Biogenic Aerosols in the UTLS:
A New Challenge for Water Vapor Raman Lidars (and other techniques?)

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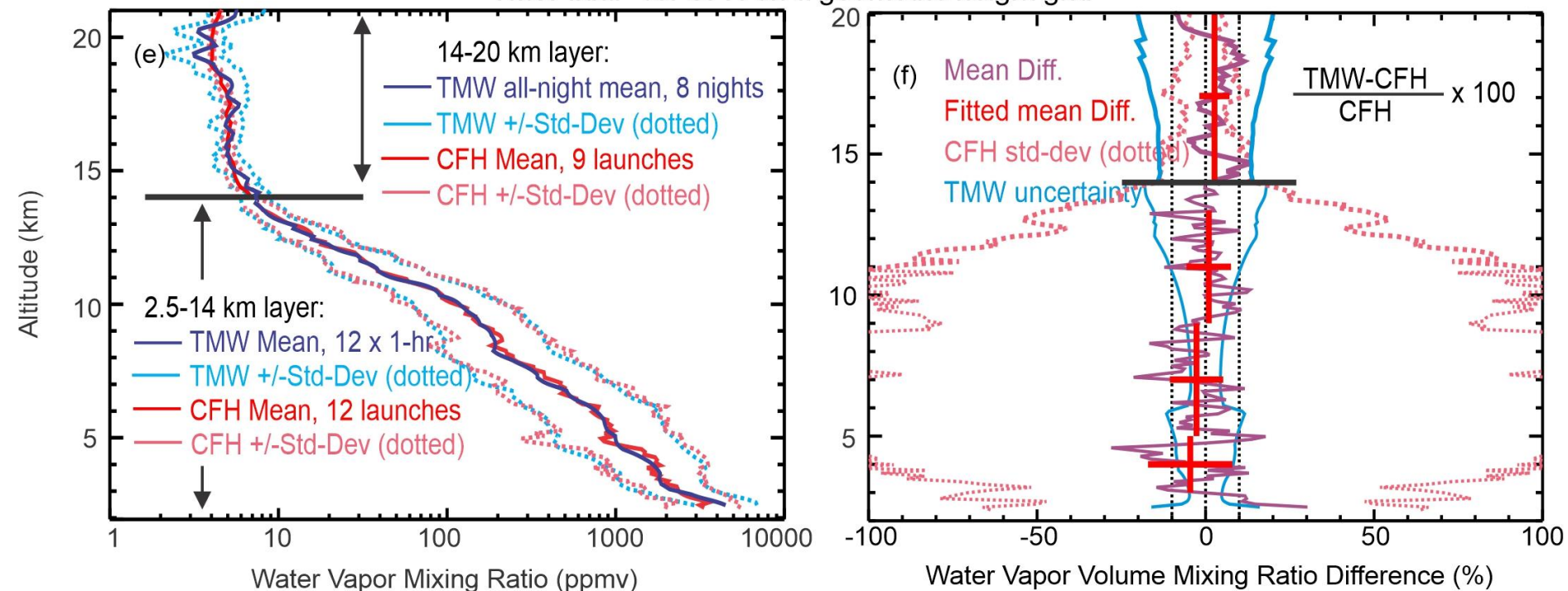
Jet Propulsion Laboratory, California Institute of Technology
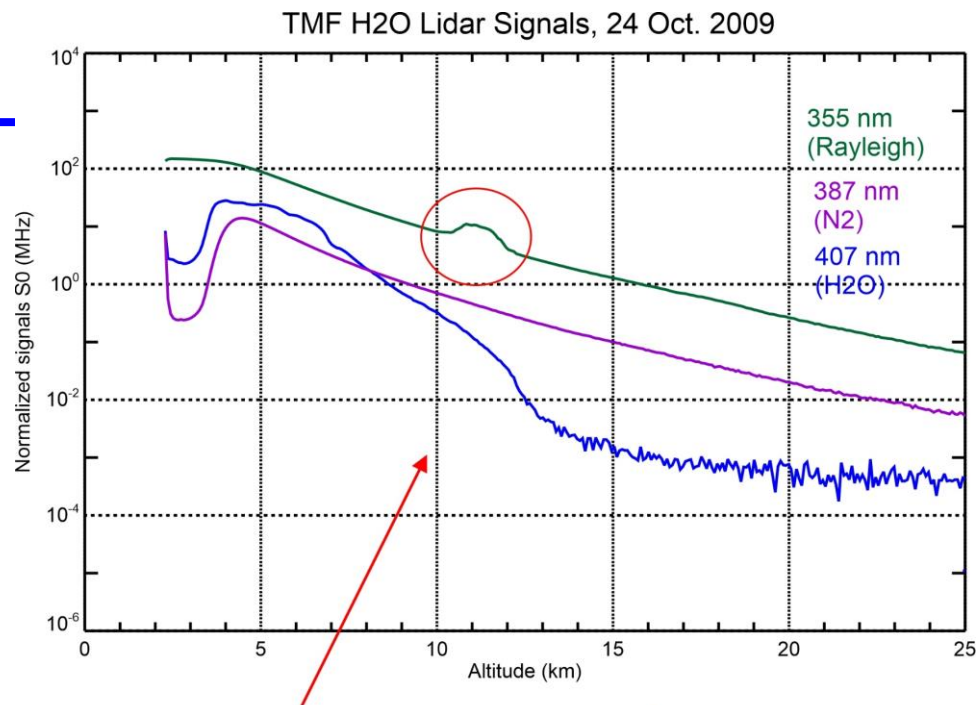
Wrightwood, California, USA

