High-altitude cold cirrus clouds

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Aim of the lecture is

- to give a short review of the current knowledge about cold cirrus clouds including observations, properties, impact on climate, modeling, problems, and laboratory studies.

Content

- Introduction
  - What are cold cirrus clouds?
- Observations and properties
  - types of the observations
  - macro- and microphysical properties
- Impact on climate
- Modeling and problems
- Laboratory methods of the study
  - solution-in-oil emulsions
  - differential scanning calorimeter (DSC)
- Summary

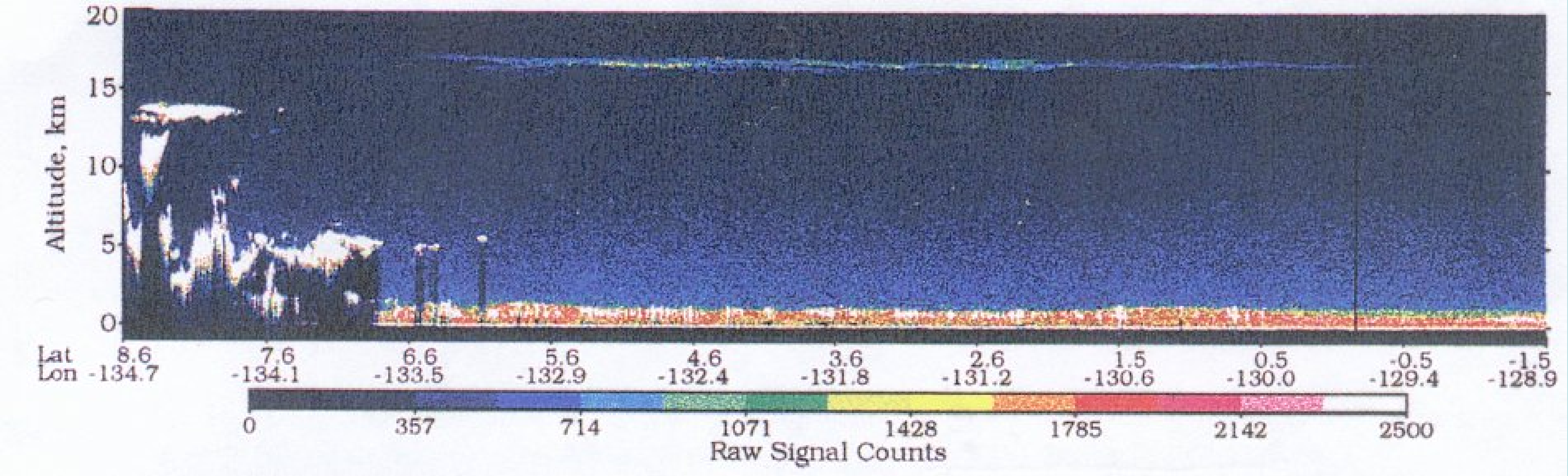
High-altitude cold cirrus clouds:

- Temperature range:
  - between ~ 210 and 185 K

- Altitude:
  - between ~ 8 - 20 km, near the tropopause
  - water concentration ~1 - 12 ppmv.

- Optical thickness τ:
  - τ < 0.03 - sub-visible cirrus (SVC) clouds
  - 0.03 < τ < 0.3 - thin cirrus clouds

Low-temperature sub-visible cirrus (SVC) cloud in the tropical tropopause at ~17 km



(From Winker and Trepte, GRL, 25, 3351-3354 (1998).)

Location and frequency of occurrence

- Observations show that high-altitude cold cirrus clouds are widespread both near the tropical and midlatitude tropopause.

- They cover approximately 20 % of the Tropics and ~20 - 30 % of the globe.

- The largest frequency of occurrence of cold cirrus is near the Tropics.

- Types of cloud observations include (i) satellite, (ii) in situ, and (iii) remote ground-based and airborne observations

(i) Satellite observations

- provide accurate information on the global distribution of cold cirrus

- are usually limited to resolution of several kilometers

- cover only the uppermost cold cirrus clouds

(ii) In situ observations

- are performed using different instrumentations placed on the airborne platforms

- are aimed on gaining the information about the ice crystal size distributions, number density, crystal habits, ice water content (IWC), water vapor concentration or relative humidity (RH_i) etc.

- provide information with special resolution on the order of meters

- But the in situ aircraft measurements are limited in time and space.

(iii) Remote ground-based and airborne cloud observations

- are intermediate between the satellite and the in situ observations

- provide measurements at the scales between tens of meters and tens of kilometers

- The ground-based remote observations sample only those high cirrus clouds that are not obscured by the lower clouds.

- A key sensor of the remote cloud observations is polarization LIDAR (Light Detection And Ranging).

- The LIDAR discriminates between liquid droplets and ice crystals. The discrimination is based on scattering theory which predicts that the liquid droplets generate no depolarization ratio of the incident laser light in the exact backscattering direction. In contrast, the ice crystals, generate a significant depolarization.

- Fluctuations of the depolarization ratio indicate about the inhomogeneity of cloud particles in terms of the size/shape/orientation.

- Other remote sensors used in the study of cold cirrus clouds are

- visible, infrared, and microwave radiometers

- all-sky photographic and video imaginary

- polarimetric Doppler radar etc.

Physical properties of cold cirrus

(i) To cloud macrophysical properties are attributed the cloud top and base heights and temperatures, separation from the tropopause level, geometrical thickness (< 1 km), all-sky high-cloud coverage, cloud-center height, cloud structure (may be layered structure), uniformity (may be present liquid layers).

