Interactive comment on "Retrievals of formaldehyde from ground-based FTIR and MAX-DOAS observations at the Jungfraujoch station and comparisons with GEOS-Chem and IMAGES model simulations" by B. Franco et al.

### **Anonymous Referee #1**

Received and published: 2 December 2014

# We thank Referee#1 for his/her review and the constructive comments. Please find below our responses written in blue.

General comments. Two new sets of formaldehyde measurements at the Jungfraujoch station retrieved from ground-based Fourier Transform Infrared solar spectra and UV-Visible Multi-AXis Differential Absorption Spectroscopy are presented in the paper. These sets are indirectly compared to check their consistency with each other using as intermediate step two global Chemistry Transport Models, GEOS-Chem and IMAGESv2. As pointed out in the text the two sets are complementary since the vertical sensitivity of the retrieval is different for the IR and the UV technique. The data sets presented here can be of interest to many users for example to assess the accuracy of space based retrievals of formaldehyde. The paper is well written and clearly structured. The scientific methods used and the assumptions made are sound and well described. The subject of the paper suits well the scope of AMT. I recommend publication after minor revisions.

There are some aspects where I would appreciate a bit more information. Since the comparison between the models and the measurements is carried on using daily averages I would like to see some discussion about the daily variation of the measurements and the models and how well it is captured by those. It would be interesting to discuss how the model output has been matched to each particular measurement (I assume it is plus minus 3 and 4 hours for GEOS-Chem and IMAGES respectively but it is not clear for me when reading the text).

The model outputs have not been matched to particular measurements within a day of observation. In the framework of this paper, we investigated the overall consistency between FTIR, MAX-DOAS and model data sets by considering the daily averages only. Moreover, the time series used in this study span 2.5 years only and hence do not provide enough statistics to investigate reliably the intra-day variations of HCHO. Such intra-day variations will be the topic of a manuscript currently in preparation, using more statistics from the long-term time series of FTIR spectra (1988-2014) involving more than 9500 individual measurements, and preliminary results have indeed revealed intra-day variations of HCHO at Jungfraujoch. Their precise characterization and representation by the models remain to be determined and verified.

The discussion about FTIR retrievals mentions the large residuals introduced by the interfering species. I would suggest to add to figure 1 another panel showing the residuals when HCHO is not included in the forward model. It will give an idea of the HCHO signal in the spectra.

This is indeed an excellent suggestion. We have added to Fig. 1 the residuals (in dark grey, see below) when HCHO is not taken into account during the fitting; the corresponding RMS for both fits is reported in the boxes (same colour key than for the residuals). The impact on the residuals is obvious in the first 3 microwindows, but not in the last one. This is partly due to the fact that the HCHO absorption is very weak within this domain range. Nonetheless, the

use of this fourth microwindow is crucial as it stabilizes the retrievals and considerably helps to reduce the residuals in the first 3 microwindows.



It is also mentioned that experiments to use HITRAN 2012 instead of HITRAN 2008 produced significant changes in the retrievals when using it for the interfering species. It will be extremely useful if you could provide any further thoughts about it. I don't think changes in HITRAN 2012 with respect to HITRAN 2008 in the 2700-2850 cm-1 region can justify such an important change in the retrieved concentrations.

As quoted in the original manuscript (Page 10732, line 26), HCHO changes in HITRAN 2012 with respect to HITRAN 2008 (which mainly consist in new self- and N<sub>2</sub>-broadening coefficients) have a limited impact on the retrieved columns of HCHO over the 2010 - 2012 period. Changes related to the interfering species are therefore mainly responsible for the significant differences in retrieved columns between HITRAN 2012 and HITRAN 2008. Among the line intensity uncertainties linked to the interfering species and evaluated in the FTIR error budget (Table 2), CH<sub>4</sub> is by far the larger contributor to the errors affecting the retrieved columns of HCHO. Hence we hypothesize that CH<sub>4</sub> changes in HITRAN 2012 with respect to HITRAN 2008 could be responsible for the large differences.

Since the publication of the HCHO cross sections by Meller and Moortgat in 2000, Chance and Orphal (doi:10.1016/j.jqsrt.2011.02.002) published new cross sections in 2011. Have you considered using these newer cross-sections? If not, why?

