Links between stratospheric aerosols, polar stratospheric clouds and meteor smoke particles

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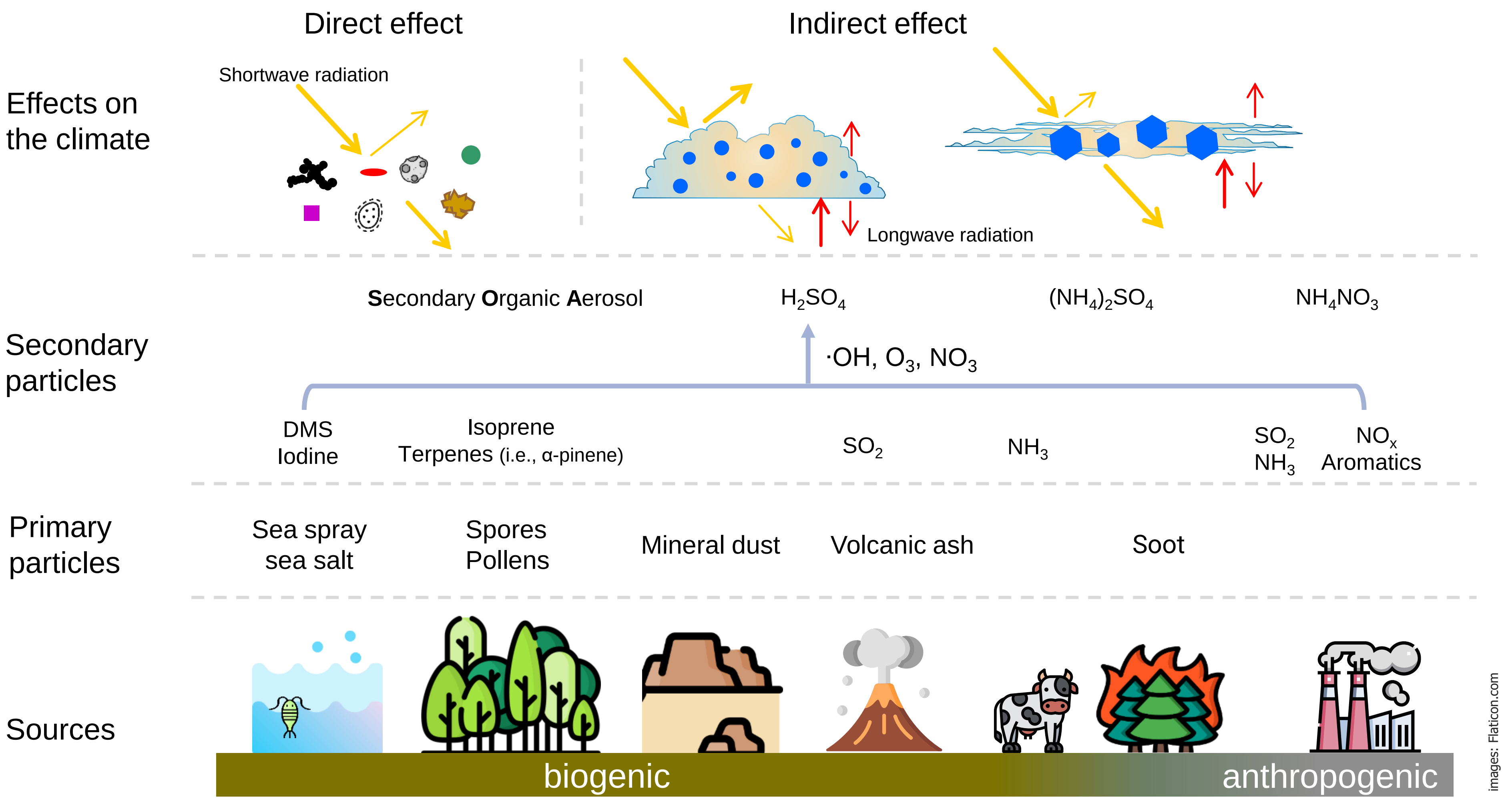
Talk outline

- Introduction to tropospheric and stratospheric aerosols
- The Junge aerosol layer in the stratosphere
- Formation and properties of Polar Stratospheric Clouds (PSCs)
- Detection and potential role of meteoric smoke particles
- Detection of tropospheric aerosol in the stratosphere

Introduction to tropospheric aerosols



Effects on the climate

Direct effect

Shortwave radiation

Indirect effect

Longwave radiation

Secondary particles

Secondary **O**rganic **A**erosol

H_2SO_4

$(NH_4)_2SO_4$

NH_4NO_3

$\cdot OH, O_3, NO_3$

DMS
Iodine

Isoprene
Terpenes (i.e., α-pinene)

SO_2

NH_3

SO_2
NH_3

NO_x
Aromatics

Primary particles

Sea spray
sea salt

Spores
Pollens

Mineral dust

Volcanic ash

Soot

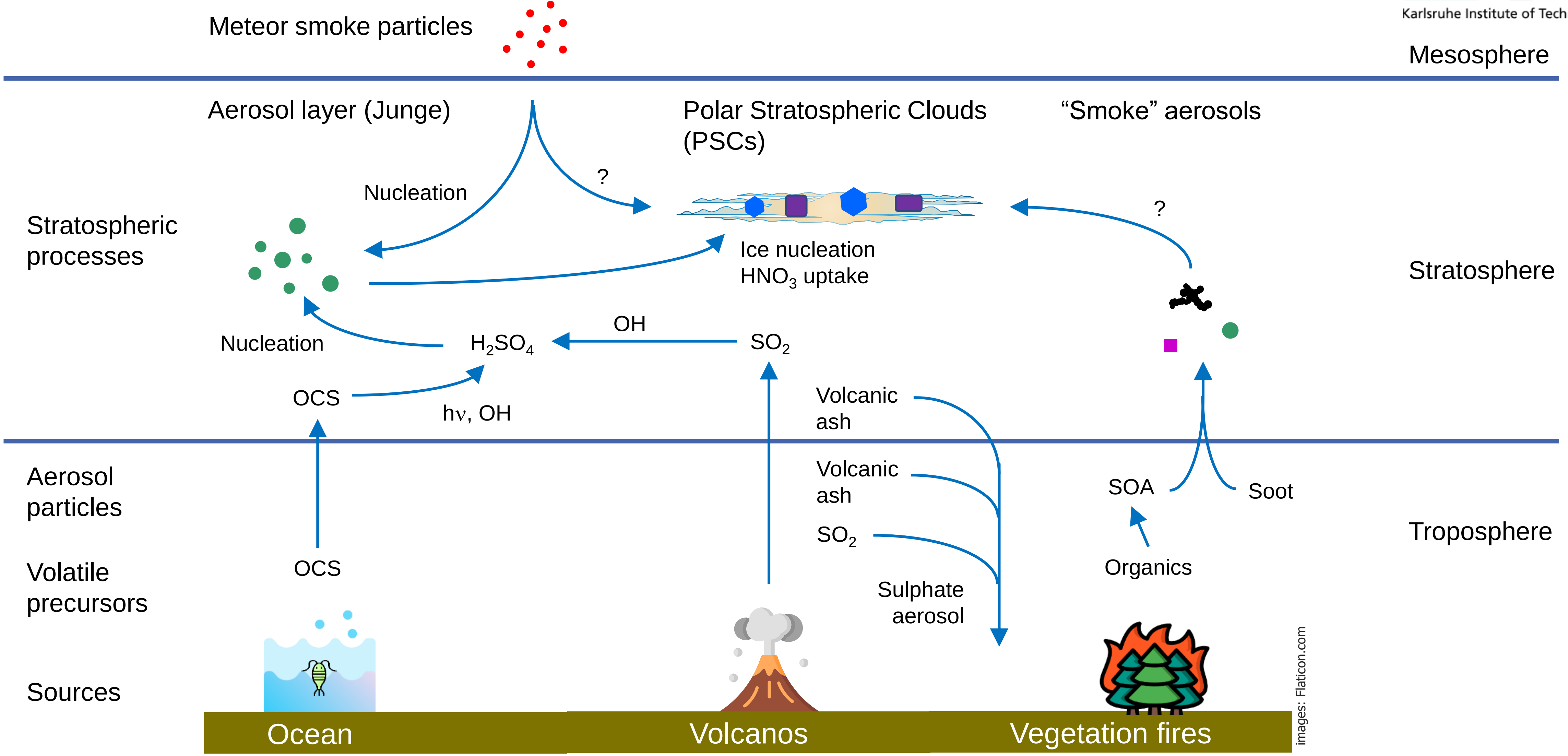
Sources

biogenic

anthropogenic

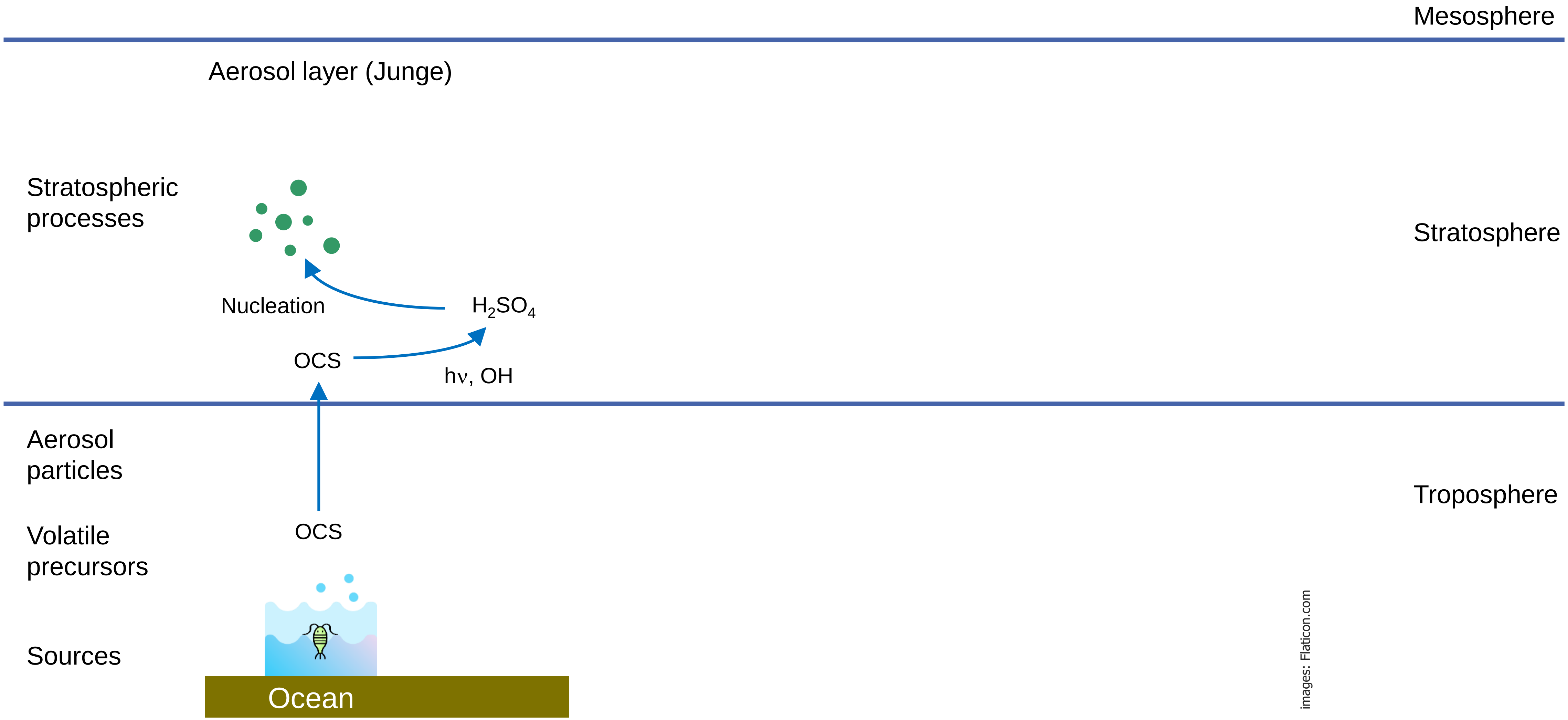
images: Flaticon.com

Introduction to stratospheric aerosols



images: Flaticon.com

The stratospheric sulphuric acid aerosol layer



Mesosphere

Aerosol layer (Junge)

Stratospheric processes

Stratosphere

Nucleation

H_2SO_4

OCS

hν, OH

Aerosol particles

Troposphere

Volatile precursors

OCS

Sources

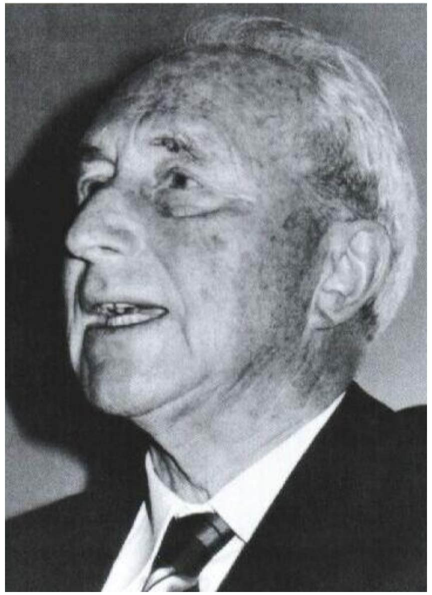
Ocean

images: Flaticon.com

The discovery of the stratospheric aerosol layer

Christian Junge (1912-1996)

1968 to 1979 Director of the newly established Department of Atmospheric Chemistry and
Physical Chemistry of Isotopes at the
Max-Planck-Institute for Chemistry in Mainz.



Science, 133, 1478-1479, 1961

A World-wide Stratospheric Aerosol Layer

Christian E. Junge, Charles W. Chagnon, and James E. Manson
Atmospheric Circulations Laboratory, Air Force Cambridge Research Laboratories, Bedford, Massachusetts

An aerosol layer has been identified by a stratospheric balloon and aircraft aerosol collection program. Measurements of horizontal extension and vertical distribution indicate that this layer is a world-wide phenomenon, displaying little variation with time and latitude. The particles in this layer range in size between 0.1 and 2 µm in radius, are water soluble, and contain sulfur as the predominant constituent. It is most likely that they are formed within the stratosphere.