- Cloud macrophysical properties are usually derived from the remote aircraft, ground-based, and satellite observations.

(ii) To cloud microphysical properties are attributed the ice crystal size distribution, number concentration, crystal habits, ice water content (IWC), water vapor concentration etc.

- Detailed knowledge of the microphysical properties require the *in situ* measurements which often require more elaborated aircraft instrumentations than that used in the remote sensing.

- The aircraft instrumentations are usually optical particle detectors and counterflow virtual impactors. They collect all particles, evaporate them, and measure the evaporated water and the residual aerosol particles.

- The main drawbacks of the existing instrumentations for the in situ measurements are their inability to properly characterize the small (< 20 μm) ice crystals.

- Recent in situ observations reveal that in cold cirrus clouds, the size and shape of ice crystals changed in comparison with those which were ~30 years ago. The ice crystals became smaller and of simpler, quasi-spherical shape.

Radiative properties

- In warm liquid clouds, the relations between the cloud particles and cloud radiative properties are relatively simple because of homogeneity of the cloud droplet shape. Also the amount of soluble substances, which can change the refractive index of the droplets, is relatively small.

- In the case of cirrus clouds the situation is more complicated because the radiative properties strongly depend on the size, shape and orientation of ice crystals.

- The shape and size of cirrus ice crystals depend on altitude, geographical location, mechanisms of the formation, and growth history.

- In cold cirrus clouds, the ice crystals are smaller than those observed in lower warmer cirrus clouds, and thus backscatter more efficiently the solar radiation, i.e. the small ice crystals increase the cloud albedo.

- Uncertainties about the shape of the ice crystals can produce large errors in the retrieved radiative properties of cirrus clouds.

Impact on climate

- Since the cold cirrus clouds are globally widespread they are important modulators of solar and terrestrial radiation and consequently important climate regulators.

- For the shortwave solar radiation, the cirrus cloud forcing is a difference between the reflected solar radiation for clear sky and the reflected radiation for cloudy sky.

- The solar cloud forcing is always negative because the cirrus clouds increase the reflected solar radiation.

- For the longwave terrestrial radiation, the cloud forcing is a difference between the outgoing to space infrared radiation for the clean sky and the outgoing to space radiation for cloudy sky.

- The infrared cloud forcing is always positive because the cirrus clouds reduce the outgoing terrestrial radiation.

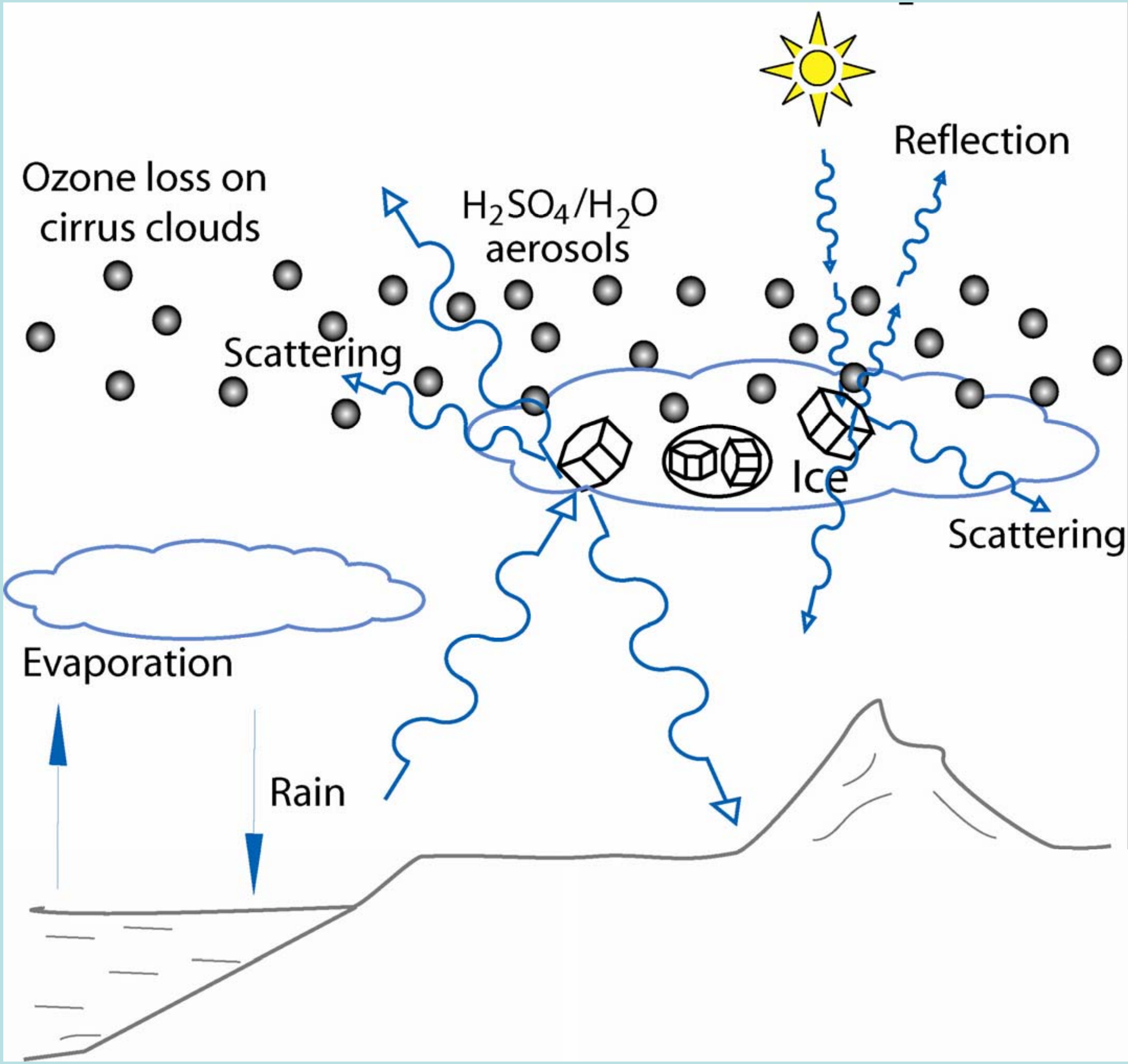
- The net cloud radiative forcing (CRF) is the sum of the two terms and can change sign from positive to negative.