The Chance and Orphal (2011) HCHO cross sections have not been considered here. They correspond to the Cantrell et al. (1990) cross sections rescaled in intensity to Meller and Moortgat (2000). This rescaling was done because it has been demonstrated that Cantrell et al. (1990) cross sections are biased systematically low (Chance and Orphal, 2011). We should therefore expect very similar HCHO SCD values using Meller and Moortgat (2000) as in the present study and Chance and Orphal (2011). This has been recently confirmed from sensitivity tests on OMI HCHO retrievals using both cross sections data sets (I. De Smedt, private communication). In the error budget on the retrieved HCHO vertical columns (see Table 3), we report an uncertainty of 9% related to the HCHO cross sections, which was derived from the mean bias between the Cantrell et al. (1990) and Meller and Moortgat (2000) cross sections in the 336.5-359 nm wavelength interval (Pinardi et al., 2013). This value of 9% should be considered as an upper limit, given the fact that Cantrell et al. (1990) is biased low and Meller and Moortgat (2000) and Chance and Orphal (2011) are in good agreement. It should be noted that the Meller and Moortgat (2000) cross sections are used in all our MAX-DOAS and satellite nadir HCHO retrievals (see e.g. De Smedt et al., 2012), which allows a more direct comparison between the ground-based and satellite HCHO data sets.

# I would appreciate a comment about the daily variations of the AK.

As explained previously, investigation of the daily variations (as well as of the inter-annual and long-term variability) of HCHO will be the subject of a follow-up paper. However, we can indicate that despite slightly varying during the day (typically in the range from 0.7 to 1.3 for the FTIR data set), the DOFS remains sufficient around noontime to allow for the investigation of the intra-day variability of HCHO. Regarding MAX-DOAS, the daily DOFS is typically varying between 1.6 and 2.2.

# Suggested technical corrections:

Thank you for your corrections, the manuscript has been amended accordingly. We have responded in blue to your comments when needed.

- Page 10716: Line 2, Correct "As a"
- Page 10717: Line 1, could you please add to the list of papers Fu et al., 2007 (doi:10.1029/2006JD007853)
- Page 10718: Line 20, include reference to Bey et al., 2001
- Page 10719: Line 2, change "trend" for "record" since the record may or may not show a trend
- Page 10720: Line 15, remove "the" in "in the northern Italy"
- Page 10721: Line 2, substitute invert by inverse
- Line 9, possible rephrasing of "and allows for characterizing. . ." to "and allows the characterization of the vertical information content of the measurement"
- Line 25, add Dufour et al., 2009 (www.atmos-chem-phys.net/9/3893/2009/) for ACE-FTS measurements
- Line 26, possible rephrasing "The simulated HCHO concentration profiles shows a. . " to "The concentration of the simulated HCHO profiles shows. . ."
- Page 10723: Line 20, correct "Finally, an S/N..." to "Finally, a S/N..."

- Line 23, I would change in for of "retrieve HCHO at ISSJ consists in. . ." to "retrieve HCHO at ISSJ consists of. . ."
- Line 27, I would remove "and" after parenthesis.
- Line 28, I would change "as such" for something like "These spectrum-specific scaled profiles are the used as a priori. . ."
- Page 10724: Line 18, is it the only MAX-DOAS in Xianghe. I think it will be better to say something like "instead of 400-720 nm as described in Wang. et al., 2014 using the reference instead of the city.

Page 10724, line 18: In the revised manuscript, "(400–580nm here, instead of 400-720nm in Xianghe)" is replaced by "(400–580nm here, instead of 400-720nm for our instrument operating in China; see the three references here above)".

- Page 10725: Line 22, I would rephrase "remote station with therefore low. . ." to "remote station with low. . ."
- Page 10726: Line 15, I would rephrase "These are calculated with by utilizing the. . ." to "These are calculated using the. . ."
- Page 10728: In the description of the GEOS-Chem model it may be worth to add the following reference which describes the isoprene oxidation mechanism by Mao et al., 2013 (doi:10.1002/jgrd.50817)
- Page 10729: Line 11, I would suggest to remove also to have "The model uses anthropogenic..."
- Line 15, I would change "examples" to "example"
- Page 10731: Line 2, I would change "may be obtained from the MAX-DOAS averaging kernels" to "may be obtained from the MAX-DOAS measurements".
- Page 10732: Line 7, suggested rephrasing "Most of the error terms have been dealt with using perturbation methods applied to all solar spectra recorded during year 2011, according to specifics given in the last column of Table 2, while the respective contributions. . ." to "While most of the error terms have been dealt with using perturbation methods applied to all solar spectra recorded during the year 2011 (details are given in third column of Table 2) the contributions of measurements. . ."
- Page 10734: Line 5, suggested rephrasing "aerosol extinction and a priori HCHO profiles, albedo)" to "aerosol extinction, HCHO a priori profiles and albedo)".
- Line 21, I would remove "In this purpose"
- Page 10735: Line 11, rephrase suggestion, "for HCHO at Jungfraujoch, we have also added to the following figures involving the FTIR and MAX-DOAS columns, the HCHO concentration retrieved by MAX-DOAS. . ." to "for HCHO measurements at Jungfraujoch, we have added to figures 4, 6 and 7 involving the FTIR and MAX-DOAS columns the HCHO concentration retrieved by MAX-DOAS. . ." because with the original sentence is not clear which figures are affected.
- Page 10736: Line 27, The sentence starting with "It also appears that other. . ." needs clarification.

