Stratospheric aerosol of extra-terrestrial origin

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Jan Leitner, Peter Hoppe

AGU Chapman Conference
Stratospheric aerosol in the post-Pinatubo era: Processes, Interactions, and Importance

18-23 March 2018

→ ERC grant “EXCATRO”: FP7 agreement no. 321040
→ *StratoClim*: FP7 grant agreement no. 603557
→ *~SPITFIRE~* within ROMIC: BMBF no. 01LG1205A

„Extra-terrestric dust“

„Cosmic Dust“

„Interplanetary Dust Particles **IDPs**“

„Stardust“

„Meteoric Dust“

„Meteoric Fragments“

„Meteoric Smoke“

Plane, John M. C., Chem. Soc. Rev., 2012, **41**, 6507–6518:

“The solar system is full of dust: if all the dust in the inner solar system (between the sun and Jupiter) were compressed together it would form a moon 25 km in diameter.”

Main sources of dust particles:

- collisions of asteroids (asteroid belt between the orbits of Mars and Jupiter),
- sublimation of comets (dust-laden ice) orbiting the sun → dust trails → origin of meteor showers
- long-decayed cometary trails.

85–95% of inner solar system dust: Jupiter-family comets with ~20 yrs orbital period.

5-15%: from asteroid belt and Halley family and Oort cloud comets.

(Plane, Chem. Soc. Rev., 2012, **41**, 6507–6518
& Plane, Chem. Rev. 2003, 103, 4963-4984)

Meteoroid → Small (sub-km) rocky or metallic body in outer space.
Meteor → Light phenomenon ("shooting star" or "falling star“), visible passage of a frictionally heated and glowing body from outer space.
Meteorite → Solid piece or debris from outer space which has survived the passage through the atmosphere and has hit the surface → **Micrometeorite** if D_p < 1 mm.

from a Meteoroid to a Meteorite

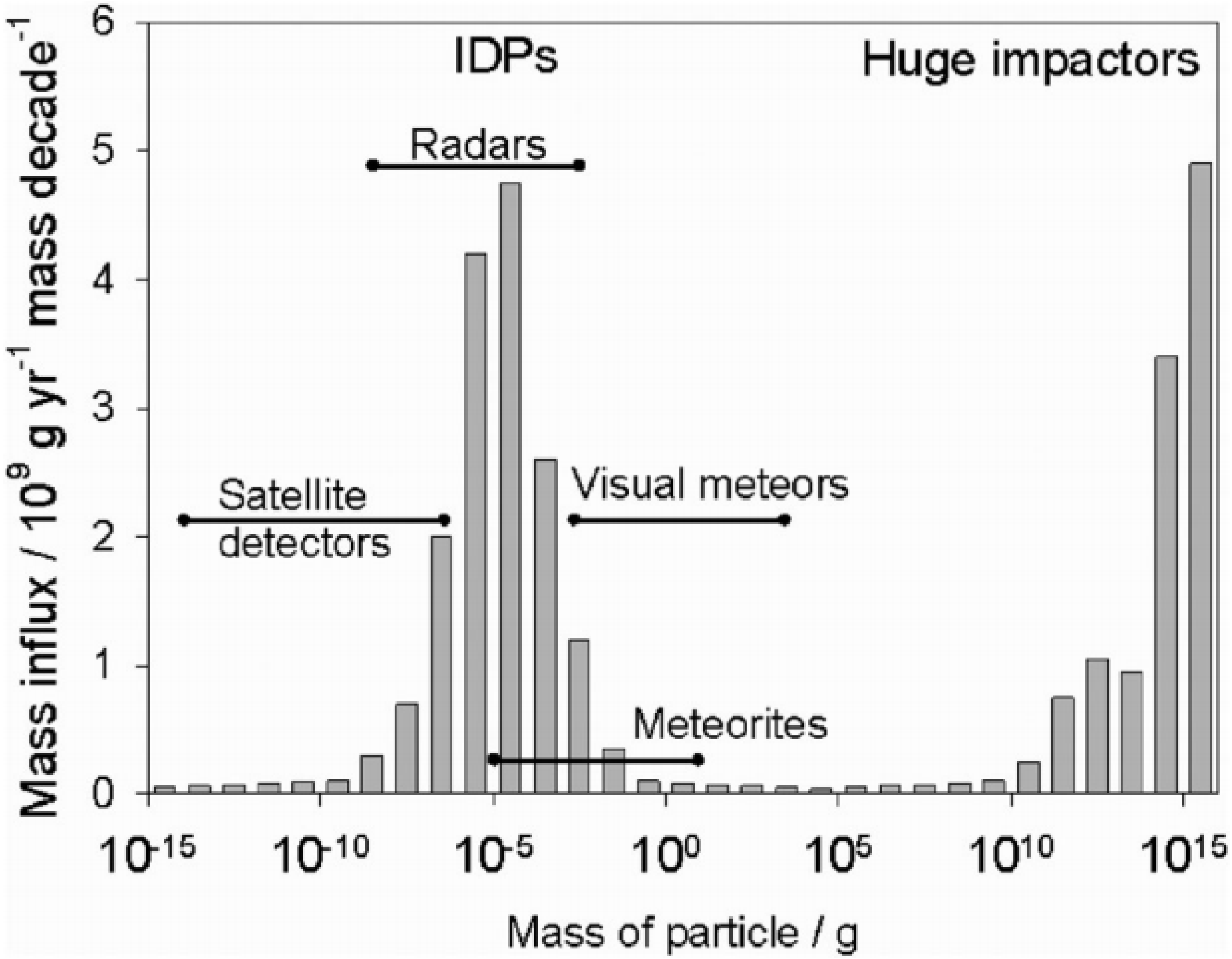


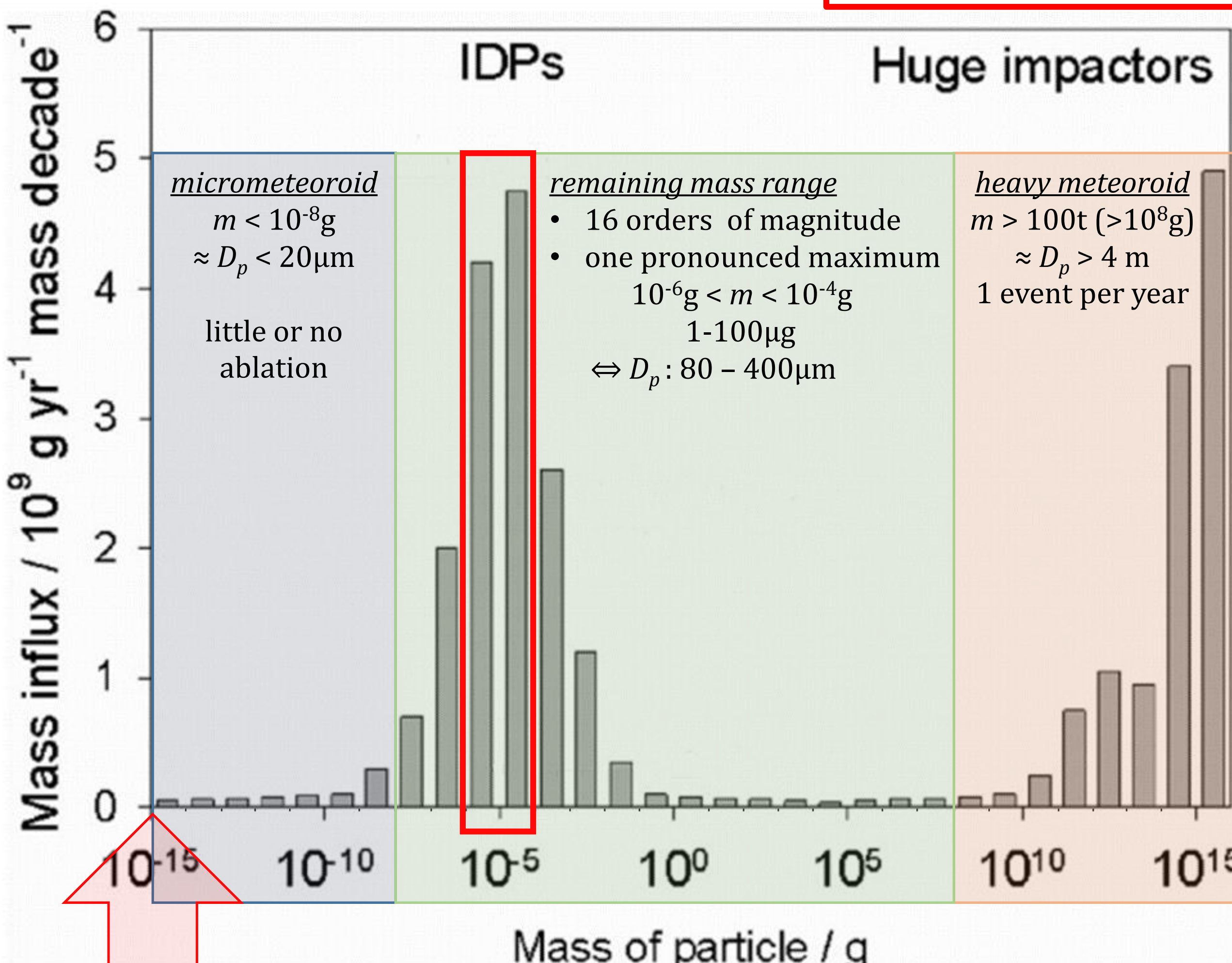
Fig. 2 Mass influx (per decade of mass) plotted against particle mass

[Data taken from: G. J. Flynn, in **Meteors in the earth's atmosphere**, ed. E. Murad and I. P. Williams, Cambridge University Press, Cambridge, 2002]

John M. C. Plane, Chem. Soc. Rev., 2012, **41**, 6507–6518

from a Meteoroid to a Meteorite

$\rho_{Met}: 2-3\ g\ cm^{-3}$ (ordinary meteor)
up to $> 8\ g\ cm^{-3}$ (iron meteor)



