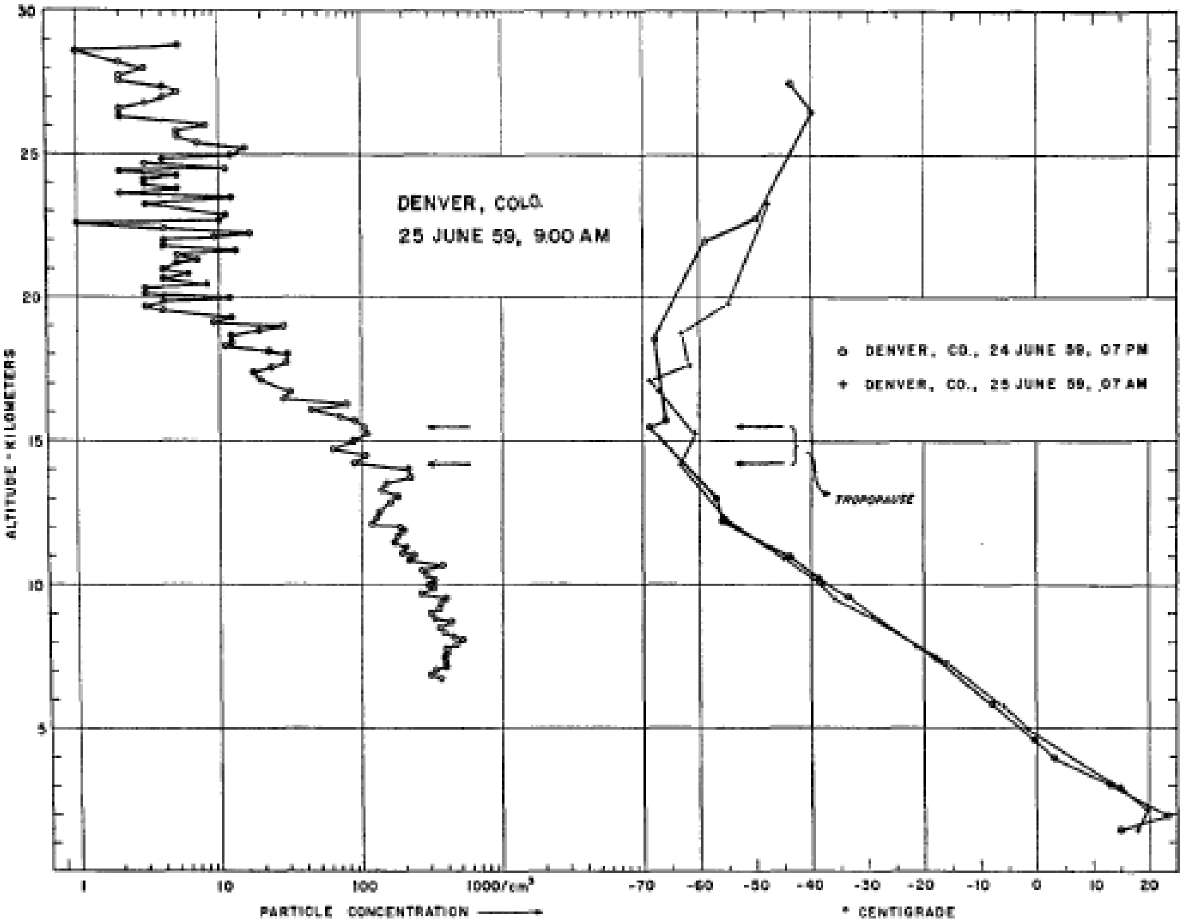


FIG. 10. Measured vertical profiles of Aitken nuclei and the available temperature soundings closest in space and time.

Junge et al. (1961)
Expansion-type Aitken particle counter
(working fluid = water)