A simple Lidar technique:

1) Depending on laser type, emit laser beam at 355 nm or 532 nm

2) Receive Raman-shifted backscatter by atmospheric Nitrogen
   (at 387 nm or 608 nm)

3) Receive Raman-shifted backscatter by atmospheric water vapor
   (at 407 nm or 660 nm)

4) Calculate ratio of H2O to N2 signals

5) Calibrate to obtain a H2O VMR profile between ground and UTLS

Within 5% of CFH up to 20 km

TMW lidar vs. CFH on a geometric height grid

(e)

14-20 km layer:
— TMW all-night mean, 8 nights
— TMW +/-Std-Dev (dotted)
— CFH Mean, 9 launches
— CFH +/-Std-Dev (dotted)

2.5-14 km layer:
— TMW Mean, 12 x 1-hr
— TMW +/-Std-Dev (dotted)
— CFH Mean, 12 launches
— CFH +/-Std-Dev (dotted)

Altitude (km)

Water Vapor Mixing Ratio (ppmv)

(f)

Mean Diff.
Fitted mean Diff.
CFH std-dev (dotted)
TMW uncertainty

$$\frac{TMW - CFH}{CFH} \times 100$$

Water Vapor Volume Mixing Ratio Difference (%)

TMF H2O Lidar Signals, 24 Oct. 2009

355 nm (Rayleigh)

387 nm (N2)

407 nm (H2O)

Cloud signature at 11 km
(on Rayleigh channel only)

No cloud signature
on Raman water vapor

Lidar H2O VMR
(~407/387 ratio)
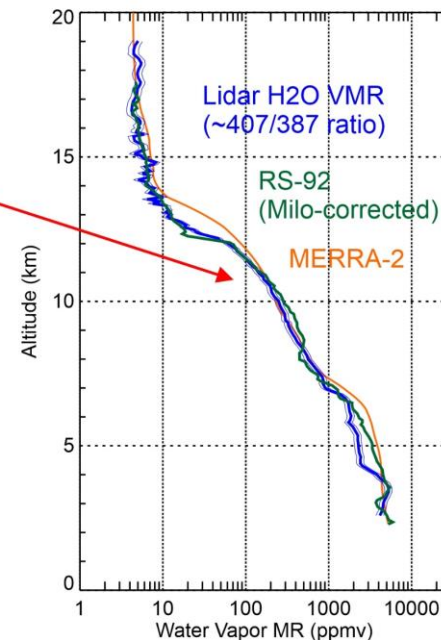
RS-92
(Milo-corrected)

MERRA-2

A good example, 2009 and many years after that:

On this day, thin clouds OK

➔ H2O profiles up to 20 km at TMF on a routine basis from 2009 to 2017

and then…..