allmost all of these become meteorites → „touch down“ at ground

According to these numbers (von Zahn, 2005): $D_p \approx 90$nm

John M. C. Plane, Chem. Soc. Rev., 2012, **41**, 6507–6518 **modified**

von Zahn, 2005

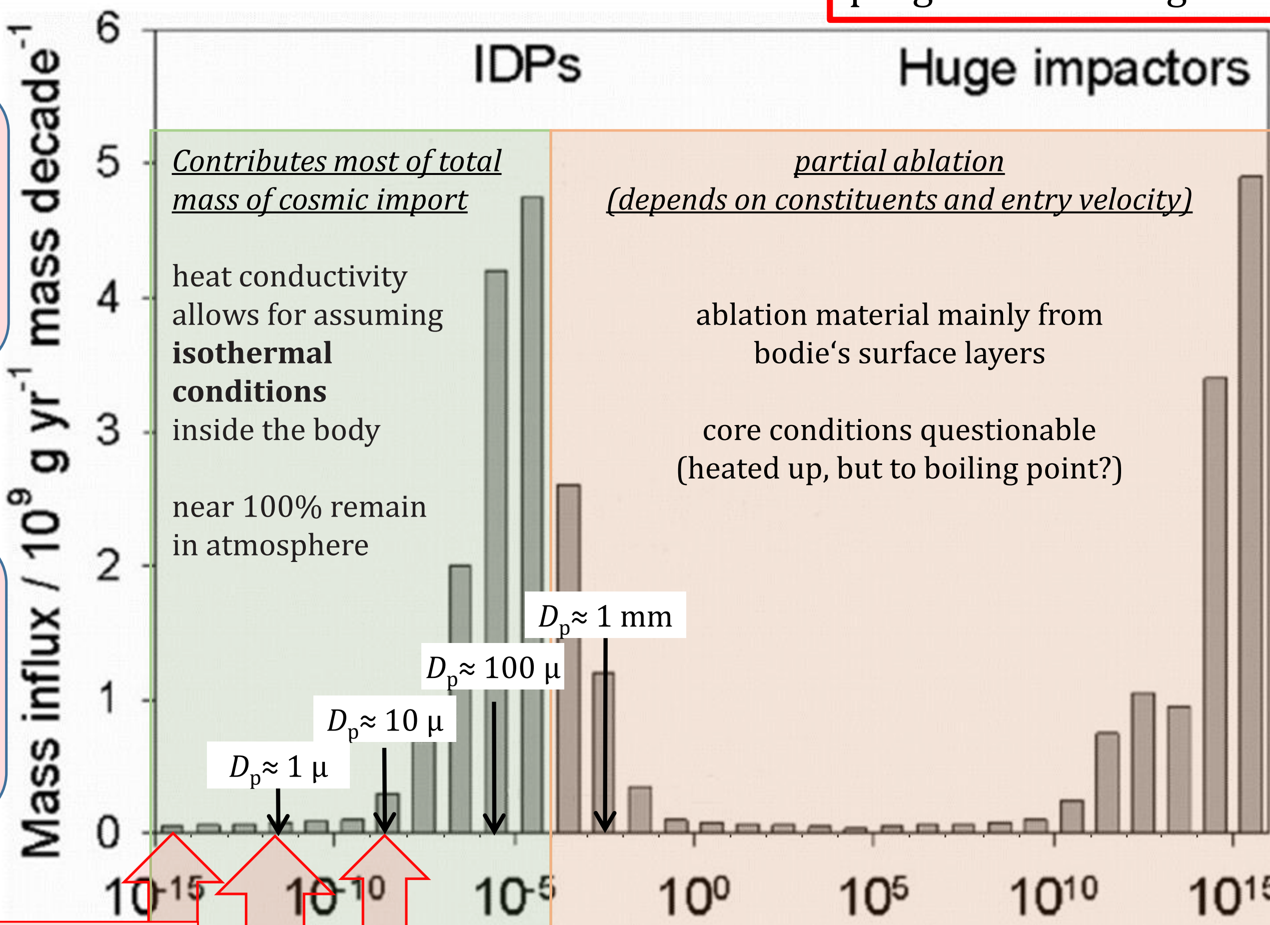
Proceedings of the 17th ESA Symposium on European Rocket and Balloon Programmes and Related Research, Sandefjord, Norway, 30 May – 2 June 2005 (ESA SP-590, August 2005)

from a Meteoroid to a Meteorite

v_{entry}: 11.5 - 72 km s^{-1}, depends on prograde or retrograde orbit

rapid frictional heating by collision with air molecules

the constituent minerals vaporize.



for D_p < 100 nm :
no frictional heating
→ no ablation*

for D_p >10 μm ablation (boiling) temps T ≈ 2000 K are reached
→ further frictional heat input is balanced by vaporization and radiative cooling, no further temperature rise occurs.*

for $D_p \approx$ 1 μm a $v_{entry} \approx$ 40 km s^{-1} required to heat up to 1000 K (sodium, *Na*, begins to evaporate).*

John M. C. Plane, Chem. Soc. Rev., 2012, **41**, 6507–6518
modified

*John M. C. Plane (Chem. Rev. 2003)

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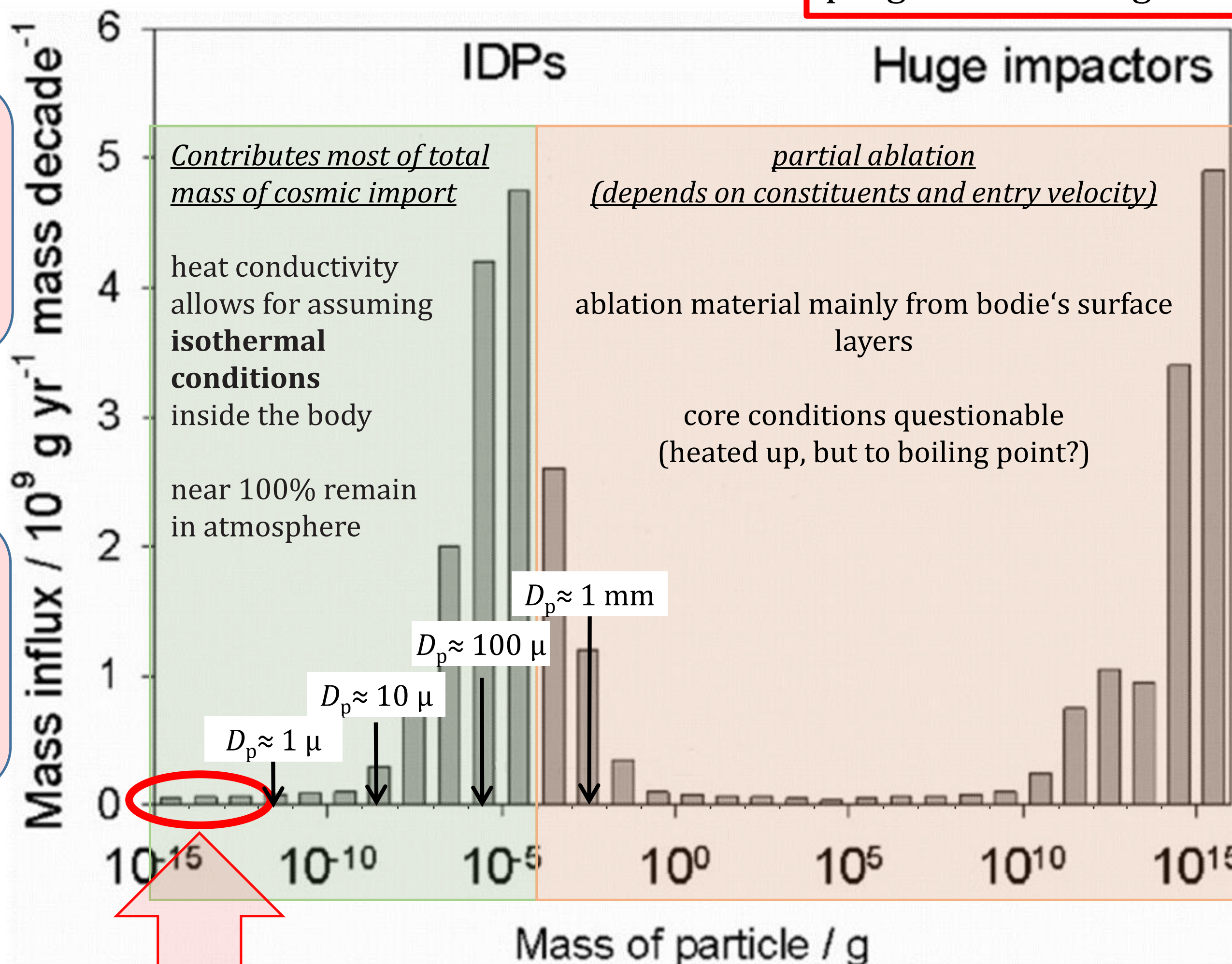


$\Sigma \approx 150$ t yr^{-1} ($\rho = 3$ g cm^{-3})
presuming mean D_p of 100, 300 and 700 nm per mass decade, respectively
→ influx of ~ 10^{20} particles per day

John M. C. Plane, Chem. Soc. Rev., 2012, **41**, 6507–6518 **modified**
*John M. C. Plane (Chem. Rev. 2003)

The total mass influx of cosmic material - the great unknown -

Table 1 Estimates of the global IDP input rate to the Earth's atmosphere (deep blue = extra-terrestrial estimate; light blue = middle atmosphere estimate; grey = ice core/deep-sea estimate)

Technique	IDP input t d^{-1}	Reference	Potential problem of technique
Zodiacal dust cloud observations and modelling	270	Nesvorný et al. [1]	Needs to be constrained by terrestrial meteor radars
Long Duration Exposure Facility	110 ± 55	Love & Brownlee[9]	Sensitive to IDP velocity distribution
High performance radars	5 ± 2	Mathews et al.[10]	Possible velocity bias / selective mass range
Conventional meteor radars	44	Hughes[11]	Extrapolation, selective mass/velocity range
Na layer modelling	20±10	Plane[12]	Sensitive to vertical eddy diffusion transport
Fe layer modelling	6	Gardner et al.[13]	Depends on vertical transport
Fe/Mg in stratos. sulphate layer	22 – 104	Cziczo et al.[14]	Data has limited geographic extent
Optical extinction measurements	10 – 40	Hervig et al.[15]	Particle refractive indices undertain
Fe in Antarctic ice core	15 ± 5	Lanci et al.[16]	Very little wet deposition by snow
Fe in Greenland ice core	175 ± 68	Lanci & Kent[17]	Uncertain atmospheric transport/deposition
Ir and Pt in Greenland ice core	214 ± 82	Gabrielli et al.[18]	Uncertain atmospheric transport/deposition
Os in deep-sea sediments	101 ± 36	Peuker-Ehrenbrink[19]	Focusing by ocean currents
Ir in deep-sea sediments	240	Wasson & Kyte[20]	Focusing by ocean currents

John M. C. Plane, Chem. Soc. Rev., **41**, 6507–6518, 2012.

- and its atmospheric lifetime

Simulations of transport & deposition of ^{238}Pu-oxide nanoparticles released due to a satellite's re-entry and entire ablation of its power unit in the upper stratosphere (above 25 km, ~11°S) in the year 1964.

→ removal of ^{238}Pu (half-life: 88yr) towards surface **within 4-5 years** due to atmospheric circulation (rather than sedimentation).

Dhomse et al., GRL, **40**, 4454–4458, 2013.

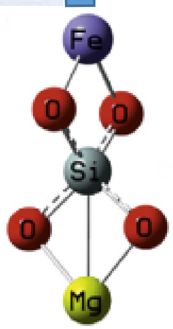
Meteoric Smoke Particles

(cf., e.g., Kremser et al., Rev. Geophys., 2016)

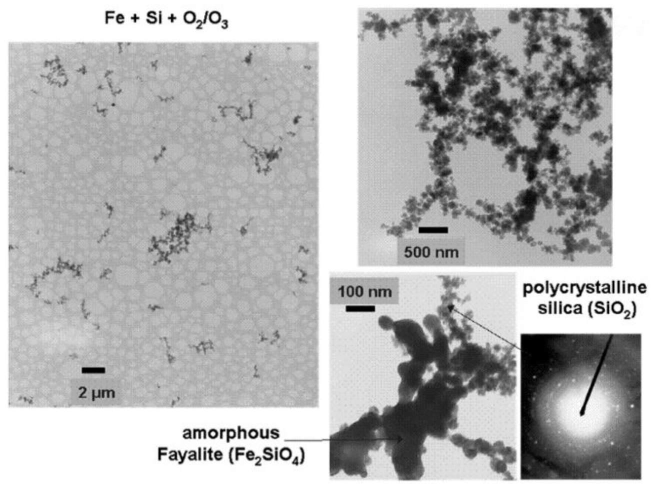
MSPs

Metallic compounds polymerize together with silicon oxides (due to dipole moments) quite rapidly
→ Metal-rich particles ("meteoric smoke" particles) with $D_p \approx 4$ nm (at ~ 80 km altitude).

