

# REMOTE SENSING OF STRATOSPHERIC TRACE GASES BY TELIS

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## ABSTRACT

TELIS (TErahertz and submillimeter Limb Sounder) is a balloon-borne cryogenic heterodyne spectrometer with two far infrared and submillimeter channels (1.8 THz and 480–650 GHz developed by DLR and SRON, respectively). The instrument was designed to investigate atmospheric chemistry and dynamics with a focus on the stratosphere. Between 2009 and 2011, TELIS participated in three winter campaigns in Kiruna, Sweden. The recent campaign took place in 2014 over Timmins, Canada. During previous campaigns, TELIS shared a stratospheric balloon gondola with the balloon version of MIPAS (MIPAS-B) and mini-DOAS. The primary scientific goal of these campaigns has been to monitor the time-dependent chemistry of chlorine and bromine, and to achieve the closure of chemical families inside the polar vortex. In this work, we present retrieved profiles of ozone ( $O_3$ ), hydrogen chloride (HCl), carbon monoxide (CO), and hydroxyl radical (OH) obtained by the 1.8 THz channel from the polar winter flights during 2009–2011. Furthermore, the corresponding retrieval algorithm is briefly described. The quality of the retrieval products is analyzed in a quantitative manner including: error characterization, internal comparisons of the two different channels, and external comparisons with coincident spaceborne observations. The errors due to the instrument parameters and pressure dominate in the upper troposphere and lower stratosphere, while the errors at higher altitudes are mainly due to the spectroscopic parameters and the radiometric calibration. The comparisons with other limb sounders help us to assess the measurement capabilities and instrument characteristics of TELIS, thereby establishing the instrument as a valuable tool to study the chemical interactions in the stratosphere.

Key words: ill-posed inverse problem, limb sounding, stratospheric trace gases, TELIS.

## 1. INTRODUCTION

Atmospheric environment is a research hotspot, with an emphasis upon its interrelation to other parts of the Earth

system. In particular, studies of gas phenomena are important to make valid evaluations on current air quality and to make reliable predictions of climate change. To have knowledge of temporal and spatial behavior of molecular concentration, remote sensing techniques on various platforms can be used to measure these atmospheric quantities.

In contrast to spaceborne observations, although a balloon can only be operated on a limited temporal and spatial coverage, balloon observations are used as a good alternative because of their flexibility and lower operational cost. Besides, a good understanding of balloon experiment performance has been proved to be valuable for the validation of spaceborne missions, and the balloon-borne instrument provides a prototype for new solutions to overcome the potential technical difficulties in the design of coming spaceborne instruments. As a state-of-the-art heterodyne spectrometer, TELIS (TErahertz and submillimeter Limb Sounder) has been mounted on a stratospheric balloon together with two sophisticated instruments MIPAS-B and mini-DOAS. The TELIS measurements are performed in the infrared and microwave region where the thermal radiation emitted by trace gases at various altitudes is detected. The combination of the three instruments has completed four scientific joint flights in the past years: over Kiruna, Sweden on 11 March 2009, 24 January 2010, and 31 March 2011; over Timmins, Canada on 7 September 2014.

This work focuses on the analysis of the TELIS far infrared spectra and presents the retrieved profiles of  $O_3$ , HCl, CO, and OH from the Kiruna flights during 2009–2011. The TELIS profiles are compared against other spaceborne limb sounders SMILES, MLS, and SMR. Furthermore, the corresponding error characterization is illustrated.

