Chilled mirror hygrometers and their „Golden Points“

A new interpretation and correction method for chilled mirror data

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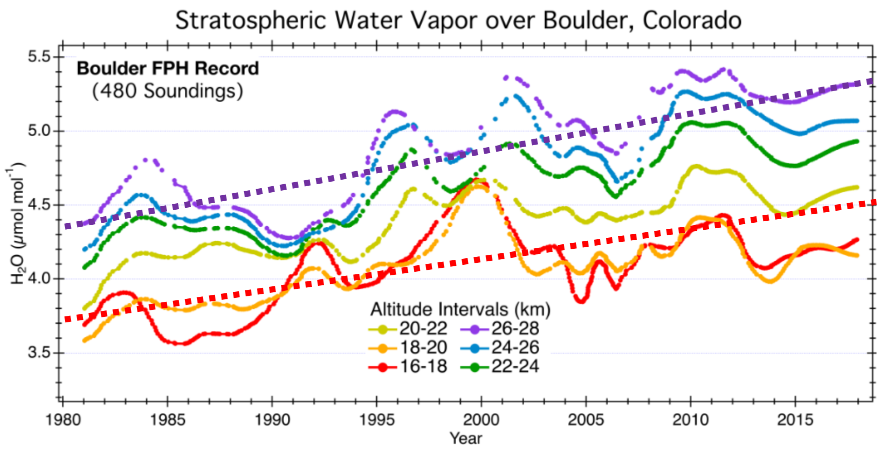
Outline

- Introduction
  - Motivation: need for accurate measurements of water vapor
  - Chilled Mirror Hygrometers (CMH): reference grade instruments
- Golden points concept
  - Example I: stationary conditions (pre-launch)
  - Example II: dynamic conditions (tropopause)
- Out-of-equilibrium correction
  - Theory
  - Example I: CFH and RS41
  - Example II: CFH ascent vs. descent
  - Example III: CFH and FLASH-B
  - Statistics of the out-of-equilibrium correction from 70 nighttime CFH-RS41 flights
- Conclusions

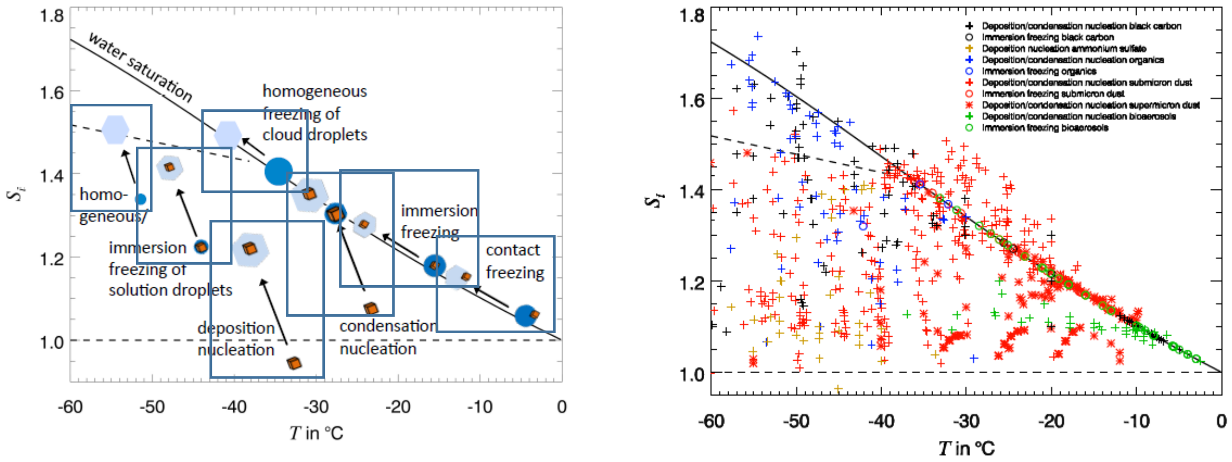
Motivation: need for accurate measurements of H_2O

Accurate measurements of atmospheric H_2O are important, e.g.:



Adapted from Hurst et al., 2018

Hoose and Möhler, 2012



Monitoring climate change

Small trends in stratospheric water vapour (few hundreds of ppbv/decade) can have a significant impact on the rate of global warming and on the rate of ozone recovery (e.g. Solomon et al., 2010).

Investigating cloud processes in the atmosphere

Understanding of ice nucleation mechanisms and cloud microphysical processes requires an accuracy often better than 10% (e.g. Hoose and Möhler, 2012).

GCOS and CIMO **“goal”** requirements are set at an ambitious level of **4-5 % uncertainty in H_2O** in the troposphere and stratosphere.

Chilled Mirror Hygrometers (CMH)

Working principle:

i) a mirror is cooled until a dew or frost layer forms onto the mirror.
ii) a feedback controller manipulates the temperature of the mirror such that the condensate *neither grows nor shrinks*.
iii) the (averaged) mirror temperature gives an estimate of the dew/frost point

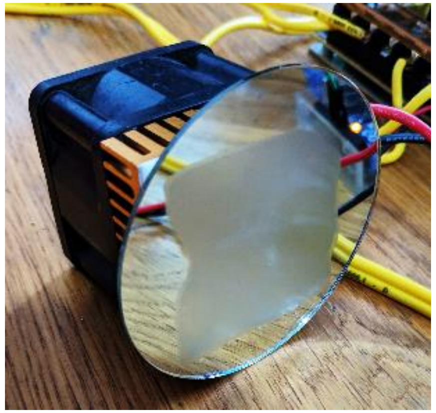
Cooling technique:
Electric (Peltier device), Cryogenic (R23, liquid N_2, ethanol+dry ice).

Condensate Growth Feedback technique:
Specular reflectivity from mirror, or forward/backward scattering from the condensate. PID control of mirror heater / Peltier current.

Metrological water vapor standard:
Rely solely on measuring temperature (SI-traceable). Used as transfer standard for humidity in many applications.

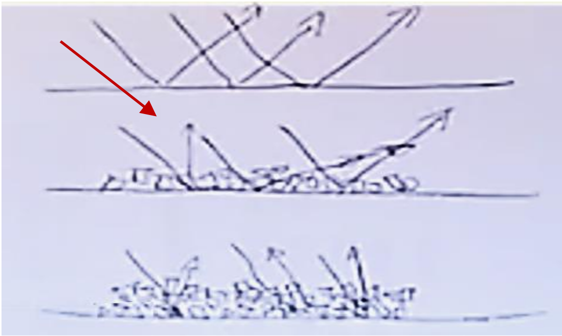
Balloon-borne CMH: (e.g. CFH, FPH, ELHYSA, SnowWhite, SkyDew, PCFH, ….) is the "*only technique capable of measuring with high quality, high vertical resolution and excellent long-term stability from the ground to the middle stratosphere*" (ndacc, 2017).