Example:

- In the nearly 'invisible' cold cirrus clouds, which are near the tropopause in middle latitudes, the small ice crystals (effective radius smaller than 7μm) absorb almost 50 % of the upwelling infrared terrestrial radiation. This absorption can have a potential contribution to the 'greenhouse warming'.
- Small ice crystals have little effect on short-wave solar radiation which almost freely passes through them.

- For the cold cirrus clouds the average value of the cloud radiative forcing (CRF) was calculated to be 1.58 Wm-2, with 2.19 Wm-2 occurring at the infrared and -0.61 Wm-2 at solar wavelength.

Cold cirrus clouds are important climate regulators.



Modeling and Problems

- Usually the radiative effects of cold cirrus are not considered in most climatic model simulations, because their average optical depth is small. However, the frequent occurrence of the cold cirrus, particularly in the Tropics, may result in noticeable impact on the radiative budget of the Earth.

- According to the calculations, the net radiative effect of the cold cirrus produced on the top-of-atmosphere is ~0.5 W/m^2. This greenhouse warming is similar in magnitude, but opposite in sign, to the direct forcing estimated for aerosol of anthropogenic origin.

- One of the numerous problems of general circulation models (GCMs), which include cold cirrus clouds, is a lack of the detail knowledge of the cloud microphysics.

- In these models, the ice crystals size distribution does not include ice crystals smaller than 20 μm that stems from the limitations in the used measuring technique.

- The Mie theory shows that ice crystals of diameter ~3 μm scatter visible radiation ~30 % more efficiently than the ice crystals of diameter 20 μm. Neglecting the ice crystals smaller than 20 μm introduces the systematic uncertainty in the calculation of the impact of cirrus on whether and climate.

- Thus the general circulation models (GCMs) have to use more reliable microphysical parameters in order to better estimate the impact of cold cirrus on whether and climate.

- The improved in situ cirrus cloud measurements as well as better understanding the microphysical processes occurring on the sub-micrometer and micrometer scales may improve our knowledge about the microphysical properties of cold cirrus clouds.

- The laboratory studies, which could cast a light on the physics and chemistry of the small aqueous solution droplets, are needed.

- The laboratory studies may also account for the recent problem concerning the unusual large humidity in the upper troposphere.

Recent cloud observations show that at $T \approx 185$ K

RH_i can reach as large as 250 %.

Nature of the high upper troposphere humidity (UTH) is not clear yet.

According to the present knowledge the large RH_i cannot exist. At the large RH_i the ice phase easily nucleates and the formed ice particles would rapidly deplete RH_i down to 100 %.

Relative humidity with respect to ice:

$$\mathrm{RH_i} = \frac{p_{env}^{w}}{p_i^{w,sat}} \times 100\%$$

p_{env}^{w} - environmental water vapor pressure

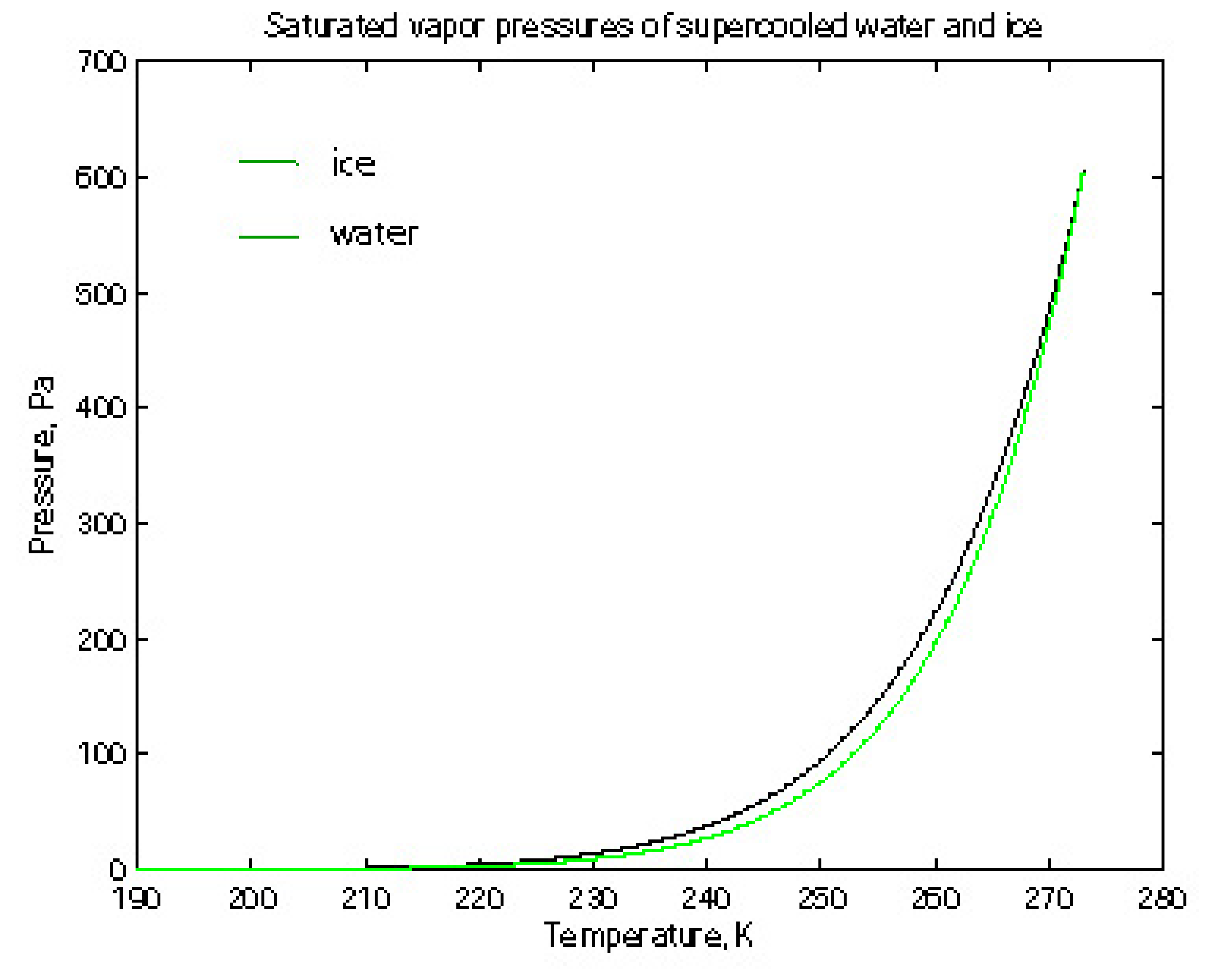
$p_i^{w,sat}$ - saturated water pressure of ice

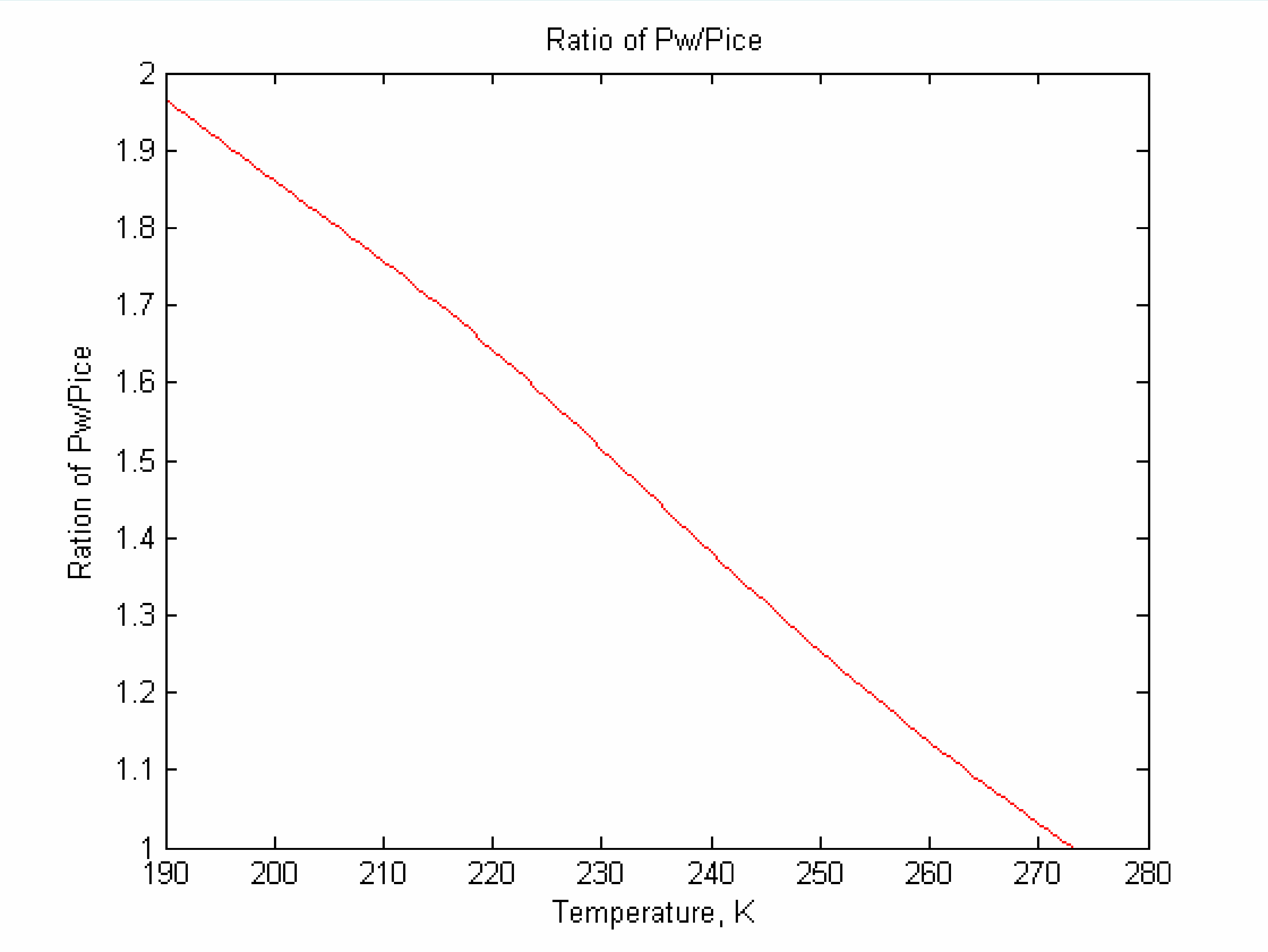
Saturated water vapor pressure is a **function of temperature only** and independent on the presence of other gases. The temperature dependence is exponential. For water vapor the semi empirical dependence reads as

$$p_{w,s} = e^{A+\frac{B}{T}+C\ln T+DT}$$

The similar exponential dependence of saturated water vapor pressure also exists for ice.

