

The authors like to thank Referee#2 for his comments and suggestions. The Referee's comments and questions are bold, the authors' replies are formatted as plain text, and excerpts from the manuscript as well as changes to the manuscript are given in italics.

Reply to Anonymous Referee #2

Review of 'Intercomparison of in-situ NDIR and column FTIR measurements of CO₂ at Jungfraujoch' by Schibig et al.

The paper by Schibig et al., shows a comparison of two very different measurement time series at Jungfraujoch station in Switzerland. Ground-based as well as FTIR column measurements from 2005-2013 are presented. The authors report a consistent trend for both data sets which are in agreement with other stations on the northern hemisphere. The FTIR data set is biased low by 13 ppmv since the stratospheric column reduces the mean column value.

We would have expected the FTIR dataset to be slightly lower, because of the lower CO₂ mole fraction in the stratosphere, but since the FTIR data set is biased high by 13 ppm, we think this is caused by the uncertainty in the HITRAN compilation, which leads to a systematic error on the retrieved total column values. Further, we expect the influence of the stratosphere on the FTIR measurements to be significantly less than 13 ppm.

The data are filtered for pollution events and clear sky conditions and evidence is provided, that the variability of both data sets is partly due to local CO₂ variations. The seasonality is shows very interesting differences between both data sets, which are not explained fully. Both data sets show the seasonal minimum at the same time in August, but different times for the maximum, which occurs in January for the FTIR data set and in March for the NDIR in-situ measurements. This is explained by different source regions for the respective months on the basis of FLEXPART footprint calculations for 2009-2011. Differences in the vertical distribution are mentioned and particularly the role of the CO₂ gradient at the tropopause is not really discussed. In general the manuscript is well written and should be published in ACP, but the analysis of the seasonal differences and the footprint analysis should be sharpened.

Main comments:

The NDIR shows the minima in August as well as the FTIR, but the maxima show differences in their time of occurrence. The FTIR shows the maximum for January whereas the NDIR exhibits its maximum in March. I'm not sure if the FLEXPART footprints in Figs. 6-8 do provide meaningful results for the free tropospheric partial columns. I don't see for a long-lived tracer like CO₂ any reason why a ten day backward footprint for the free troposphere should provide an indication of sources and sinks. For the lowest layer this might be valid, but how does the respective footprint explain the seasonal differences in the free troposphere?

The vertical transport time scale in the troposphere is usually smaller than 10 days (as used in our FLEXPART simulations). Therefore, the model particles are usually widely dispersed in the troposphere after 10 days of transport. Although they won't be well mixing within the whole

northern hemispheric troposphere, the influence of surface source regions beyond the 10 day transport is usually sufficiently diluted and one does not find distinct signals from any specific source region. This is also true for free tropospheric release (receptor) locations since horizontal transport is faster in the troposphere and despite the absence of significant turbulent dispersion the particle plumes disperse due to wind field divergences. Therefore, we are convinced that the 10 day transport scale and derived surface residence times are sufficient to allow a qualitative interpretation of the contribution from different potential source areas.

How different are the footprint distributions in January, March and August from the other months? The different time of occurrence of the respective winter maxima is also not explained by the footprints. Is it maybe caused by seasonality of e.g. warm conveyor belts and therefore seasonality of the vertical tropospheric column? I suggest to analyze the FLEXPART output for this.

The footprints for the selected months are fairly representative for the respective season, with the exception of the January footprints which revealed strong influence from northern Africa at different vertical levels, which was not observed in other winter months. In order to further analyze the influence on transport on the observed seasonal cycle, we analyzed the timing of surface influence for different land regions and present this as a new figure and section in the revised manuscript. This extended transport analysis is able to explain the observations in the sense that we find an increased decoupling between the free troposphere and the land surface north of 30°N during the winter months, whereas the influence from tropical land surfaces south of 30°N was increased in winter. Both suggests lower CO₂ in the FT (free troposphere) than at the surface and an interruption of the wintertime increase in the FT above JFJ due to the onset of the decoupling and tropical influence just following the observed maximum in February.”

The following section was added at page 9, line 29:

“In general, the decoupling between the FTIR columns and possible surface fluxes of CO₂ from land surfaces north of 30°N was strongest during the winter month (January to March), when especially low surface residence times were simulated by FLEXPART for the free tropospheric FTIR columns (Figure 9). From April to September larger surface residence times were seen also for the FTIR columns and a stronger coupling between surface fluxes and the free troposphere can be expected. At the same time residence times over tropical land surface (south of 30°N) were generally larger for the FTIR columns and were especially increased from February to April (see Figure 9).

and page 13, line 6:

“...2009). The findings based on Figure 9 can help to understand the shift in the observed wintertime maximum of CO₂ between FTIR (January) and NDIR (March-April) The land surfaces of northern hemispheric mid-latitudes act as a net CO₂ source during the winter half year, since photosynthesis is largely reduced and respiration and anthropogenic emissions of CO₂ dominate the budget, hence, the observation of maximum CO₂ at the end of the winter half year and close to the surface. For the free troposphere above JFJ as observed by the FTIR the direct link to these wintertime releases of CO₂ is weakened due to generally reduced vertical transport. At the same time more frequent transport from and land surface contact in the tropics can be deduced, an area that even during the winter half year may act as a net CO₂ sink due to photosynthetic

uptake. An earlier onset of decreasing CO₂ in the FT above Jungfraujoch could thereby be explained by different seasonality of transport and vertical mixing. Additionally... ”

And the following figure with caption was added as Figure 9:

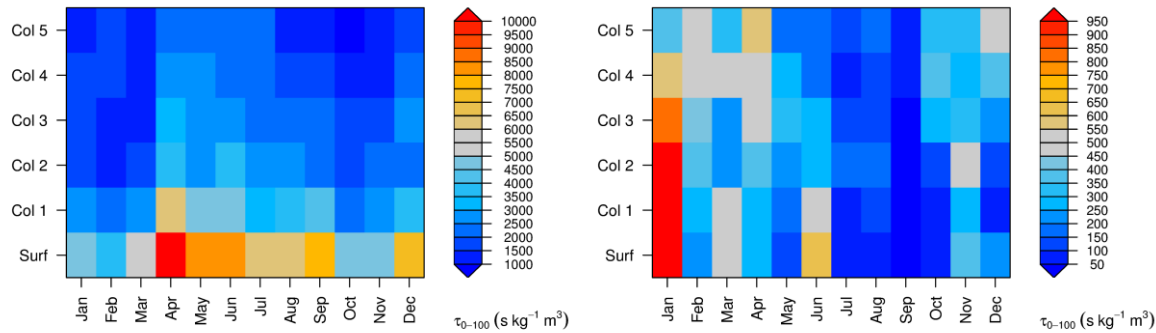


Figure 9, Annual cycle of FLEXPART derived total surface residence time over land for different vertical arrival columns above Jungfraujoch: (left) for land surfaces north of 30°N and (right) for land surfaces south of 30°N.

Which role plays the seasonality of different tropopause height occurrence frequency over JFJ in winter and summer for the interpretation of the CO₂ columns and the summer - winter difference between FTIR and NDIR? Further as mentioned in the manuscript also the seasonality in the UTLS modifies the column. Is it possible to quantify this a bit more?

Indeed, this is an interesting, important, and valid point that hasn't been addressed in the present work, therefore we cannot adequately reply to it. Generally, we would expect a lower tropopause could potentially lower the column integrated CO₂ value due to the expected lower stratospheric CO₂ mole fraction. A detailed analysis regarding this issue requires substantial additional modeling, which was not possible within this work.

p.5. 1. 13: Please specify the long-term stability (i.e. error due to drift) and the total uncertainty of the NDIR.

The value given in the manuscript corresponds to the standard deviation of several cylinder measurements each lasting at least one hour. The gas from the cylinders was treated, calibrated, and evaluated exactly the same way as outside air, which is why we consider this standard deviation as the precision of our system. The long term stability is taken care of by frequent measurements of calibration gases (see section 2.2). To make this clearer, we changed the sentence at page 5, line 14 from:

“Cylinder measurements with a known mole fraction showed a precision better than 0.04 ppm for 1 hour analysis.”

to:

“Cylinder measurements with a known mole fraction showed a long-term precision for hourly averages better than 0.04 ppm. The accuracy of our target cylinder corresponds to less than 0.1 ppm (WMO target value for CO₂ measurements) calculated as standard deviation of the mean considering the number of independent calibration set (high span, low span, working gas).”

Technical: Fig.3: The caption refers to black lines or dots, which I can't find. Please correct.

That's correct, the caption refers to an older version of the figure. It was changed to:

“Figure 3. A: Histogram of all NDIR residuals (yellow) and the filtered NDIR residuals representing the background values (red) of the in-situ measurements; B: Histogram of all FTIR residuals (light blue) and the filtered FTIR residuals representing the background values (blue) of the column.”

Changes on the authors' behalf:

The wavelength of the NDIR analyzer was added, p. 5, line 8 was changed from:

“...NDIR spectrometer (Maihak S710) with a frequency ...”

To:

“...NDIR spectrometer (Maihak S710) measuring at a wavelength of 4.26 μm with a frequency...”

The Figures' numbers were updated because of the additional figure.

For more clarity, page 15, line 27 was changed from:

“...or (c) since the FTIR retrievals has little vertical sensitivity the measured column signal contains mixed information from the troposphere and the stratosphere.”

to:

“...or (c) since the FTIR vertical sensitivity was not exploited in the present retrievals the measured column signal contains mixed information from the troposphere and the stratosphere.”

The reference of Rothman et al., (2005) at page 22, line 19 was moved down after Revelle et al. (1957), to maintain the correct alphabetical order.