dew on mirror



Arduino Chilled Mirror Hygrometer © MIT

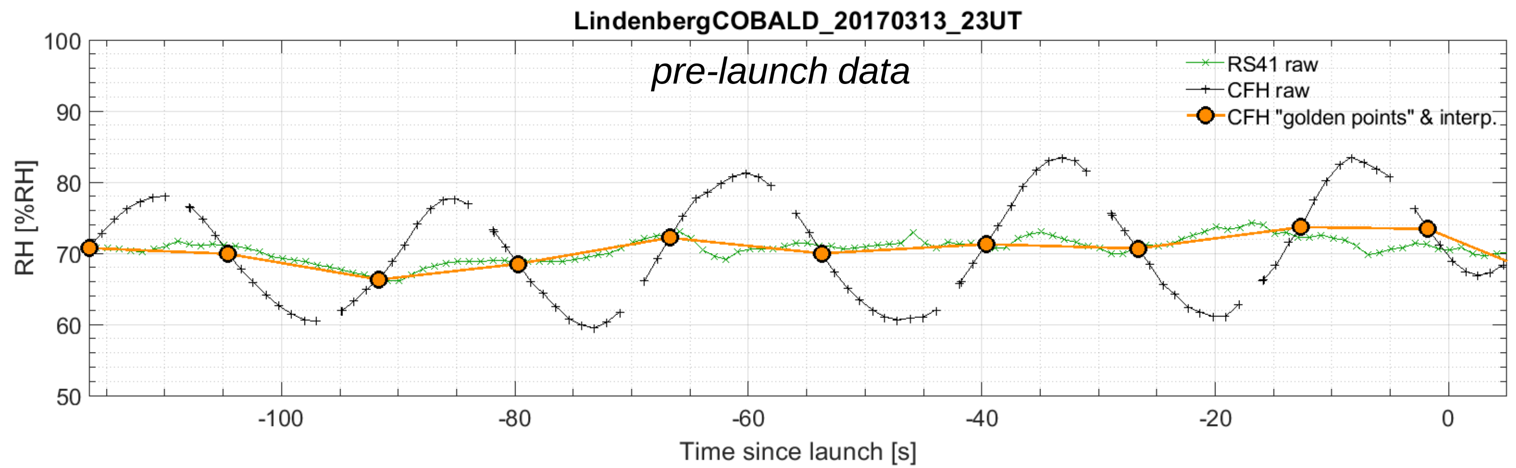
specular reflected light feedback

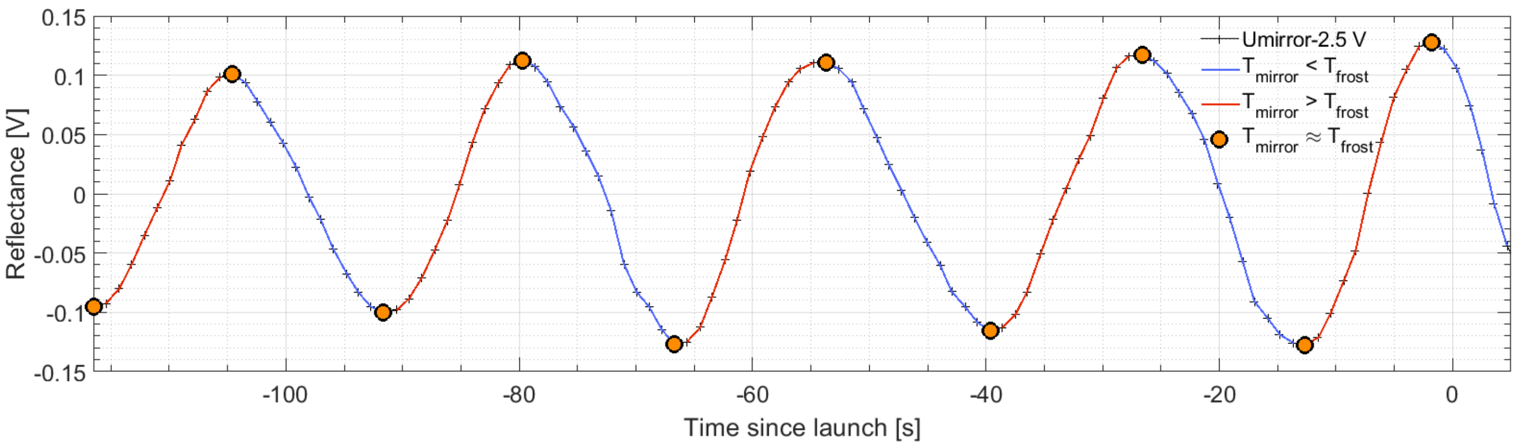


The specular "bulk reflectivitiy" decreases as mirror coverage increases and vice-versa (because of diffusion-scattering losses). Tom Peter's sketch.

Mirror Temperature, Mirror Reflectivity and “Golden Points” I

Chilled mirror instruments measure the **mirror temperature**.
However…
what we want is the **frost point temperature**.





Chilled mirror instruments measure also the **mirror reflectivity**.

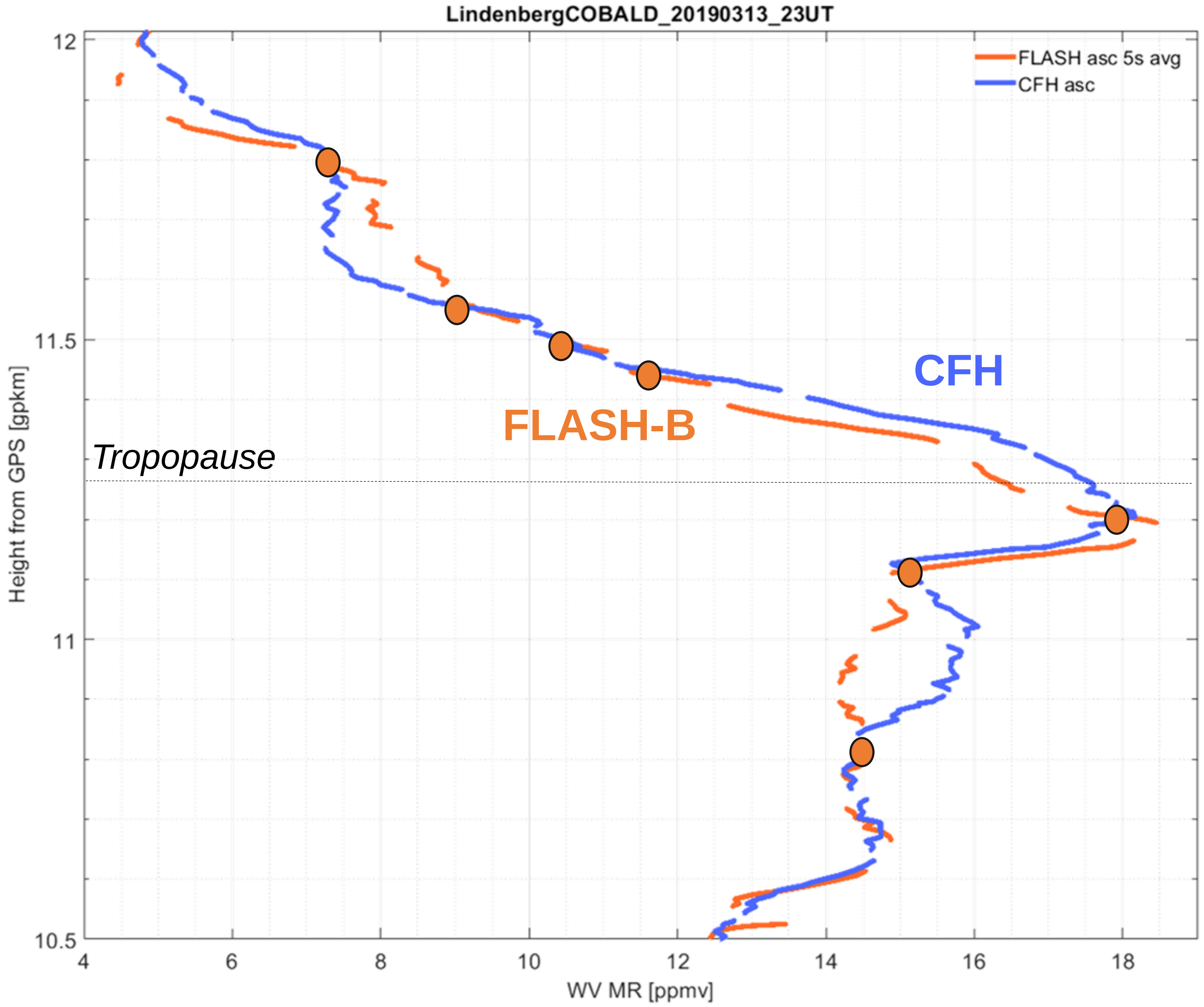
mirror reflectivity increases
-> mirror coverage decreases
-> condensate evaporates
-> **mirror too warm**