Line 10736, line 27: The sentence has been reformulated: "It is difficult to compare the ISSJ with other background sites because many of them experience enhancements in HCHO concentration resulting from distant anthropogenic activity and biomass burning."

- Page 10738: Line 21, remove "-" from highly-industrialized and –populated
- Page 10743: Line 3, change equals to equal
- Line 5, change equals to equal
- Line 24, change "such" to "these"
- Legend Table 2, rephrase suggestion, ",excepting" by "with the exception of"

- Legend Table 3, it would be nice to mention that it is for the MAX-DOAS measurements
- Figure 6, y-label of lower panel is not completely visible. This problem has been fixed.

Interactive comment on "Retrievals of formaldehyde from ground-based FTIR and MAX-DOAS observations at the Jungfraujoch station and comparisons with GEOS-Chem and IMAGES model simulations" by B. Franco et al.

M.K. Sha (Referee) mahesh.sha@kit.edu

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We thank Dr Sha you for his review and the constructive comments. Please find below our responses written in blue.

# General comments:

This paper presents a study of the retrieval strategies for the formaldehyde profiles retrieved from the measurements performed using ground based Fourier Transform Infrared (FTIR) solar absorption spectra and the UV-Visible Multi-AXis Differential Optical Absorption Spectroscopy (MAX-DOAS) scans at the high altitude remote Jungfraujoch station. A comparison of the measurement data with two state-of-the-art global Chemistry Transport Models (CTMs), GEOS-Chem and IMAGESv2 has been shown. The seasonal variability of formaldehyde has been shown from the retrievals using the CTMs output as intermediate input. Furthermore, it is well shown how one can get complementary information about the vertical distribution of formaldehyde by using the two data sets of the IR and UV-VIS instruments which have different vertical resolution and sensitivity. The retrieval results have the potential to improve the accuracy of formaldehyde measurements performed from space based instruments. The paper describes the work very well and in a structured manner. Therefore I recommend it for the AMT publication with some minor additions as outlined below in the specific and technical comments.

Specific comments:

Out of the six microwindows used by Vigouroux et al.only four of them have been selected for the analysis of the FTIR data in this study. It is pointed out in the paper that the reason for discarding the two other microwindows is due to the presence of systematic residuals or very strong interferences blinding the weak formaldehyde absorption. I would suggest including the plots for the two discarded microwindows in figure 1 as this will show the amplitude of the interference in the residual.

We certainly concur with this suggestion but would like to point out that the simultaneous use of 6 microwindows to fit HCHO also impacts the residuals in the first 4 microwindows (most prominently in MW3 around 2778 cm<sup>-1</sup>). We have therefore prepared an additional figure similar to Fig. 1, but showing the fitting of HCHO with the 6 microwindows (see below), where the larger residuals in MW5 and MW6 (the two bottom frames) with respect to the first four microwindows are obvious. We thought of adding this figure in the Supplementary Material as Fig. S2 and a reference to it has been added in the manuscript. Furthermore, the residuals when assuming no HCHO absorption during the fitting process are also shown as grey curves, similarly to the new Fig. 1.



Additionally you may spare few words on why you have the selected spectrum measured at SZA of  $80^{\circ}$  for figure 1.

We have selected this spectrum because it has been recorded on the same observational day as the example of MAX-DOAS scan fit presented in Fig. 2. In addition, such a geometry of observation can be considered as typical since a SZA of  $80^{\circ}$  corresponds approximately to the  $60^{\text{th}}$  percentile value of the SZA distribution from the entire FTIR data set.