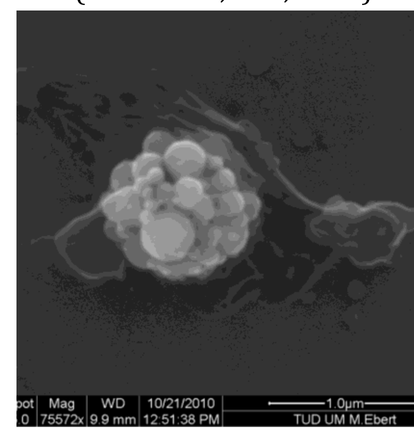
ablation at 80-100km



Aggregates of lab-grown polymerized nano-MSPs
(Plane et al., Chem. Rev., 2015)



Iron-oxide sampled in polar winter stratosphere:
~20% of all particles sampled
(Ebert et al., ACP, 2016)



Vapour/Molecules
Fe-Mg-SiO_4 composition (olivin-type)
(Saunders and Plane, 2011;
A. D. James et al., Atm. Sol. Terr. Phys., 2017
Frankland et al., Icarus ,296, 150–162, 2017)

Re-combination of spherules as „bunch of grapes"
(Bigg et al., Tellus, 1970)

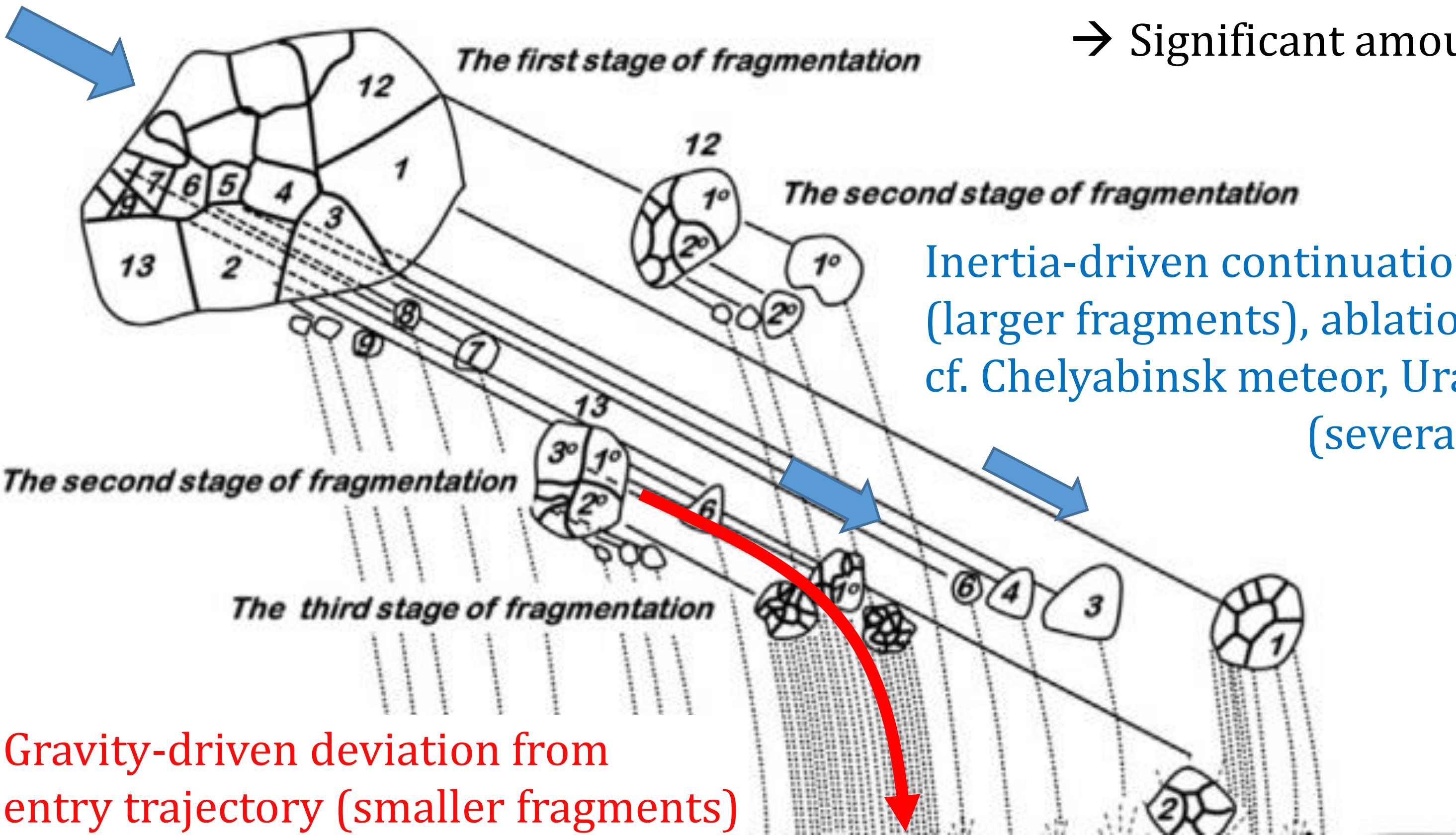
Over several days: growth due to condensation and coagulation
→ sediment into the lower mesosphere.

Alternatively: Meteoric Fragmentation

MFs : 95 % of meteoroids (D_p > 1 mm) fragment on atmospheric entry (Subasinghe et al., MNRAS, **457,** 1289–1298, 2016).

The volatile “glue” evaporates (tarry material, holding together the refractory grains in the meteoroid).

→ Significant amount of unablated MFs.



Inertia-driven continuation of entry trajectory (larger fragments), ablation may continue.
cf. Chelyabinsk meteor, Ural region, Russia
(several videos in the web)

Gravity-driven deviation from entry trajectory (smaller fragments)

Graphic from: Sears et al., Planetary and Space Science, 124, 2016.

Meteoric Fragmentation:

Check youtube links:

https://www.youtube.com/watch?v=-EVeGwe3TJc
https://www.youtube.com/watch?v=xCHh97iSksY

...for watching a very exciting example of a particular kind of meteric fragmentation

Impact of meteoric aerosol material in the atmosphere:

- Noctilucent clouds[1)] & polar mesospheric summer echos, both due to presence of ice particles in the Mesosphere. (e.g. Megner et al., ACP 2006; Rapp und Lübken, ACP 2004)

- Heterogeneous chemical reactions/conversions due to provision of surface, no matter if coated (H_2O/ice, H_2SO_4, HNO_3,…) or not.

- Polar stratospheric cloud[2)] formation → denitrification/dehydration of polar stratosphere. (e.g. T. Peter & J.-U. Grooß, in *Stratospheric Ozone Depletion and Climate Change*, ed. R. Müller, Royal Society of Chemistry, Cambridge,2012)

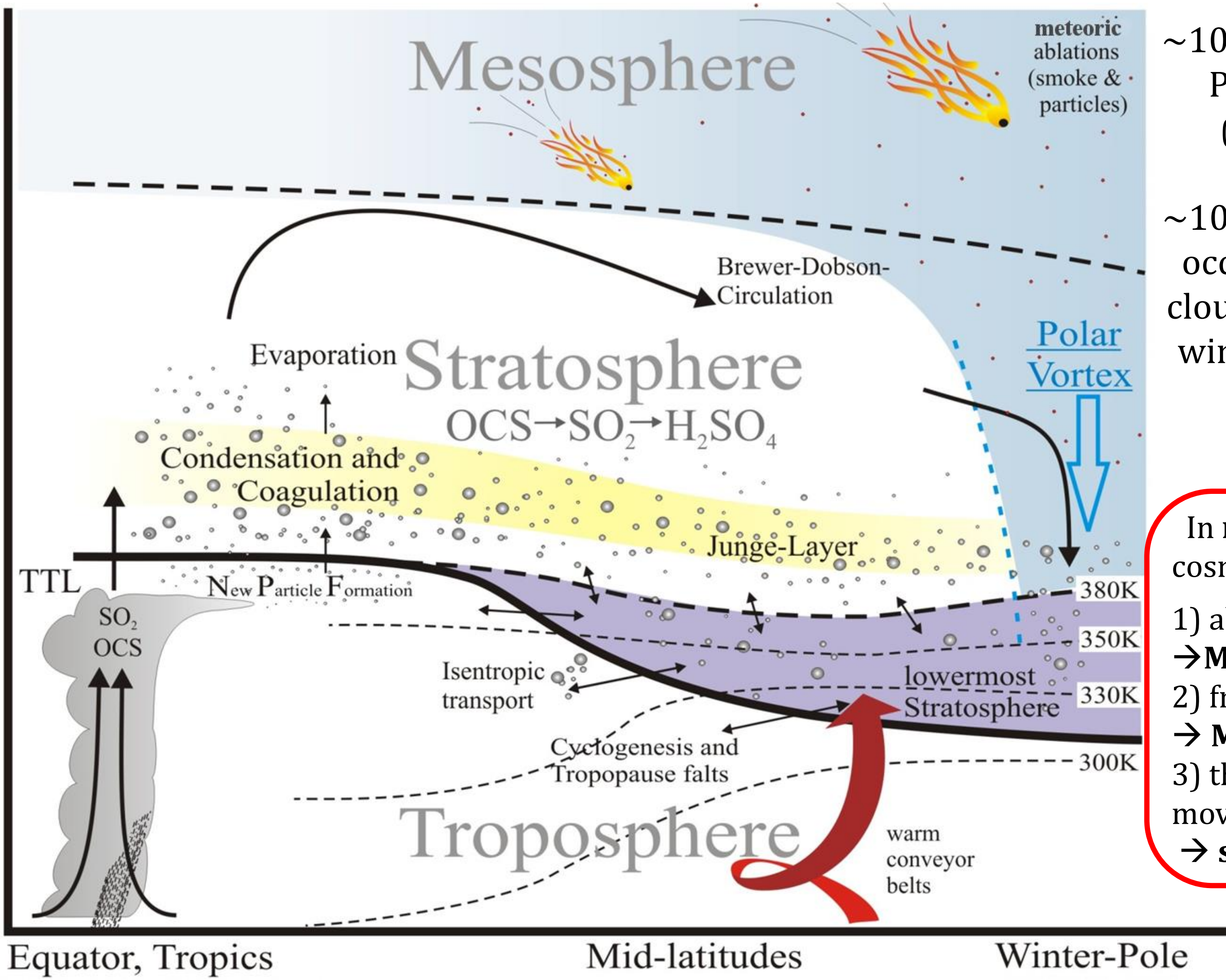
Image examles:

[1)] https://de.wikipedia.org/wiki/Leuchtende_Nachtwolke#/media/File:Helkivad_%C3%B6%C3%B6pilved_Kuresoo_kohal.jpg

[2)] https://www.meteoros.de/themen/atmos/beugung-interferenz/polare-stratosphaerenwolken/

Impact of meteoric aerosol material in the atmosphere:

For the aspect of surface provision: not neccessarily the particles` mass, but number matters.