Expansion chamber for measuring stratospheric aerosols

Junge, Cagnon, Manson, J. Meteorology, 18, 81, 1961.

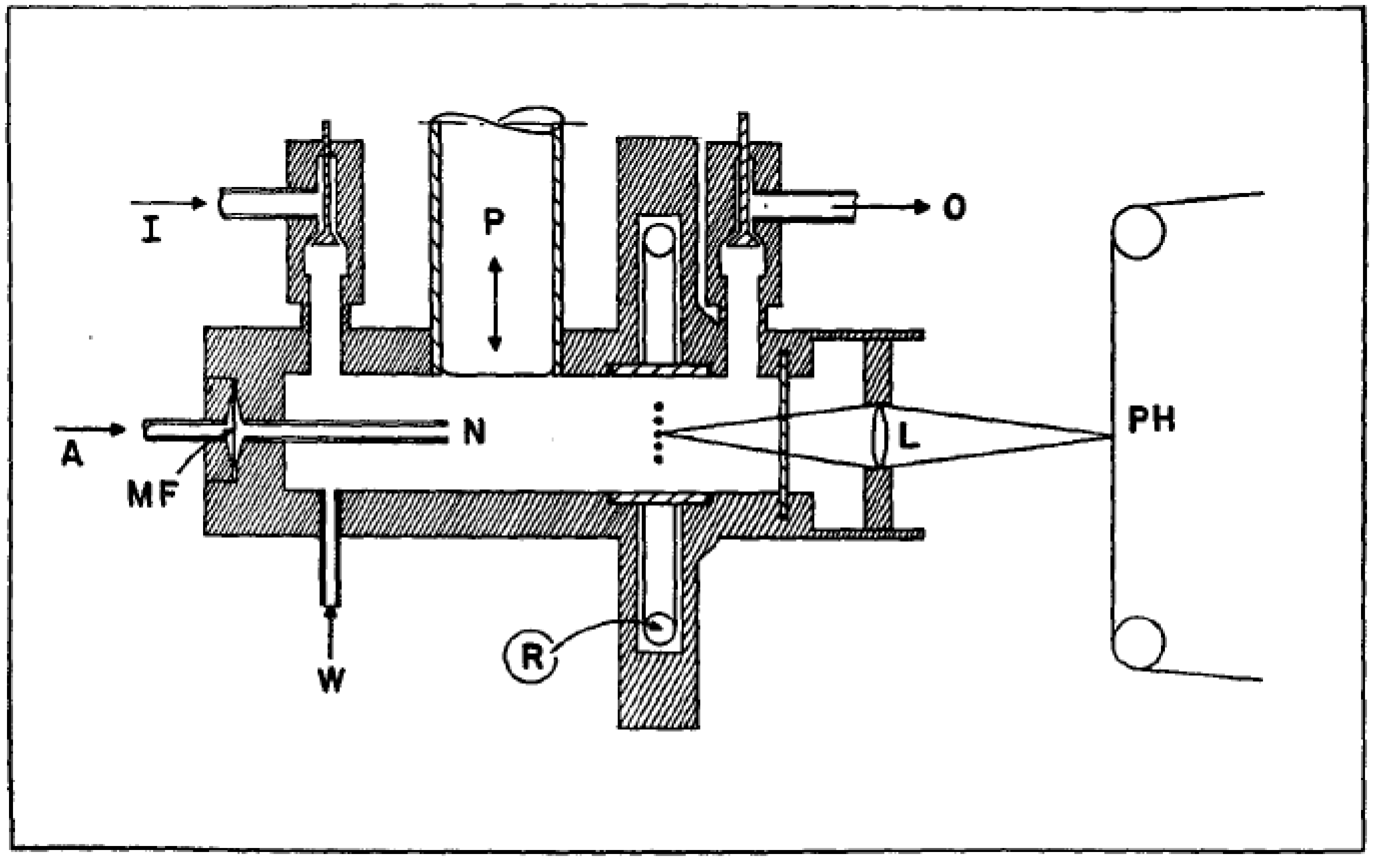


FIG. 8. Schematic diagram of the automatic recording Aitken Nuclei Counter. *I* = inlet tube for sample. *O* = outlet tube to pump. *P* = expansion piston. *A* = inlet for air injection. *MF* = millipore filter. *N* = nozzle to facilitate good mixing in chamber. *R* = ring flashlight. *L* = lens. *PH* = photographic film. *W* = water injector.

Latest development: Mobile and automated expansion-type cloud chamber PINE for measuring ice nucleation and Ice-Nucleating Particles (Möhler et al., AMT, 2021)

https://www.noell.bilfinger.com/pine/#c167514
https://www.imk-aaf.kit.edu/PINE_INP_monitoring.php

First height profiles of stratospheric aerosols (Junge et al., 1961)

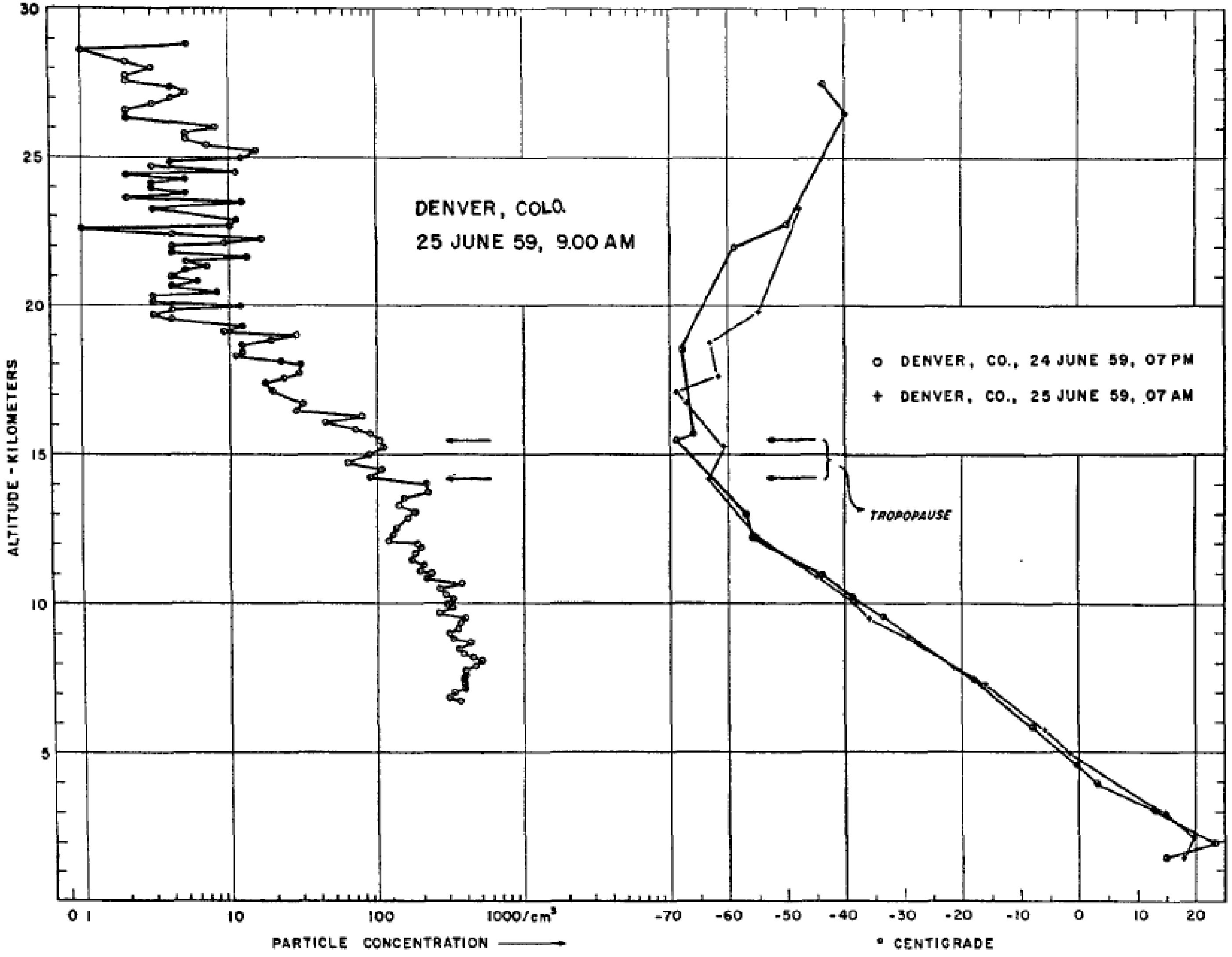
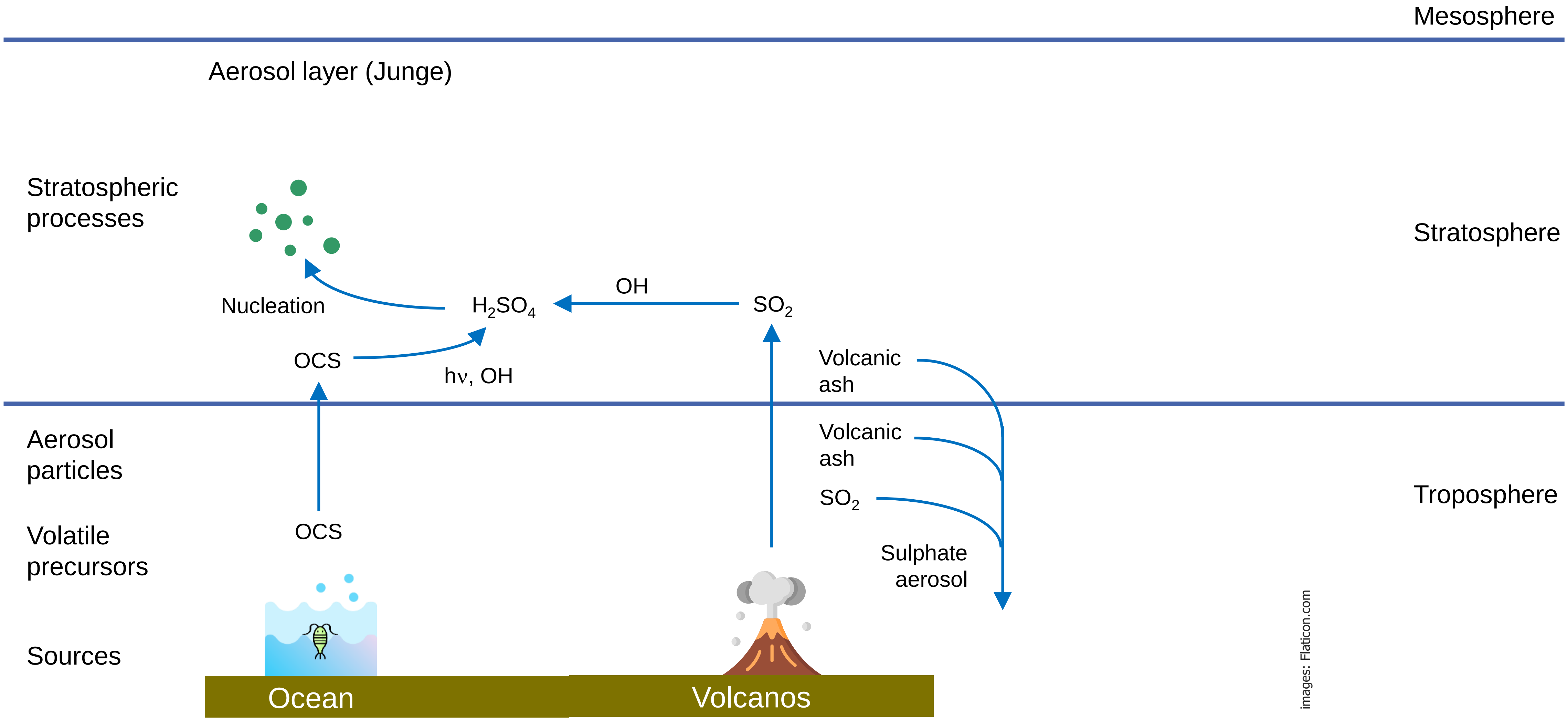


Fig. 10. Measured vertical profiles of Aitken nuclei and the available temperature soundings closest in space and time.