Saturated vapor pressures of ice and supercooled water





This is not what is in the atmosphere

- Nucleation
- Condensation - growth of droplets
- Evaporation - decreasing of droplet size

- Freezing of droplets
- Melting of ice crystals

- Vapor deposition - growth of ice crystals
- Sublimation - evaporation of ice crystals

These transitions are the 1-st order phase transitions.

They are accompanied by the consumption and emission of specific heat (enthalpy) of the transitions.

Laboratory studies

Method: differential scanning calorimeter (DSC).

Advantage:

- DSC provides a precise thermal picture of phase transitions occurring during the cooling/warming of samples

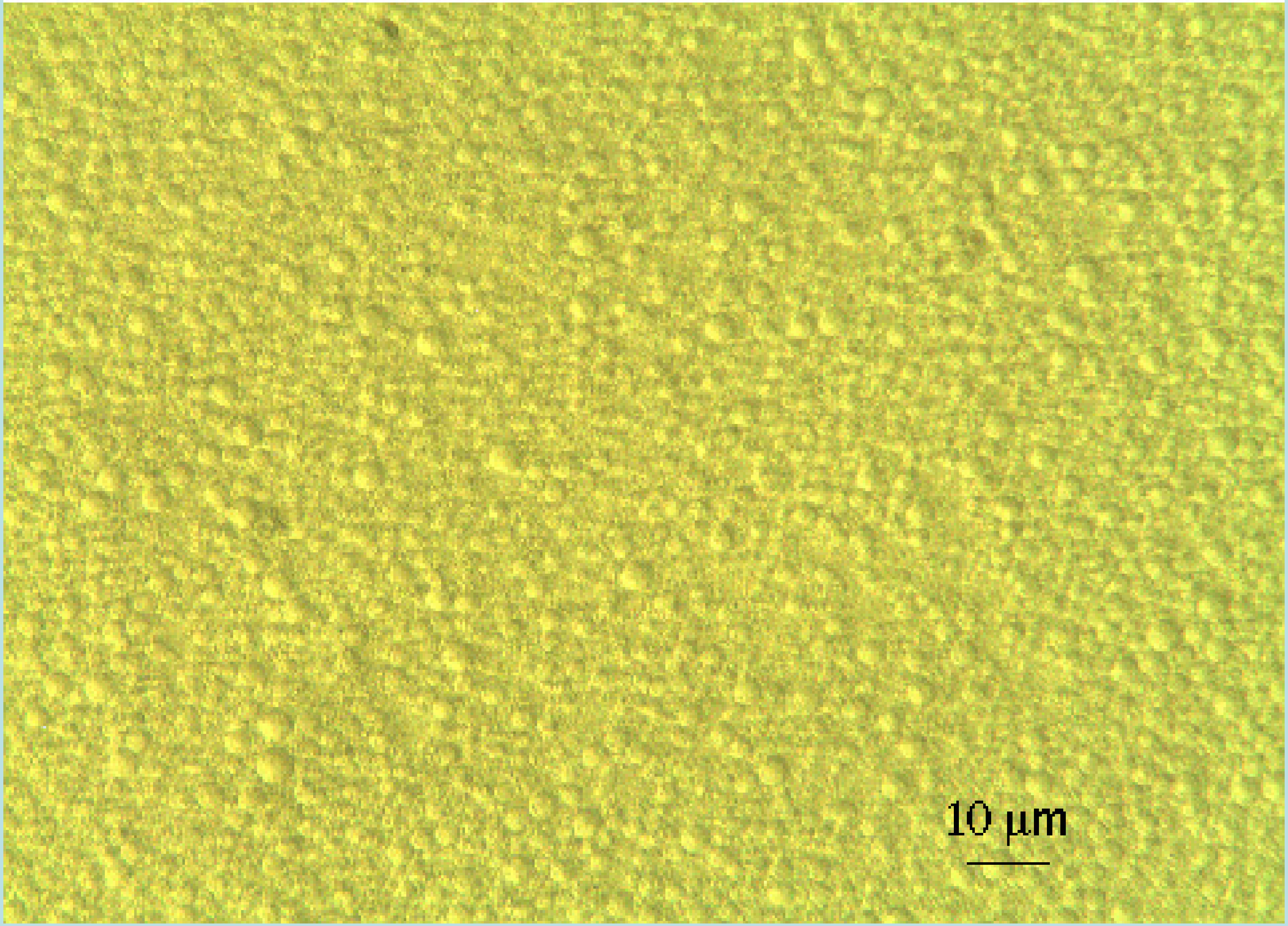
- Samples for the calorimetric study:

  - bulk droplets of diameter d ≈ 1.5 mm

  - very large populations of droplets d < 5 µm

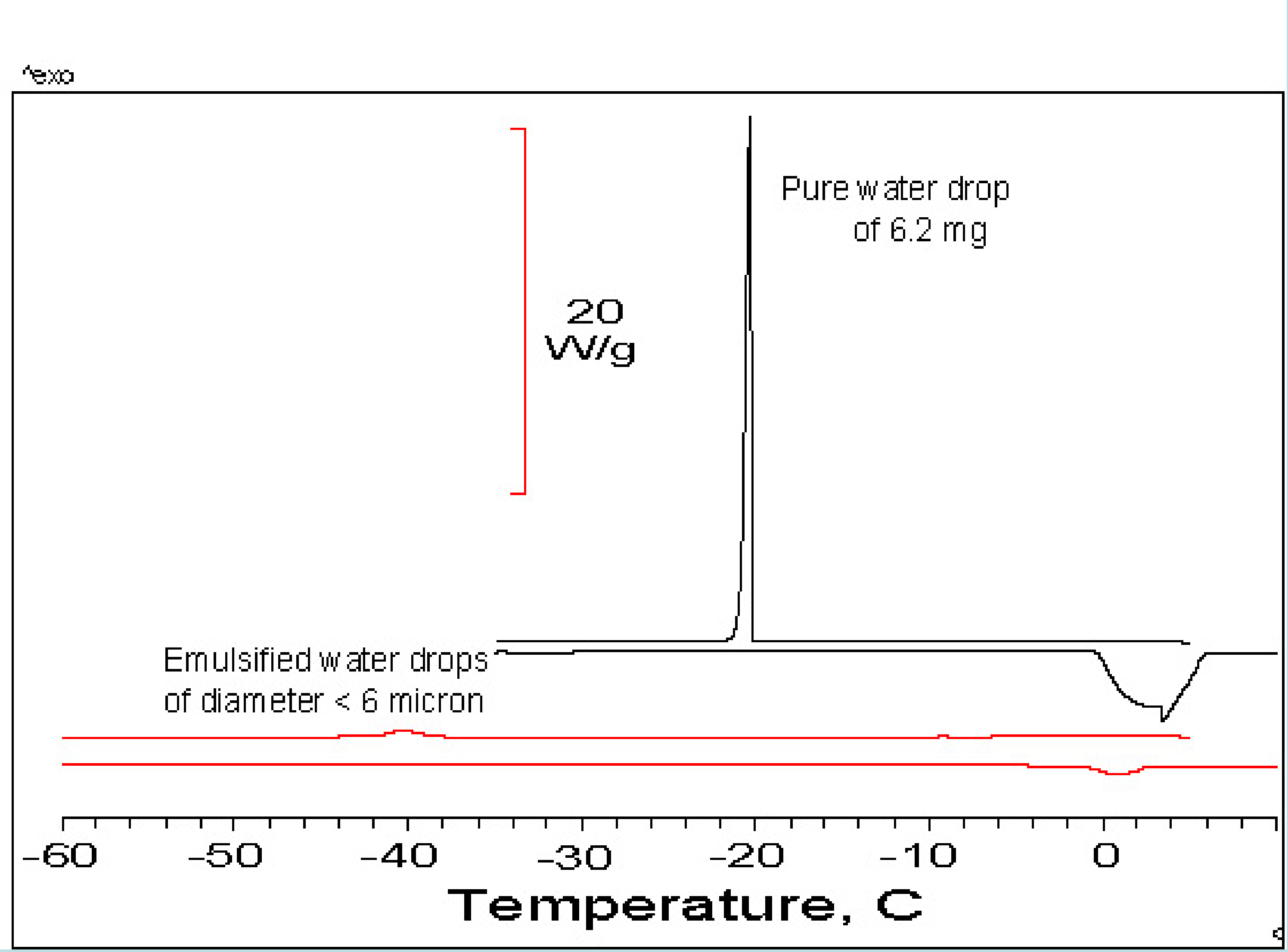
- The calorimetric study of these two types of samples shows that the knowledge obtained from the study of only bulk samples may not be sufficient for the understanding of the formation and microphysical properties of cold cirrus clouds.

An example of the emulsion.