## 2. TELIS

The TELIS instrument operates in a multi-channel mode that has been developed cooperatively by several European research institutes. DLR is responsible for a tunable 1.8 THz channel [1] with enhanced stability, whereas SRON is responsible for a tunable 480–650 GHz channel [2] based on the Superconducting Integrated Receiver

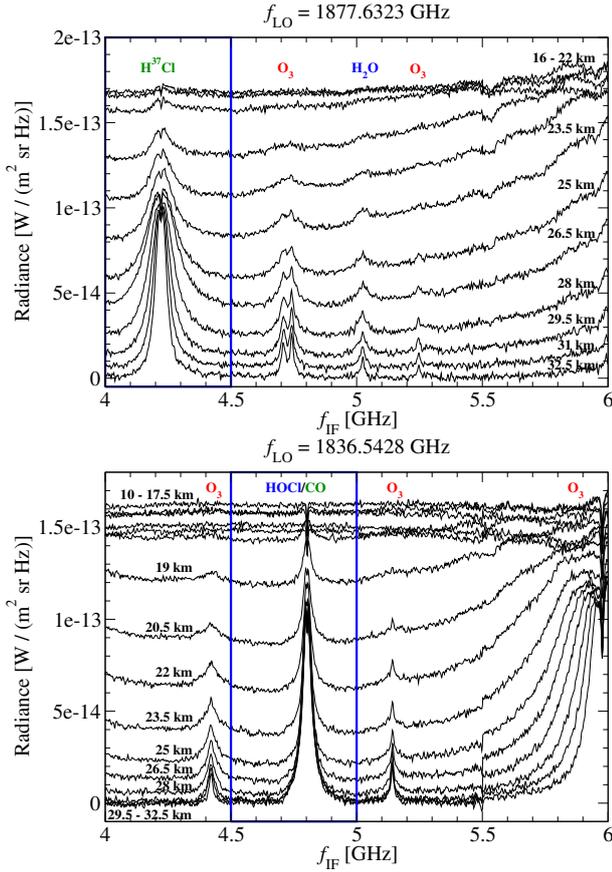


Figure 1. Far infrared limb spectra of atmospheric HCl and CO observed by the TELIS 1.8 THz channel during the 2010 flight.

(SIR) technology. The TELIS 1.8 THz channel measures the signal at a local oscillator (LO) frequency  $f_{LO}$  between 1790 GHz and 1880 GHz. Because the instrument operates in a double sideband (DSB) mode, the recorded spectrum is then generated from the two sidebands with respect to  $f_{LO}$ , i.e.  $f_{LO} - f_{IF}$  and  $f_{LO} + f_{IF}$  with  $f_{IF}$  ranging from 4 to 6 GHz.

In contrast to the balloon-borne Fourier transform spectrometer MIPAS-B, the different target molecules to be detected cannot be simultaneously covered by the TELIS instrument due to the requirement of different local oscillator configurations. A set of optimal LO frequencies  $f_{LO}$  has been determined by modelling the spectra in a double sideband mode using standard atmospheric constituent profiles and a common molecular spectroscopic database. The selection criteria of the TELIS instrument was to have isolated spectral lines of the target species with insignificant overlapping contributions from other interfering species.

A single limb-scanning sequence comprises a series of radiance spectra with equidistant steps between two consecutive tangent points. Most of the limb sequences are characterized by tangent heights range from 10 or 16 km up to 32.5 km discretized in 1.5 km steps (see Fig. 1). In

the case of a few weak molecules (e.g. OH), the vertical spacing rises to 2 km, leading to a broader averaging kernel in the retrieval. Each spectrum was measured for 1.5 s and the calibrated spectra were measured approximately every 30 s. At the tangent point of the line-of-sight, the vertical resolution is estimated to be 1.5–3 km for observational frequencies around 1.8 THz. The horizontal (azimuth) resolution is roughly a factor of two worse due to the anamorphicity of the telescope.

According to the processed antenna beam profile measurements, the tangent offset with respect to the commanded tangent height (taken from AHRs pitch  $0^\circ$ ) is  $3.4 \text{ arcmin}$  for the 1.8 GHz channel, which shows that the actual tangent height of a pencil beam is a bit higher than the commanded one. The antenna beam profile of the THz-channel is Gaussian shaped with a full width half maximum (FWHM) in the vertical direction of  $0.1043 \pm 0.0008^\circ$ . Moreover, the on-ground laboratory measurements show that the sideband ratio is estimated to lie in the range of 0.95 to 1.05.

