Evolution of Arctic springtime trace gas species between 2006 and 2017

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Motivation and the PARIS-IR Instrument

The chemical composition of the Arctic atmosphere is influenced by both natural and anthropogenic effects. As atmospheric composition is tied to climate, studying the changes in trace gas species can lead to a more thorough understanding of the changing Arctic conditions[1]. As part of the Canadian ACE/OSIRIS Arctic Validation Campaigns, which began in 2004, remote sensing measurements of trace gases have been made on a yearly basis at the Polar Environment Atmospheric Research Laboratory (PEARL; 80.05° N, 86.42° W; shown below) located in Eureka, Nunavut. These yearly validation campaigns commence in late February, shortly after polar sunrise, and continue for approximately six weeks into early April. In addition to serving to validate the ACE-FTS, ACE-MAESTRO and Odin-OSIRIS instruments, these missions provide invaluable opportunities to measure changing polar trace gas concentrations.

Figure 1 (left): The Polar Environment Atmospheric Research Laboratory (PEARL)
Figure 2 (right): Location of PEARL Ridge Lab in Eureka, Nunavut.

One of the instruments involved in these campaigns is the Portable Atmospheric Research Interferometric Spectrometer for the InfraRed (PARIS-IR; shown below). PARIS-IR is a compact portable high-resolution Fourier Transform Spectrometer built as a terrestrial version of the Atmospheric Chemistry Experiment - Fourier Transform Spectrometer (ACE-FTS)[2-4]. PARIS-IR records solar absorption spectra within the range of 750 - 4400 cm^{-1} with 0.02 cm^{-1} of spectral resolution. Since 2004 PARIS-IR has been brought to the PEARL Ridge Lab as part of the Canadian ACE/OSIRIS Arctic Validation Campaigns, and over the campaign it makes a measurement, consisting of 20 co-added scans, every approximately seven minutes, weather permitting[2]. A suntracker on the roof of PEARL and a pickoff mirror in the lab directs sunlight into PARIS-IR. The focus of this study is to build upon the work of Griffin et al. (2017) who analyzed changes in eight trace gas species (O$_3$, HNO$_3$, HCl, HF, CH$_4$, C$_2$H$_6$, N$_2$O and CO) retrieved from the PARIS-IR springtime measurements for the eight-year period spanning 2006 to 2013[2]. To this end, an additional four years, 2014 to 2017, of trace gas measurements for these eight species will be examined, and progress towards the retrieval of a ninth species, water vapor, will be shown.

Sunlight from suntracker
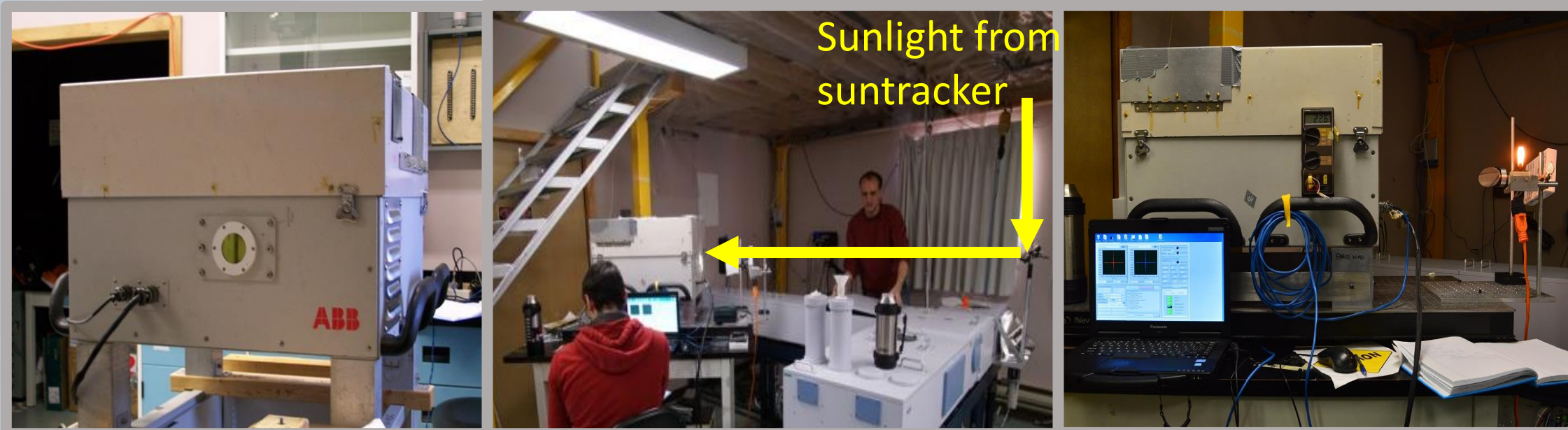
Figure 3 (left): PARIS-IR
Figure 4 (middle): PARIS-IR making solar spectra measurements in the PEARL Ridge Lab (Photo credit: Erik Lutsch)
Figure 5 (right): PARIS-IR recording glo-bar test measurements