~100 cm^{-3} to explain effecting PMSE and NC formation (Megner et al., ACP, 2006)

~10 cm^{-3} to explain largescale occurrence of stratospheric clouds during the 2010 arctic winter (Engel et al., ACP, 2013)

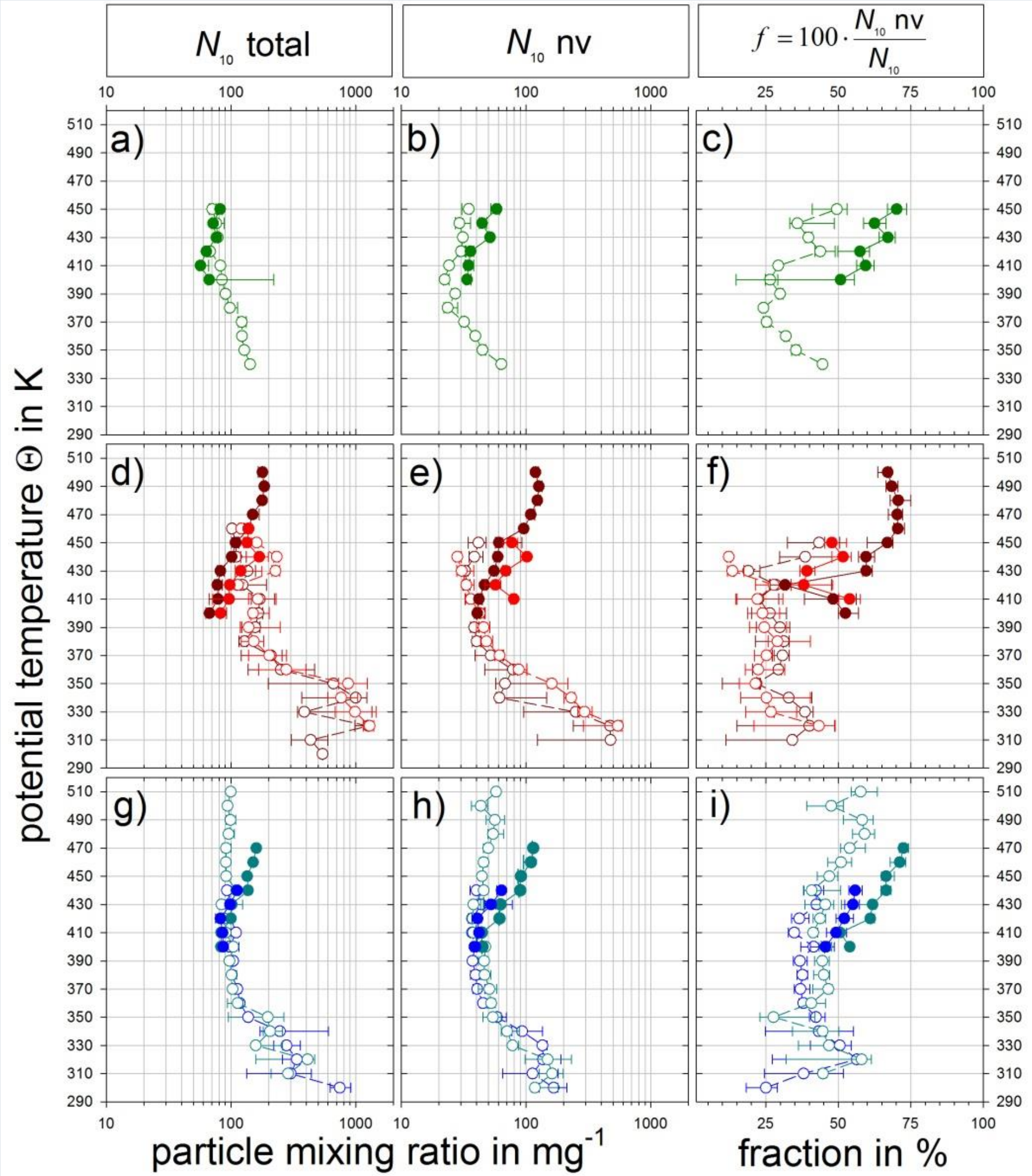
In **number** terms, the cosmic contributions are:

1) ablation & polymerisation
→**MSPs**
2) fragmentation
→ **MFs**
3) the fact that planet Earth moves through a dusty space…
→ **sub-μ IDPs**

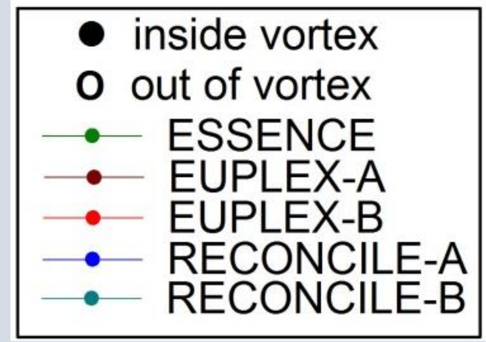
High altitude measurements aboard the M-55 *Geophysica* :

Length: 22.87 m
Height: 4.83 m
Wingspan: 37.46 m

Max ceiling	69 000 ft
Usual ceiling (measurements)	67 000 ft
Max payload	2 553 kg
Scientific payload (measurements)	2 000 kg
Endurance at max scientific payload	4 h
Max range	3 450 km
Take-off runway length	1 800 m
Crew (pilots + operators)	1



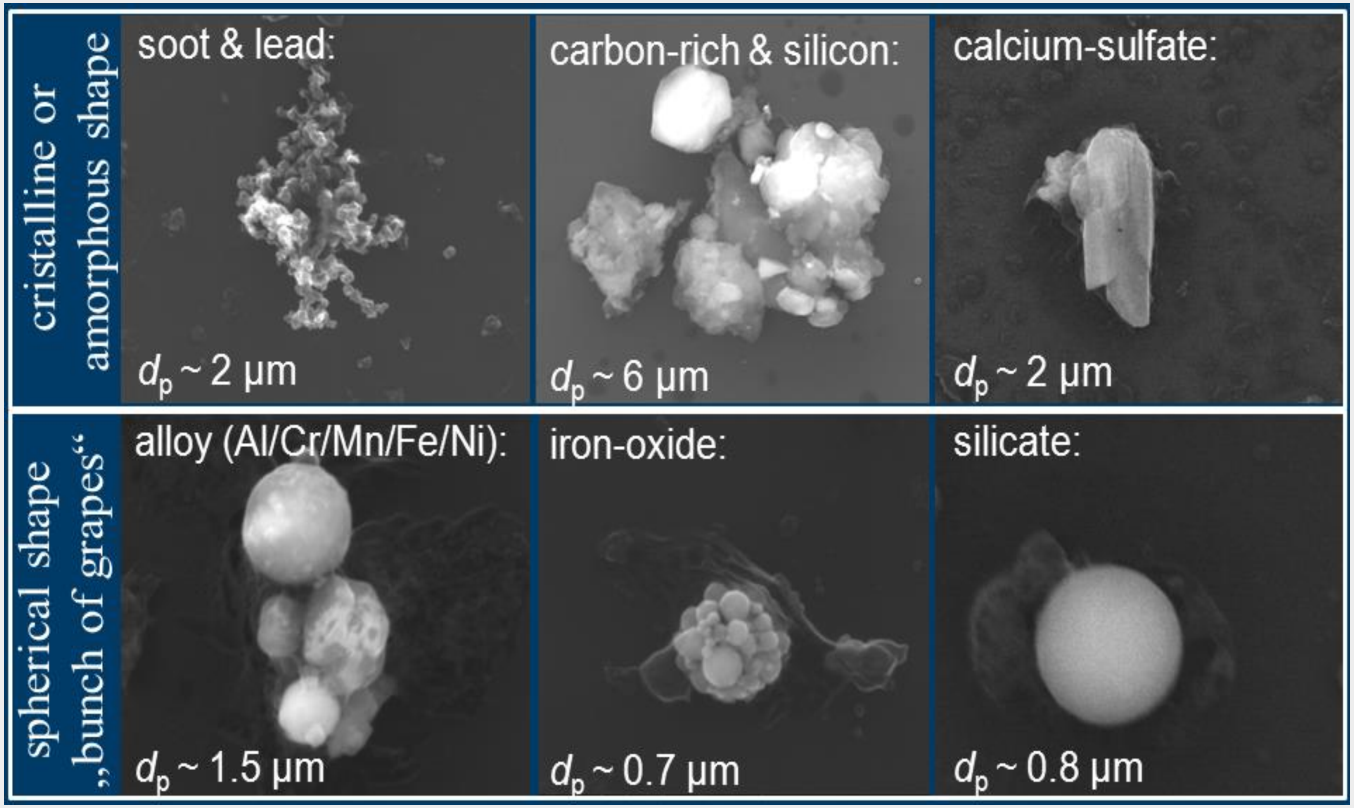
Weigel et al., ACP 2014



Aerosol in the Arctic winter vortex

- Significant increase in number with increasing altitude, inside the vortex
- Predominantly consisting of non-voltile material, thermo-stable at ~250°C

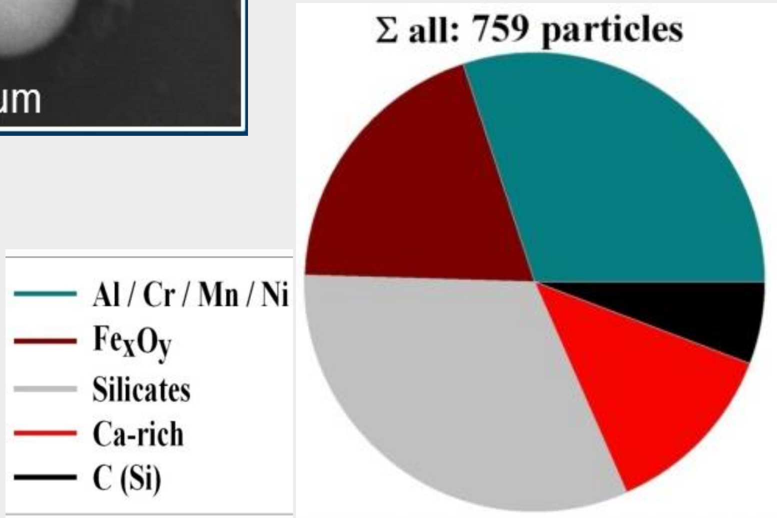
Impactor samples from the arctic winter stratosphere (2010)



all samples
were taken at
Θ > 410 K

More details:
Ebert et al., ACP 2016

Partly indicative for meteoric source
BUT
origin at surface or even anthropogenic
influences not excluded!



AIDA Cloud-Chamber , KIT – Karlsruhe

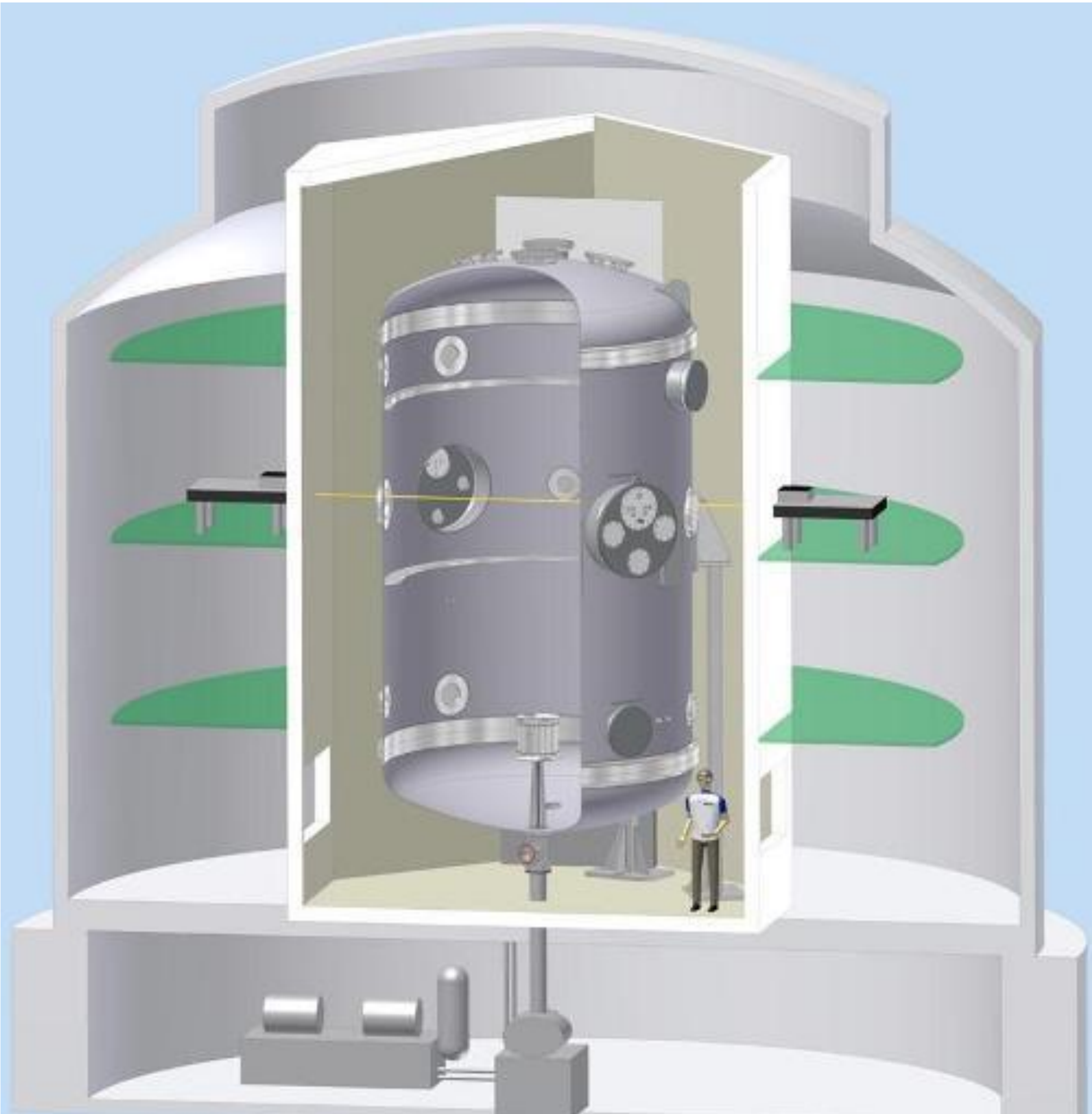


Image grabbed from http://www.imk-aaf.kit.edu/73.php

ASCONA, December 2012

(AIDA Study COncerning NAx formation)