Volcanic aerosols



Mesosphere

Aerosol layer (Junge)

Stratospheric processes

Stratosphere

Nucleation

H_2SO_4

OH

SO_2

OCS

$h\nu$, OH

Volcanic ash

Aerosol particles

Volcanic ash

SO_2

Troposphere

Volatile precursors

OCS

Sulphate aerosol

Sources

Ocean

Volcanos

images: Flaticon.com

Volcanic aerosols: Major eruptions

Direct emission of SO_2 into the stratosphere

→ Conversion to H_2SO_4

→ Nucleation/condensation to aerosol particles

→ Periodoc enhanmcement of aerosol volume

→ Enhanced light scattering and radiation cooling

$SO_2 + OH + M \rightarrow HSO_3 + M$

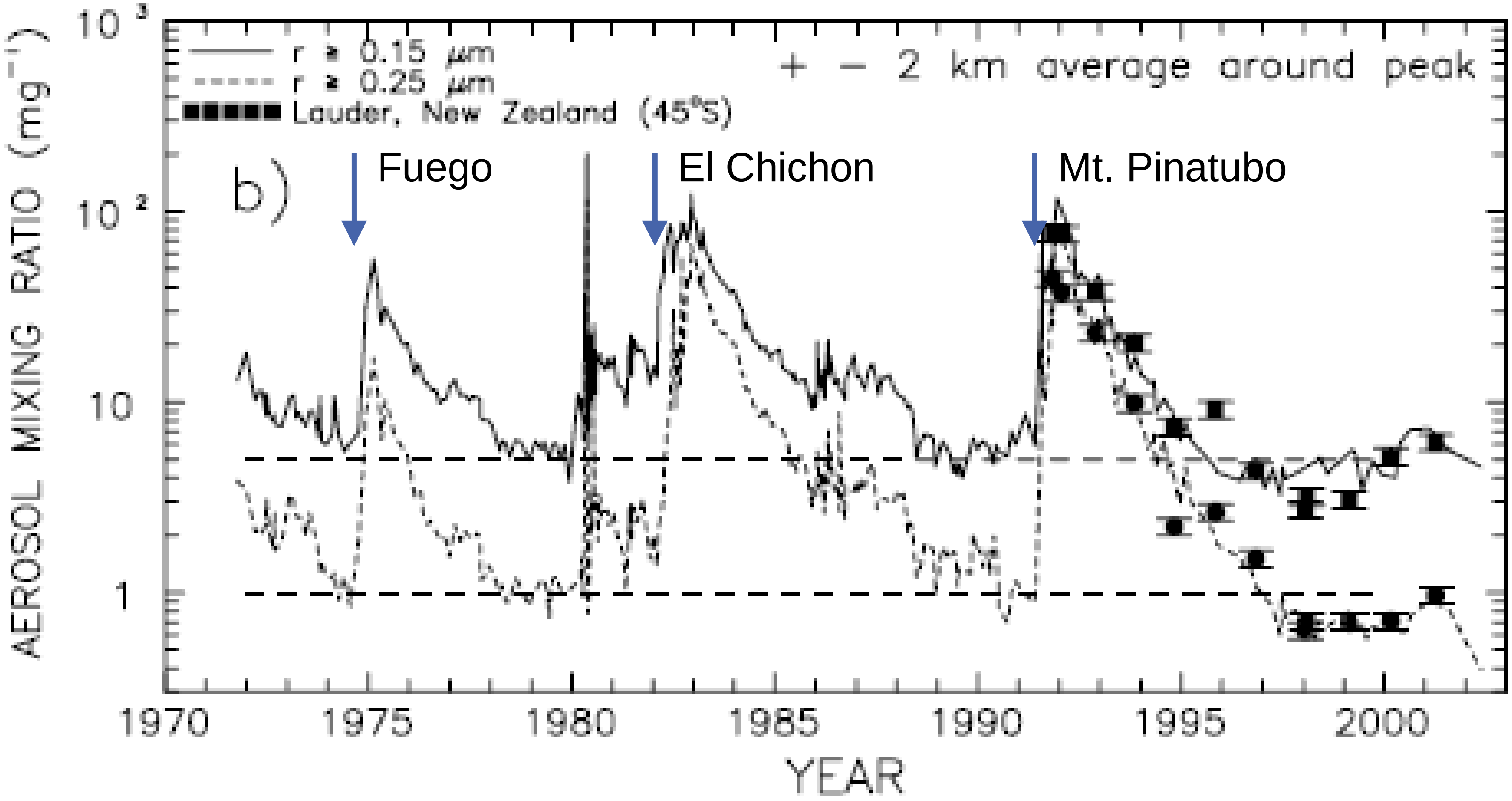
$HSO_3 + O_2 \rightarrow SO_3 + HO_2$

$SO_3 + H_2O + M \rightarrow H_2SO_4 + M \rightarrow \text{Aerosol}$

Volcano	Eruption date	Stratospheric aerosol impact (Tg)
Background	1979	< 1
Katmai	June 1912	20
Agung	March 1963	16-30
Fuego	October 1974	3-6
El Chichon	April 1982	12
Mt. Pinatubo	June 1991	30
Cerro Hudson	August 1991	3

Volcanic aerosols: Impact on stratospheric aerosols

Balloon borne OPC (Optical Particle Counter) measurements over Laramie, Wyoming (lines) and New Zeeland (symbols)



Deshler et al., 2003

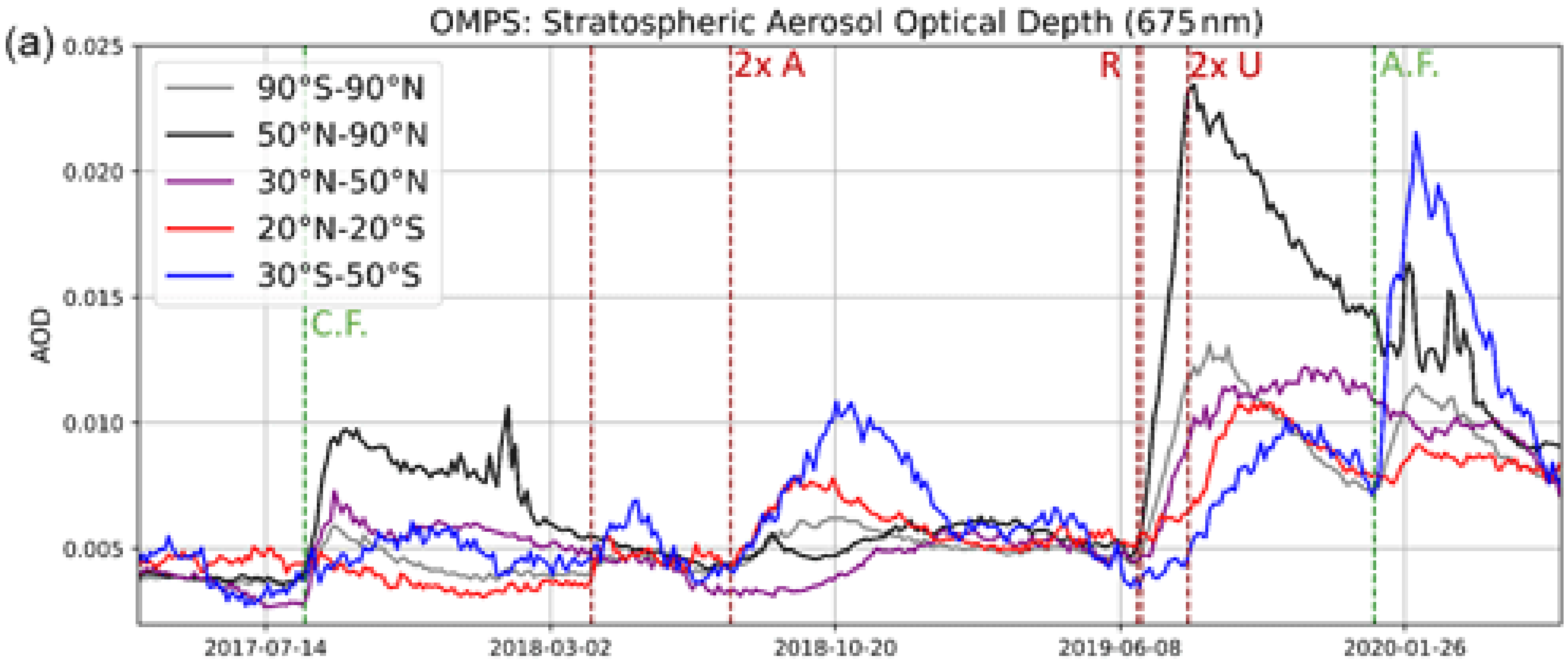
Volcanic aerosols: Impact on stratospheric aerosols

Enhanced stratospheric aerosol optical depth (from OMPS aerosol extinction) after

- ➢ Raikoke eruption, June 2019
- ➢ Ulawun eruptions June and August, 2019

Signatures from fire plumes are also visible:

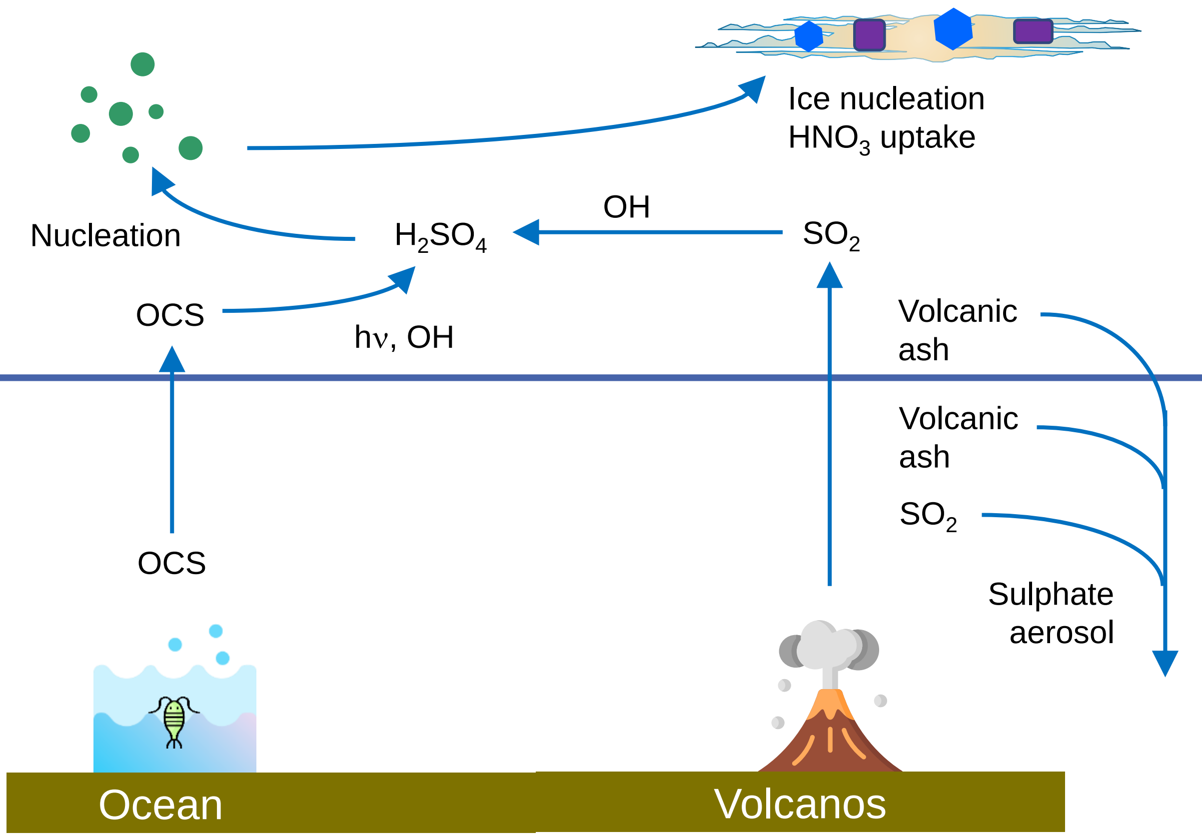
- ➢ Canadian fires 2017
- ➢ Australian fires 2020



Kloss et al., ACP, 2021

Polar Stratospheric Clouds (PSCs)

Mesosphere



Aerosol layer (Junge)

Polar Stratospheric Clouds (PSCs)

Stratospheric processes

Ice nucleation
HNO_3 uptake

Nucleation

H_2SO_4

OH

SO_2

OCS

hν, OH

Volcanic ash

Aerosol particles

Volcanic ash

SO_2

Volatile precursors

OCS

Sulphate aerosol

Sources

Ocean

Volcanos

Mark R. Schoeberl, NASA GSFC

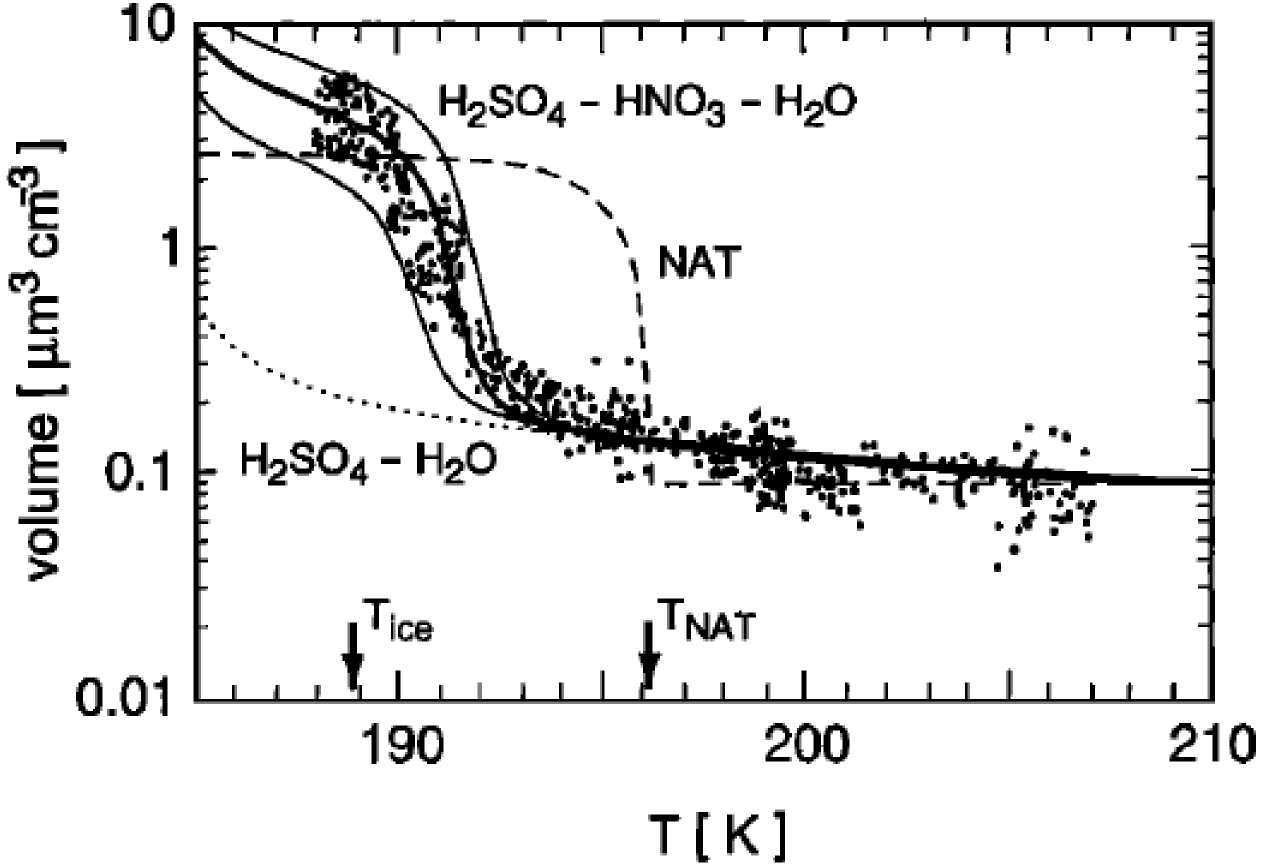
Stratosphere

Troposphere

images: Flaticon.com

PSC: Formation of ternary solutiom particles (STS)

STS = Stratospheric Ternary Solution particles



Carslaw et al., Modeling the composition of liquid stratospheric aerosols, Rev. Geophys., 35, 125-154, 1997.