### 3. INVERSION

For the interpretation of the TELIS measurements, a forward model  $F$  is required that simulates the radiance measurement as function of the atmospheric state vector  $x$  by taking into account the forward model parameters  $b$  and the instrument model parameters  $c$

$$y^\delta = F(x, b, c) + \delta, \quad (1)$$

where  $y^\delta$  and  $\delta$  represent the noisy data vector and the noise vector, respectively.

Inverse problems in atmospheric science are inherently ill-posed and the least squares solution is not a reliable estimate of the true solution. In order to obtain a solution with physical meaning, additional constraints have to be imposed on  $x$ , a process which is defined as regularization. By imposing the so-called regularization matrix  $L$ , the estimation of the state vector  $x$  from the measurement vector  $y^\delta$  can be formulated as a minimization problem involving the objective function

$$\mathcal{F}(x) = \|F(x) - y^\delta\|^2 + \lambda \|L(x - x_a)\|^2, \quad (2)$$

where  $x_a$  and  $\lambda$  denote the a priori state vector and the regularization parameter, respectively. The selection of an appropriate regularization parameter providing at the same time a small residual and a moderate value of the penalty term is crucial in the method of Tikhonov regularization. Alternatively, iterative regularization methods use an iteration-dependent  $\lambda$  and the number of iteration steps  $i$  plays the role of the regularization parameter. The discrepancy principle as an a posteriori stopping rule is used in order to avoid an uncontrolled explosion of the noise error.

The retrieval code PILS (Profile Inversion for Limb Sounding) [3] has been developed to derive atmospheric

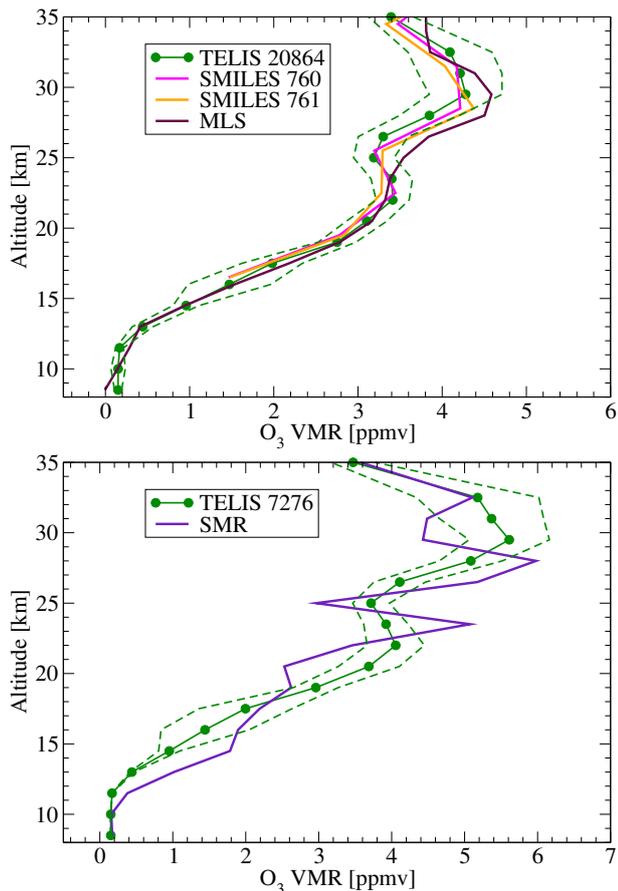


Figure 2. Top: comparison of  $O_3$  retrievals from TELIS, SMILES, and MLS on 24 January 2010. The lowest tangent height is 10 km and the retrieval results below this altitude have little physical meaning. The dashed green lines indicate the overall accuracy in the TELIS profile. Bottom: comparison of  $O_3$  retrievals from TELIS and SMR on 24 January 2010.

parameters from infrared and microwave limb measurements. The code is based on a line-by-line program GARLIC [4] which is capable of performing high resolution infrared/microwave radiative transfer calculations soundly and efficiently, and on a nonlinear least squares framework with direct and iterative numerical regularization methods.