Methods for Filtering, Retrievals, and Analysis

The interferograms recorded by PARIS-IR are Fourier transformed into raw spectra, which are filtered for: detector saturation; varying cloud cover during the measurements; overall cloud cover; and other instrumental errors that contribute to poor spectral quality. After filtering, an Optimal Estimation Method (OEM) based technique is applied to the data in order to retrieve trace gas profiles. This technique iteratively fits a calculated spectrum to the measured spectrum by adjusting target trace gas profiles[2, 5, 6]. The microwindows used for each of the eight main trace gases, along with interfering species for each region, are taken from Griffin et al. (2017).

The retrieval is performed with the SFIT4 0.9.4.4 retrieval package and the HIgh-Resolution TRANSmission (HITRAN) 2008 spectroscopic database on a 29 layer grid[2, 7, 8]. SFIT4 requires input parameters for the temperature, pressure, and an a priori profile for the trace gases. Below ~1.0 mbar daily temperature and pressure profiles are calculated from National Centers for Environmental Prediction (NCEP) profiles that have been interpolated to PEARL[2, 9]. Above this monthly temperature and pressure profiles from the Whole Atmosphere Community Climate Model (WACCM) v6 for Eureka are used[2, 10]. The a priori gas profiles come from a 40 year run (1980-2020) of WACCM v6 for Eureka[2]. Figure 6 shows the retrieved vertical profiles for CO from the 2017 campaign along with the a priori profile used in retrievals. Retrieved volume mixing ratio profiles are integrated in order to obtain total column measurements, and a root-mean-square degrees-of-freedom-for-signal (RMS/DOFS) filter is applied to the retrieved data to ensure its quality.

Figure 6: Plotted retrieved vertical mixing ratio profiles for CO from 2017 (blue) and the a priori used (red)
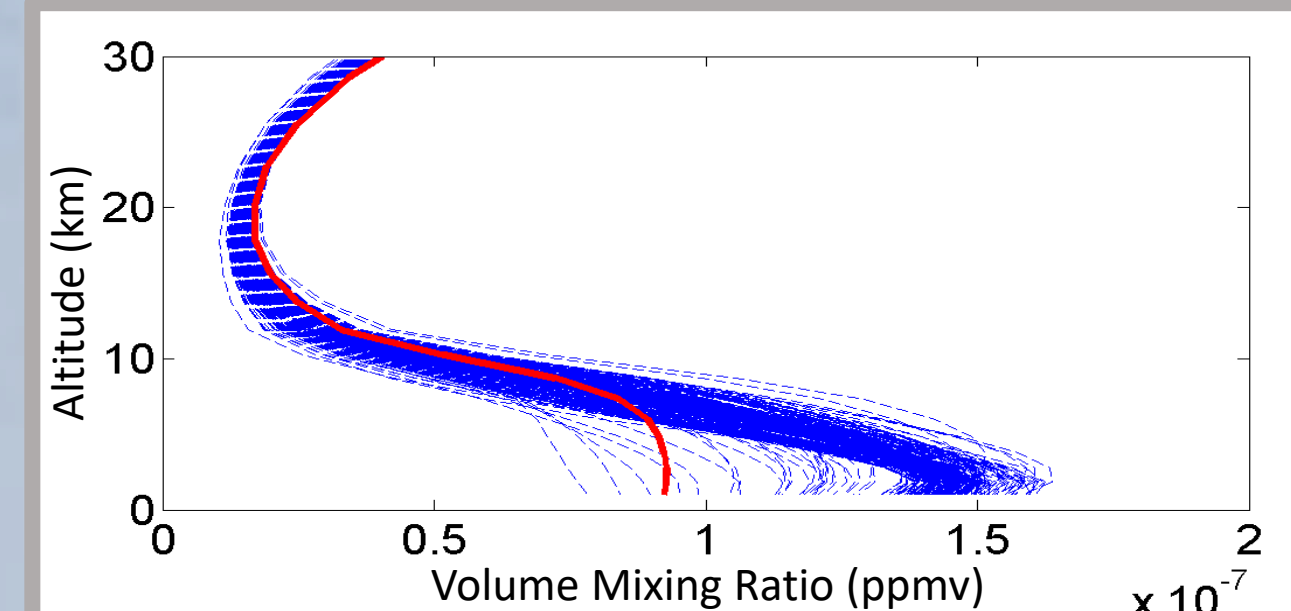
To examine the trace gas species for trends, the annual mean of each trace gases is fit with a first order polynomial[2, 11]. To determine if the rate of change is statistically significant, the number of years to determine a trend is calculated for each gas. Following Weatherhead et al. (1998) we assume a model function of the form[11]:

$$Y(t) = \mu + \omega t + N = \mu + \omega t + \varphi N_{t-1} + \varepsilon_t$$

where $Y(t)$ is the measurement, μ is a constant, ω is the trend magnitude, t is time, and N is the noise with autocorrelation φ and random noise ε. Then we can use:

$$n^* = \left[\frac{3.3\sigma_N}{|\omega|} \sqrt{\frac{1+\varphi}{1-\varphi}} \right]^{2/3}$$

to find the number of years (n^*) of data required to statistically determine a trend with probability 0.90[11].

For stratospheric trace gas species, due to the variable nature of the stratospheric polar vortex, only measurements taken outside of the polar vortex can be considered for trend analysis[2]. The vortex edge is determined from the scaled Potential Vorticity (sPV) at 20 km along the line of sight, with 1.2×10^{-4} s^{-1} defining the outer edge. The sPV for each measurement is generated as part of the Derived Meteorological Product (DMP) file produced for each year's measurements, however the input required for the 2014-2017 DMP files is currently under revision, limiting our capability to analyze stratospheric species here.

Results

The fitted annual mean timeseries for all eight of the trace gas species studied in Griffin et al. (2017) are shown below. The stratospheric trace gas time series below are not filtered for whether the measurements are taken inside or outside of the polar vortex, as we are currently still in the process of producing the necessary input required for the production of the DMP files. For those four stratospheric species, the variations in the plotted time series illustrate the dynamical and chemical variability associated with the polar vortex. Fits of observed changes in each of the four tropospheric species are summarized in Table 1 below, along with the minimum number of years required to detect a trend in the data. Both the values found by Griffin et al. (2017) for the 2006 to 2013 period and those found for the expanded dataset covering 2006 to 2017 are provided. It was noted in Griffin et al. (2017) that after 2009, C$_2$H$_6$ sharply increased for a number of years, and so an additional fit of C$_2$H$_6$ starting in 2009 is also provided. However with the low total column values found for C$_2$H$_6$ in 2016 and 2017 very little difference is evident between the two fits.
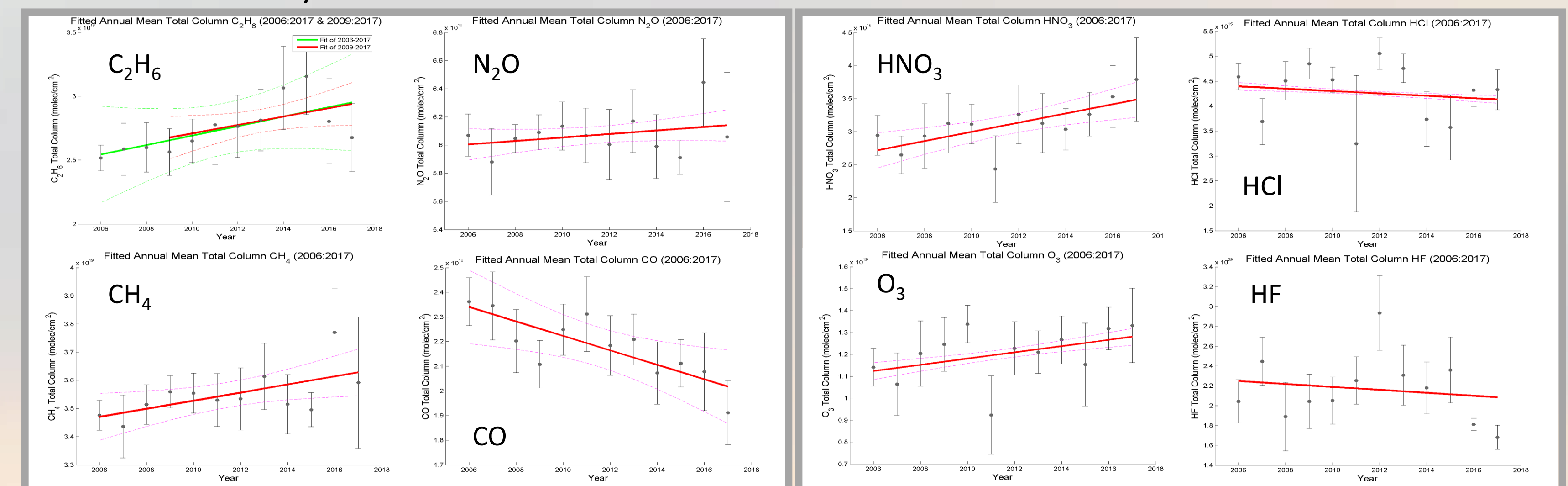
**Figure 7:** Fits of the four tropospheric trace gas species studied