Figure 1. Particle volume measurements of *Dye et al.* [1992] (January 24, 1989) compared with model calculations for the growth of different types of stratospheric particles [*Carslaw et al.*, 1994]: dotted line, liquid H_2SO_4-H_2O droplets; dashed line, nitric acid trihydrate (NAT; $HNO_3 \cdot 3H_2O$) particles [*Hanson and Mauersberger*, 1988]; solid line, HNO_3-H_2SO_4-H_2O droplets. Calculated volumes are for 55 mbar altitude, with 5 ppmv H_2O and 10 ppbv total HNO_3 (thin solid lines correspond to 5 and 15 ppbv HNO_3). Further details of the calculation are given by *Carslaw et al.* [1994]. Similar calculations have been made by *Drdla et al.* [1994] and *Tabazadeh et al.* [1994a].

PSC – MIPAS result (from EU project POSTA)

MIPAS-B on CNES balloon flight along edge of polar vortex (unpublished result)

January 11, 2001

Courtesy of Hermann Oehlhaf, IMK-ASF

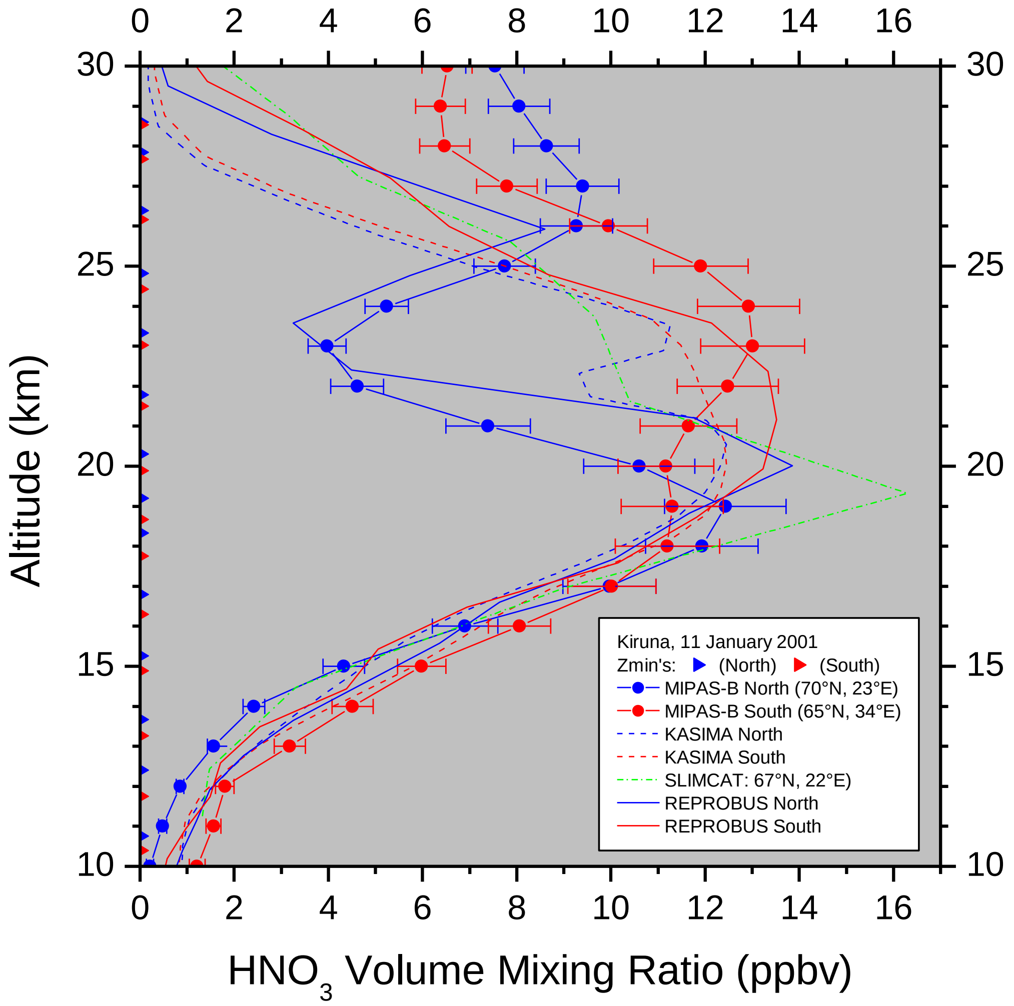
PSC – MIPAS result (from EU project POSTA)

MIPAS-B on CNES balloon flight
along edge of polar vortex
(unpublished result)

January 11, 2001

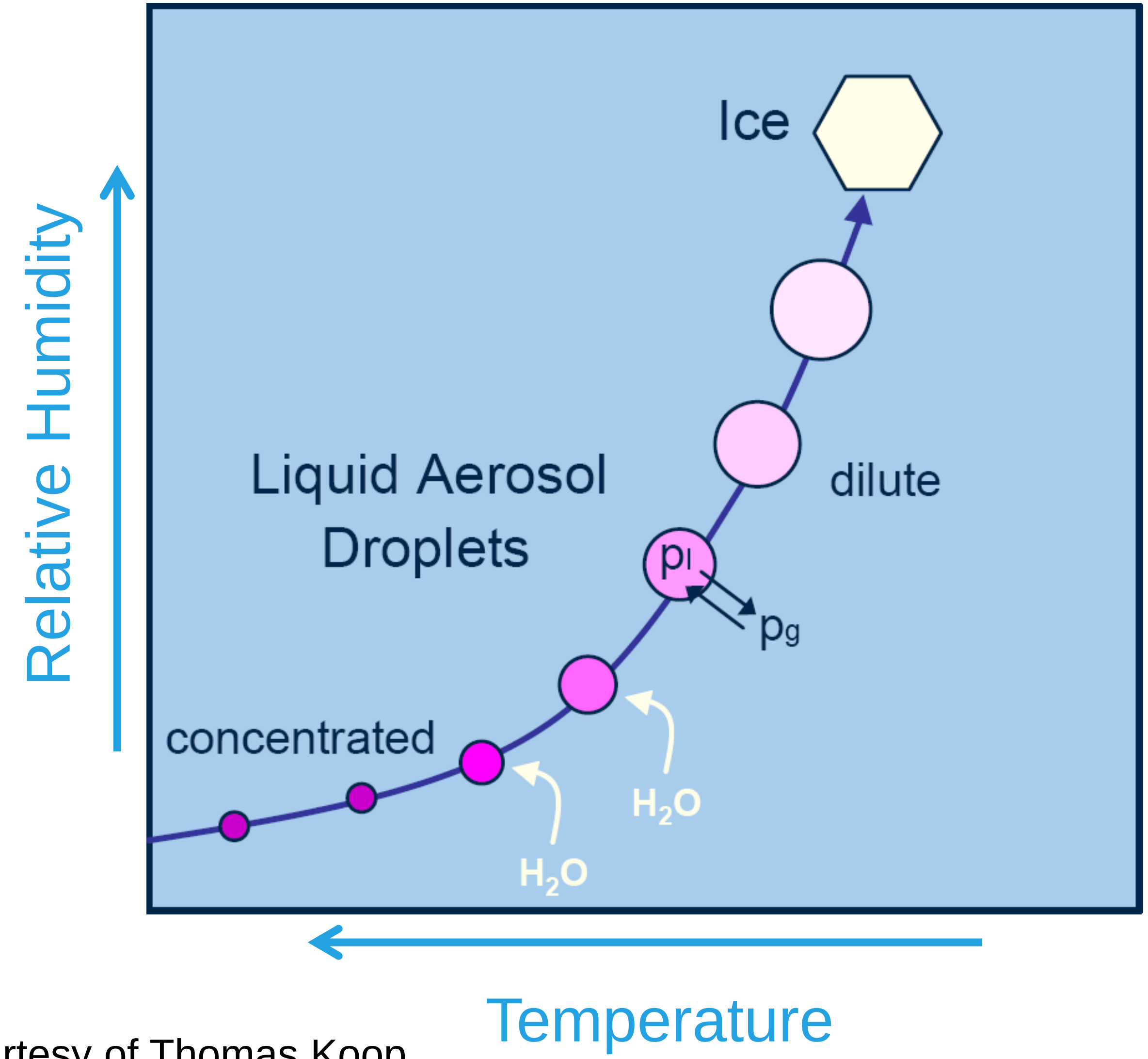
Various HNO3 height profiles from measurements (symbols) and
models (solid and dashed lines)

- Red: outside vortex → regular profile
- Blue: inside vortex → depleted by uptake into STS



Courtesy of Hermann Oehlhaf, IMK-ASF

PSC: Ice formation



Courtesy of Thomas Koop

<u>Increase of relative humidity (e.g. by updrafts in lee waves)</u>

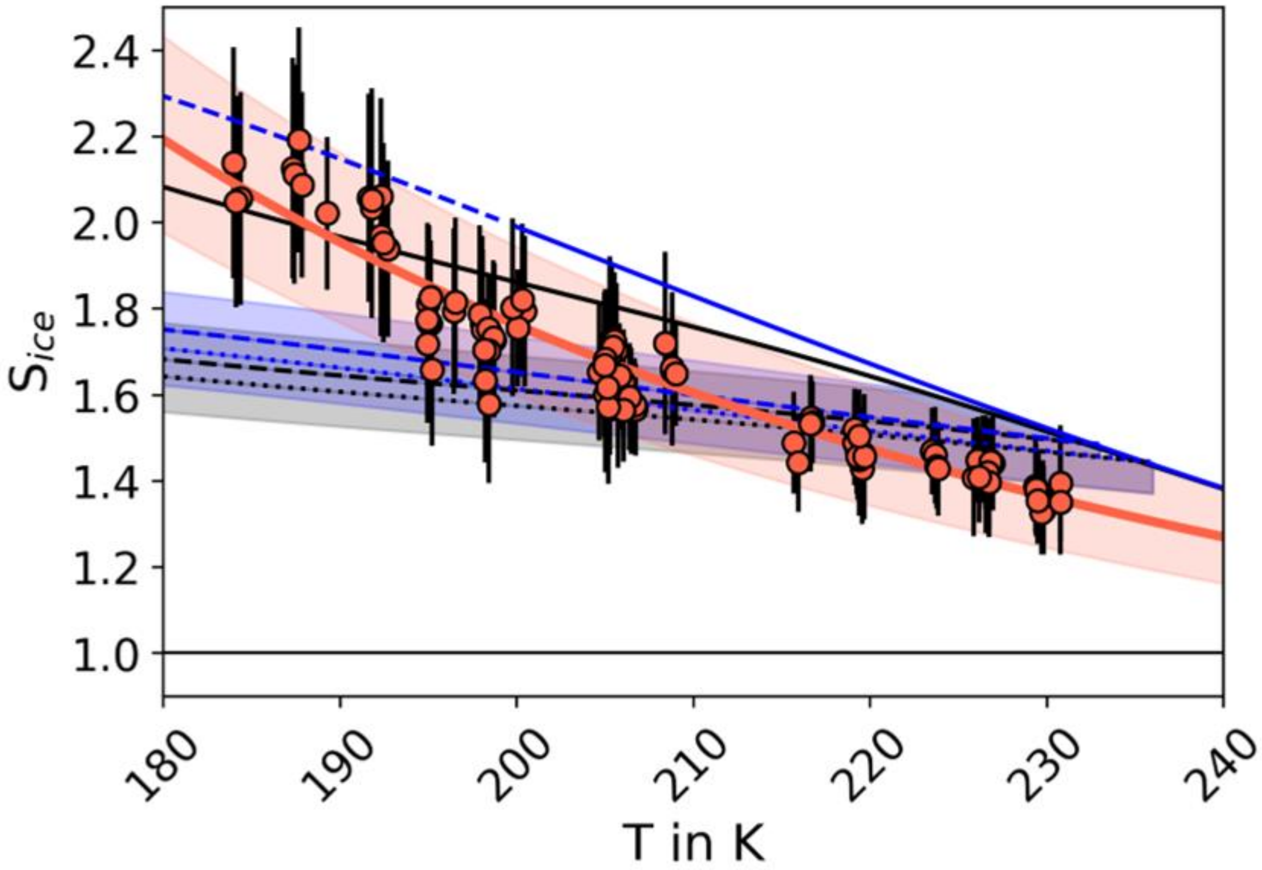
→ Water uptake by aqueous aerosol particles (Rault`s law)

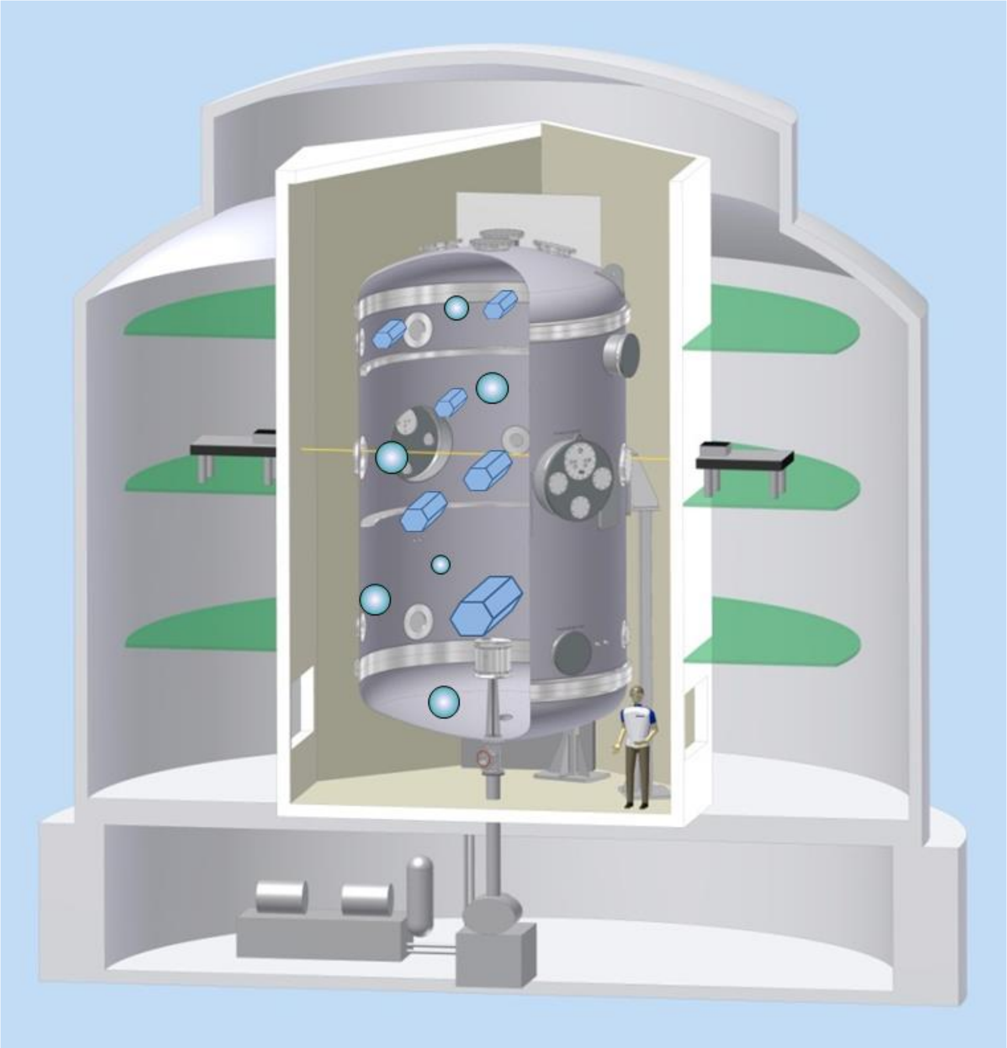
→ Dilution of solutes

→ Homogeneous freezing (Koop et al., 2000)