Stratospheric Aitken particle concentration profiles

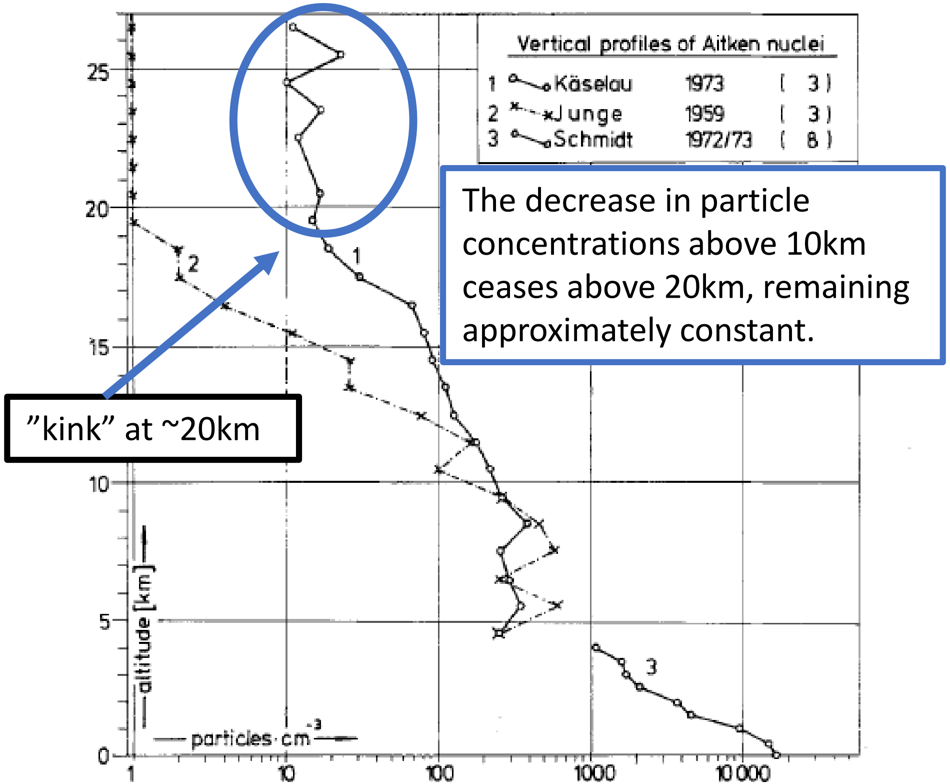


Figure 6
3 Aitken nuclei profiles in the atmosphere

<u>Optical particle counters</u> and particle collectors such as <u>impactors</u> or on <u>filters</u> measure **<u>only particles larger than ~100nm in radius</u>**.

<u>Aitken nuclei counters</u> measure particles to **<u>smaller sizes down to ~10nm in radius</u>**.

Balloon-borne measurements with Lindau Aitken nuclei counter designed specifically to measure stratospheric Aitken particle concentrations **<u>(radius between 10nm and 100nm)</u>**.

Kaselau et al. (Oct 1973, Germany, 53N) (1974, Pure. Appl. Geophysics)

All the condensation nucleus counters indicated generally decreasing mixing ratios with increasing altitude between the tropopause and about 20 km, although this decrease was most marked for the Junge and Rosen results. Above 20 km, which the Sands did not reach, the Junge and Rosen instruments indicated roughly constant mixing ratios, and the Käselau instrument indicated increasing mixing ratios with increasing altitude.

THEORIES OF SOURCES

Three general theories of the origin of Aitken particles in the stratosphere have been proposed: (1) they are formed in the stratosphere by gas to particle conversion, (2) they are of tropospheric origin, and (3) they are of extraterrestrial origin.

Considerable support is given to the theory that heterogeneous nucleation is the major mechanism for the nucleation of sulfate in the stratosphere by *Mossop* [1963, 1965]. Particles were collected by impaction at an altitude of 20 km on eight occasions prior to the eruption of Agung in 1963 and thus following a long time period of little highly explosive volcanic activity. The collected particles were examined by electron microscopy and electron diffraction. Unlike most recent workers Mossop concluded that most of the particles consisted of ammonium sulfate. He found that within each large particle there were one or more electron dense water-insoluble particles. These were best revealed by floating the electron microscope grid specimen uppermost on 'carefully purified water.' Their radii varied from 0.005 to 0.35 μm. Mossop theorized that they were of extraterrestrial origin and acted as nuclei on which ammonium sulfate deposited. Apparently, no other studies of quite this type have been attempted.

<u>Cadle and Kiang (1977, Reviews of Geophysics).</u>

The Junge layer, which occurs at a height of 20 km and consists of particles bigger than 0.1 μm, seems to be the sink for the Aitken nuclei which reach this altitude. The constant concentration of the Aitken nuclei above 20 km is a hint that there may be another source for these particles and this would have to be in the stratosphere. On the other hand, the concentration of the Aitken nuclei is too small to serve as the only, or at least the dominant, source of Junge particles. It is now generally assumed that the important source of big particles is a growth out of the gas phase. In this growth process there must be a period when the particles are in the Aitken nuclei size range. This would therefore serve as a stratospheric source of Aitken nuclei and would account for the nearly constant concentration above 20 km, that is observed.

Kaselau et al. (1974, Pure Appl. Geophys.)

The profiles of *Käselau et al.* [1974] of Aitken particle concentrations, which were obtained up to 27 km, indicated nearly constant concentrations in terms of numbers of particles per cubic centimeter, that is, increasing mixing ratios with increasing altitude, above 20 km. Possibly, above 20 km, extraterrestrial material became dominant. The Sands measurements did not reach this altitude, but neither the earlier work by *Junge* [1961] nor that of *Rosen and Hofmann* [1975] indicated such constant concentrations above 20 km. Since the errors associated with the data of Figure 1 have not been evaluated except in the general manner already discussed, firm conclusions with regard to extraterrestrial material cannot be drawn.

Cadle and Kiang (1977, Rev. Geophys.)