I would appreciate if you could include a figure showing the formaldehyde profile distribution from the combined results of both instruments.

Unfortunately combining the profile distributions from both instruments is not an easy task given the respective vertical sensitivity and resolution of FTIR and MAX-DOAS products and would require further efforts. This would require "nearly-simultaneous" measurements to correct for the bias and to smoothly connect the FTIR and MAX-DOAS profiles, which are not strictly complementary because the FTIR retrievals are also sensitive in the lower troposphere (although less than MAX-DOAS).

Figure 6 c, lower panel has the y label cut. Please increase the size of the axis-labels and the legends.

The problem of the y-label has been fixed. In the final version of the manuscript, the figure will be presented on a page with portrait orientation and hence will be bigger and easier to be read.

Technical comments:

Thank you for your corrections, the manuscript has been amended accordingly. We have responded in blue to your comments when needed.

- Page 10717: Line 22, "various" – please give the name of the instruments.

These are SCIAMACHY, GOME and ACE-FTS instruments. We have added the names in the manuscript, Page 10717, line 22.

- Page 10718: Line 16, please modify "IR" to "infrared (IR)".
- Page 10719: Line 4, here you may include the names of some of the presently orbiting and future proposed satellite missions which can measure formaldehyde.
  We have included the previous instruments as well as TROPOMI in the text, Page 10719, line 4.
- Page 10720: Line 24, I would change "Bruker IFS-120 HR" to "Bruker IFS 120 HR".
- Page 10720: Line 24, I would change "InSb" to "Indium Antimonide (InSb)".
- Page 10720: Line 24, I would change "HgCdTe" to "Mercury Cadmium Telluride (HgCdTe).
- Page 10721: Line 2, inverse of twice the maximum optical path difference is the spectral sampling. Do you apodize the interferograms? What is the reason for the varying spectral resolution?

There is no apodization involved in the treatment. The spectral resolution is selected according to the time of the day: higher resolution (2.85mK, typically 5 scans of 69 s each) for high sun, lower resolution (4.96mK and typically 3 scans (but as low as 1)) of 40 s each for low sun. In addition, if the weather conditions are not fully optimal (e.g., wet conditions), we only measure at lower resolution to avoid modulation of the signal at higher resolution. Beware that the total recording time is significantly longer than the cumulative duration of the scans (mirror back and FT computation).

Note that there was a mistake Page 10721, line 3: "alternating between 0.004 and 0.006  $cm^{-1}$ " has been corrected by "alternating between 0.003 and 0.005  $cm^{-1}$ ". Thank you for bringing our attention on this.

Page 10721: Line 4, how many spectra are co-added here to improve the S/N? Generally three consecutive individual spectra are co-added.

- Page 10723: Line 15, a figure showing the formaldehyde profile would be helpful here.

We have added as Fig. S1 to the Supplementary Material a figure showing the HCHO a priori profile used for the FTIR and MAX-DOAS retrievals, and the profile of relative standard deviations used as diagonal elements of the covariance matrix for the FTIR retrievals. Please find this new figure here below. We now refer to it in the updated manuscript, Page 10723, line 15.



- Page 10724: Line 10, I would change "nadir satellite" to "nadir viewing satellite".
- Page 10725: Line 23, I would change "At ISSJ, a remote station with therefore low HCHO content, these two settings . . ." to "ISSJ being a remote station has low HCHO content, as a result these two settings . . .".

Page 10725: Line 28, please spare few words on the reason to select a fifth-order polynomial for the fit.

The choice of a fifth-order polynomial for the HCHO DOAS fit is extensively described in Pinardi et al. (2013). Sensitivity tests showed that any change of the polynomial order has a strong impact on the diurnal behavior of the HCHO DSCD, especially for high elevation angles, including zenith. It was found that using a lower polynomial order (e.g. 3<sup>rd</sup> order) introduces a misfit that activates the correlation between Ring and HCHO differential absorption features. A number of combinations of polynomials and Ring effect cross sections were investigated and the sensitivity of the retrieved HCHO DSCDs to the choice of the Ring effect cross section was smallest when using a polynomial of order 5. We have added the Pinardi et al. reference on Page 10725, line 28 to justify the selection of an order 5 polynomial.

Page 10727: Line 18, please specify what is meant by thin cloud conditions? Which cloud index or threshold has been used?