Conditions

$T_{chamber} < 192K$

$p_{chamber}$ = ambient

Ingredients:

- HNO_3-H_2O vapor (45-55 wt %)
- $FeMgSiO_4$ (Univ. of Leeds, UK)
- Illite SE by Arginotec
  (cont.: Al,Fe,Mg,Si,Ca,K)
- Soot (Palas GFG-1000)
- Soot-Lead-Mix (Palas GFG-1000)

Key Instruments:

¤ FSSP-300 ¤ CCP ¤ CPCs,
¤ WELAS-OPC ¤ SID-3 ¤ Impactor
¤ SIMONE (depolarization)
¤ FTIR spectrometer

ASCONA: particle diffraction patterns (SID-3)

Homogeneous

Heterogeneous: Illite-SE

long , very thin needles:
aspect ratio: > 5

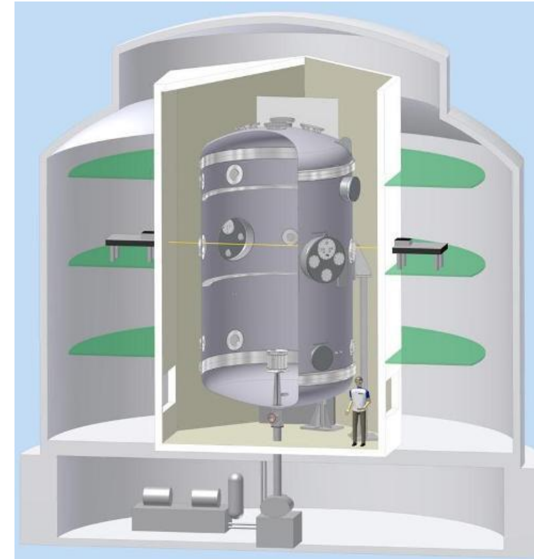
comparatively small & more compact:
aspect ratio: 1-2

ASCONA: extinction spectra (FTIR)

Infrared extinction spectroscopy was used to infer the chemical identity of the nucleated particles in the chamber.

NAD and NAT have modifications, denoted α-/β-NAX featuring characteristic spectroscopic signatures in the mid-IR range.

Preliminary Data – by courtesy of Schnaiter & Wagner, KIT



On time scales of the experiments (~6 h), no NAT formation occurred although S_{NAT} ranged at 16-26. Exclusively the nucleation of the NAD phase was detected:

- homogeneously: α-NAD, needles
- heterogeneously: β-NAD of more compact shape.

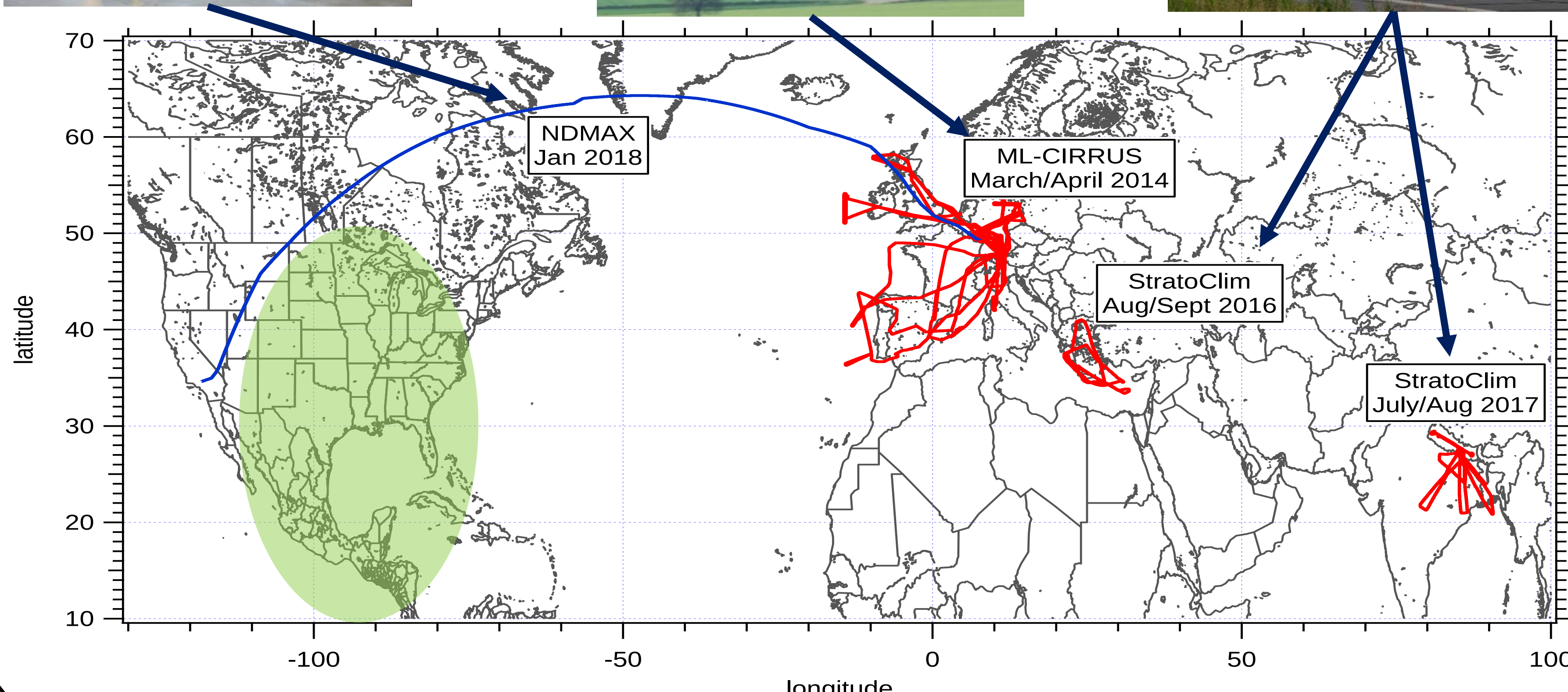
$HNO_3 \cdot \mathbf{2}H_2O$ is supposed to be "..only slightly less stable…" than $HNO_3 \cdot \mathbf{3}H_20$ (D. R. Worsnop et al., Science, 1998)

ASCONA recap

- NAT nucleation was never observed!
- HNO_3 nucleation clearly accelerated by nuclei compared to homogeneous formation.
- $FeMgSiO_4$ slightly more efficient than Illite (higher particle numbers and larger sizes).

- Re-assessment of former studies concerning limiting values for nucleation processes in the atmosphere (Hoyle et al., ACP, 13, 9577-9595, 2013).
- Observed largescale stratospheric clouds during arctic winter 2010 require hetero-geneous nucleation of ice, e.g. on meteoritic dust or preexisting NAT particles (Engel, ACP, 13, 10769–10785, 2013).
- MSP and MF analogues demonstrated to promote the nucleation of HNO_3-hydrates accounting for NAT observations of winter pole stratospheric clouds in the absence of water ice.
 (Alexander D. James et al. ACPD, https://doi.org/10.5194/acp-2017-816, 2017).

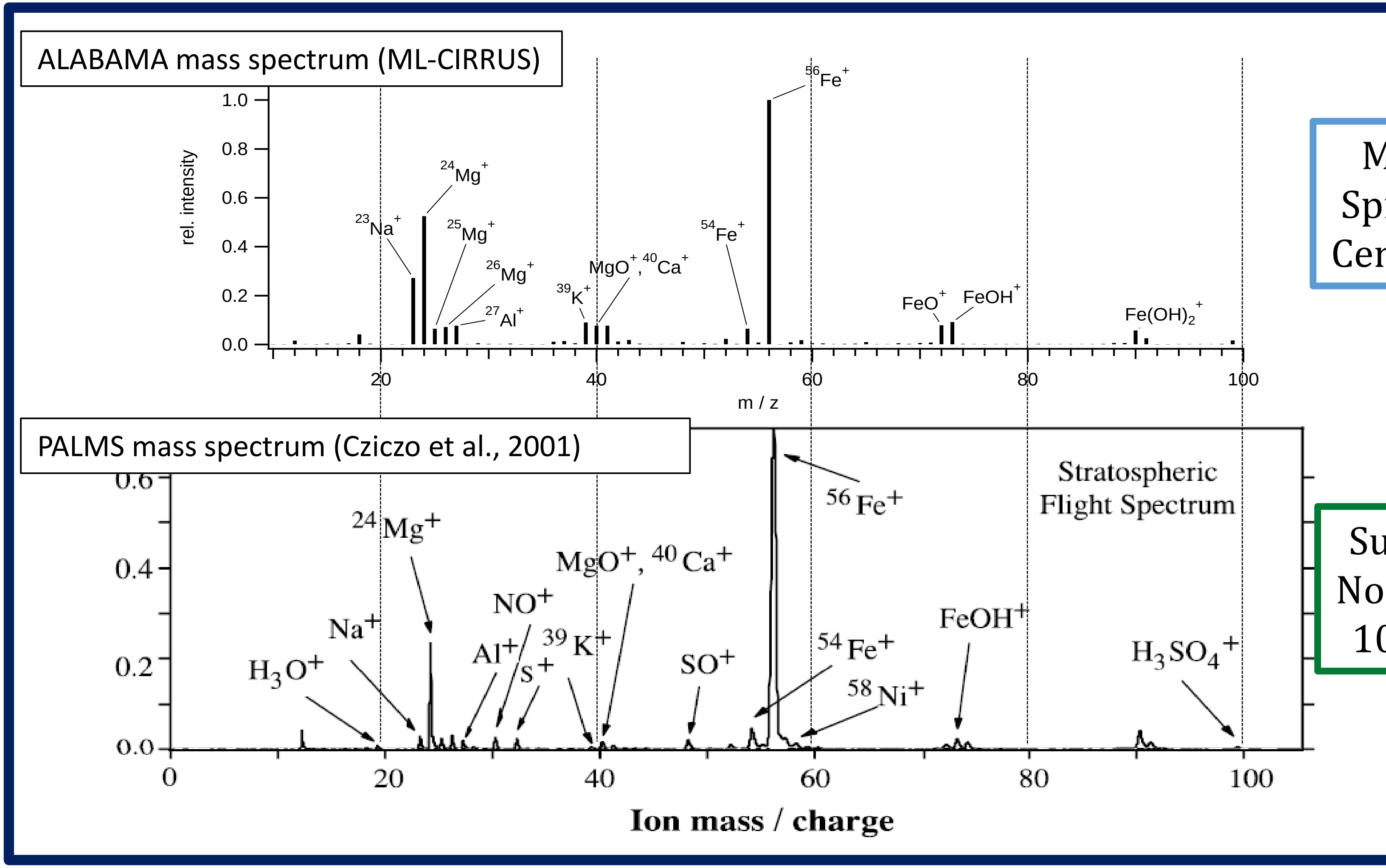
Recent aircraft missions at UT/LS altitudes



in winter (ND-MAX), spring (ML-CIRRUS), and summer (StratoClim I & II) between 20°N to 65°N.