PSC – ice crytals: AIDA cloud chamber experiments

Homogeneous freezing insets measured in 63 AIDA experiments in the years 2007 to 2020

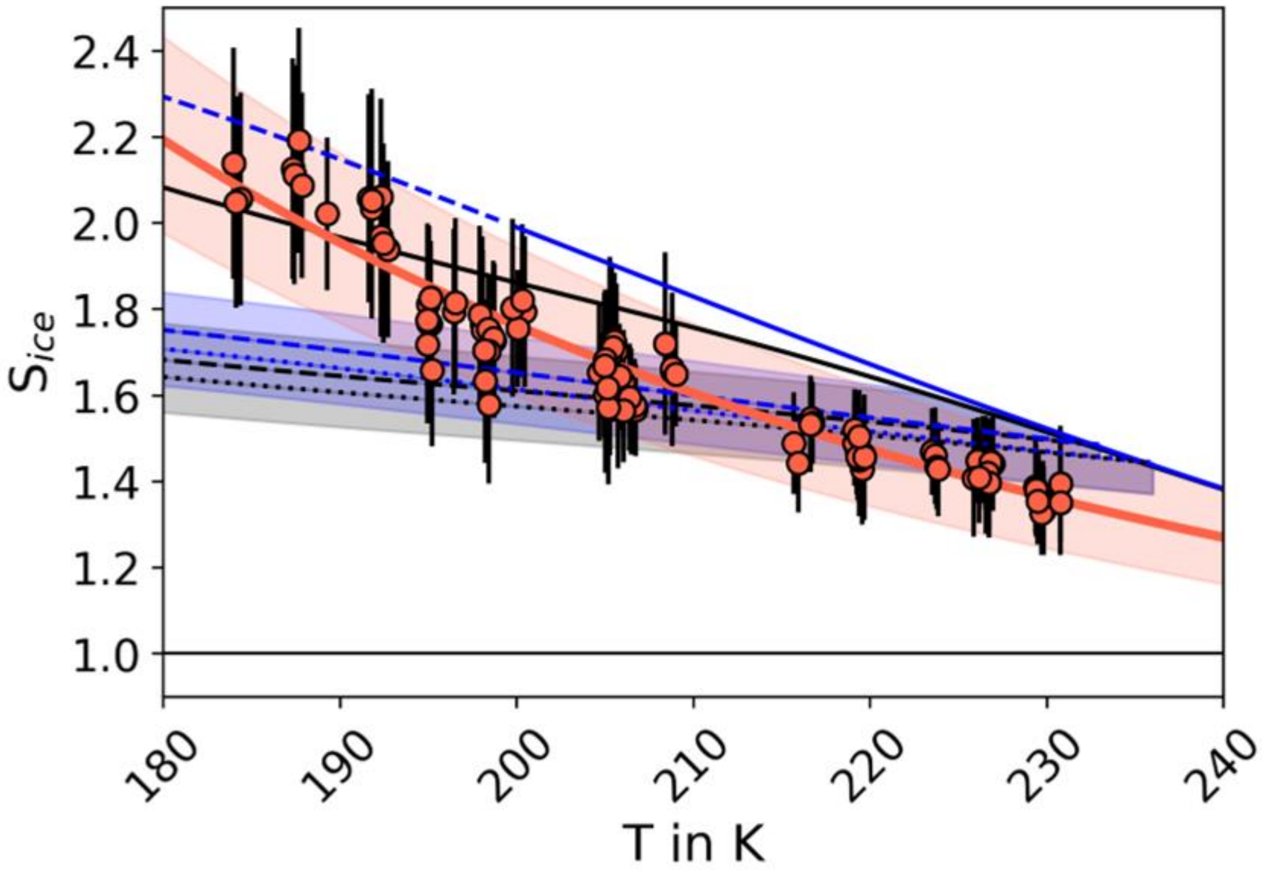




Schneider et al., ACP, 2021 (see also Möhler et al., ACP, 2003)

PSC – ice crytals: AIDA cloud chamber experiments

Homogeneous freezing insets measured in 63 AIDA experiments in the years 2007 to 2020



At low T (< 200 K), measured freezing onsets clearly deviate from line predicted by Koop et al., 2000

→ New fit line for homogeneous freezing onset of aqueous sulfuric acid aerosol:

$$\boxed{lnS_{ice} = a + b\,T^{-1};\ \ a = -1.4 \pm 0.05, b = 390 \pm 10\ K}$$

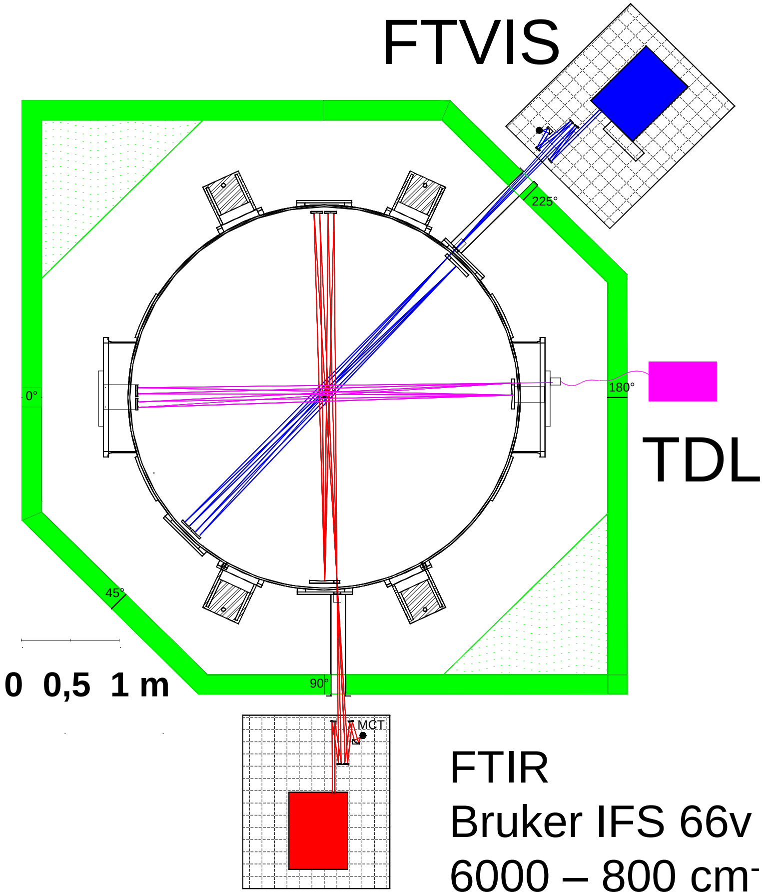
Possible explanation: Saturation for supercooled water higher than formulated by e.g. Murphy and Koop, 2005

Schneider et al., ACP, 2021 (see also Möhler et al., ACP, 2003)

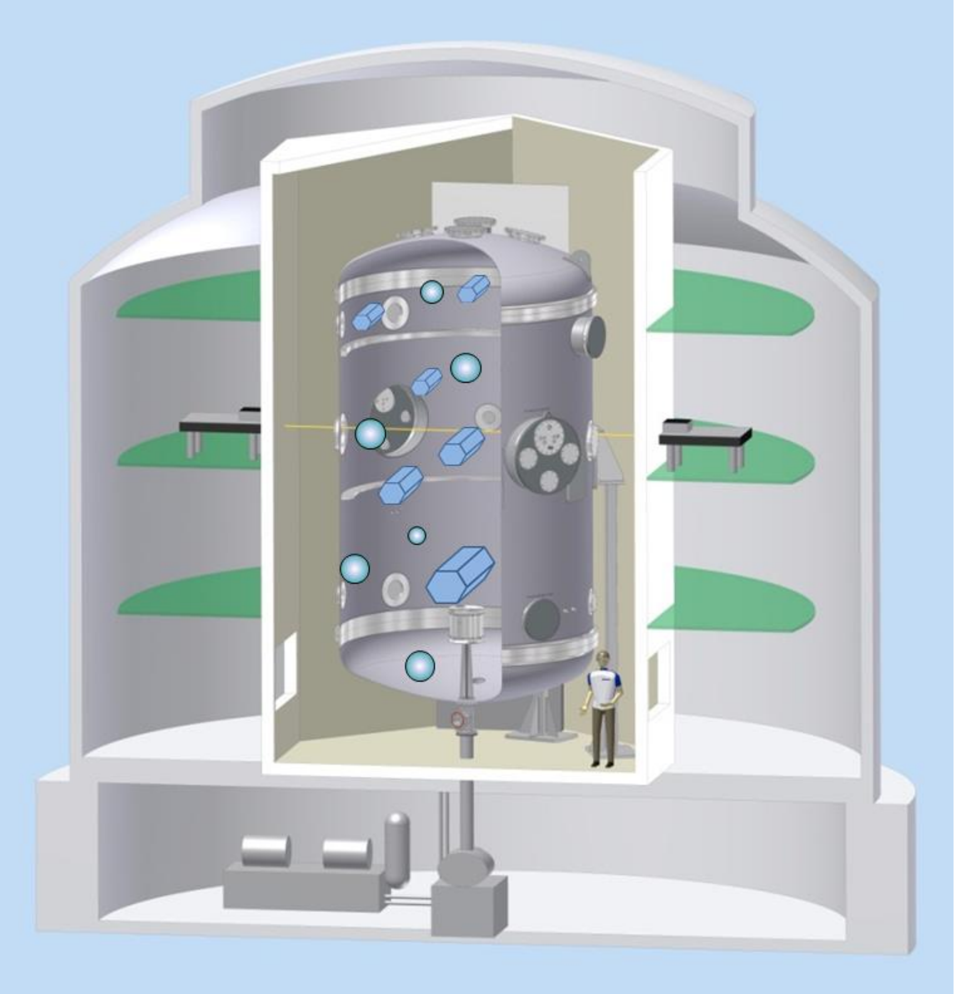
PSC – NAD: AIDA cloud chamber experiments