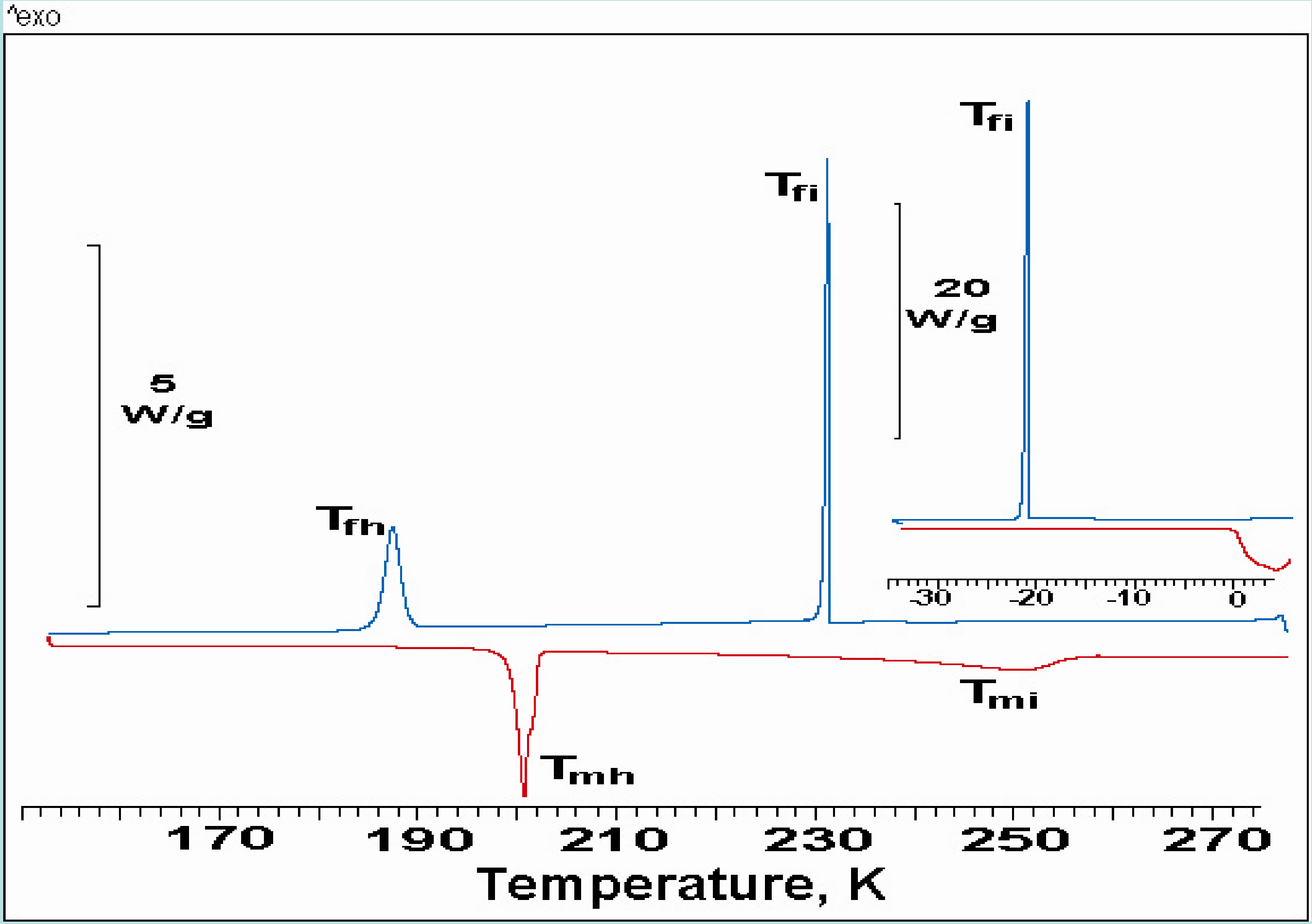


In the emulsion, we can simultaneously study the populations of several millions droplets of the sizes and compositions similar to those encountered in the upper troposphere.

An example of how freezing temperature reduces with the size of pure water droplets