TMF H2O Lidar Signals, 9 Sept. 2017
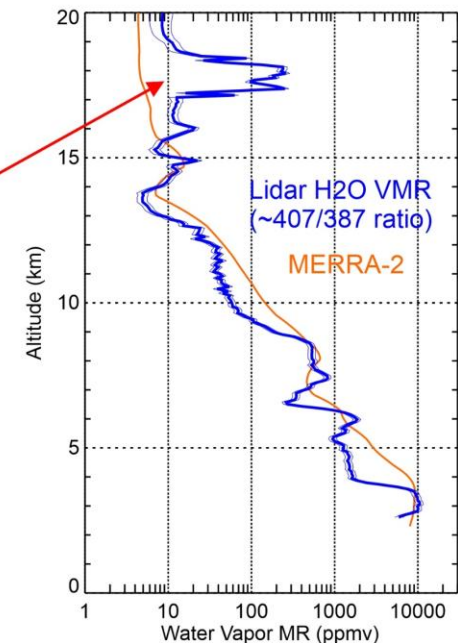

355 nm (Rayleigh)
387 nm (N2)
407 nm (H2O)
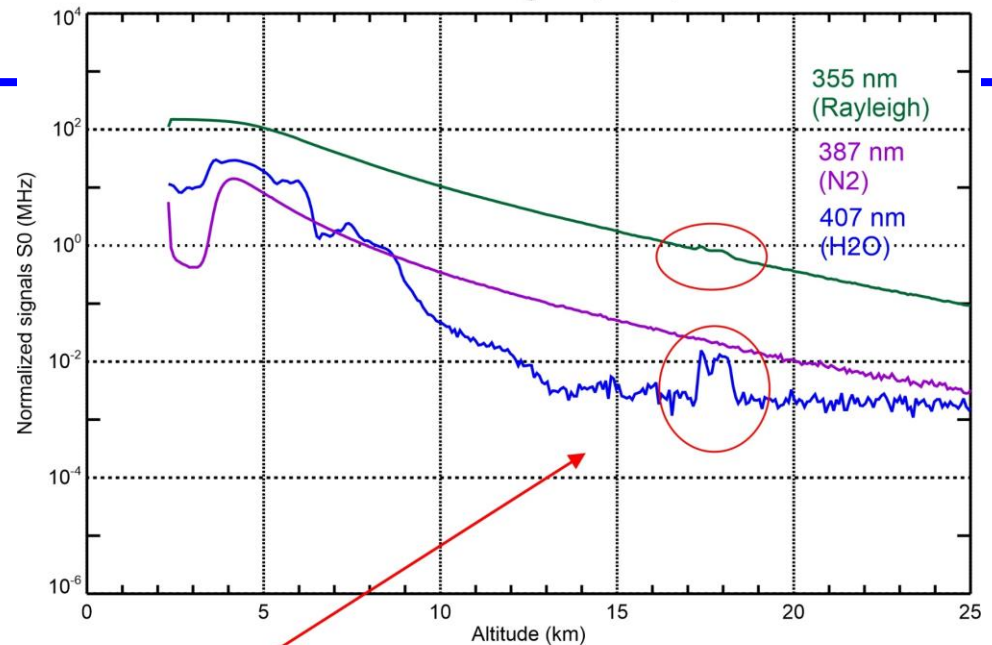
Sept. 2017, trouble really started:

No clouds

Only thin smoke,

but huge consequences

➜ H2O profiles must be cut-off at 15 km on even lower

This time, signature at 18 km = Smoke

Barely seen on Rayleigh channel but
Large on H2O channel!

Huge signature on Raman water vapor

Lidar H2O VMR (~407/387 ratio)

MERRA-2

Nothing new about this

Immler et al., 2005

And a few others

But the worse was yet to come....

Fluorescence from atmospheric aerosol detected by a lidar indicates biogenic particles in the lowermost stratosphere

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Abstract. With a lidar system that was installed in Lindenberg/Germany, we observed in June 2003 an extended aerosol layer at 13 km altitude in the lowermost stratosphere. This layer created an inelastic backscatter signal that we detected with a water vapour Raman channel, but that was not produced by Raman scattering. Also, we find evidence for inelastic scattering from a smoke plume from a forest fire that we observed in the troposphere. We interpret the unexpected properties of these aerosols as fluorescence induced by the laser beam at organic components of the aerosol particles. Fluorescence from ambient aerosol had not yet been considered detectable by lidar systems. However, organic compounds such as polycyclic aromatic hydrocarbons sticking to the aerosol particles, or bioaerosol such as bacteria, spores or pollen fluoresce when excited with UV-radiation in a way that is detectable by our lidar system. Therefore, we conclude that fluorescence from organic material released by biomass burning creates, inelastic backscatter signals that we measured with our instrument and thus demonstrate a new and powerful way to characterize aerosols by a remote sensing technique. The stratospheric aerosol layer that we have observed in Lindenberg for three consecutive days is likely to be a remnant from Siberian forest fire plumes lifted across the tropopause and transported around the globe.