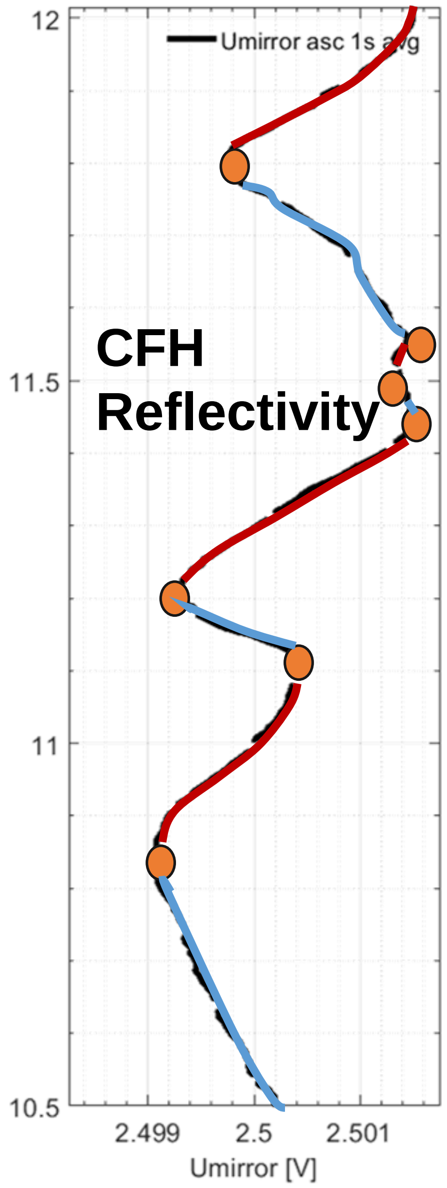
mirror reflectivity decreases
-> mirror coverage increases
-> condensate grows
-> **mirror too cold**

mirror reflectivity has a min/max
-> condensate transitions from growing-to-evaporating or evaporating-to-growing.
-> **Tmirror = Tfrost**

At those transition points, a.k.a. **“golden points”**, we obtain a *precise estimate* of the true atmospheric frost point.

Mirror Temperature, Mirror Reflectivity and "Golden Points" II





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In this example, the "**golden points**" allowed to:

- perform in-flight calibration of FLASH-B (see also Krämer et al. 2009)
- discover the existence of a fluorescence counting offset in this particular FLASH-B instrument.

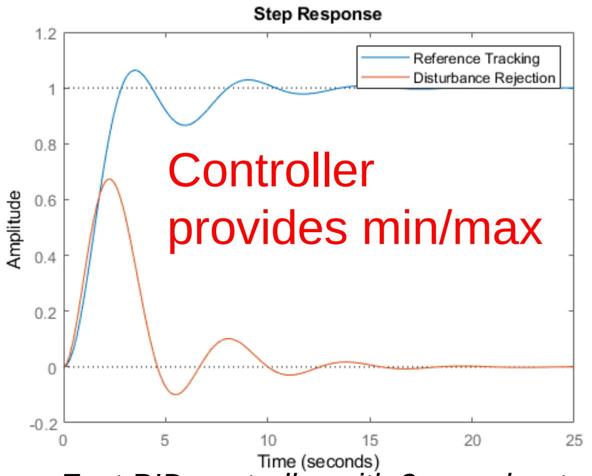
Golden points: summary

In principle, **any chilled mirror hygrometer** with an *accurately calibrated mirror temperature* provides several **accurate frost point temperature measurements** ("golden points") per balloon sounding.

- Useful to detect calibration, time-lag and/or offset errors, e.g. in RS41 or FLASH-B.
- Allows slow-responding CMH instruments to provide useful measurement points.

The number of golden points depends on:

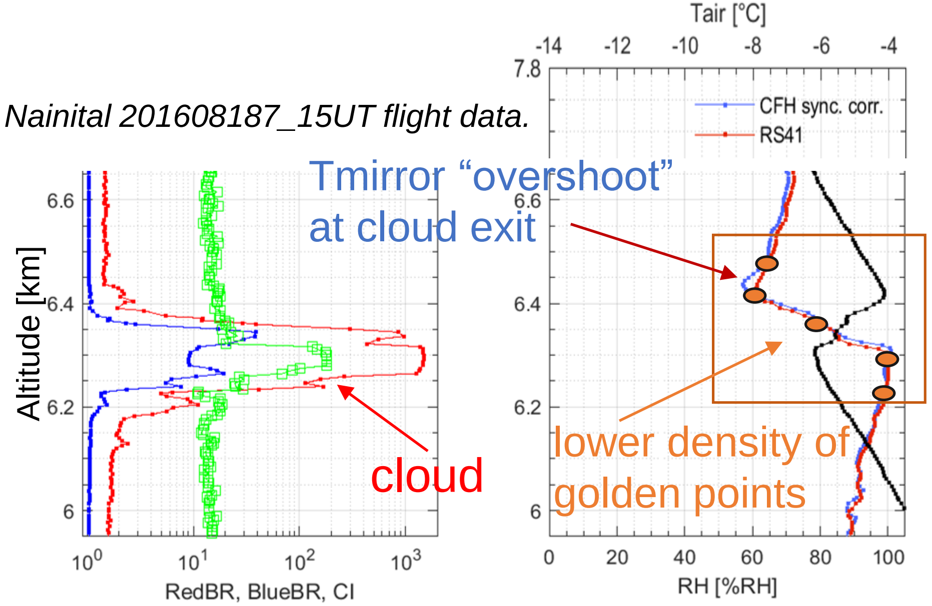
- the performance of the feedback controller (fast-responding vs. slow-responding)
- the nature of the condensate (fine frost vs. coarse frost)
- the state of the atmosphere (slowly-varying frost point / fast-varying frost point, good ventilation / poor ventilation).



Fast PID controller with 3 overshoots.

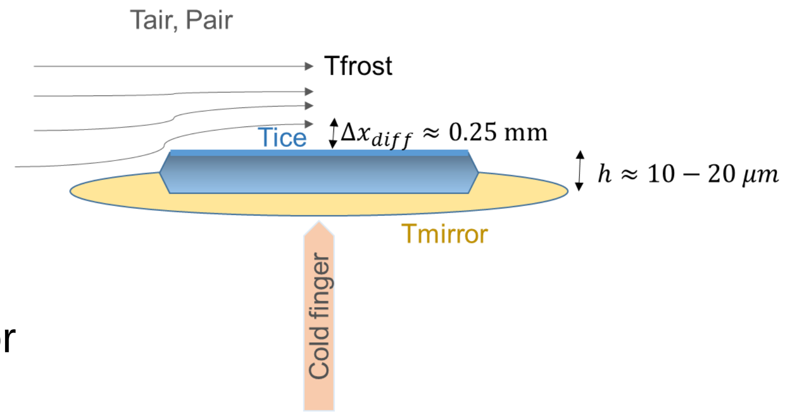
Polycrystalline frost layer on CFH. From Vömel et al. 2016

Nainital 201608187_15UT flight data.



Out-of-equilibrium theory I

We consider an infinite reservoir of water molecules (continuously renewing airflow)



<u>Fick's first law of diffusion, first order approximation,</u> gives:

$$\frac{dM}{dt} \approx D_{H_2O} \cdot \frac{\left(n_{H_2O}(T_{frost}) - n_{H_2O}(T_{ice})\right)}{\Delta x_{diff}}$$

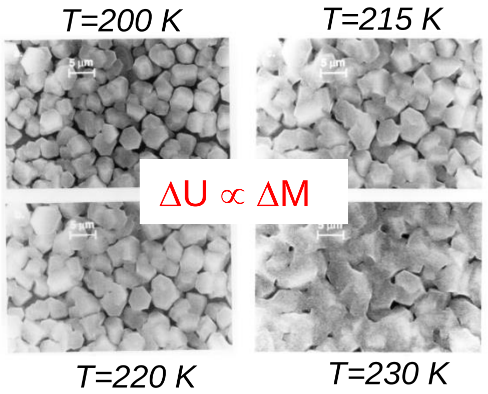
- $\frac{dM}{dt}$ in $\frac{mol}{m^2 s}$ the molar flux on the mirror.
- Δx_{diff} in m is the thickness of the boundary layer (aerodyn.) between the mirror and the ambient airflow.
- $n_{H_2O}(T) = \frac{e_{sat}(T)}{RT_{air}}$ in $\frac{mol}{m^3}$ the saturation molar number density at the temperature T.
- $D_{H_2O} \approx 2 \cdot 10^{-5} \cdot \frac{1013}{P_{air}} \cdot \left(\frac{T_{air}}{273.15}\right)^2$ in $\frac{m^2}{s}$ the gas-phase diffusion constant of water in air.