## 4. RETRIEVALS

The concentration profiles of  $O_3$ , HCl, CO, and OH derived from limb spectra in the TELIS 1.8 THz channel are discussed in this section.

### 4.1. $O_3$

In practice, the TELIS consortium did not define any dedicated ozone frequency microwindow during previous balloon campaigns, because ozone appears in almost all observed microwindows. Here,  $O_3$  is retrieved from the transitions around 1832.12 GHz and 1828.42 GHz, respectively.

The top panel of Fig. 2 shows a comparison of  $O_3$  retrievals from TELIS, SMILES, and MLS on 24 January 2010. Both SMILES profiles fall well within the error margin of the TELIS profile, but large discrepancies are found above the observer altitude (not shown) due to the limited information of TELIS in this altitude range. Apart from that, a promising agreement between SMILES and TELIS is reached between 16 and 35 km. The MLS and TELIS VMR profiles agree within the overall accuracy of the TELIS profile excepting a region around 25–28 km, while a maximum difference of about 25% is found at 28 km.

The coincident TELIS and SMR data measured on 24 January 2010 are compared in the bottom panel of Fig. 2. The TELIS and SMR profiles reach an acceptable agreement despite some oscillations in the SMR profile. A weak regularization in the SMR retrieval can provoke this under-smoothing feature in the profile.

Fig. 3 depicts the estimated smoothing, noise, and model parameters errors in the  $O_3$  retrieval for TELIS measurement 20864 and 7276. The uncertainties in the pointing information and pressure are the most important error sources below 20 km. Above 20 km, the calibration and spectroscopic errors are dominant, which are likely to be determined by spectral lines, and thus by ozone concentrations.

### 4.2. HCl

Hydrogen chloride is the main chlorine reservoir species monitored by TELIS, the 2010 flight took place over northern Scandinavia inside the activated Arctic vortex where chlorine activation can be examined. In the 1.8 THz channel, one  $H^{37}Cl$  transition line at 1873.40 GHz ( $62.49 \text{ cm}^{-1}$ ) was detected.

The comparison of the HCl profiles retrieved from TELIS and SMILES is shown in the top panel of Fig. 4. It can be noticed that the vertical resolution of TELIS is better than that of SMILES over the plotted altitude range and a proper comparison should take this fact into account. The plotted TELIS profile is convolution of the original profile and the averaging kernels for the SMILES data product. The TELIS and SMILES products agree well within the accuracy domain, excepting the small disagreements at 17.5, 23.5, and 32.5 km.

The bottom panel of Fig. 4 shows the comparison between the MLS and TELIS profiles. Within the plotted