The cloud screening method developed by Gielen et al. (2014) is based on the color index (CI; ratio of the radiance at two wavelengths) measured at zenith and its diurnal variation. This

empirical parameter gives information about the colour of the sky: from blue during clear skies to white/grey when clouds or aerosols are present. Based on the comparison between measured and simulated CI, the following sky conditions can be defined: clear-sky, thin (white/light grey) clouds/polluted, thick (dark grey) clouds/heavily polluted, and broken clouds. Quantitative information such as the cloud cover percentage cannot be derived from this method. In the present study, we reject MAX-DOAS scans corresponding to thick clouds and broken clouds conditions, i.e. those for which the quality of the aerosol and trace gas retrievals are potentially strongly affected by the sky conditions. The sentence "*The cloud screening method developed by Gielen et al. (2014) and based on the measured color index at zenith has been therefore utilized as an additional selection criterion: only scans corresponding to clear-sky or thin clouds conditions are further selected for the comparison with FTIR and model data.*" has been rephrased as follows, Page 10727, line 18:

"The cloud screening method developed by Gielen et al. (2014) and based on the measured color index at zenith has been therefore utilized as an additional selection criterion. This empirical parameter gives information about the colour of the sky: from blue during clear skies to white/grey when clouds or aerosols are present. Based on the comparison between measured and simulated CI, the following sky conditions can be defined for each MAX-DOAS scan: clear-sky, thin (white/light grey) clouds/polluted, thick (dark grey) clouds/heavily polluted, and broken clouds. In the present study, MAX-DOAS scans corresponding to thick clouds or broken clouds are rejected since the quality of the aerosol and trace gas retrievals can be potentially strongly affected by these sky conditions."

Interactive comment on "Retrievals of formaldehyde from ground-based FTIR and MAX-DOAS observations at the Jungfraujoch station and comparisons with GEOS-Chem and IMAGES model simulations" by B. Franco et al.

#### **Anonymous Referee #3**

Received and published: 19 December 2014

We thank Referee #3 for his/her review and the constructive comments. Please find below our responses written in blue.

#### **General Comments**

This manuscript describes the retrieval of vertical profiles of HCHO at the high-altitude Jungfraujoch station from July 2010 to December 2012 using both FTIR and MAXDOAS techniques. The retrieval methodologies are compared and the vertical sensitivity of each technique is assessed. The measurements are interpreted using the GEOS-Chem and IMAGESv2 chemical transport models. The seasonal cycle of HCHO is investigated, and is found to be consistent between the two datasets, while the models generally underestimate summertime HCHO. Overall, the manuscript provides a clear and comprehensive description and validation of an optimized FTIR retrieval strategy. The results should be of interest to the FTIR, DOAS, and CTM communities. The manuscript is well written and is suitable for publication in AMT after the minor comments below are addressed.

#### Specific Comments

Page 10721, line 2 – Isn't 1/MOPD used more often than 1/2MOPD? For boxcar apodization, the first zeros of the sinc function are separated by 1/MOPD. A separation of 0.5/MOPD implies the Rayleigh resolution criterion (sinc2 ILS of a grating spectrometer), where two sinc2 features are resolved when the peak of one is located at the first zero of the other. Using RES = 0.5/MOPD implies MOPD of 125 to 83 cm for RES = 0.004 to 0.006 cm-1, which is lower than the usual 250 cm for NDACC FTIR measurements.

In our case as well as in many other FTS studies, the spectral resolution is defined by 1/(2xOPD). This definition is simple to use and very close of the full width at half maximum. The OPD are respectively 175 cm for 2.85 mK and 100 cm for 4.96 mK and we never push the instrument to the ultimate resolution for atmospheric observations. To clarify this point, we have reformulated this definition in the manuscript, Page 10721, line 2, as follows: "It is characterized by a typical spectral resolution (defined here as the inverse of twice the maximum optical path difference) alternating between...".

However, there was a mistake regarding the resolution indicated in the manuscript: "0.004 to 0.006 cm<sup>-1</sup>" has been corrected by "0.003 to 0.005 cm<sup>-1</sup>". Thank you for bringing our attention on this.

Page 10727, line 3 - Comment on why the correlation length for MAX-DOAS (0.2 km) is so much smaller than that for FTIR (3 km).