<u>The change in reflectivity</u> is assumed to be proportional to the molar flux:

$$\frac{dU_{mirror}}{dt} = -A' \cdot \frac{dM}{dt}$$

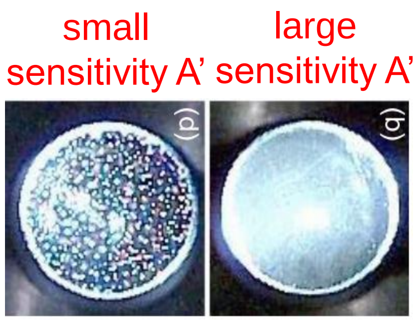
with A' a *sensitivity* value in $\frac{V}{\frac{mol}{m^2}}$.

The sensitivity value is assumed to be dependent on the morphology of the condensate layer and on the detection electronics.



Thermal annealing of an ice film formed by water vapour deposition on a polished aluminium plate at 180 K .

Leu and Kayser, 2009



Polycrystalline frost layer on CFH. Vömel et al. 2016

Out-of-equilibrium theory II

Recall:

$$\frac{dU_{mirror}}{dt} = -A' \cdot \frac{dM}{dt} \approx A' \cdot \frac{D_{H_2O}}{\Delta x_{diff}} \cdot \frac{\left(p_{sat}(T_{ice}) - p_{sat}\left(T_{frost}\right)\right)}{RT_{air}}$$

Approximations:

- $T_{ice} \approx T_{mirror}$ (because of assumed small thickness (~10-20 μm) and mass (~350-700 μg) of the condensate)
- $p_{sat}(T_{mirror}) - p_{sat}\left(T_{frost}\right) \approx \frac{H_{ice}}{R} \cdot \frac{p_{sat}(T_{mirror})}{T_{mirror}^2} \cdot (T_{mirror} - T_{frost})$ (as long as $|T_{mirror} - T_{frost}| < 2 - 5\ K$)
- $\Delta x_{diff} = \Delta x_{diff}(Re)$ ($\approx 0.25mm$ during balloon ascent, thicker in situations of poor ventilation such as during pre-launch)
- $A' \approx \text{constant}$ unique to the condensate once it has been formed and equilibrated.

Rewriting:

$$\frac{dU_{mirror}}{dt} = A \cdot \left(\frac{0.25mm}{\Delta x_{diff}(Re)} \cdot \frac{p_{sat}(T_{mirror})}{P_{air}} \cdot \frac{T_{air}}{T_{mirror}^2}\right) \cdot \left(T_{mirror} - T_{frost}\right)$$

$$T_{frost} = T_{mirror} - \left(\frac{\Delta x_{diff}(Re)}{0.25mm} \cdot \frac{P_{air}}{p_{sat}(T_{mirror})} \cdot \frac{T_{mirror}^2}{T_{air}}\right) \cdot \frac{1}{A} \cdot \frac{dU_{mirror}}{dt}$$

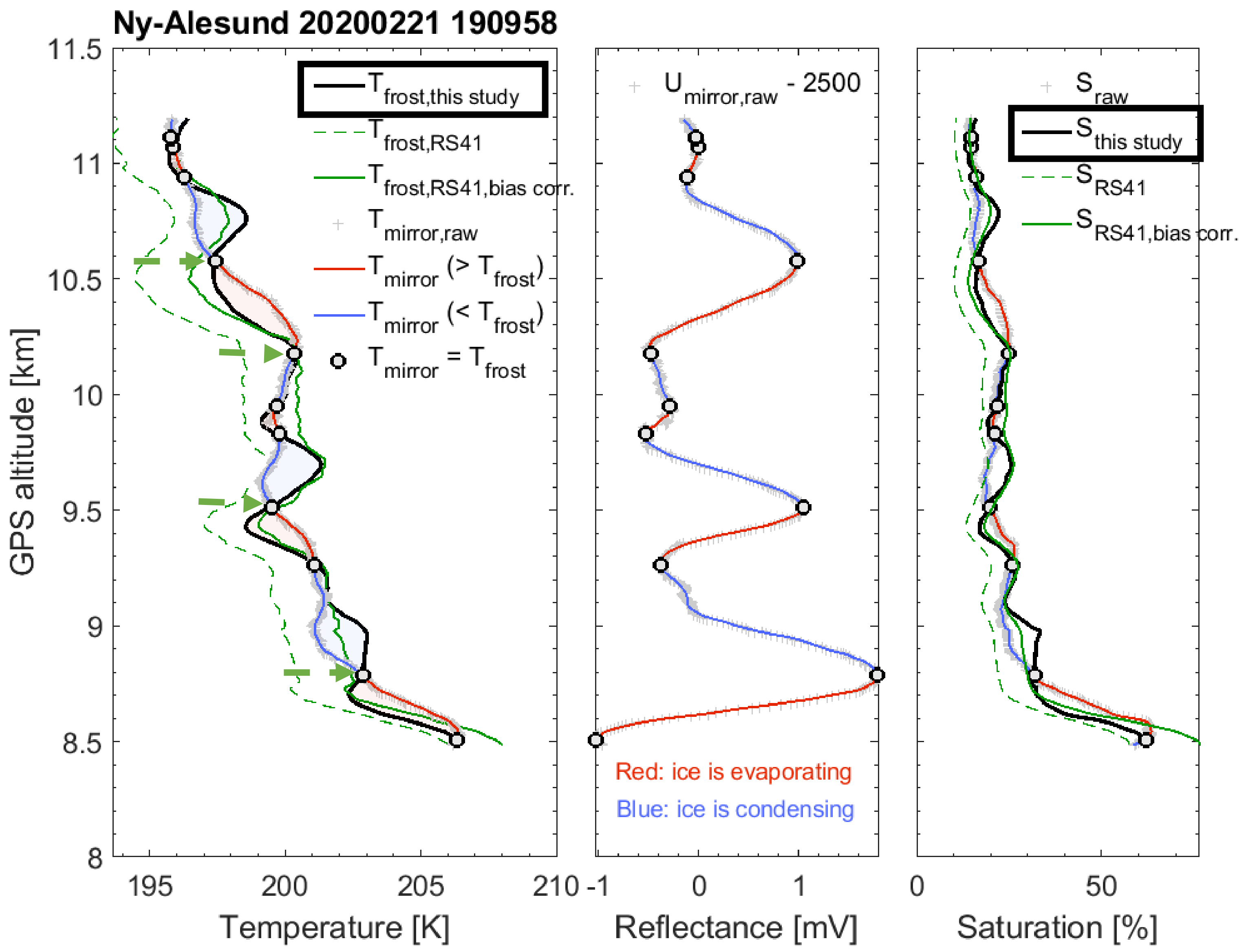
$$= T_{mirror} - B(T_{mirror}, T_{air}, P_{air}, w) \cdot \frac{1}{A} \cdot \frac{dU_{mirror}}{dt}$$

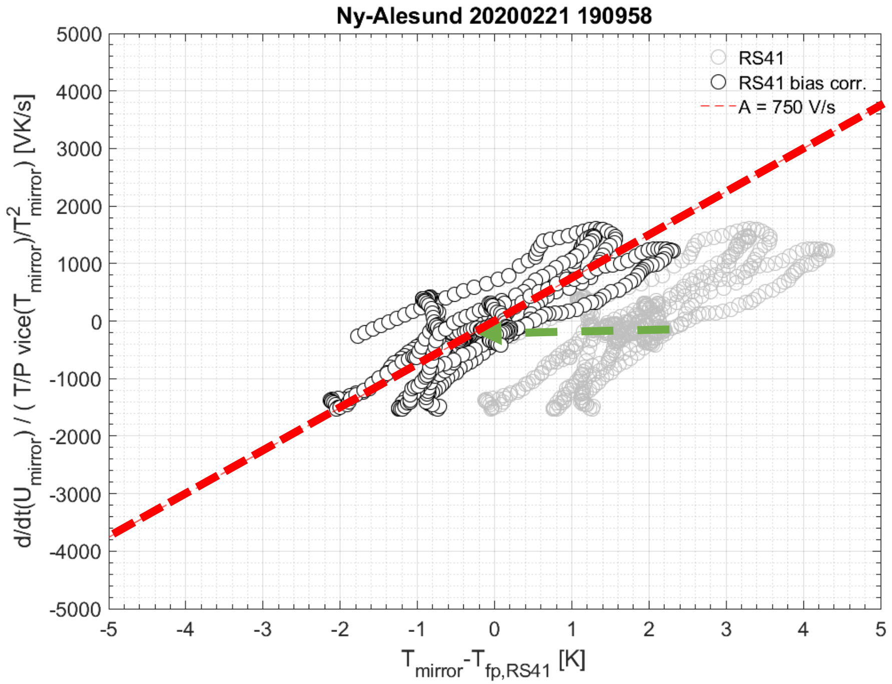
$A > 0$ sensitivity constant in $\frac{V}{s}$

→ If we have an estimate for A, the **frost point temperature can be corrected**

We can use the companion measurement (RS41, FLASH-B), or ascent/descent CFH data, to estimate A.