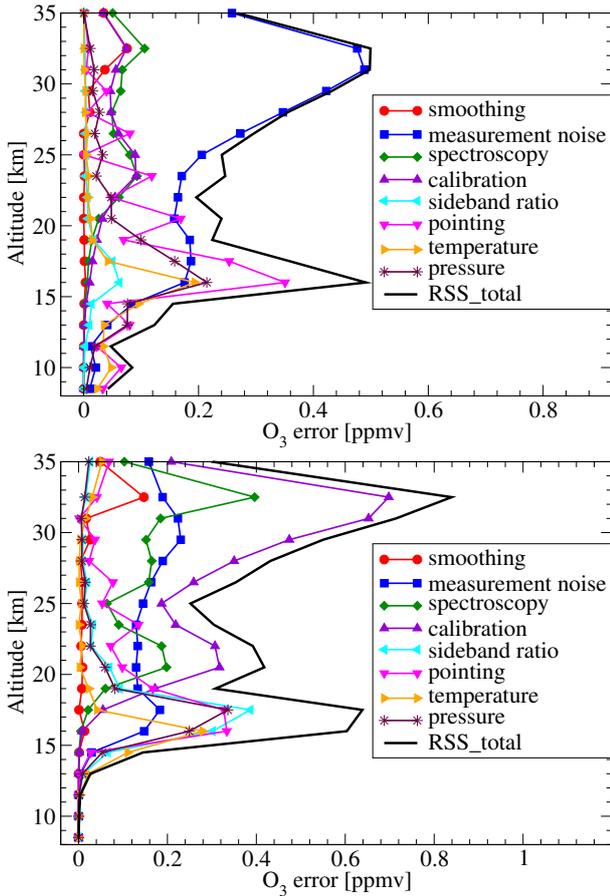


Figure 3. Smoothing, noise, and model parameters errors for the  $O_3$  retrieval using TELIS's far infrared measurement 20864 (top) and 7276 (bottom) during the 2010 flight. The solid black line (RSS\_total) refers to the total retrieval error represented by the RSS of all error components.

overall accuracy of the TELIS profile, the HCl concentrations agree over almost the entire altitude range of 15–35 km. The MLS profile only falls outside the accuracy domain at 23 km and below 16 km.

Fig.5 depicts the estimated retrieval error budget for the HCl retrieval during the 2010 flight. Between 22 and 32.5 km, the total retrieval error reaches 0.3 ppbv and exceeds 0.5 ppbv below 22 km. All errors steeply increase for altitudes up to 20 km and from 30 km upwards. One reason for larger errors around the lowest tangent height (16 km) can be that the nominal abundances ( $< 1$  ppbv) below 20 km are discovered and a reasonable retrieval below this altitude is difficult.

At higher altitudes, maximum errors of 0.18 and 0.22 ppbv arise from the uncertainties in the calibration and the spectroscopic parameters, respectively. Because the nonlinearity effect has the highest impact on altitude levels with larger concentrations, this explains why the calibration error is dominant in this altitude range.

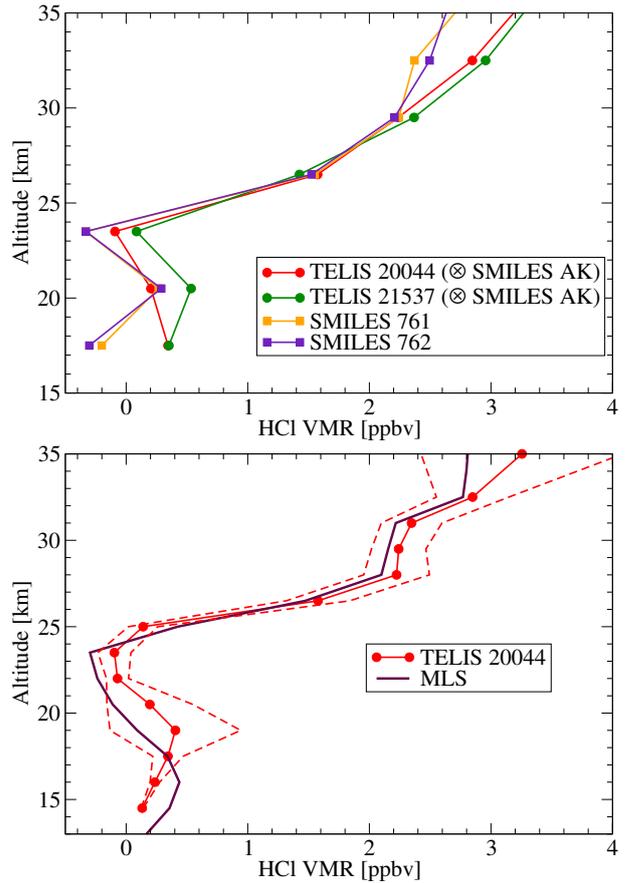


Figure 4. Top: comparison of  $O_3$  retrievals from TELIS and SMILES on 24 January 2010. Bottom: comparison of  $O_3$  retrievals from TELIS and MLS on the same day.

The noise error is found to be larger than the other error components above 20 km, which is different for the HCl retrieval using the TELIS submillimeter spectra ( $\approx 0.01$  ppbv) [5].

### 4.3. CO

Due to a photochemical lifetime in the troposphere, the data of carbon monoxide inside the polar vortex measured by the TELIS instrument helps to understand regional-scale transport of pollution. TELIS probed CO at the transition frequency of 1841.36 GHz.