(soot), or they form in situ by gas to particle conversion, like sulphate aerosol. While particles may enter the stratosphere in the tropics (Brock et al., 1995), the tropopause in the mid latitude efficiently suppresses tropospheric-stratospheric transport (TST) (Holton et al., 1995). However, recent observation of stratospheric aerosol layers by satellite-borne instruments and ground-based lidar suggest that strong thunderstorms are able to inject smoke from forest fires into the stratosphere at high latitudes (Fromm et al., 2000). Latest in situ measurements also proof the presence of forest fire particles in the stratosphere (Jost et al., 2004). Even though these layers are optically thin, they are highly relevant because when aerosols leave the troposphere they escape their most efficient removal mechanism which is wet scavenging and consequently their lifetimes are strongly increased. The implications of particles in the stratosphere on the radiative balance and chemistry crucially depend on their physical and chemical properties. Ground or satellite based sensors may measure the global distribution of the optical depth of aerosols but usually provide little information on the exact type and source of the particles (Kaufman et al., 2002).

Lidar is a powerful tool to investigate atmospheric aerosol since it makes it possible to measure its distribution from near to the ground up to the upper atmosphere with high vertical and temporal resolution. The different lidar techniques applicable to aerosol measurements like multi-wavelength
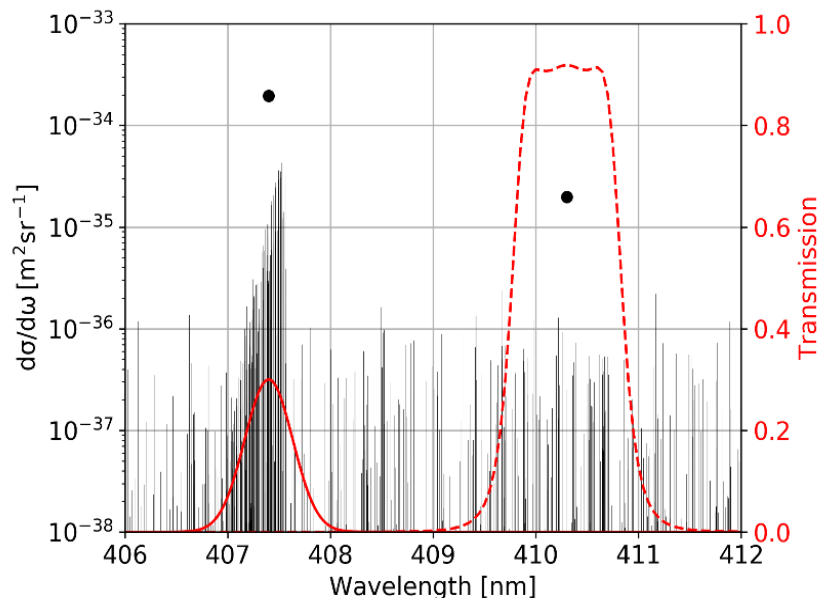
2009-2021 timeseries
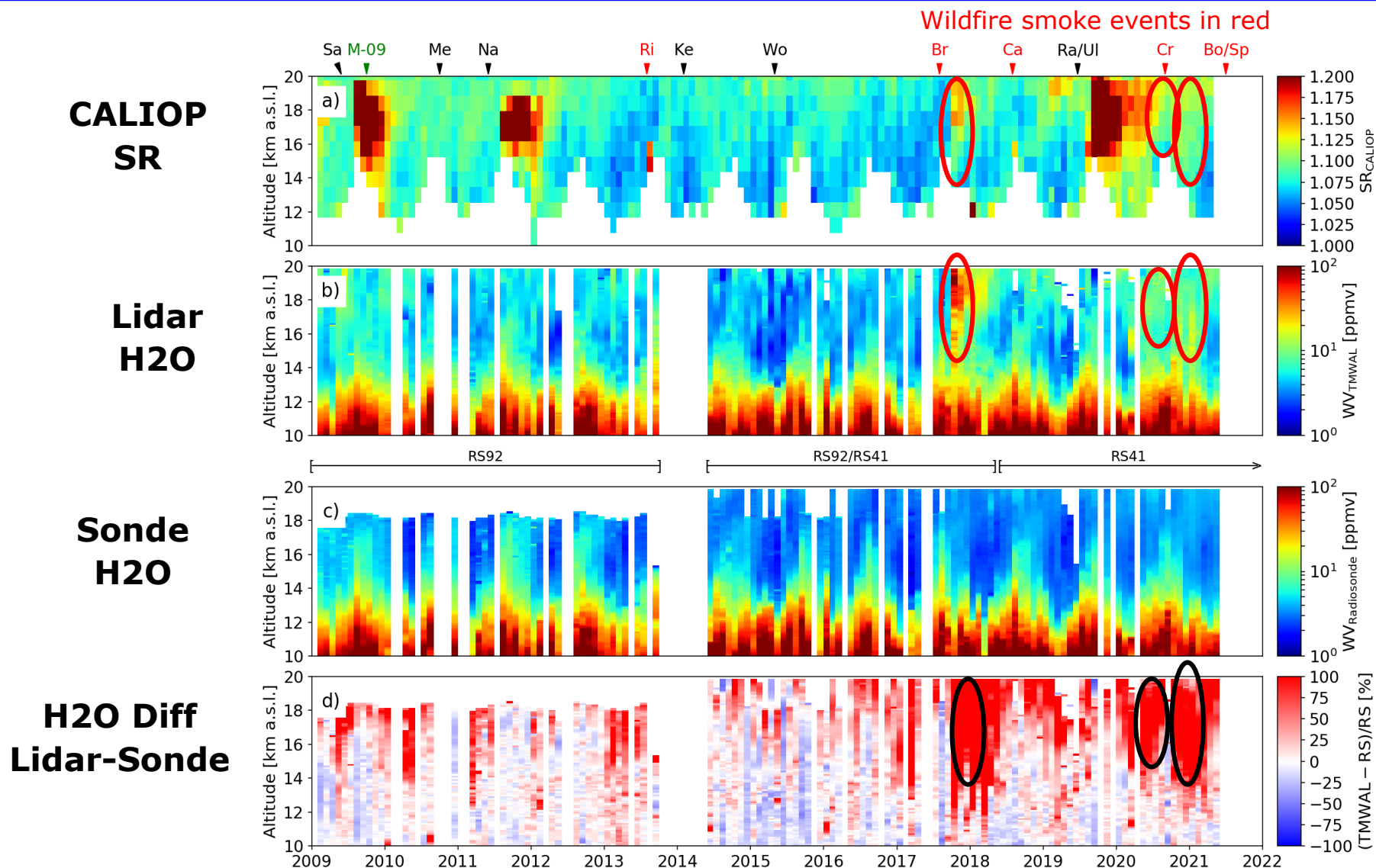
Large positive bias ONLY when smoke is detected