Correction example I (a): CFH and RS41





i) Bias correction of RS41 @ golden points:
$T_{f,rs41,corr.} = \alpha \cdot T_{f,rs41}^2 + \beta \cdot T_{f,rs41} + \gamma$

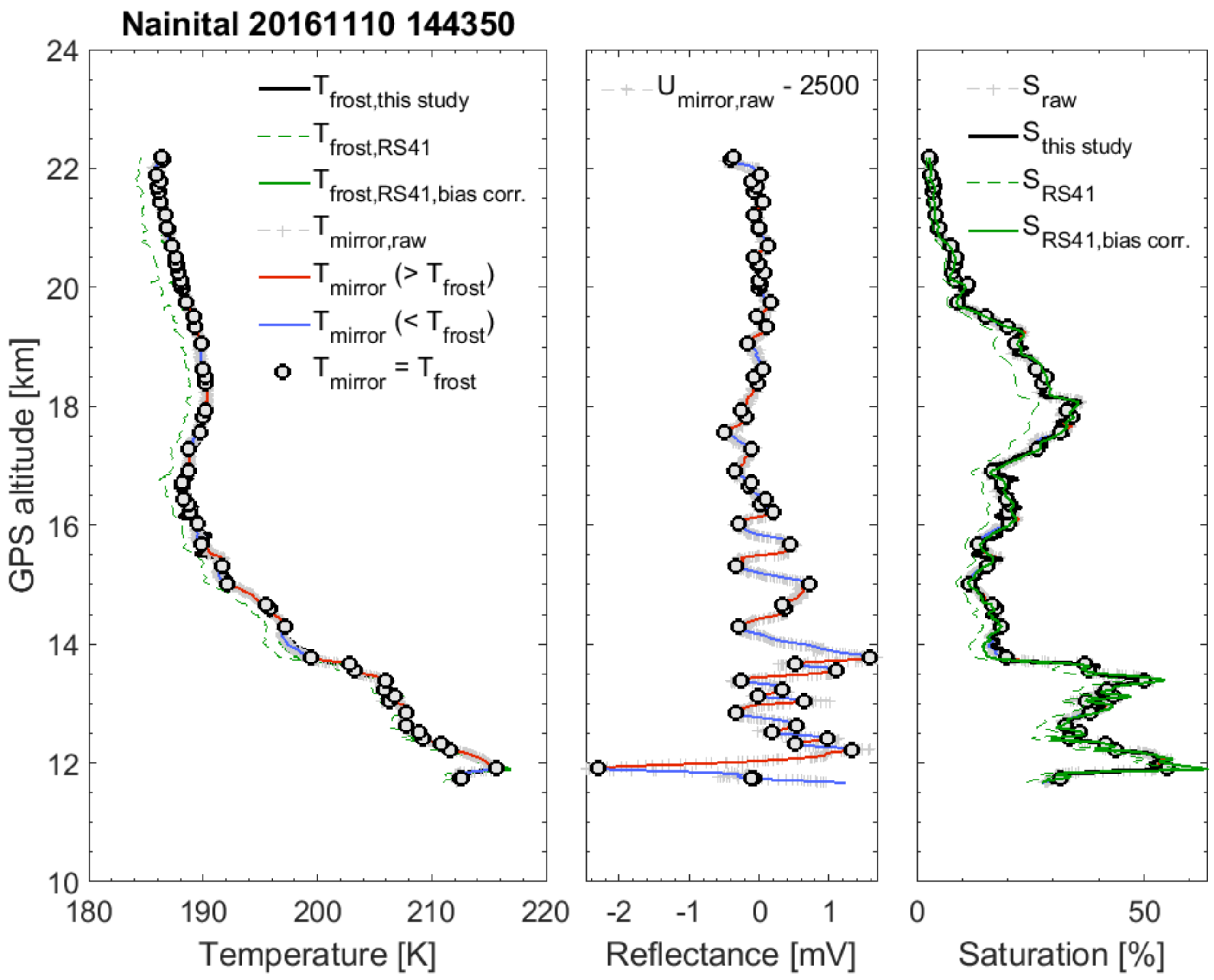
ii) Calculate the film sensitivity (slope) A

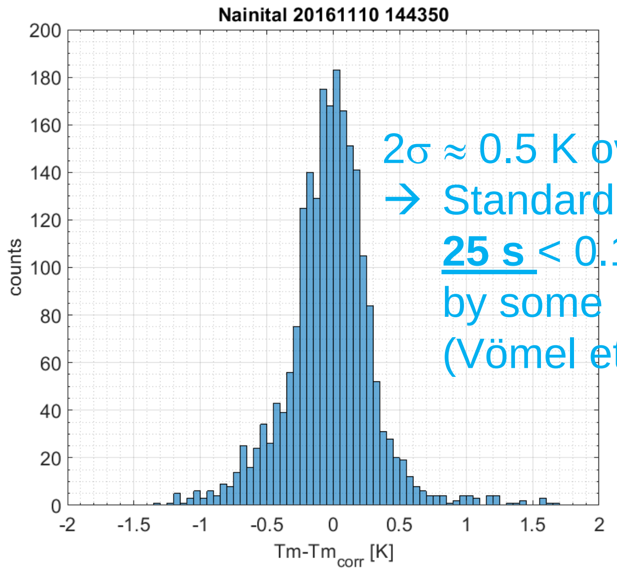
iii) Correct CFH:

$$T_{frost} = T_{mirror} - \frac{B}{A} \cdot \frac{dU_{mirror}}{dt}$$

with $B = B(T_{mirror}, T_{air}, P_{air}, w)$

Correction example I (b): when ice film sensitivity is high: small deviations Tmirror - Tfrost