in situ optical measurements

AIDA = Aerosol Interactions and Dynamics in the Atmosphere

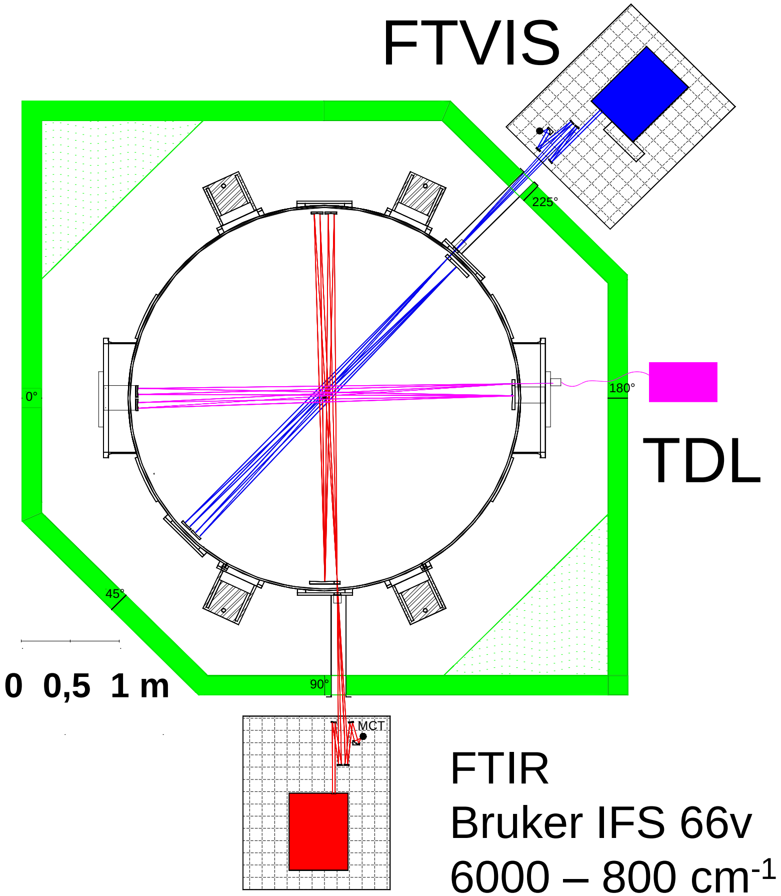


FTIR
Bruker IFS 66v
6000 – 800 cm^{-1}
0,1 cm^{-1} resolution



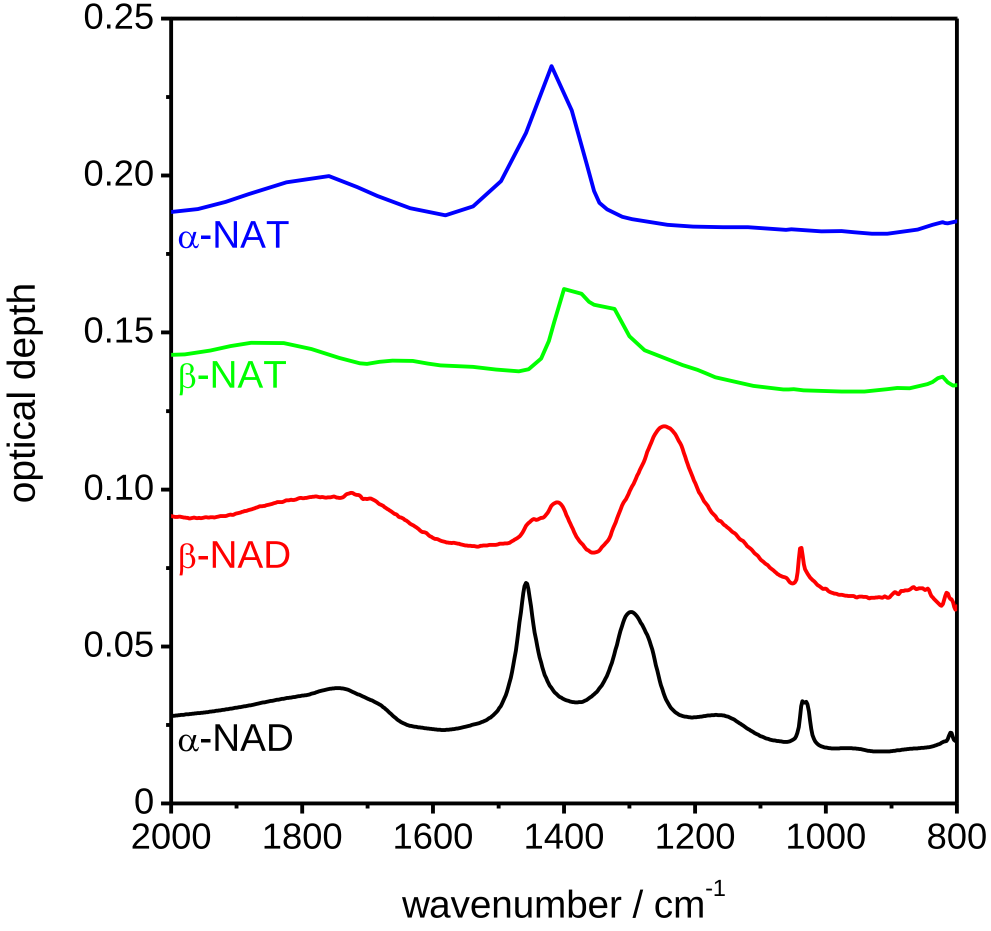
PSC – NAD: AIDA cloud chamber experiments

in situ optical measurements



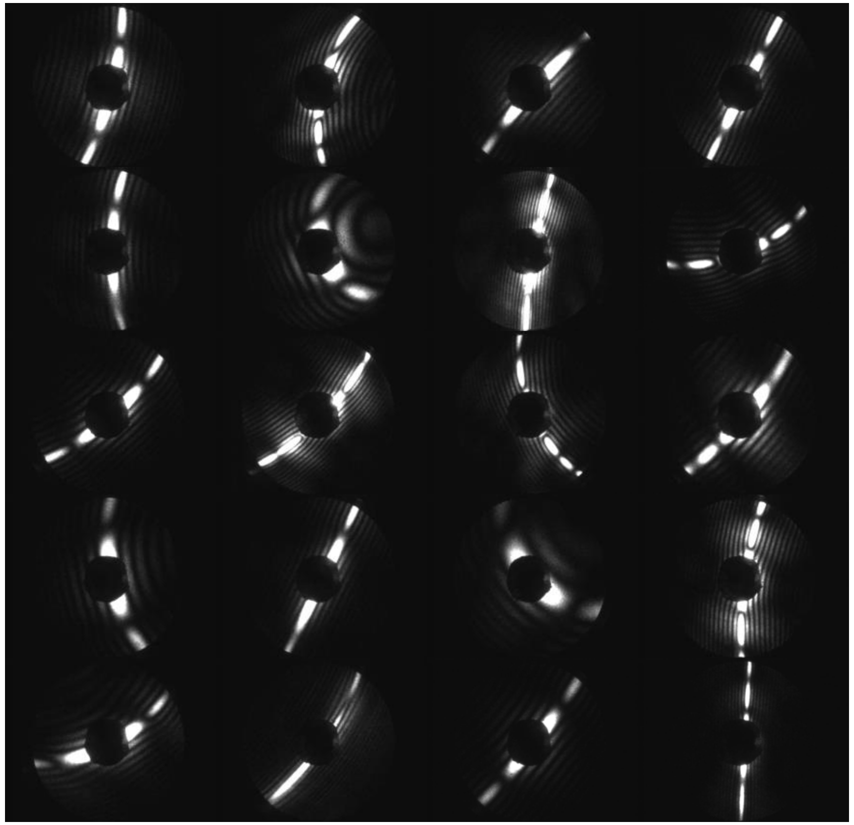
FTIR
Bruker IFS 66v
6000 – 800 cm^{-1}
0,1 cm^{-1} resolution

Extinction spectra



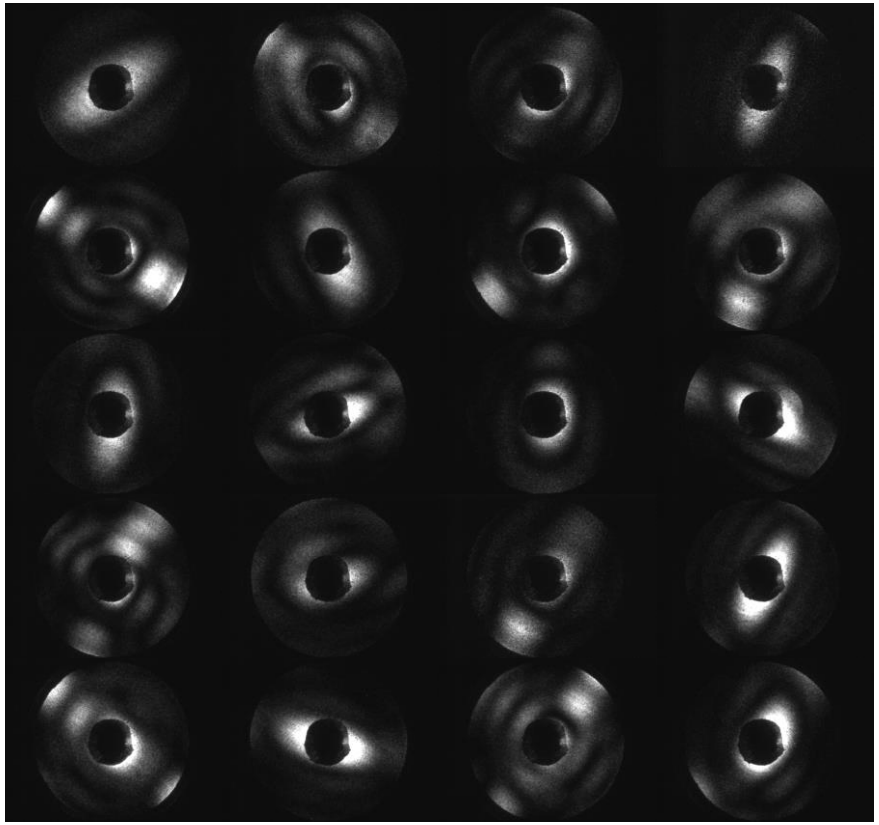
PSC – NAD: AIDA cloud chamber experiments

Hom. nucleation → α-NAD



long, very thin needles:
aspect ratio: > 5

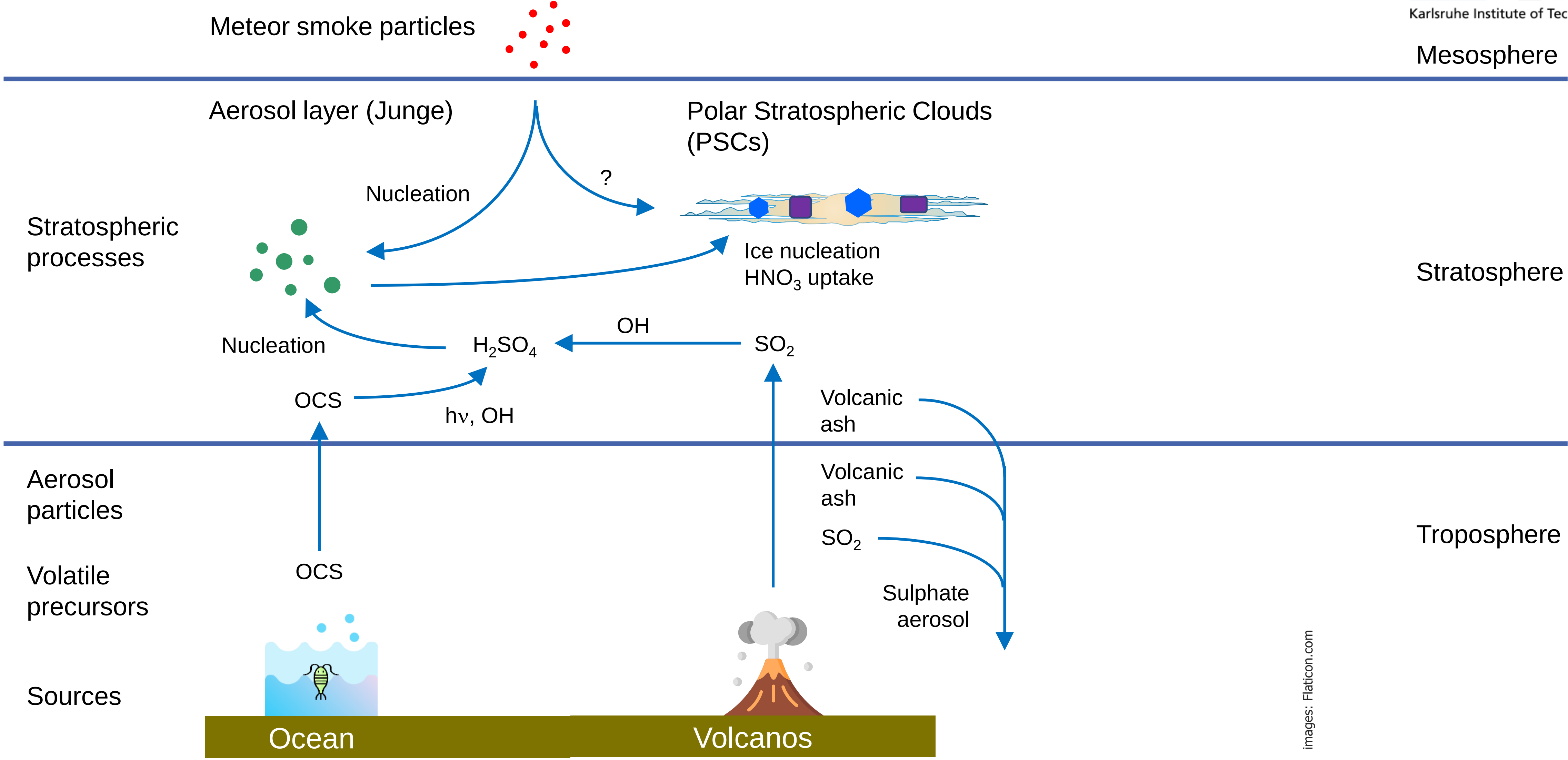
Het. nucleation → β-NAD



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

→ Nucleation impacts particle morphology and thereby vertical settling velocity

Meteor smoke particles



Meteor smoke particles

Mesosphere

Aerosol layer (Junge)

Polar Stratospheric Clouds (PSCs)

Nucleation

?