In Fig. 6, the CO profile retrieved from measurement 20864 is compared against the MLS profile due to the small time difference (approximately 0.5 h) and close geolocation. An excellent agreement can be found in both retrievals and the peak value at 32.5 km captured by TELIS was also successfully retrieved by MLS. The MLS profile overall falls within the accuracy domain of the TELIS profile and both profiles show virtually identical shape.

In Fig. 7, we display the corresponding error budget of

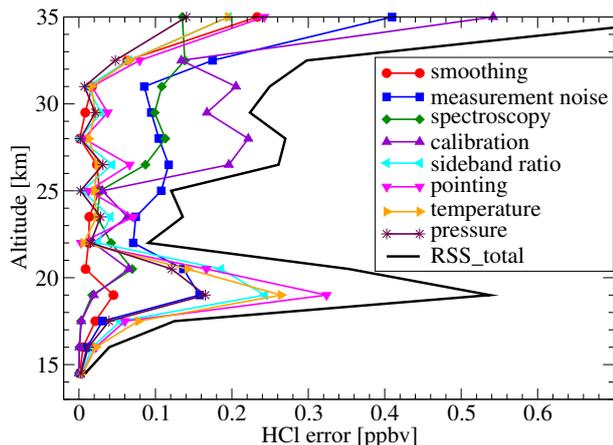


Figure 5. Smoothing, noise, and model parameters errors for the HCl retrieval. The estimates correspond to TELIS's far infrared measurement 20044 during the 2010 flight.

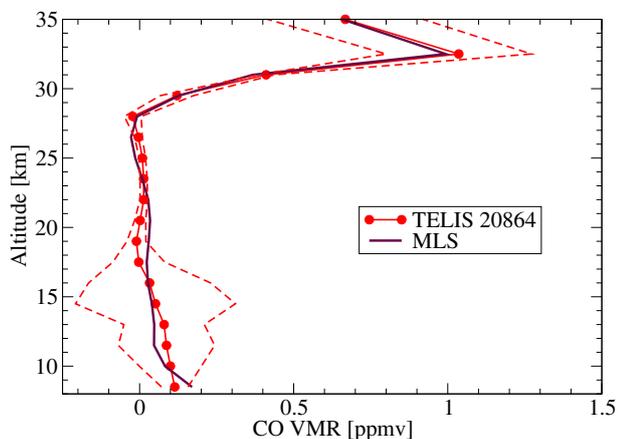


Figure 6. Comparison of CO retrievals from TELIS and MLS on 24 January 2010.

the CO retrieval. At lower altitudes, the uncertainties in the pointing information and temperature are the two important error sources, with the peak appearing near 15 km. The measurement noise dominates the error budget between 17.5 and 26.5 km, although the propagated noise error is only slightly larger than others. At higher altitudes, the spectroscopic parameters appear to be the most critical error source. The total retrieval error is of about 0.01–0.25 ppmv.

#### 4.4. OH

OH is one of the most interesting species observed in TELIS far infrared spectra. TELIS is the first instrument which uses the OH transitions around 1.8 THz (e.g. 1834.75 and 1837.80 GHz).

The OH observation on 24 January 2010 is displayed in

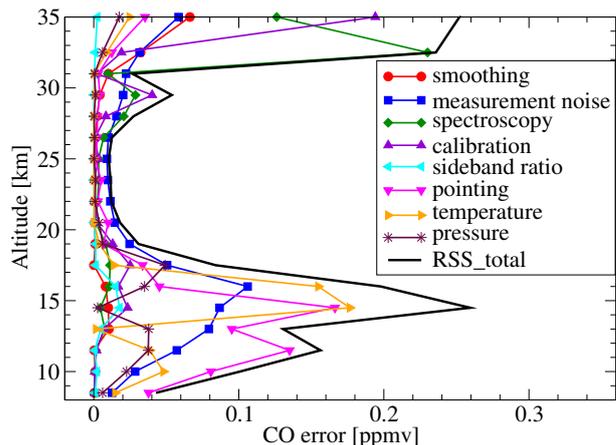


Figure 7. Smoothing, noise, and model parameters errors for the CO retrieval. The estimates correspond to TELIS's far infrared measurement 20864 during the 2010 flight.