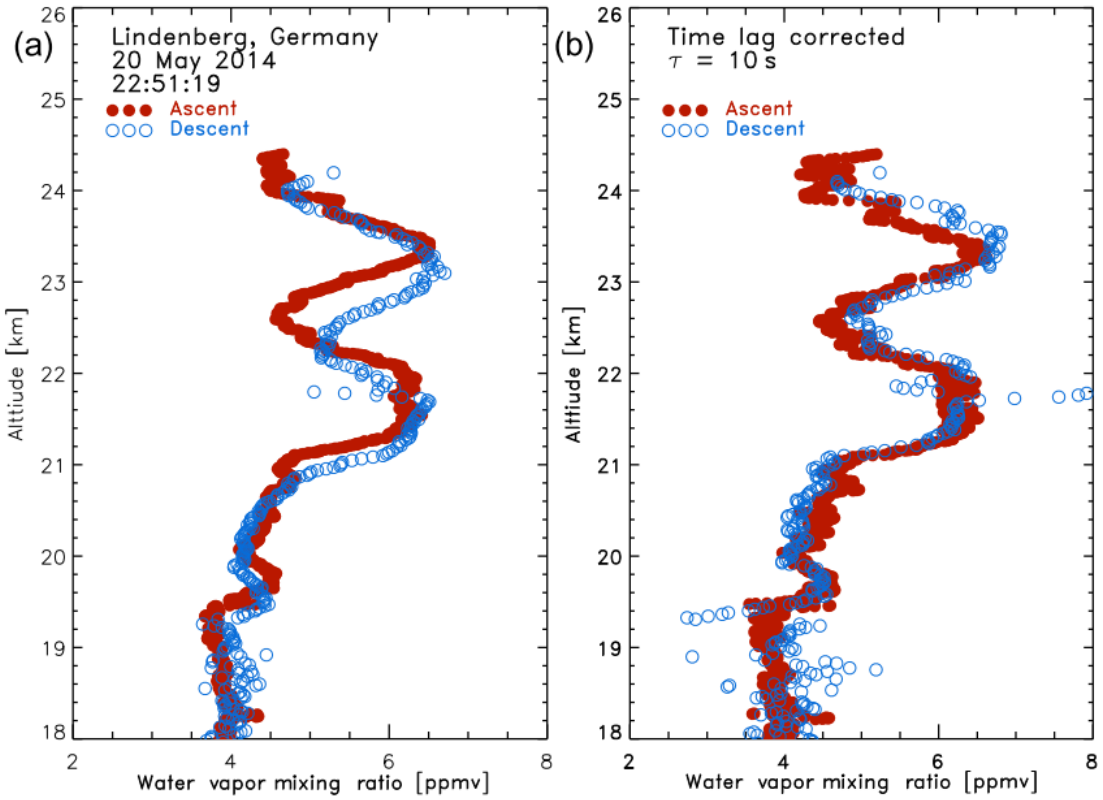




2σ ≈ 0.5 K over 1 s

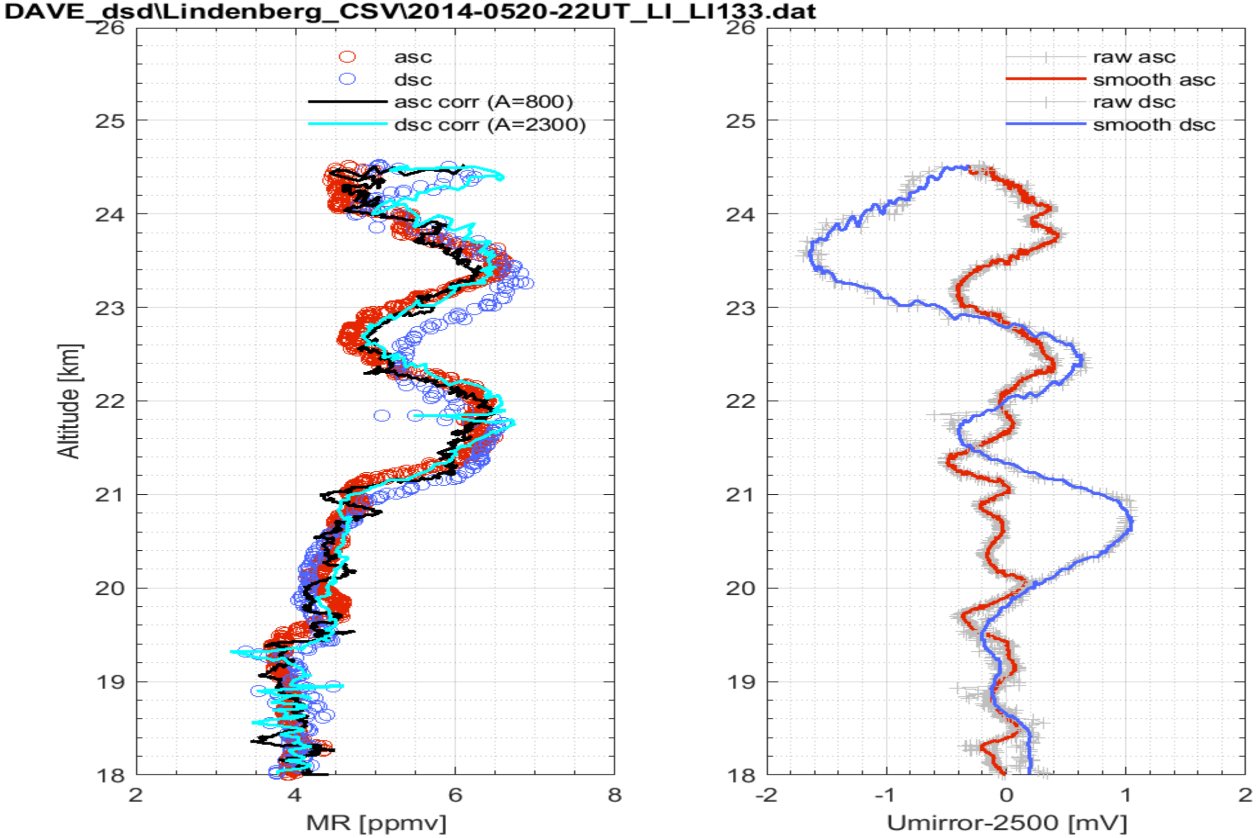
→ Standard error **over 25 s** < 0.1K achieved by some instruments (Vömel et al., 2007)

Correction example II: CFH ascent and descent



Vömel et al. 2016

Figure 2. Water vapor profile launched at Lindenberg, showing a pronounced vertical feature, which is again detected on descent. Left panel: raw data at 1 s resolution. Right panel: ascent and descent data time lag, corrected using a 10 s time lag constant.



