

Final author comment for “Long-term evolution and seasonal modulation of methanol above Jungfrauoch (46.5°N, 8.0°E): Optimisation of the retrieval strategy, comparison with model simulations and independent observations.”

To Referee #1

	<i>Comment</i>	<i>Answer</i>
1	For the bulk of the analysis, the authors state that only spectra with solar zenith angles between 65 and 80 degrees were used. However, the authors then go on to devote a section of the paper to the diurnal variation of methanol. For the analysis of diurnal variation, the authors use cases with SZA as low as 30 degrees. The authors do state that the diurnal variation analysis was limited to cases where the DOFS > 1. However, the use of different subsets of the dataset for different parts of the analysis is not well justified. Are the cases with SZA between 30 degrees and 65 degrees of questionable quality or not? If these cases are OK, then why exclude them from the main analysis? More justification/explanation is needed here.	The first goal of this paper is to present results retrieved with a strategy that significantly improves the information content and to exploit them to study the long-term trend and seasonal modulation of CH ₃ OH as well as to compare our total, lower-tropo and UTLS columns with independent observations and model. Spectra with solar zenith angle (SZA) between 65 and 80 degrees were selected since they provide sufficient information content and corresponding pieces of information allowing performing such investigations. However, it also seemed interesting to push further our study on methanol diurnal variation. To cover the broadest possible range of hours during the day, we included all data with DOFS of at least 1. The quality of this subset is certainly as good, only its usability is limited to total column investigations.
2	The paper would benefit from improvements to Section 3, “Data characterization and error budget”. On pages 5/6, the authors state that two different values for the SNR were used in the two different fitting windows, “since the fitting quality is significantly different in both windows”. This SNR is presumably what feeds into the “measurement noise error” listed in Section 3. Based on the text here, I’m not sure that the “noise” here is truly noise. Is there no way to quantify the instrument noise independently of retrieval fitting quality? Couldn’t the difference in fitting quality arise from interference from ozone (or some other molecule)? If so, then presumably it shouldn’t really be characterized as “noise”?	In order to optimize the information content and since there is a difference of fitting quality between both windows (due to the ozone interference), we had to tune the signal to noise ratio (SNR) for inversion either to 140 or 80 depending on the spectral range. It is an ad-hoc parameter allowing us to reduce systematic residuals while maximizing the information content. The difference in fitting quality in the two windows has been taken into account in the error budget. Also, it should be noted that the actual SNR of the spectrum is evaluated in regions that are always fully saturated and ranges between 250 and 1800 as mentioned in the introduction. To summarize, there is the noise characterizing the spectrum, dependent on the instrument performance and on the observing conditions, and the noise on the measurement, which is also function on the quality of the spectroscopic parameters and on the strength of the interferences.