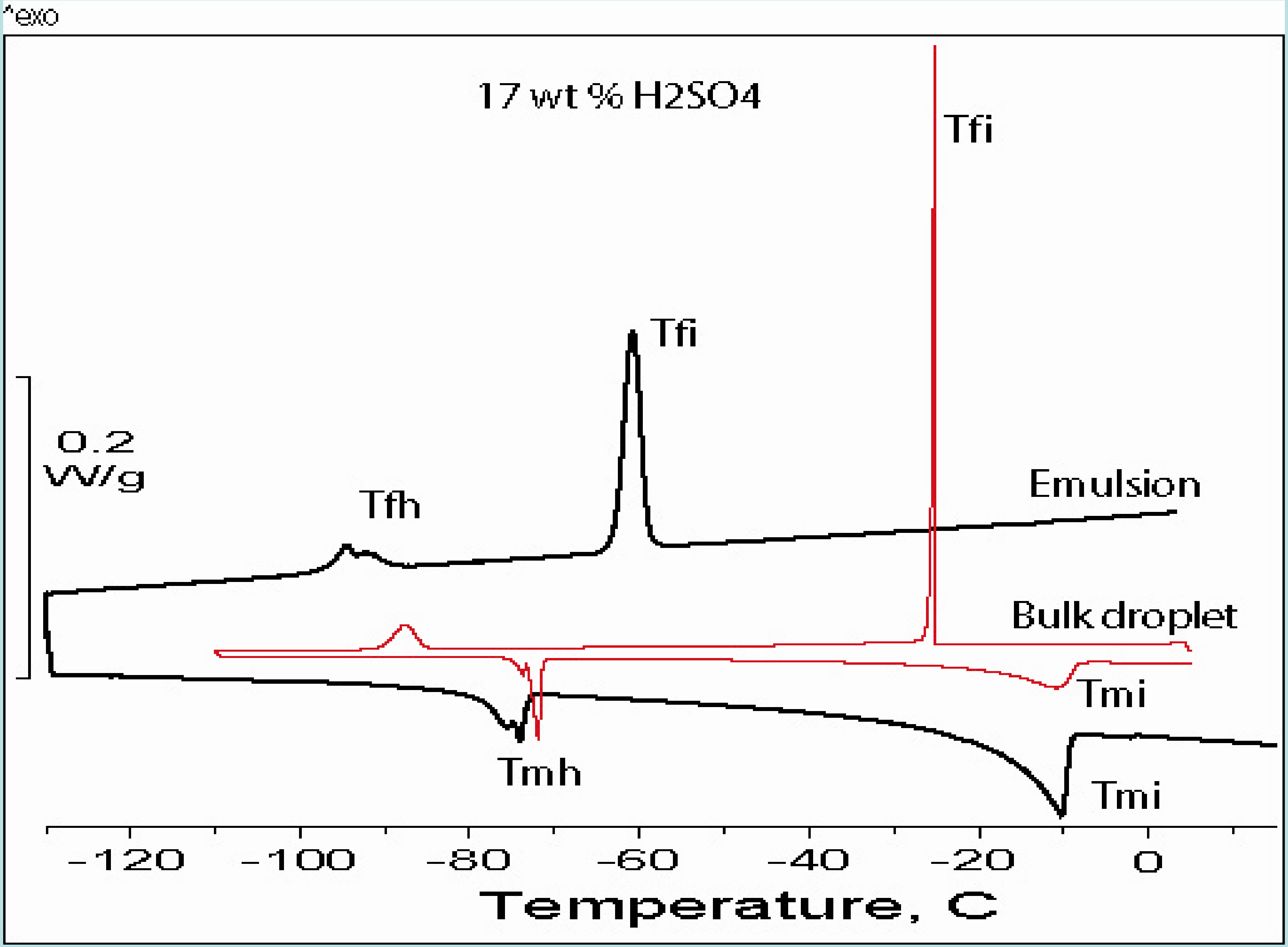


Addition of solutes to water reduces not only the freezing and melting temperatures of ice but also the number of the freezing/melting events. (Bulk water and 25 wt% H_2SO_4).

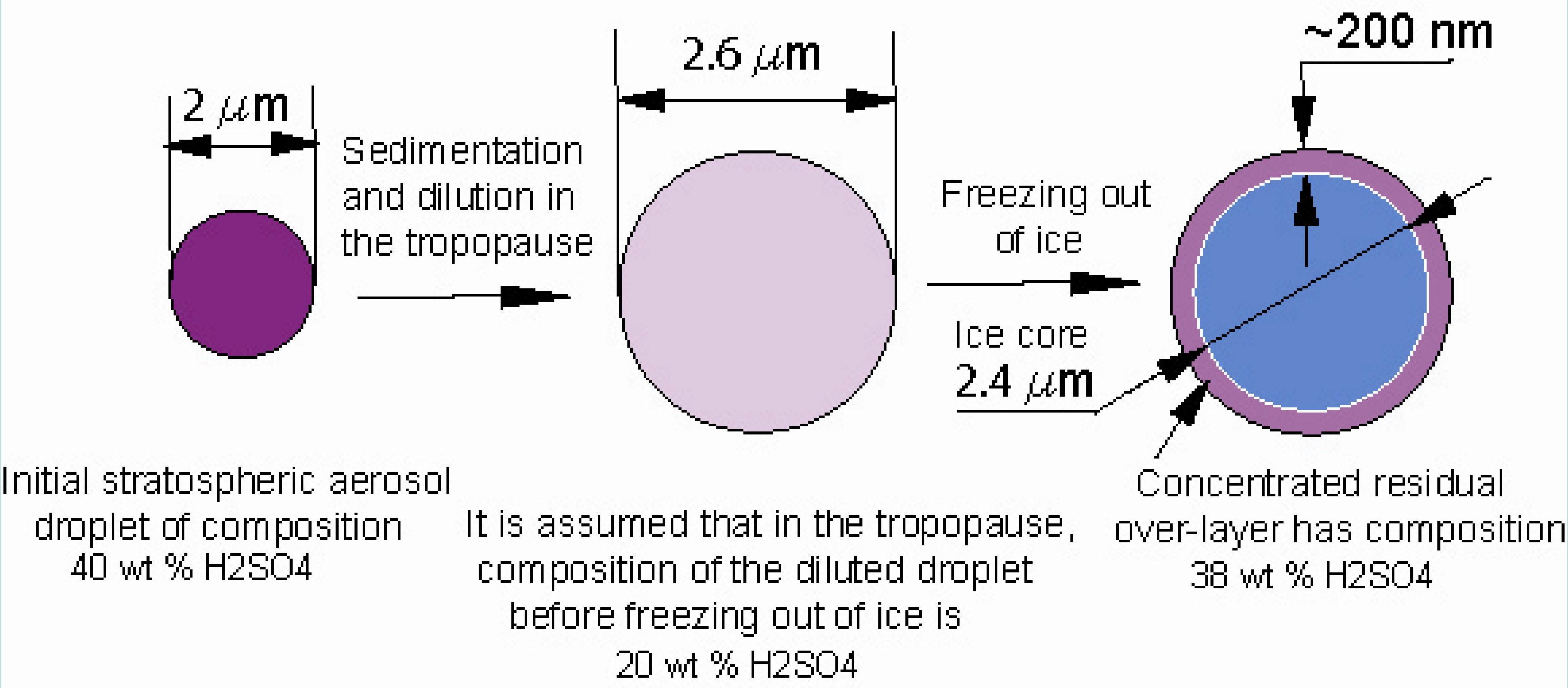


As ice freezes out within solution droplets the acid molecules (ions) are expelled from the ice lattice to form a residual solution.

Both the freezing temperatures of ice and residual solution reduce with the size of droplets.



What may happen in the upper troposphere (hypothesis)



Initial stratospheric aerosol droplet of composition 40 wt % H2SO4

It is assumed that in the tropopause, composition of the diluted droplet before freezing out of ice is 20 wt % H2SO4

Concentrated residual over-layer has composition 38 wt % H2SO4

Dilution and freezing of H_2SO_4/H_2O aerosol drop.

Summary

- Calorimetric measurements confirms the hypothesis that in cold cirrus clouds, ice crystals can be coated with liquid solution coating

- the solution coating around ice particles can change the radiative properties and the rate of development and dissipation of cold cirrus clouds

- since the rate of ozone loss is larger in/on liquids than on solids, the presence of the coating can increase the rate of ozone loss

- the coating may be responsible for the persistent high in-cloud RHi