Stratospheric processes

Ice nucleation
HNO_3 uptake

Stratosphere

Nucleation

H_2SO_4

OH

SO_2

OCS

hν, OH

Volcanic ash

Aerosol particles

Volcanic ash

SO_2

Troposphere

Volatile precursors

OCS

Sulphate aerosol

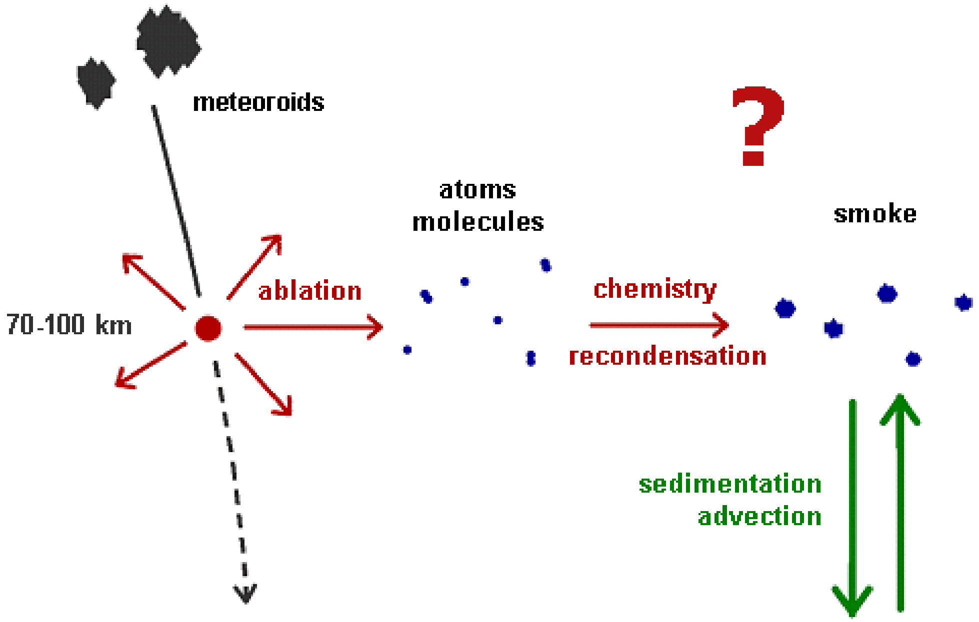
Sources

Ocean

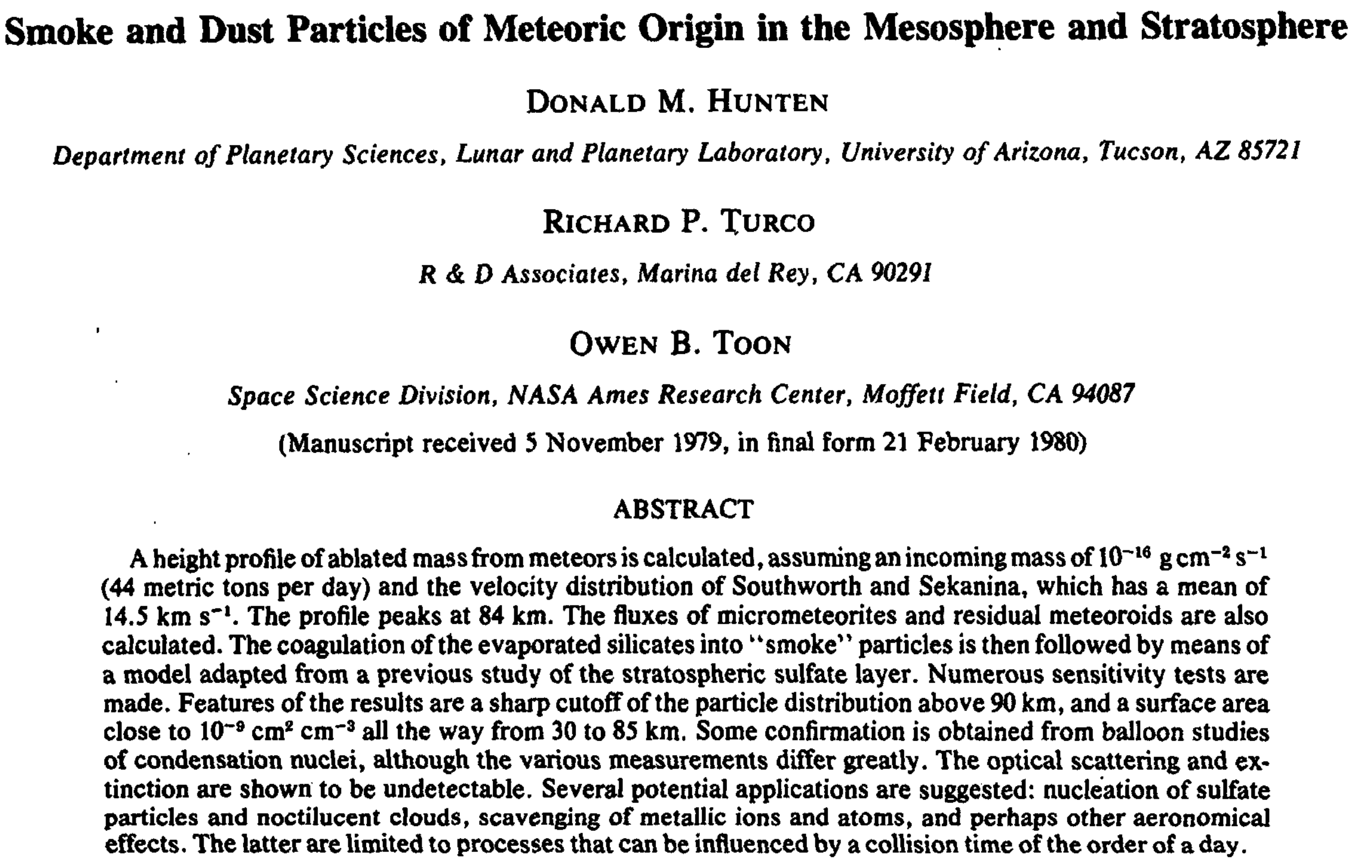
Volcanos

images: Flaticon.com

Meteor smoke: early observations and modelling



Hunten et al., J. Atmos. Sci., 1980



Smoke and Dust Particles of Meteoric Origin in the Mesosphere and Stratosphere

Donald M. Hunten

Department of Planetary Sciences, Lunar and Planetary Laboratory, University of Arizona, Tucson, AZ 85721

Richard P. Turco

R & D Associates, Marina del Rey, CA 90291

Owen B. Toon

Space Science Division, NASA Ames Research Center, Moffett Field, CA 94087

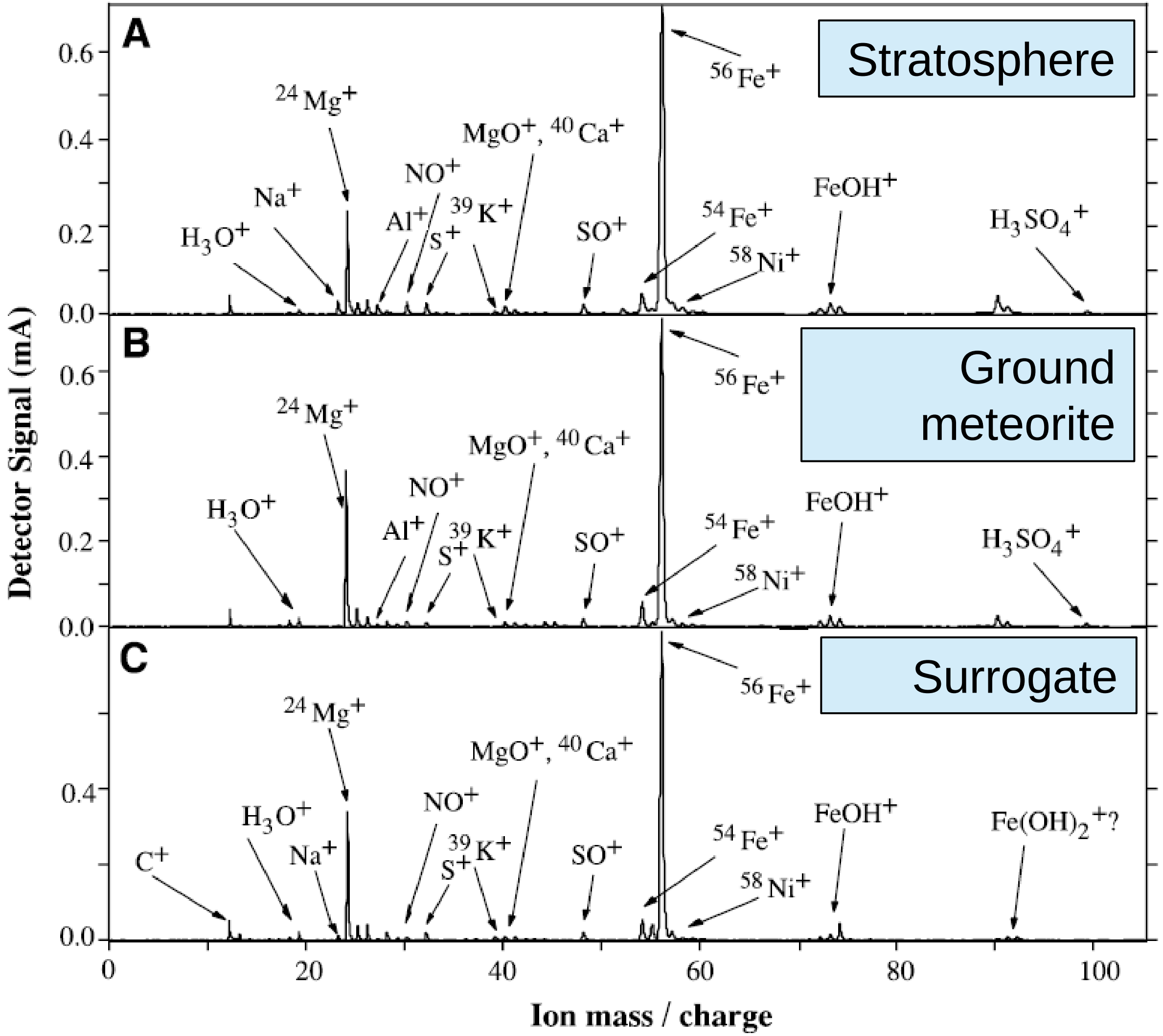
(Manuscript received 5 November 1979, in final form 21 February 1980)

ABSTRACT

A height profile of ablated mass from meteors is calculated, assuming an incoming mass of 10^{-16} g cm^{-2} s^{-1} (44 metric tons per day) and the velocity distribution of Southworth and Sekanina, which has a mean of 14.5 km s^{-1}. The profile peaks at 84 km. The fluxes of micrometeorites and residual meteoroids are also calculated. The coagulation of the evaporated silicates into "smoke" particles is then followed by means of a model adapted from a previous study of the stratospheric sulfate layer. Numerous sensitivity tests are made. Features of the results are a sharp cutoff of the particle distribution above 90 km, and a surface area close to 10^{-9} cm^2 cm^{-3} all the way from 30 to 85 km. Some confirmation is obtained from balloon studies of condensation nuclei, although the various measurements differ greatly. The optical scattering and extinction are shown to be undetectable. Several potential applications are suggested: nucleation of sulfate particles and noctilucent clouds, scavenging of metallic ions and atoms, and perhaps other aeronomical effects. The latter are limited to processes that can be influenced by a collision time of the order of a day.

Meteor smoke: icentification by single particle MS

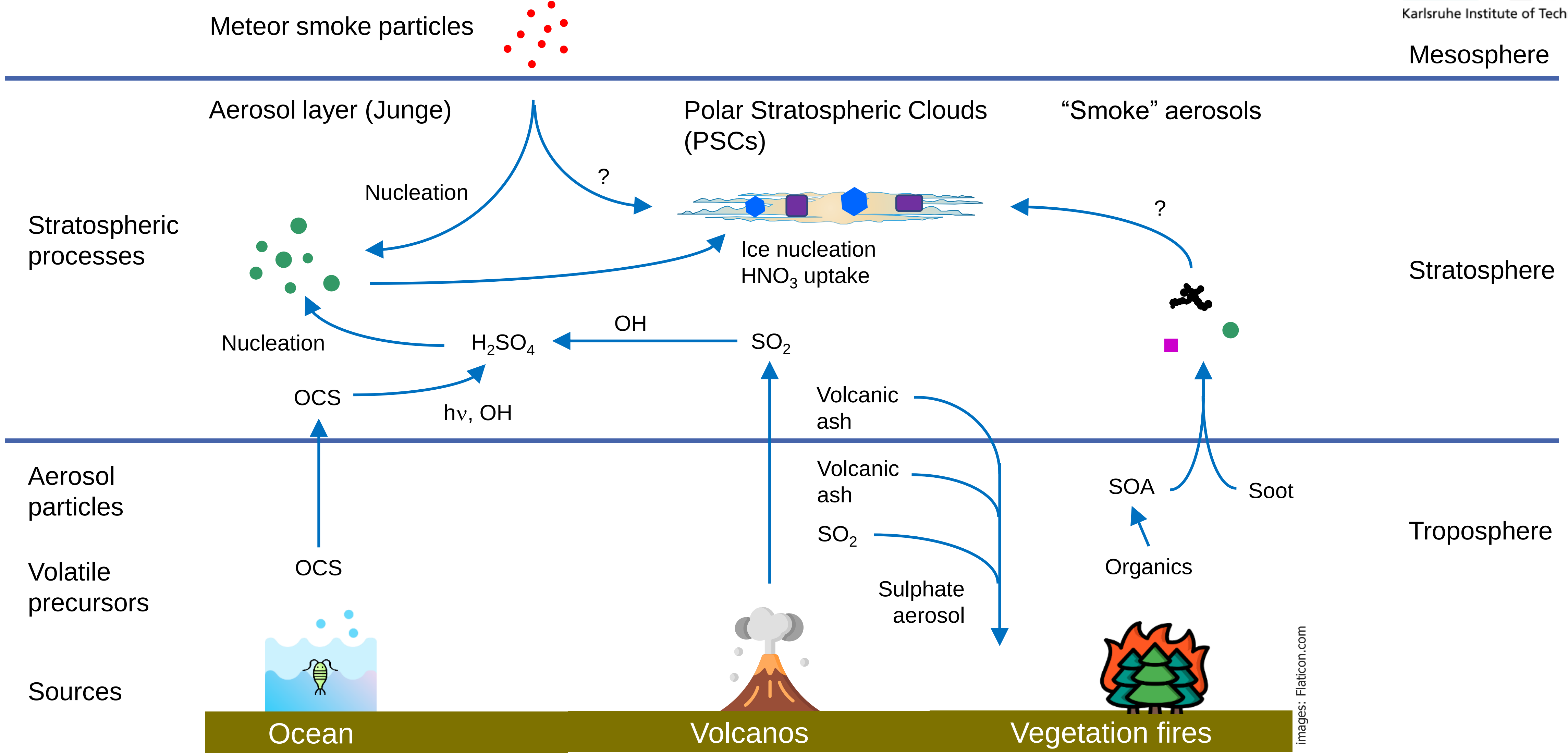
Positive ion spectra from single particles:



Cziczo et al., Science, 2001

NASA research aircraft WB-57

Soot and „smoke“ particles

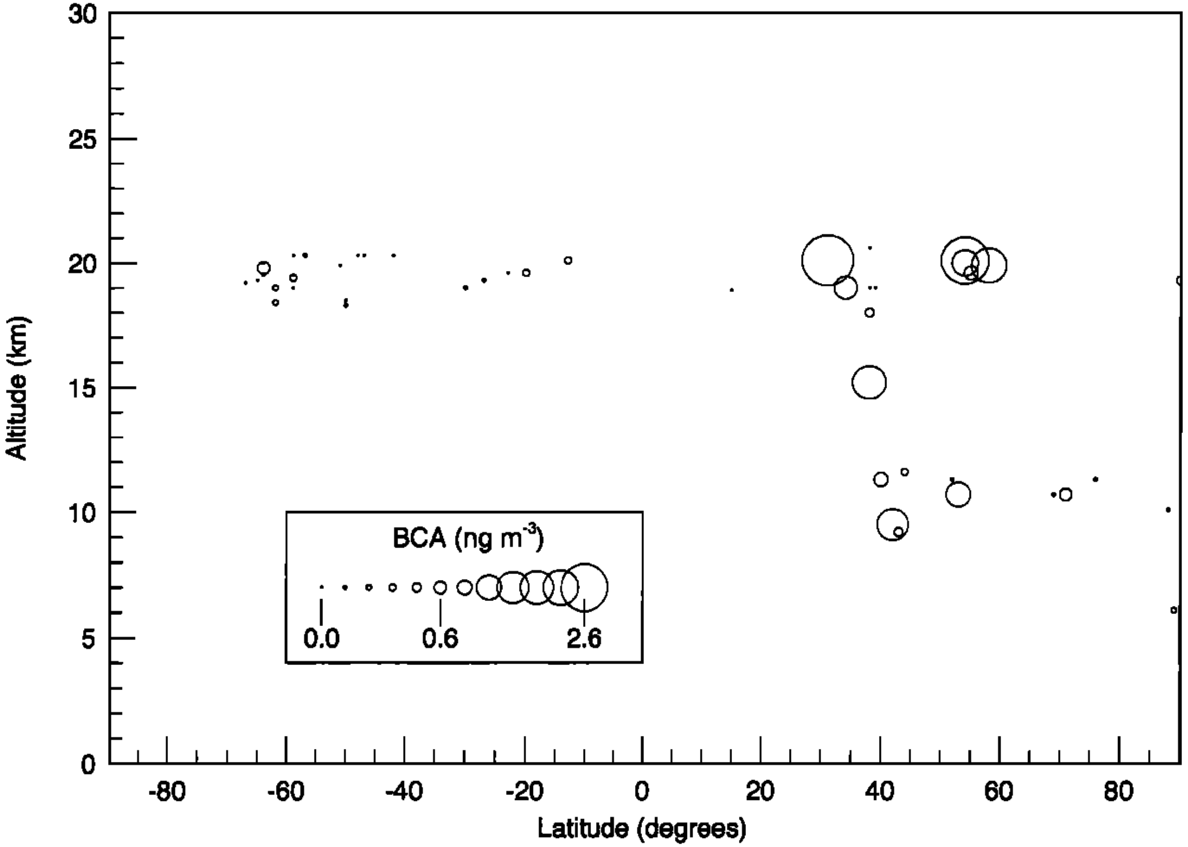


images: Flaticon.com

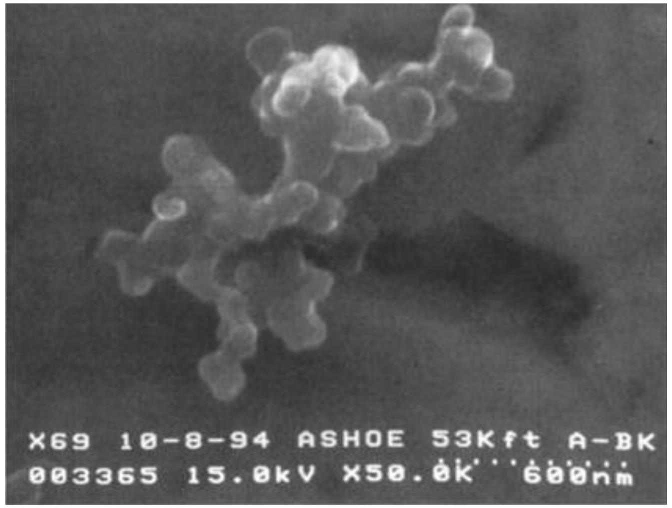
Soot and „smoke“ particles

Soot particles collected on wire impactors on ER-2 aircraft (Pueschel et al., JGR, 1997, 2000)

→ Lifted by gravito-photophoresis? (Rohatschek et al., J. Atm. Chem.,1984)



Pole-to-pole variability of soot mass concentrations (Pueschel et al., 1997)

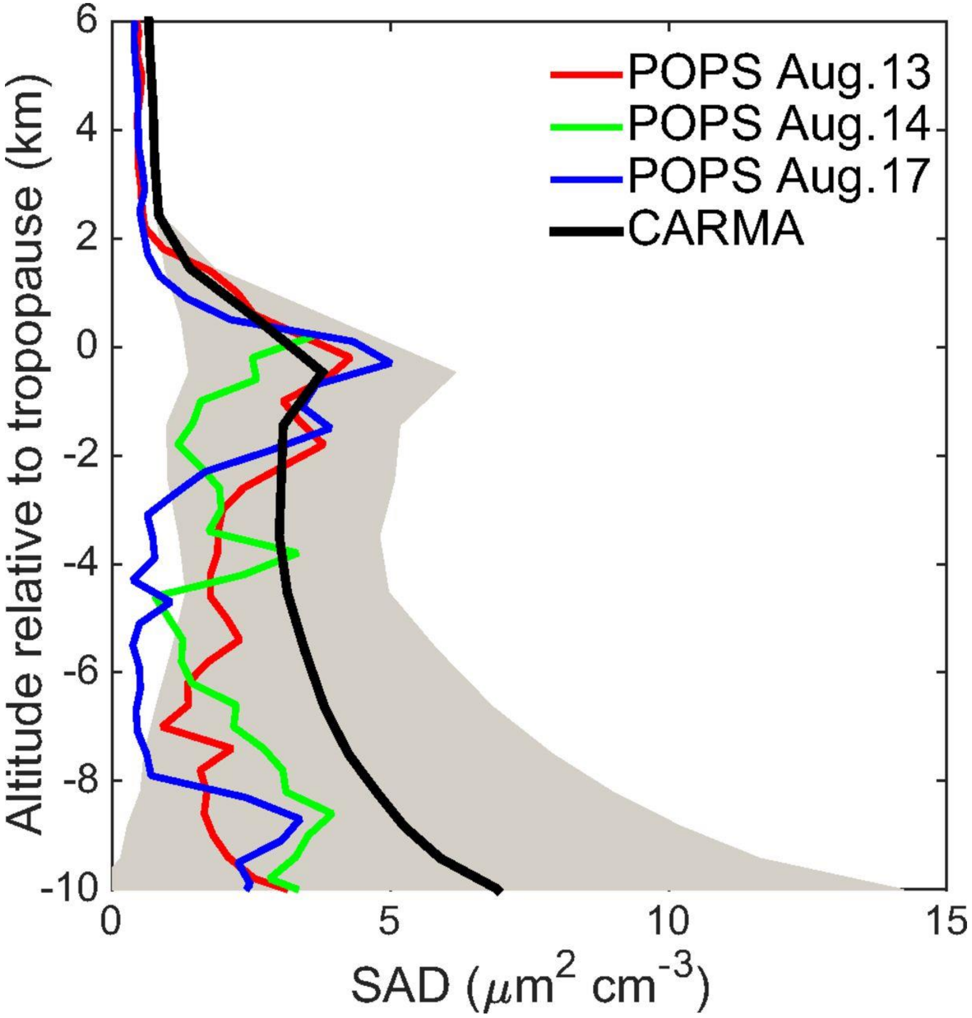


Soot particle collected in the wake of Concorde at 16.3 km altitude (Pueschel et al., 1997)

Soot and „smoke“ particles: Asian summer monsoon (ASM)

Yu et al., PNAS, 2017 (www.pnas.org/cgi/doi/10.1073/pnas.1701170114).

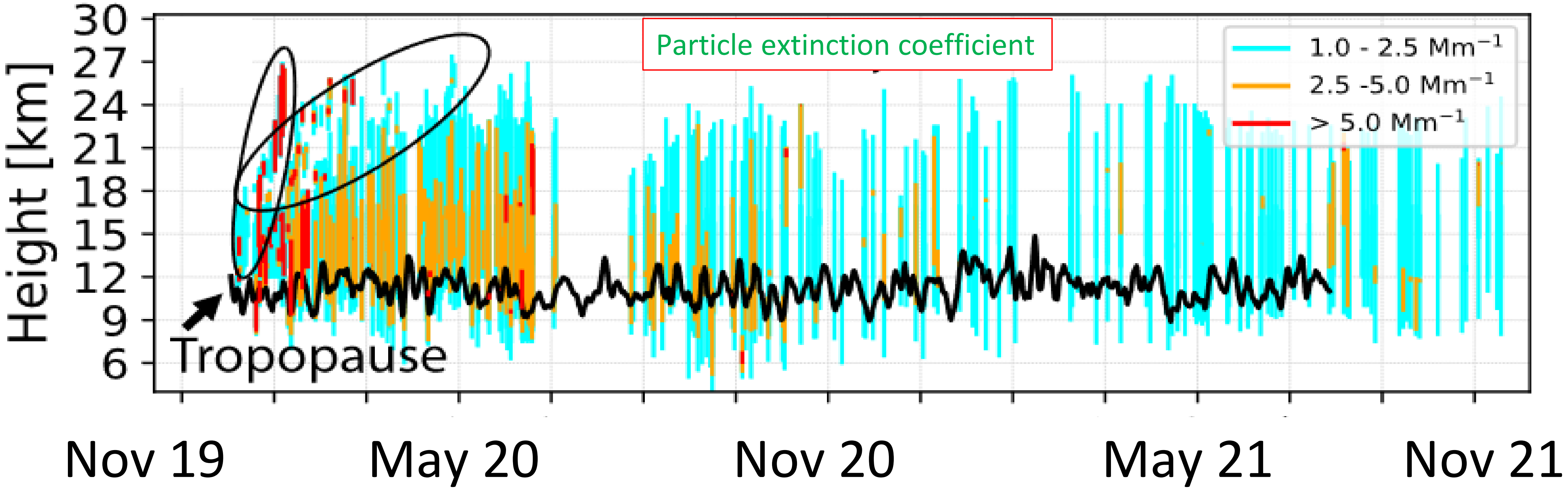
- Increasing aerosol optical depth (AOD) was retrieved from satellite observation (Vernier et al., JGR, 2015)
- Aerosol profiles measured with balloon-born leightweight OPC POPS (Gao et al., AST, 2016)
- Aerosol transported in the ASM anticyclone is exported to the entire Northern Hemispheric stratosphere.
- About ~15% of the Northern Hemisphere column stratospheric aerosol surface area originates from the Asian summer monsoon anticyclone region



Soot and „smoke“ particles

Polly Lidar measurements, Southern South America (50°-60°S)
Ohneiser et al., ACP, 2020, 2022

See also recent MOZAiC results (Engelmannm et al., 2021; Ohneiser et al., 2021)



Courtesy of Albert Ansmann, TROPOS, Leipzig. Germany

Summary

Tropospheric aerosol:

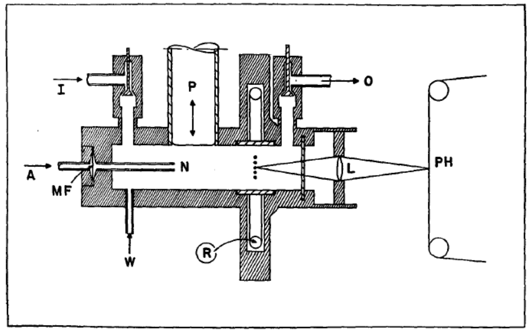
- ➢ Complex mixture of various particle types
- ➢ Natural and anthropogenic sources
- ➢ Primary emission and secondary formation processes

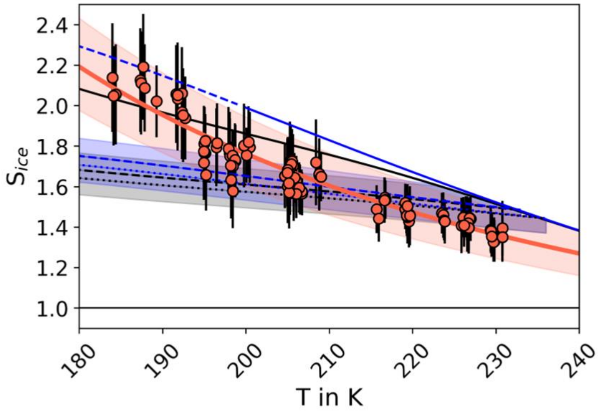
Stratospheric aerosol:

- ❖ Aqueous sulphuric acid particles (Junge layer) from OCS
- ❖ Periodic enhancement by volcanic SO_2
- ❖ Source of PSCs in wintertime polar stratosphere
- ❖ Includes meteoric smoke particles (also act as nuclei?)
- ❖ Soot and smoke particles detected (wire impactors)

Recent findings:

- ○ Freezing onset higher than predicted by Koop et al., 2000
- ○ Liquid water saturation may be higher than currently formulated
- ○ Het./hom. NAD nucleation forms particles of different morphology
- ○ Lofting of tropospheric aerosols into the stratosphere: impact on PSCs, ozone, …





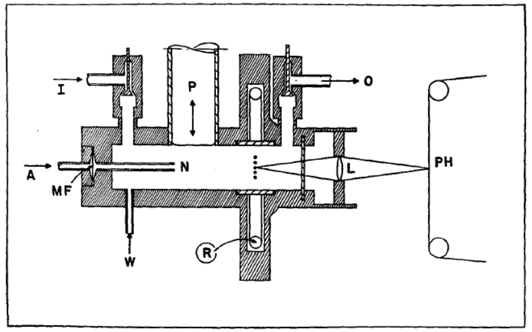
Ongoing work and future challenges

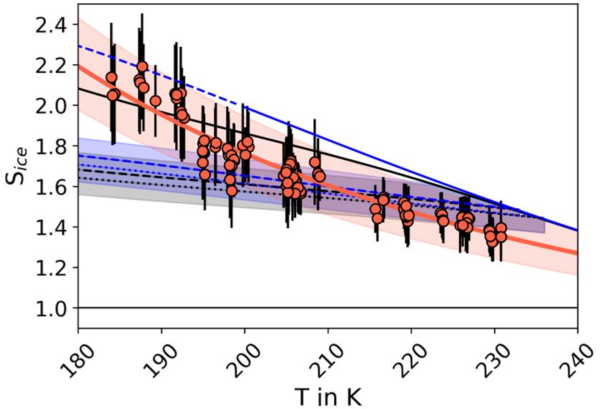
Laboratory process studies:

- Better understand homogeneous freezing of solutes
- Quantify saturation point of liquid water at low T
- Quantify nucleation rates of nitric acid hydrates (NAT, NAD)

Stratospheric in situ measurements:

- Morphology of solid PSC particles
- Sources and distribution of different aerosol types
- Quantify contribution and impact of non-volcanic tropospheric aerosols (smoke)





Thanks for listening (ottmar.moehler@kit.edu)