3	<p>The discussion of the spectroscopic uncertainty associated with ozone is also rather vague. When the authors discuss the uncertainty associated with ozone in the HITRAN 2008 compilation, are they referring to intensity uncertainties? Width uncertainties? Which line parameters were “incremented” in the sensitivity test?</p>	<p>We only included the uncertainty on the line intensity for methanol and for each interfering gas. When available, the uncertainties on other parameters such as line width and line positions are not well enough documented or appear to be very conservative. To clarify this, the sentence (Page 4668, line 2) will be rewritten as follows:</p> <p>“We noted that the SFIT-2 algorithm fails to perform a satisfying retrieval when using spectroscopic parameters with ozone lines intensity incremented by 10 %, suggesting that the error on the concerned lines intensity is more likely to be closer to 5 (or even lower) than to 10%.”</p>
4	<p>The value of the comparisons of the Jungfraujoch data with ACE-FTS 10 degree zonal monthly means is unclear. The authors clearly state that the ACE-FTS zonal means are capturing events that are not seen at the Jungfraujoch. The differences in March-May are huge. In this case, why bother to compare with the zonal mean? Why not be more selective in the ACE-FTS cases used in this comparison, and make some attempt to select the ACE-FTS cases that sample airmasses that are also likely to be sampled by the Jungfraujoch observations?</p>	<p>Unfortunately there is not much occultations available in the UTLS (upper-troposphere lower-stratosphere) altitude range for us to be more selective. (For your information, there is only 12 ACE-FTS occultations available at a distance of less than 500km from the Jungfraujoch.) Therefore we had to settle for a wide latitudinal band and to make comparisons with only monthly means. Despite this settlement we managed to show a good agreement for the seasonal cycle.</p>
5	<p>Page 4662, Line 25 : “uncertainties in our knowledge of the methanol global sources and sinks in the atmosphere”. Firstly, I would suggest removing “in the atmosphere” from the end of this sentence. Secondly, this paper focuses on the Jungfraujoch dataset. While this is an interesting dataset, it represents a very specific location, and is not going to shed much light on the global sources and sinks. This paragraph is a list of statements. The authors might consider re-writing it to emphasize what their dataset brings to the suite of existing data.</p> <p>What is the main point of this sentence? What is the “first” here? The first ground-based time series? (Presumably not, based on other references cited here.) The first time series at the Jungfraujoch? Perhaps, but that is perhaps not what makes this dataset special. I would have thought that what makes this dataset special is the length of the record, and the location of the Jungfraujoch station, ie where the airmasses come from and what sources are represented in these airmasses.</p>	<p>The paragraph will be modified as follows (Page 4662, Line 24):</p> <p>“In addition, previous studies have reported the measurement of methanol from ground-based infrared solar absorption observations performed at Kitt Peak (31.9 N, 111.6W, 2090ma.s.l.; Rinsland et al., 2009) and at Saint-Denis (Reunion Island, 21 S, 55 E, 50ma.s.l.; Stavrakou et al., 2011; Vigouroux et al., 2012). However, there still remain large uncertainties in our knowledge of the methanol global sources and sinks, as indicated by the large discrepancies existing between different measurement-based estimates of the total sources (Galbally and Kirstine, 2002; Tie et al., 2003; von Kuhlmann et al., 2003a, b; Jacob et al., 2005; Millet et al., 2008; Stavrakou et al., 2011).</p> <p>In this paper, we report the first long-term methanol time series (17 years) derived from ground-based high-resolution infrared spectra recorded with a Fourier Transform InfraRed (FTIR) spectrometer operated under clear sky conditions at the high-altitude International Scientific Station of the Jungfraujoch (ISSJ, Swiss Alps, 46.5 N, 8.0 E, 3580ma.s.l.; Zander et al., 2008), providing a valuable tool for model and satellite validation and complementing the NDACC measurements at northern mid-latitudes.”</p>
	<p><i>Other minor comments are also much appreciated and will be taken into account for the final version of this paper.</i></p>	