Figure 8: Fits of the four stratospheric trace gas species studied

Table 1: Rate of change and years required to detect trends in tropospheric species

Gas	Rate of Change (2006-2013)	Years Needed for Trend (2006-2013)	Rate of Change (2006-2017)	Years Needed for Trend (2006-2017)
CH$_4$	0.5±0.1 % yr^{-1}	8	0.4±0.2 % yr^{-1}	7
CO	-0.8±0.6 % yr^{-1}	9	-1.3±0.4 % yr^{-1}	12
N$_2$O	0.3±0.3 % yr^{-1}	10	0.2±0.1 % yr^{-1}	8
C$_2$H$_6$	1.6±0.2 % yr^{-1}	8	1.3±0.5 % yr^{-1}	11
C$_2$H$_6$ (Post 2009)	2.3±0.5 % yr^{-1}	8	1.2±0.6 % yr^{-1}	9

Water Vapor Retrieval

Retrievals of a total column water vapor product from the PARIS-IR dataset are under development. Water vapor must be retrieved on a logarithmic scale since water vapor is log-normally distributed, as opposed to the normal distribution of the other trace gases retrieved from PARIS-IR[12]. This requires the atmospheric state vector, a priori profile, covariance matrix and Jacobian to all be transformed onto a logarithmic scale[12]. In the below figure, four potential water vapor microwindow regions are shown. These microwindows have been used in other FTIR water vapor retrieval schemes, and were selected for use here due to their few interfering species and for possessing features resolvable at PARIS-IR's resolution[12-15].
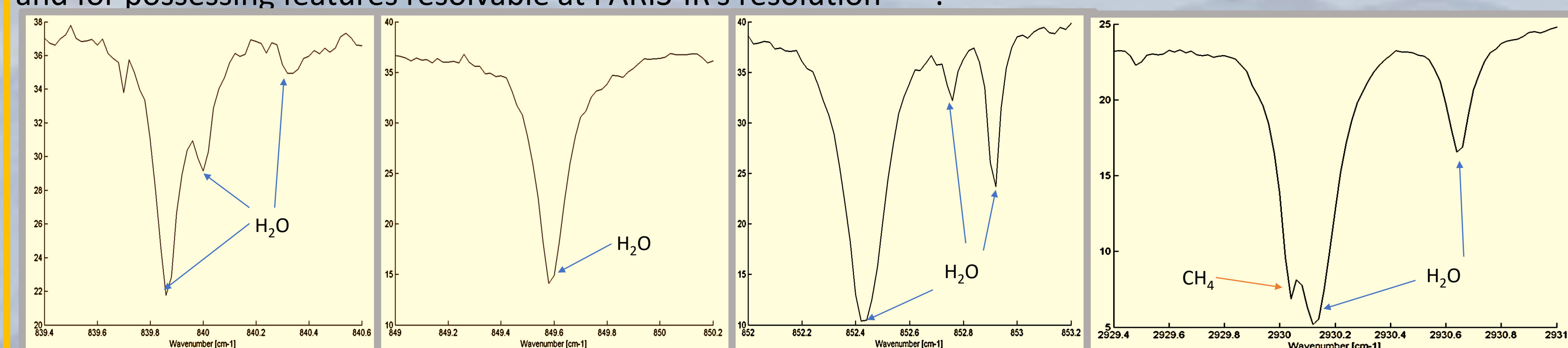
Figure 9: Potential microwindows for PARIS-IR water vapor retrievals. Taken from a typical PARIS-IR spectrum recorded March 18 2017 at the PEARL Ridge Lab

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