GRUAN ICM-13 Virtual Meeting, Nov 15-19, 2021

1) Add a "fluorescence channel" at 410 nm using a filter wheel in front of the 407 PMT


At 410 nm:

- the lidar will receive only 3-5% of the water vapor signal received at 407 nm

- assuming nearly-flat fluorescence spectrum between 407 and 410 nm,
  the lidar will receive as much fluorescence as received at 407 nm

2) Calibrate the 410 nm channel w.r.t. the 407 nm channel using either lamp
   or sky background

3) Once inter-calibrated, subtract fluorescence received at 410 nm from the 407 nm channel

4) What is left is actual H2O VMR, i.e., corrected form fluorescence
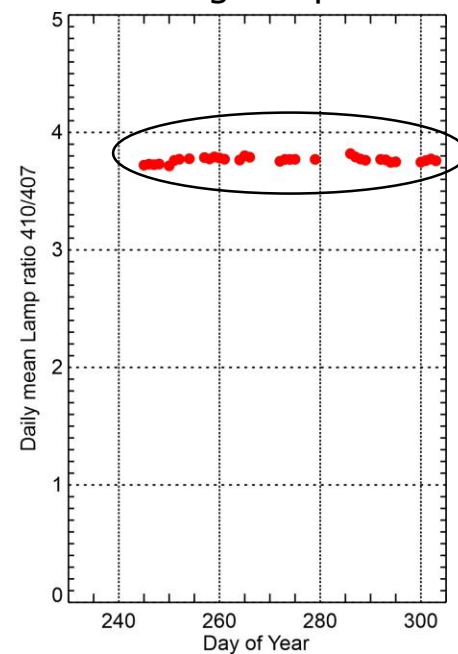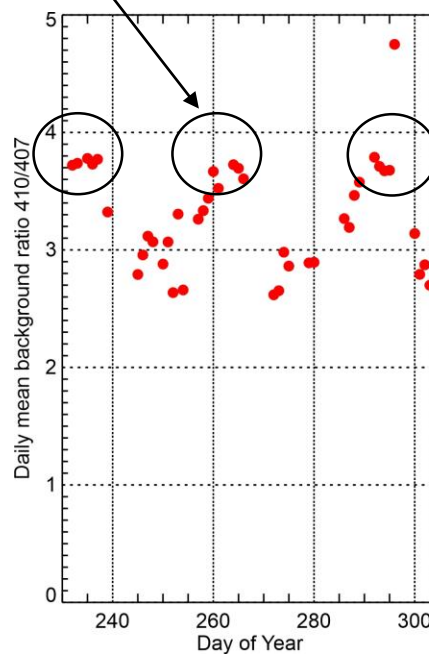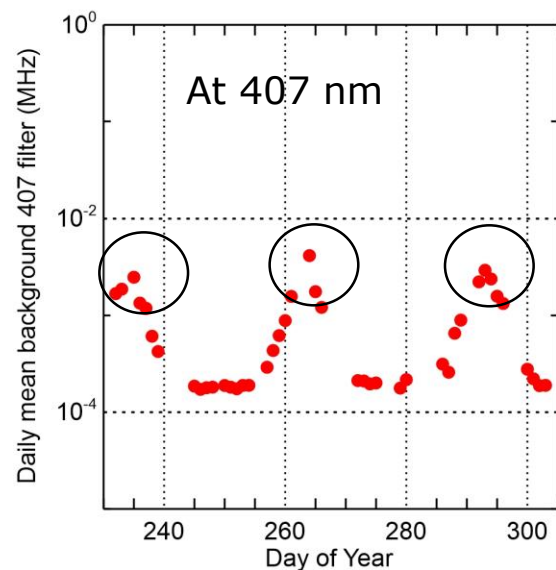
Sky background vs. day of year

At 410 nm
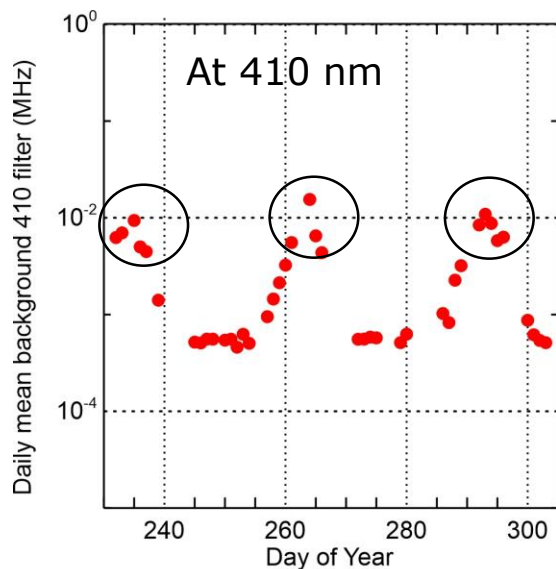
Use sky background intensity during full moon