	Comment	Answer
1	Is the 98% absorption for O3 reported in the “1037” window along a nadir or slant path? If it is a nadir path, then I would expect that along the long slant paths used for these observations that this window would become entirely opaque. Since evidently reasonable CH3OH retrievals are obtained, this does not happen. Could the authors explain? Is this due to the sufficient spectral points located between the O3 lines to allow for detection of the broad CH3OH feature? If so, the authors could highlight the importance of the high spectral resolution of the FTIR used.	The absorption numbers cited are based on the simulation of both windows illustrated in Figure 1. The simulation is based on a resolution of 6.1mK ($1 \text{ mK} = 0.001 \text{ cm}^{-1}$) and along a slant path with a solar zenith angle of 80° (see text and caption of Figure 1).
2	The total estimated error is frequently written (Worden et al., 2004) as the sum of smoothing errors, measurement errors (due to spectral noise) and finally systematic and random errors due to uncertainties in both fixed parameters (such as spectral properties) and parameters that change from observation to observation but are not retrieved (e.g., temperature). The authors have calculated systematic and random errors (as shown in Table 1), but it is not clear which, if any, were included in the total error estimate shown in Figure 2. I believe they should all be included, but am willing to see counter arguments.	It appears that the total error estimate illustrated on Figure 2 is unclear and will be removed since it does not include all error components. We prefer distinguish both types of error (random and systematic) for a proper characterization of our data for potential users.
3	This discussion on the evaluation of the uncertainties in the O3 line parameters is confusing. It appears that since a 10% uncertainty in the O3 line (intensities??, please clarify) leads to poor O3 (please confirm) retrievals, then 10% is too high an estimate, and 5% was chosen instead, but 5% does not appear in Table 1.	See answer to Referee #1, 3. The comments in the table will be more explicit on this point.
4	The sentence starting with “Even” needs to be broken up and rewritten, it uses “respectively” too many times.	The sentence will be modified as follows (Page 4672, Line 25) : “Even though we found no significant trend of methanol through the day in summer, a significant increase during winter and the rest of the year has been evaluated at 0.4 ± 0.3 and $1.1 \pm 0.2\%$ /degree in the morning. For the afternoon, the corresponding rates amount to -0.9 ± 0.2 and $-0.5 \pm 0.1\%$ /degree, showing significant decreases.”
5	Where is it shown that the IMAGES model fails to reproduce the observed diurnal variation?	For clarity, we had to only show monthly mean IMAGES results. For your information, the IMAGES “MEGAN” simulation gives a relative standard deviation (RSD) of 15 and 4% for July and December while the FTIR results give a RSD of 29 and 19% for the same months.

6	You cannot state that the seasonal amplitude of the in situ measurements appears to be significantly lower than the signal from the FTS, as there are no in situ data in June and July, when the summer peak occurs. I do not agree with this statement.	<p>The sentence will be re-written as follows (Page 4674, Line 10) :</p> <p>“The seasonal amplitude shows a good agreement on the data dispersion (see error bars) except for the fall season with more compact values. The high standard deviation in summer appears to be due to only a few days with high methanol mixing ratios.”</p> <p>And in the conclusion (Page 4676, Line 16) :</p> <p>“Although the seasonal amplitude is larger in the FTIR lower tropospheric data compared to in situ measurements, a good agreement is generally found regarding the data dispersion.”</p> <p>Will be replaced by :</p> <p>“The FTIR lower tropospheric data compared to in situ measurements generally shows a good agreement regarding the data dispersion.”</p>
7	Sentence starting with "Indeed" is too long and cumbersome.	<p>The sentence will be rewritten as follows (Page 4674, Line 15):</p> <p>“Indeed, it has been established by Legreid et al. (2008), that there is a considerable contribution of methanol from the south since methanol is emitted in large amounts from biogenic sources (Fall, 2003; Jacob et al., 2002, 2005; Singh et al., 1994) more active in the south of the Alps than in the north.”</p>
8	Is there an inconsistency here? How can the methanol variability be underestimated by IMAGES if the seasonal cycle is not significantly different: similar peak to peak variability and insignificant fractional difference; in the text it says -6 and +1, but Figure 7 shows much greater values for March and April.	<p>The methanol variability within e.g. a month is underestimated by IMAGES for the UTLS columns. For your information, the IMAGES “MEGAN” simulation gives a relative standard deviation (RSD) for UTLS columns of 15 and 6% for July and December while the FTIR results give a RSD of 43 and 40% for the same months. However, the amplitude of the seasonal cycle is well appraised by the model.</p> <p>The -6 and +1% are mean relative differences. The paragraph will be modified as follows (Page 4674, Line 26) :</p> <p>“On the other hand, the seasonal cycle of methanol UTLS columns is satisfactorily characterized by FTIR results and the IMAGES simulations in terms of absolute value with a non-significant mean fractional difference with FTIR of $-6\pm 49\%$ and $1\pm 48\%$, respectively for MEGAN and IASI.”</p>
9	Figure 5: What are the three blue lines?...	<p>These are the linear regression and its standard deviation (1-sigma) of the methanol total columns. The caption will be completed.</p>
<i>Other minor comments are also much appreciated and will be taken into account for the final version of this paper.</i>		