Fig. 8. Two OH measurements were obtained before local sunrise and 2 h before local noon, respectively. The major differences in both retrieved profiles are located around 20–25 km and are due to the fact that OH responds very quickly to solar radiation.

In Fig. 9, all errors excepting the temperature error, are smaller than 1.5–2 ppbv below 30 km. The model parameter errors due to spectroscopy and calibration stretch from 25 km upwards where OH abundances start to increase. However, all errors steeply increase above 30 km.

First OH retrievals from TELIS measurements are presented. Because of the large measurement noise, the precision in the OH retrievals is not highly satisfactory. Further investigations into cross-validations of the TELIS OH profiles will be done.

## 5. CONCLUSIONS

In this work, we present the retrieval results of TELIS far infrared spectra from recent Kiruna balloon campaigns. In case of  $O_3$ , HCl, and CO, the TELIS retrievals agree very well with profiles obtained by other spaceborne limb sounders. These comparisons further demonstrate the consistency and reliability of the TELIS profiles. First OH retrievals prove the retrieval feasibility of 1.8 THz transitions. The corresponding error analysis helps us to better understand the instrument characteristics and the measurement capabilities.

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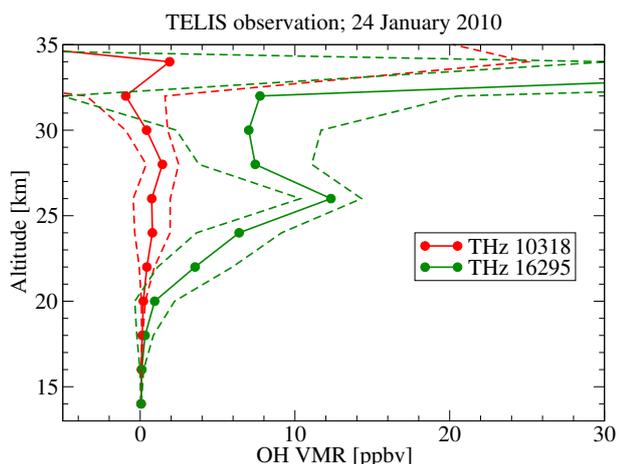


Figure 8. OH profiles retrieved from the TELIS data on 24 January 2010. The solid red and green lines correspond to the OH profiles obtained from measurements 10318 and 16295, respectively. The dashed lines refer to the overall accuracy of both OH profiles.

ature retrievals and geolocation data, A. de Lange and J. Landgraf for many fruitful discussions and suggestions on TELIS retrievals, Y. Kasai and J. Urban for sharing SMILES and Odin/SMR retrievals.

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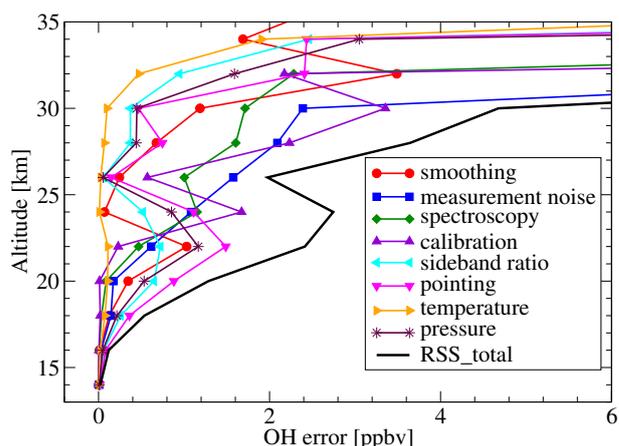


Figure 9. Smoothing, noise, and model parameters errors for the OH retrieval in the second microwindow. The estimates correspond to TELIS's far infrared measurement 16295 during the 2010 flight.

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