Ratio of sky background during full moon…   …matches well…

…ratio of 410 to 407 during lamp runs

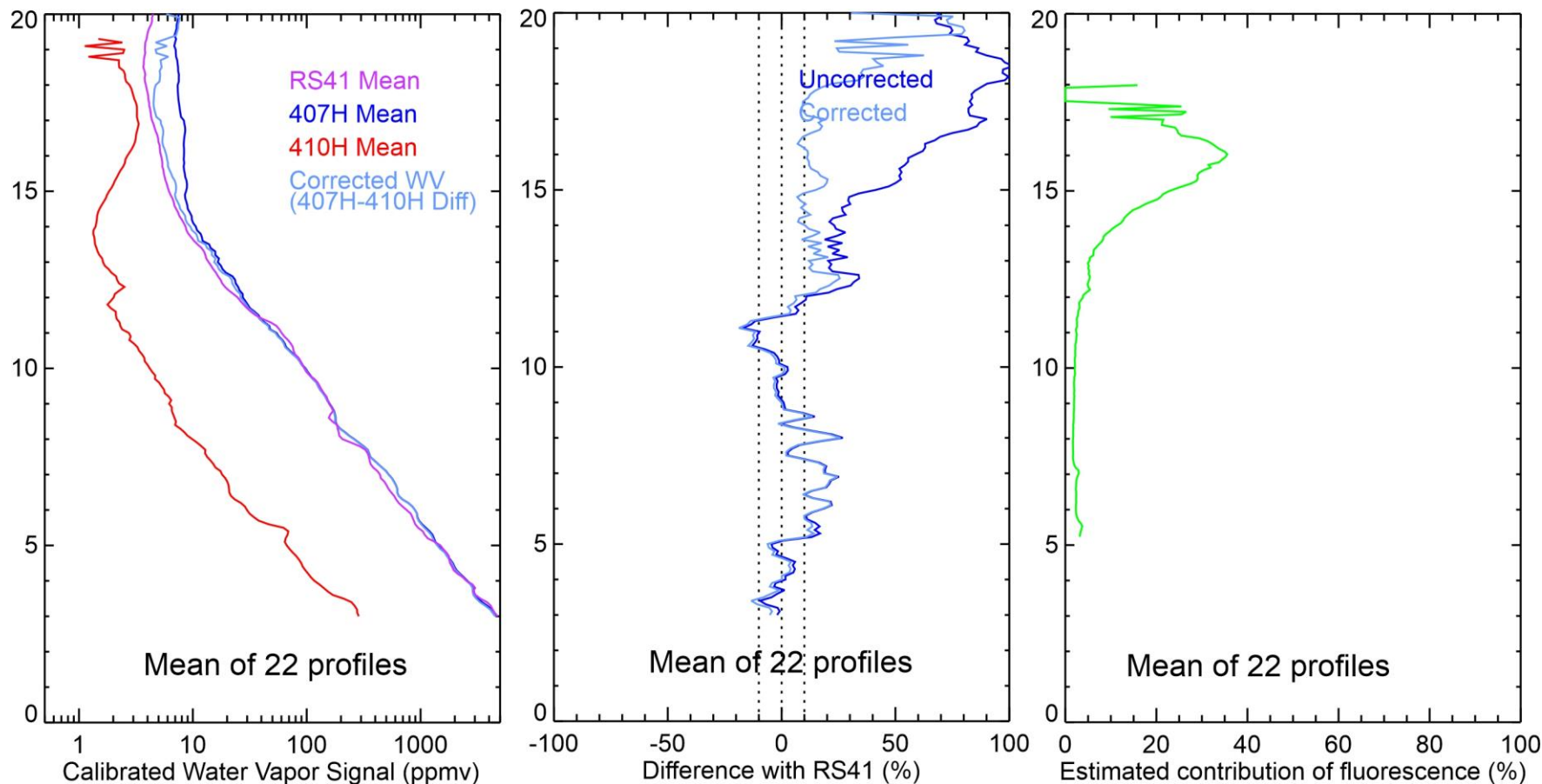At 407 nm

➔ 410/407 inter-calibration = 3.8

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Left plot:
Dark blue = Uncorrected H2O VMR
Red = "fluorescence" profile scaled to H2O VMR
Light blue = H2O VMR after fluorescence is subtracted

Middle plot: Diff with RS41
Dark blue = uncorrected
Light blue = corrected

Right plot (green curve):
Relative contribution of
fluorescence

➜ H2O VMR can be corrected to approach 5-10% accuracy

CONCLUSION

1) H2O Raman Lidar has been (and still is) a nice "Ancillary Measurement" technique for water vapor up to the UTLS, as proved over the period 2009-2017

But…

2) UTLS has become significantly "dirtier" since the recent increase of wildfire activity in Siberia and North America (also Australia)

3) This increase revealed an important caveat for the long-term monitoring of H2O by Raman lidar, with the necessity to develop a robust and accurate fluorescence correction

4) Fluorescence contamination is obvious for UV lidars (emission at 355 nm) but is yet to be demonstrated for lidars with emission at 532 nm

5) For UV lidars, fluorescence correction is possible by simply adding a 410 nm channel as a "cheap fix"

6) Adding a spectrometer instead of a single channel will likely be a more accurate solution as it will take into account the wavelength dependence of the fluorescence spectrum