Mixing ratio

original corrected

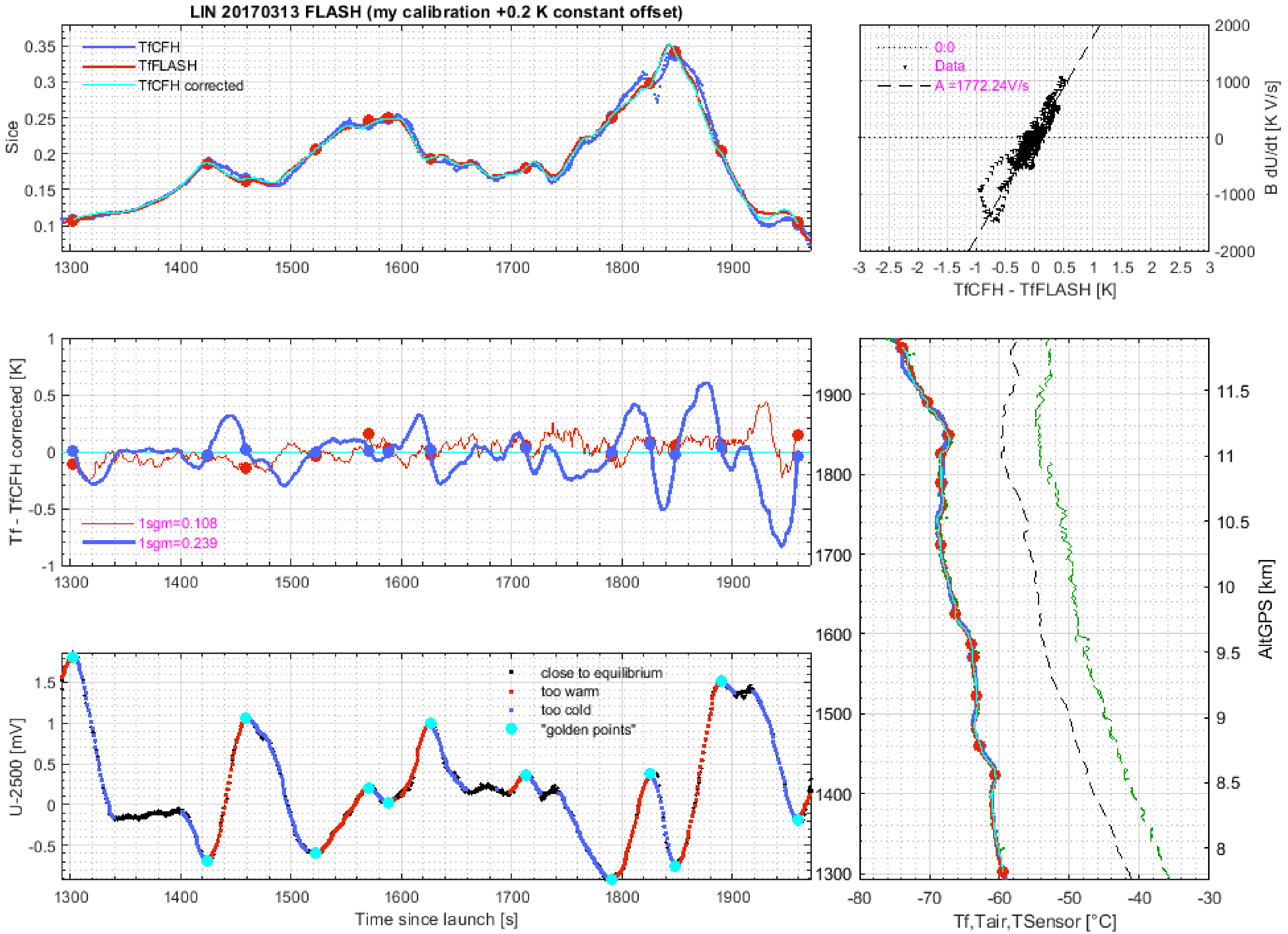
Reflectivity

$$T_{frost} = T_{mirror} + \tau_{strato} \cdot \frac{dT_{mirror}}{dt}$$

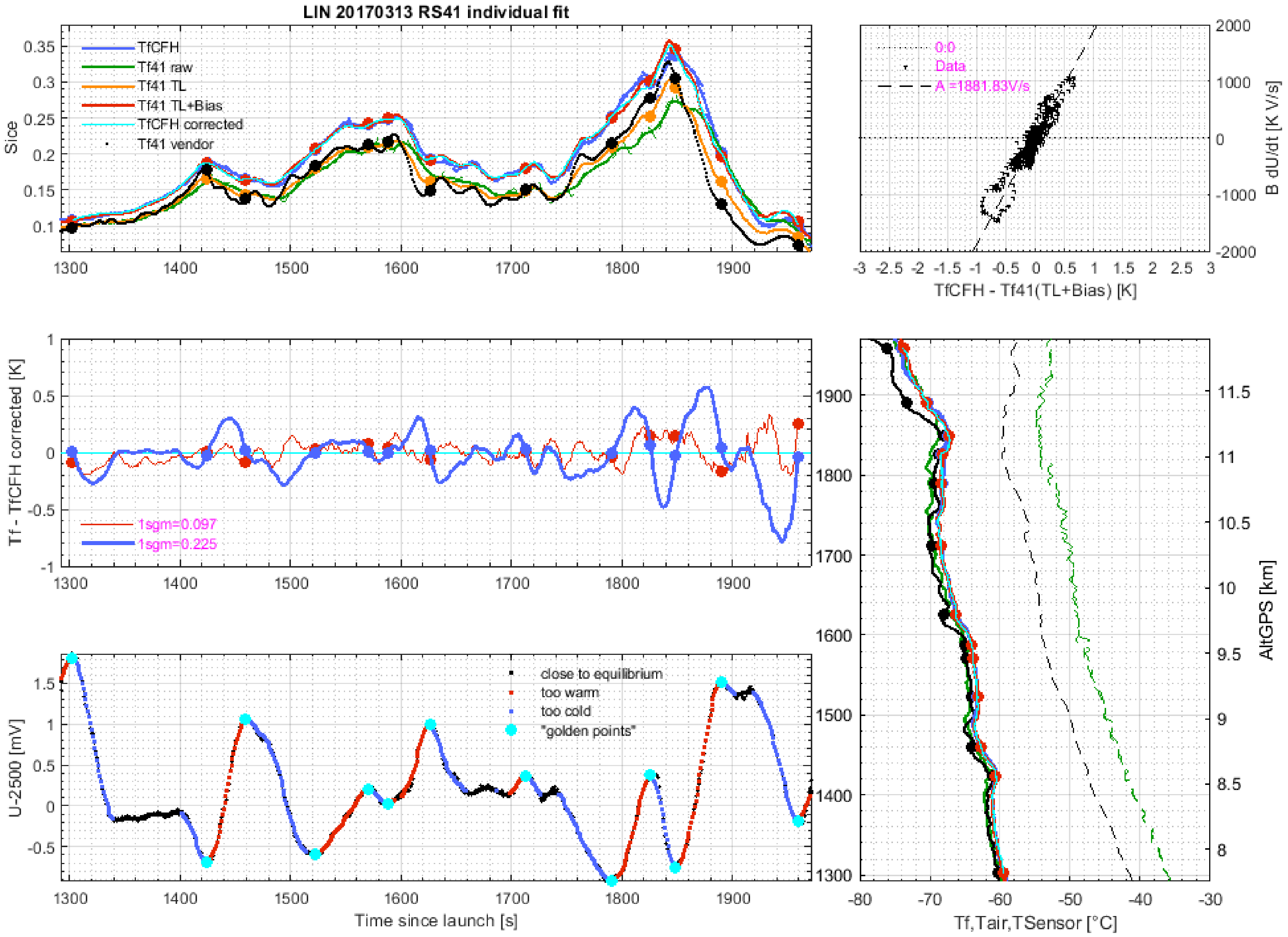
vs.

$$T_{frost} = T_{mirror} + \left(- \frac{\Delta x_{Diff}}{0.25\ mm} \cdot \frac{P_{air}}{p_{sat}(T_{mirror})} \cdot \frac{T_{mirror}^2}{T_{air}} \right) \cdot \frac{1}{A} \cdot \frac{1}{\frac{dT_{mirror}}{dU_{mirror}}} \cdot \frac{dT_{mirror}}{dt}$$

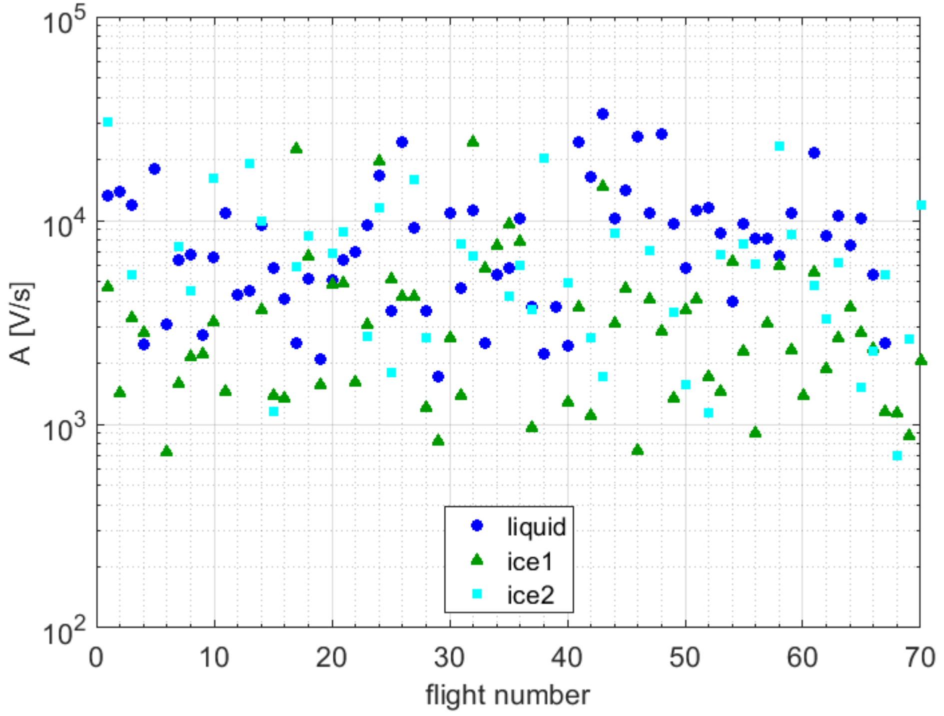
Correction example III: CFH and FLASH-B correction procedure