PALMS measurements aboard the NASA WB-57 in summer/fall of the years 1998 and 1999, from 10°N to 45°N at 350 K < Θ <465 K (Murphy et al., Science 1998; Cziczo et al., Science 2001)

Results from Single Particle Ablation Mass-Spectrometry



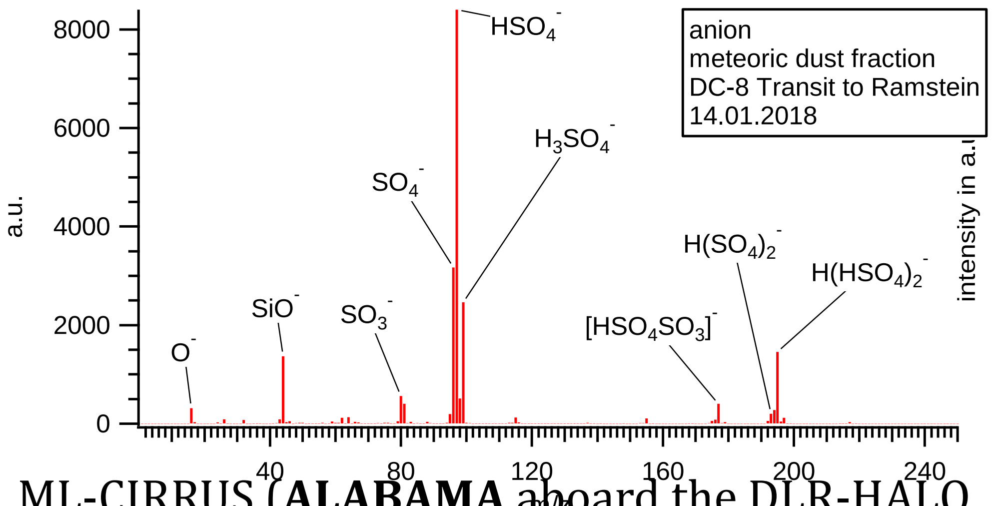
ML-CIRRUS
Spring season
Central Europe

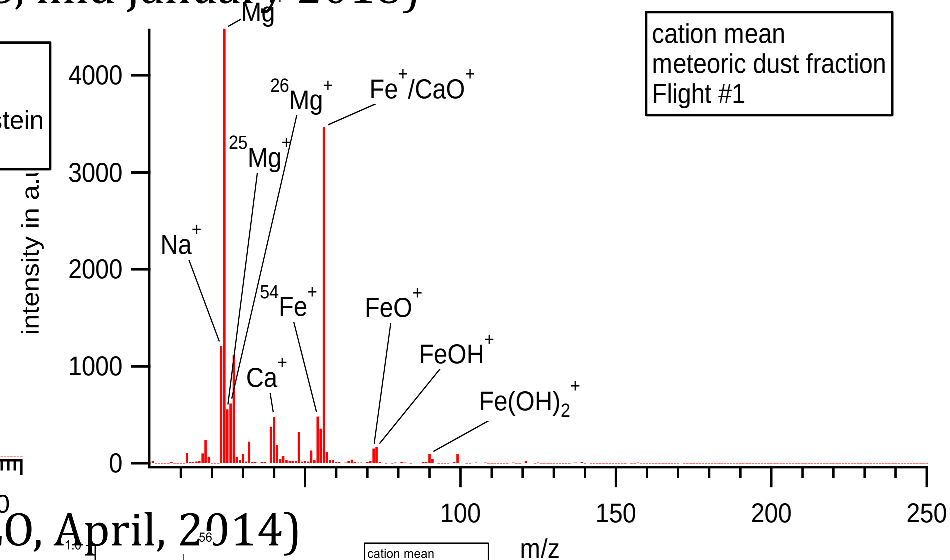
Summer/Fall
North America
10°N – 45°N

- Comparison with previous observations
  (Murphy et al., Science, 1998; Cziczo et al., Science 2001; Murphy et al., Q. J. R. Meteor. Soc.,2014).

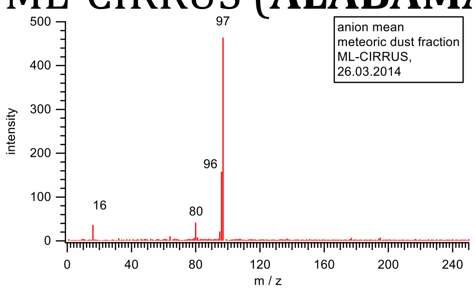
→ Distinct peaks of Mg and Fe dominate the spectra.

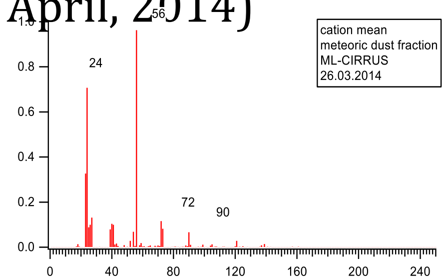
ND-MAX 2018 (**ERICA** aboard the NASA DC-8, mid January 2018)





ML-CIRRUS (**ALABAMA** aboard the DLR-HALO, April, 2014)



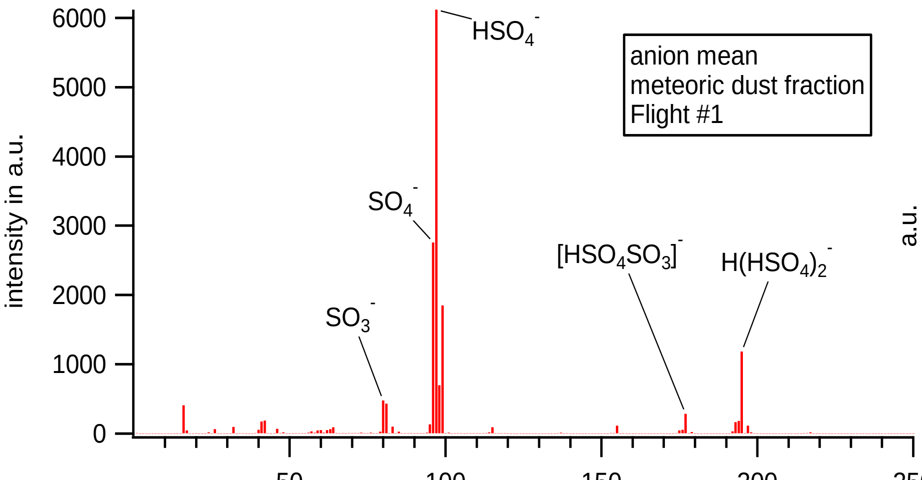


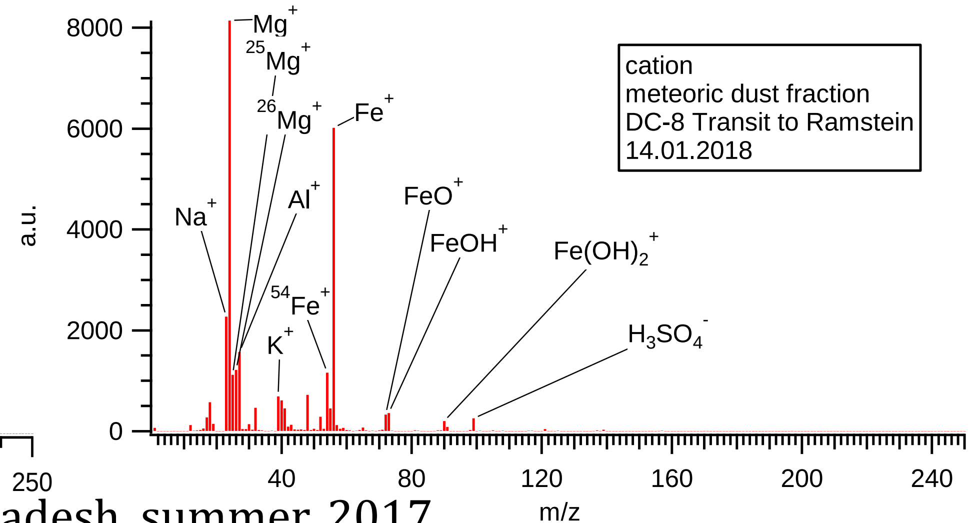
m/z

Single particle mass spectra clustered by similarities: **PART I**

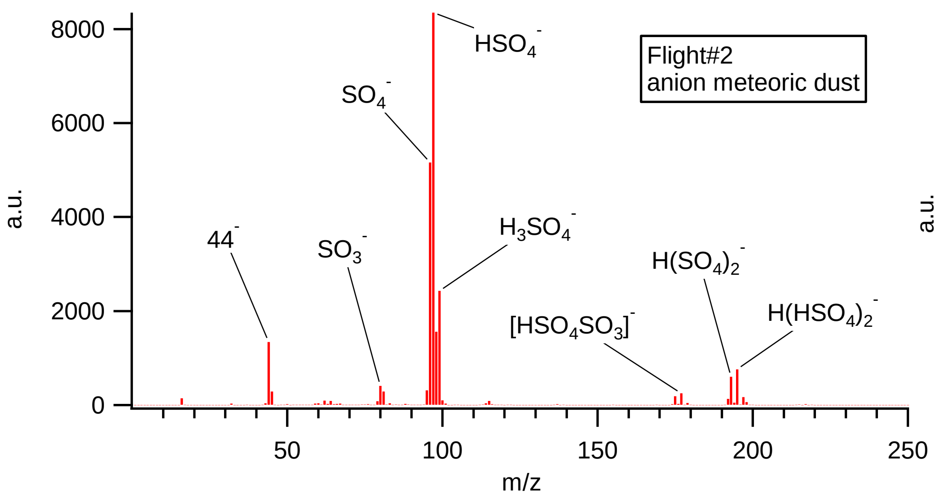
only in the stratosphere: dominating mass peaks of **Mg** (m/z 24 and isotopes at 25 and 26), **Fe** (m/z 56 and 54) as well as **oxides of iron** (FeO, FeOH, m/z 72, 73) and **sulfuric acid** (anions at m/z 96 and 97).