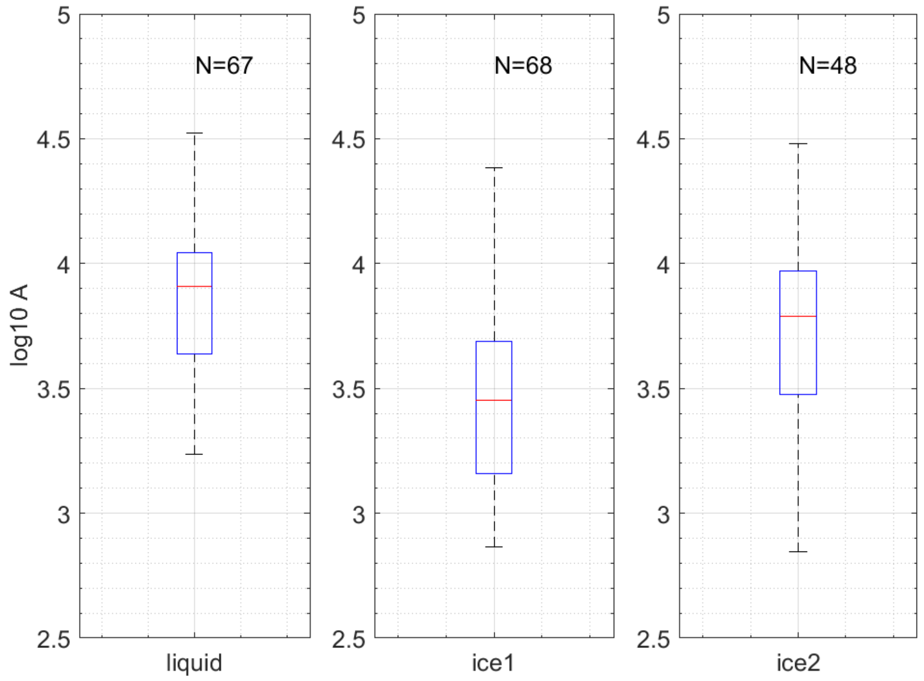


Correction example III: CFH and RS41 correction procedure



CFH-RS41 correction (70 nighttime flights): variability of A





The film sensitivities

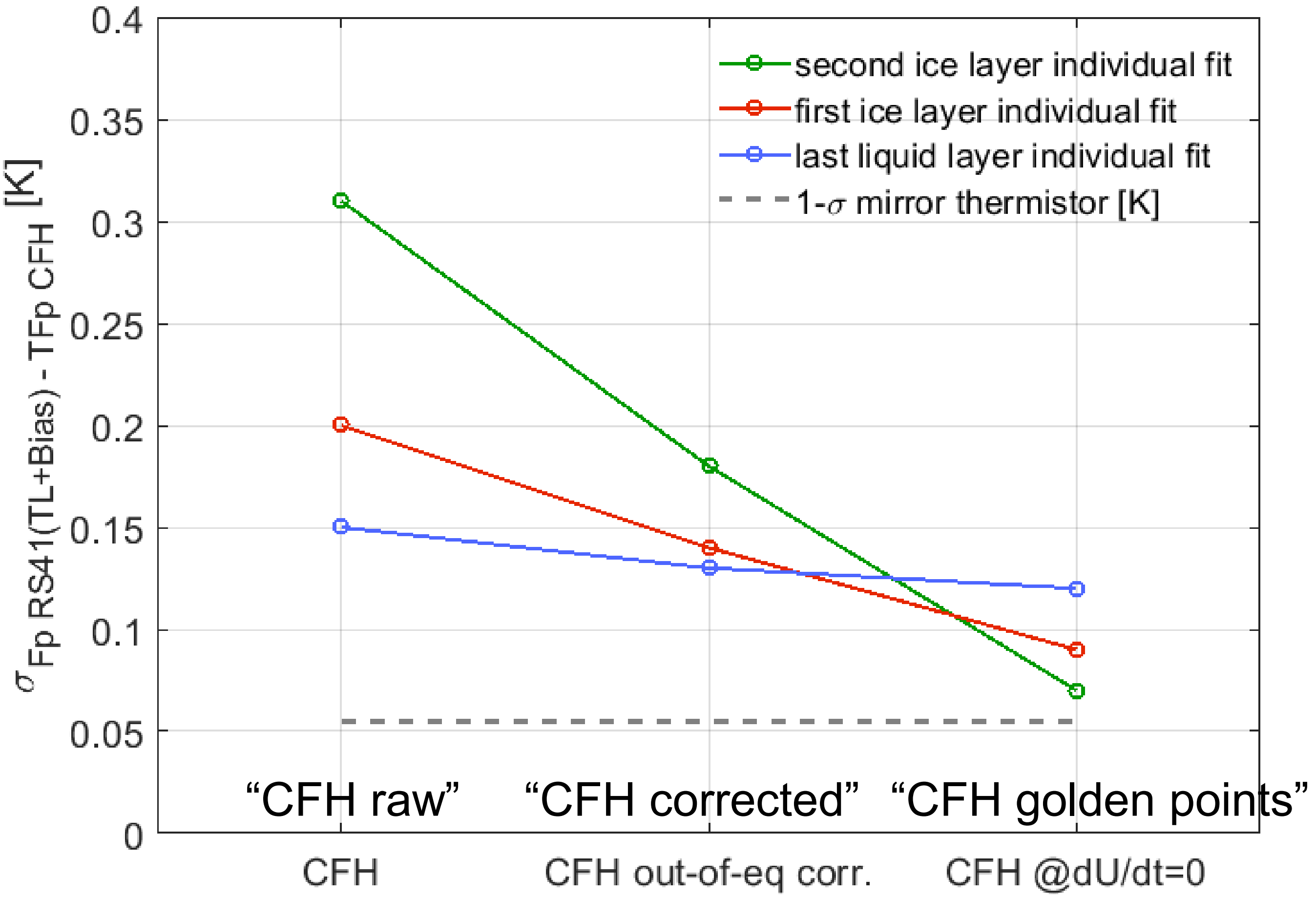
- span about 1 to 2 orders of magnitude
- vary from film-to-film and from flight-to-flight

Variability associated with different film morphologies, themselves associated to the mirror surface properties (impurities, coating, etc.) and to the prevailing temperature, ventilation and cooling rate during film formation.

- **First ice-film (formed at -15°C) has the smallest sensitivity** likely due to an ice film with larger ice-crystals and which is potentially more “patchy”.
- **Second ice film (formed at -53 °C) sensitivity has the largest scatter** likely due to large flight-to-flight differences in prevailing temperature, ventilation, cooling rates during film formation and mirror impurities.
- **Liquid film sensitivity is the highest and has the least scatter** likely due to the higher morphology consistency of dew.

CFH-RS41 correction (70 nighttime flights): residuals

residual error [K]



- UT/LS (second ice layer) benefits the most out of the out-of-equilibrium correction

  ~40% reduction of the residual error between raw and corrected, up to 80%-90% in some individual cases

- Golden Points' residual error is only slightly above the nominal uncertainty of the thermistor calibration.

Conclusions

CMH provide feedback of the thermodynamic equilibrium of their condensate through the reflectivity measurement. This allows for a new interpretation and correction method for CMH data.

At the golden points (dU/dt=0):

- The mirror temperature is the frost point with an accuracy **better than 0.2 K** (assuming 10 s smoothing to eliminate electronic noise).
- This corresponds to an uncertainty in H_2O partial pressure **better than 2-3% in the stratosphere and even less in the troposphere**.
- The golden points can be used to **calibrate other instruments** (e.g. offset, bias and time-lag correction).
- Based on these golden points, we have derived an improved time-lag and bias correction for the Vaisala RS41 radiosonde.

Outside of the golden points (|dU/dt|>0):

- A **correction can be achieved using simultaneous measurements** from a second instrument
- In the **worst cases, larger than 5K deviations** between mirror temperature and the estimated frost point are possible, but in general, they are **typically better than 0.5 K**.
- When the mirror temperature deviates significantly from the true atmospheric frost point, our **non-equilibrium correction may remove 80%-90% of the non-equilibrium error**, thereby **increasing significantly the vertical resolution** of the measurement. This happens typically for cases with coarse ice films and/or large mixing ratio changes in the atmosphere.

Thank you! Questions?