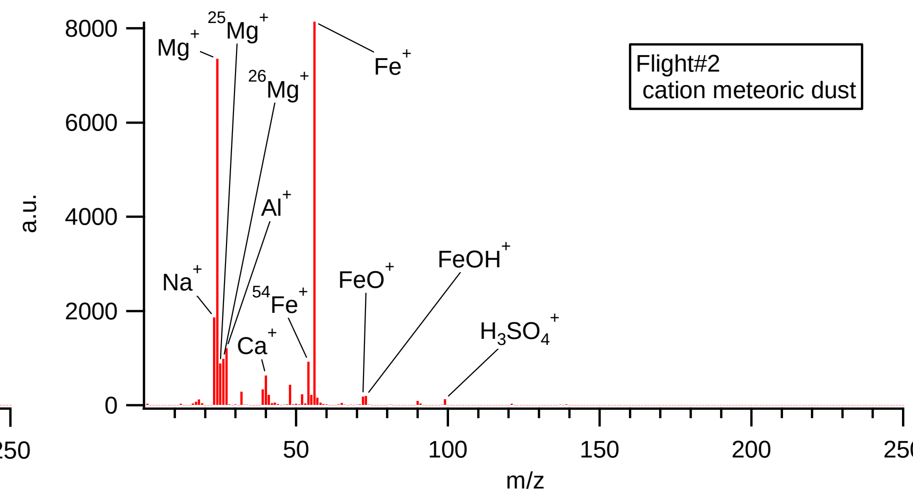
StratoClim-I; **ERICA**, Mediterranean, Aug/Sept, 2016





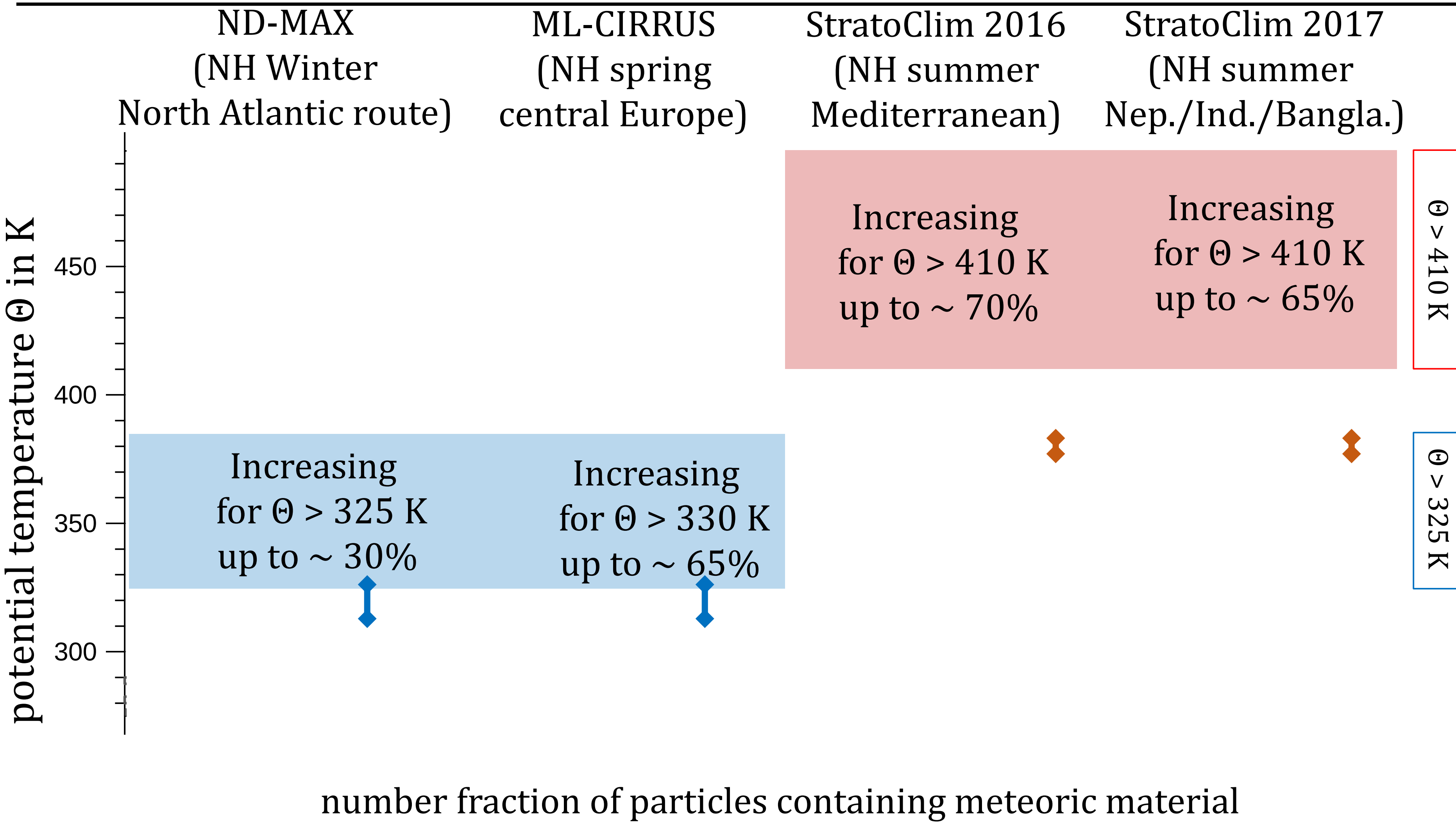
StratoClim-II; **ERICA**, Nepal/India/Bangladesh, summer, 2017





Single particle mass spectra clustered by similarities: **PART II**

only in the stratosphere: dominating mass peaks of **Mg** (m/z 24 and isotopes at 25 and 26), **Fe** (m/z 56 and 54) as well as **oxides of iron** (FeO, FeOH, m/z 72, 73) and **sulfuric acid** (anions at m/z 96 and 97).

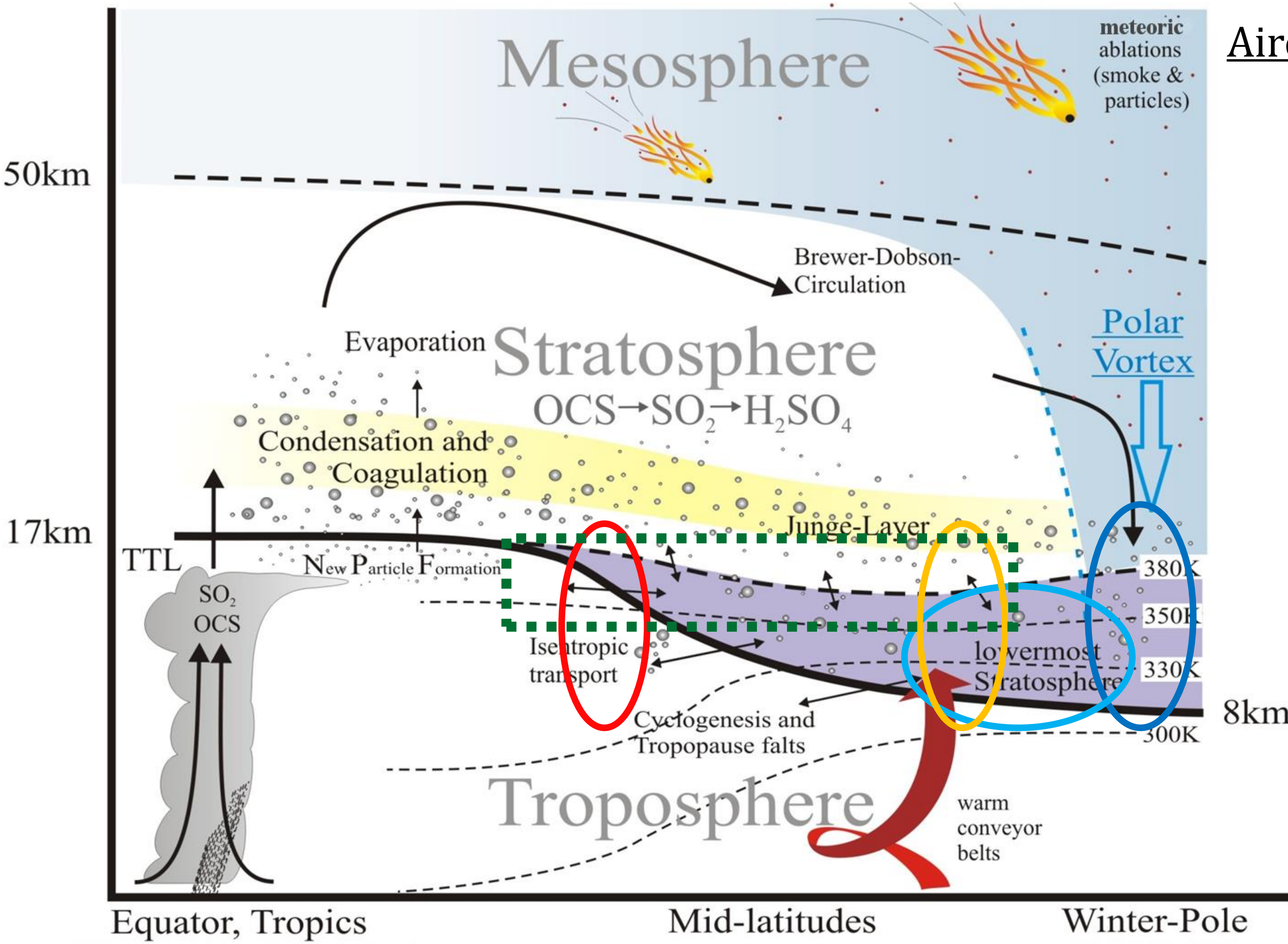


Flight-wise clustering of spectra according to meteoric fingerprints:
During winter/spring at mid-latitude, the tropopause is located at Θ ≈ 310-330 K,
during summer at lower latitudes at Θ ≈ 380 K.

→ Single-flight profiles confirm that this particle type is preferably found in the stratosphere.

→ High abundance in the LS, low abundance in UT: indicative for source higher up in the stratosphere.

Aircraft missions survey



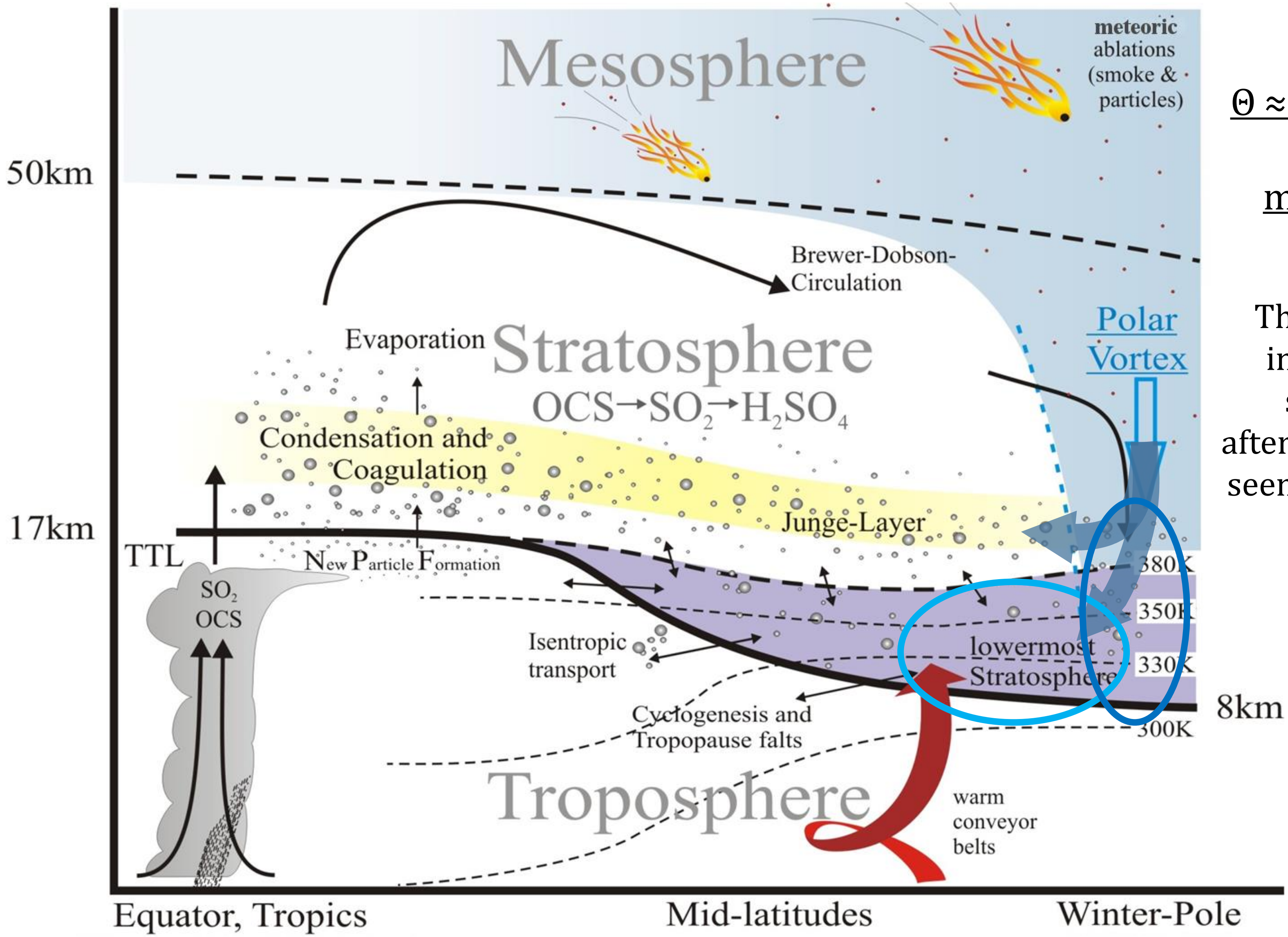
M-55 Geophysica
Θ < 500 K
EUPLEX, 2003
RECONCILE, 2010
ESSENCE, 2011

M-55 Geophysica
NH summer
StratoClim-II, 2017
Θ < 480 K

PALMS on WB-57
NH summer/fall
1998/99, 45°-10°N
350 K < Θ < 465 K

M-55 Geophysica
NH summer
StratoClim-I, 2016
Θ < 490 K

G550 HALO/NASA DC-8
ML-CIRRUS, Apr. 2014, Θ < 385 K
ND-MAX, Jan. 2018, Θ < 340 K

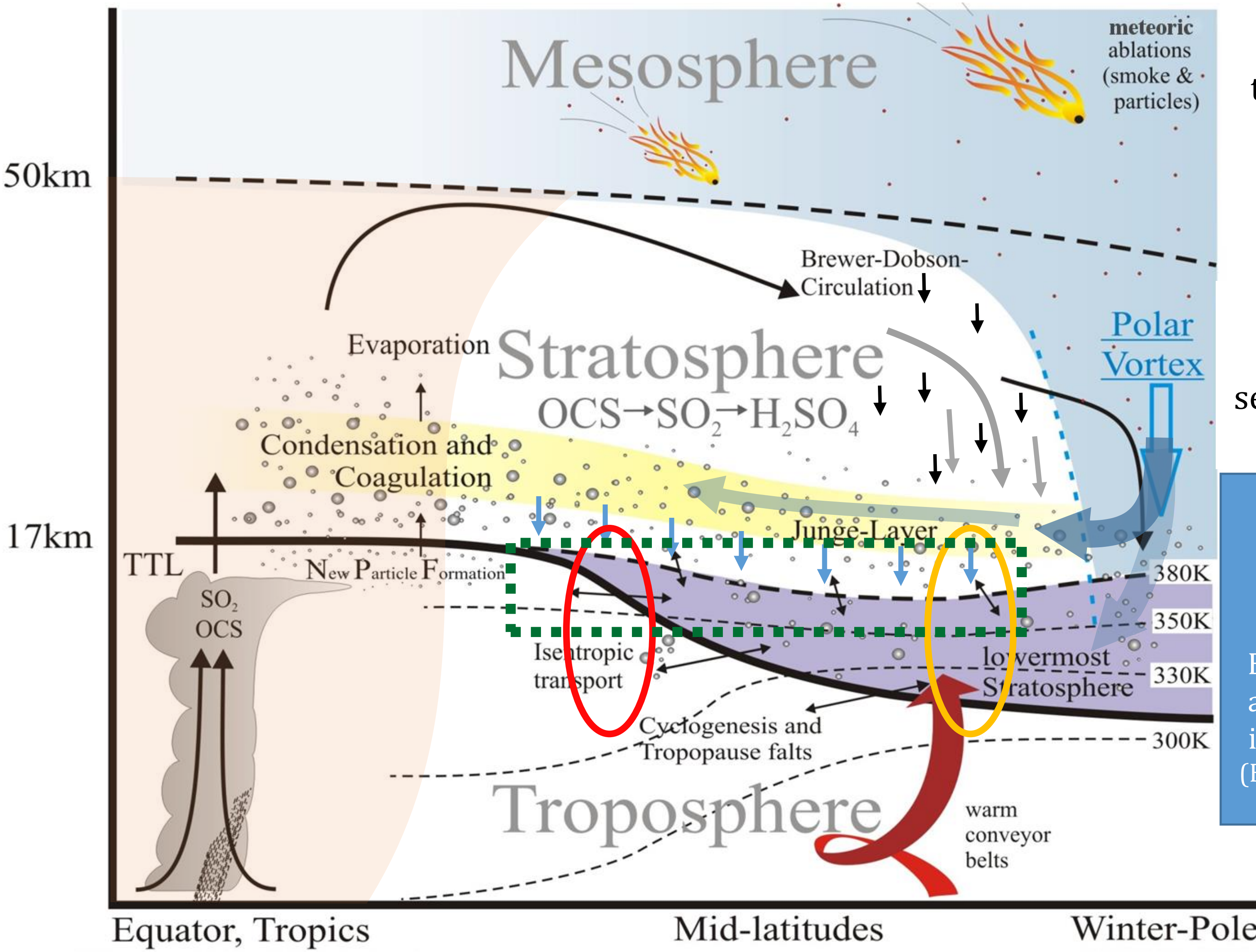


For
$\Theta \approx 325$ - 380 K at high and mid-latitudes:

The entrainment into lowermost stratosphere after vortex break-up seems most relevant

M-55 Geophysica
$\Theta < 500$ K
EUPLEX, 2003
RECONCILE, 2010
ESSENCE, 2011

G550 HALO/NASA DC-8
ML-CIRRUS, Apr. 2014, $\Theta < 385$ K
ND-MAX, Jan. 2018, $\Theta < 340$ K



<u>Towards tropics:</u>

tracers (N_2O) do not indicate recent air mass descent from high altitudes

⬇

subsidence of meteoric particles seems decoupled from air mass

Slow decent of MSPs when incorporated in H_2SO_4 below 35 km
(Neely et al., GRL, 2011)

Eventually promoted by atmospheric circulation in summer hemisphere.
(Birner & Bönisch, ACP 2011, Bönisch et al., ACP, 2011

⬇

<u>sedimentation fall-out from Junge-layer???</u>

<u>M-55 Geophysica</u>
NH summer
StratoClim-II, 2017
$\Theta < 480$ K

<u>PALMS on WB-57</u>
NH summer/fall
1998/99, 45°-10°N
$350 \text{ K} < \Theta < 465$ K

<u>M-55 Geophysica</u>
NH summer
StratoClim-I, 2016
$\Theta < 490$ K

Wrap-up

- Influx of cosmic/interplanetary material is matter of debate.
- The mass contribution of sub-µm diameter sized particles may be negligible, but in number terms ....

Fairly undeniable:

- the potential of sub-µm meteoric aerosol material to promote the mid-atmosphere's cloud formation (noctilucent clouds and polar stratospheric clouds),
- the atmospheric circulation is one of the most efficient mechanisms for removing meteoric material from the middle atmosphere (particularly the vortex-driven subsidence), and,
- additionally, ambling sedimentation together with the summer hemisphere atmospheric circulation may contribute by a slower but continuous removal.

The respective contribution of:

1) **sub-µm IDPs** (→ grains, massive, compact)
2) **MFs**, (→ mostly unblated compact grains)
3) **MSPs** (→ spherules, agglomerated spherules, porous, hollow inclusions)

to the *alien* aerosol population within the Earth's atmosphere remains largely unquantified.

Wrap-up

But, the physical habit of meteoric particles
(porous spherules versus compact grains) may determine:

- their atmospheric lifetime (particle mass to volume ratio → sedimentation speed)
- their nucleation properties and solubility in water, H_2SO_4, HNO_3, (Organics?) may be determined by surface roughness, contact angle, porosity, etc. promoting/diminishing, e.g., condensation/evaporation.

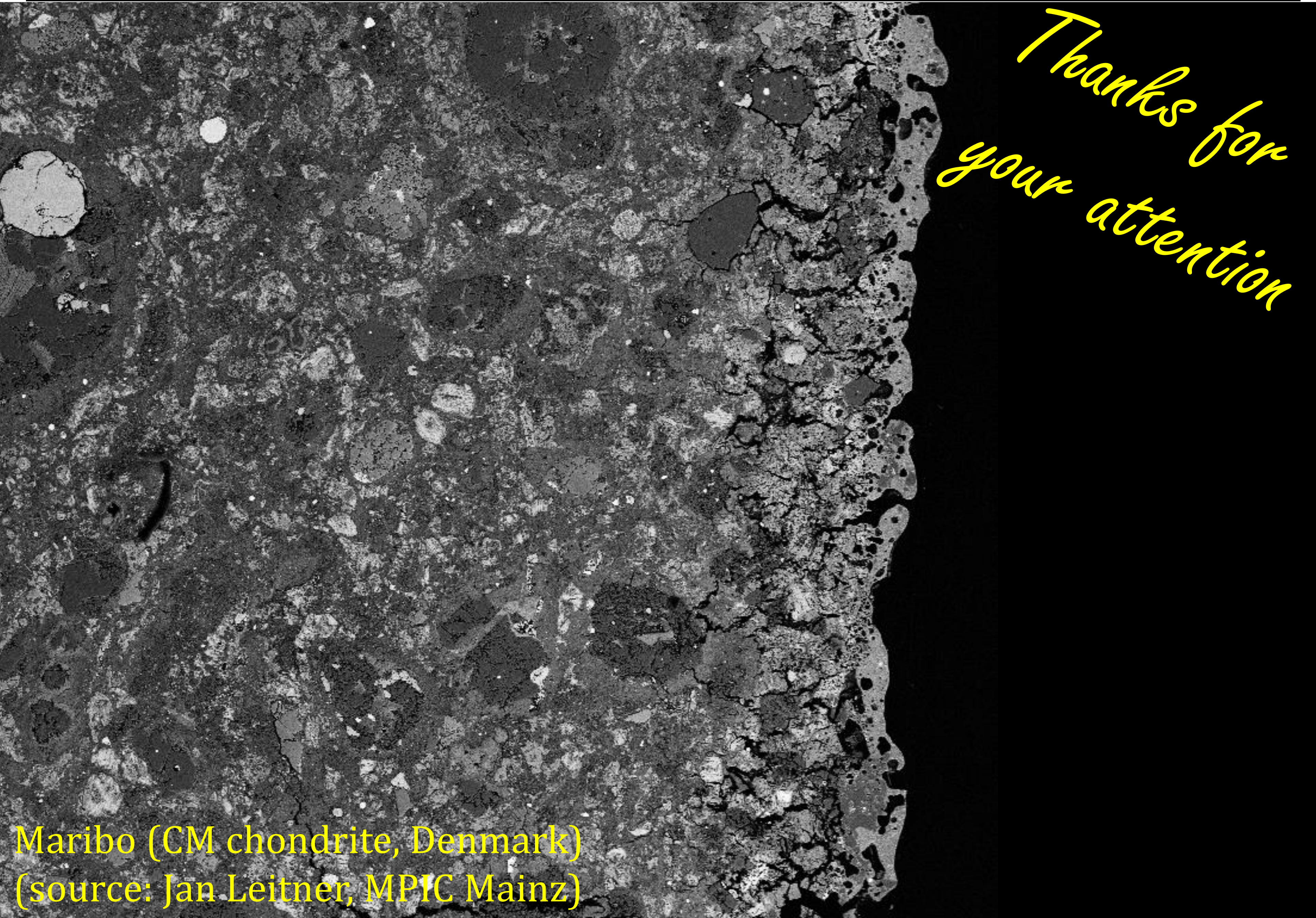
Meteoric sub-µm aerosol seems to be omni-present above the NH tropopause!

- Detected meteoric material is generally associated with H_2SO_4
- At higher latitudes, possibly the remnants of previous vortex-driven subsidence dominate.

- Observations in the LS (summer at mid-latitude and towards the tropics) indicative for a source further aloft (although, e.g., ambundant buoyancy up to 400 K associated with AM-Anti-cyclone should counteract any downwelling)

HYPOTHESIS
As the observed particle subsidence is not associated with recent (massive) air mass transport from above, sedimentation of H_2SO_4-incorporated meteoric particles from the Junge layer („fall-out“) seems most likely.

Thanks for your attention



Maribo (CM chondrite, Denmark)
(source: Jan Leitner, MPIC Mainz)