This difference on the choice of the correlation length is mainly due to the difference in vertical sensitivity between both techniques. Fig. 3 shows that FTIR HCHO averaging kernels are not vertically resolved. In contrast, MAX-DOAS measurements show sensitivity to trace

gas and aerosol vertical distributions roughly in the first kilometer above the station, with a maximum of sensitivity in the first altitude layers close to the station. Using a correlation length of 3 km as for FTIR would significantly limit the ability of the MAX-DOAS profiling algorithm to retrieve surface concentration variations independently from the upper layers. 0.2 km was found to optimize the DOFS of the MAX-DOAS retrievals.

Page 10733, para 1 – Could comment on why the HITRAN 2012 data is causing problems. Which interfering species are causing the issues?

As quoted in the original manuscript (Page 10732, line 26), HCHO changes in HITRAN 2012 with respect to HITRAN 2008 (which mainly consist in new self- and N<sub>2</sub>-broadening coefficients) have a limited impact on the retrieved columns of HCHO over the 2010 – 2012 period. Changes related to the interfering species are therefore mainly responsible for the significant differences in retrieved columns between HITRAN 2012 and HITRAN 2008. Among the line intensity uncertainties linked to the interfering species and evaluated in the FTIR error budget (Table 2), CH<sub>4</sub> is by far the larger contributor to the errors affecting the retrieved columns of HCHO. Hence we hypothesize that CH<sub>4</sub> changes in HITRAN 2012 with respect to HITRAN 2008 could be responsible for the large differences.

Page 10733, lines 18-20 – This gives a progressive increase in temperature error, but Table 2 just says "±4 K around NCEP noon profile". Which is correct?

Page 10733, lines 18-20: The progressive increase in temperature error is the correct statement. As the indication in Table 2 is misleading, we have replaced it by "Assuming the NCEP profile uncertainty pattern (see text.)".

Page 10734, para 1 – Revise this paragraph to address each of the errors in Table 3, in turn. Some error sources seem to be missing from the text (smoothing and noise errors of 9.1%). How is the 20% error on the O4 cross section related to the 6.3% uncertainty related to aerosols?

The only missing error sources in the text is the combined smoothing and noise errors of 9.1%. To address this point we have added the following sentence on page 10734, line 6: "The combined smoothing and measurement noise errors reach 9.1%.". The error on HCHO VCD related to the uncertainty on the aerosol retrieval is estimated from a sensitivity approach using retrieved aerosol extinction profiles plus their corresponding error (i.e., the sum of smoothing and noise errors plus a 20% error due to the uncertainty in the O4 cross sections; see Clémer et al., 2010 and Wagner et al., 2009) as input and comparing the results to the standard HCHO retrievals. For the sake of clarity, the following sentence on page 10734, lines 6-10: "Regarding the uncertainty related to the aerosol profile retrieval, an error of 6.3% is obtained on average on the retrieved HCHO VCD when the smoothing error and measurement noise errors on the retrieved aerosol profiles are combined to a systematic error of 20% on the O4 cross section (Clémer et al., 2010; Wagner et al., 2009)." has been replaced by "Regarding the uncertainty related to the aerosol profile retrieval, an error of 6.3% is obtained on average on the retrieved HCHO VCD based on a sensitivity test approach using aerosol extinction profiles plus their corresponding error (i.e., the sum of smoothing and noise errors plus a 20% error due to the uncertainty in the O4 cross sections; see Wagner et al., 2009 and Clémer et al., 2010) as input and comparing the results to the standard HCHO retrievals."

- Page 10734, line 24 – Are the FTIR and MAX-DOAS measurements also daily averaged?

Yes, these are daily-averaged FTIR and MAX-DOAS data that are compared to the models. We have added this information at the beginning of the same paragraph, Page 10734.

Page 10740, lines 8-10 – "Indeed, the mean fractional differences between the models and the FTIR data are generally consistent with the mean fractional differences calculated between the models and the MAX-DOAS observations (Table 4)." This sentence does not seem consistent with the results in Table 4, e.g., compared to IMAGES, the mean fractional differences for FTIR is negative and those for MAX-DOAS is positive. And for GEOS-Chem, the differences wrt FTIR are twice as large as those for MAXDOAS. Clarify this discussion here and in the Conclusions, page 10746, lines 7-11.

Indeed, these sentences are misleading. They have been reformulated as follows:

Page 10740, line 8-10: "Indeed, the mean fractional differences between the models and the FTIR data as well as the mean fractional differences calculated between the models and the MAX-DOAS observations are embedded in the respective uncertainties of the ground-based measurements (see Table 2, 3 and 4)."

Page 10746, lines 7-11: "Moreover, when compared to the FTIR and MAX-DOAS measurements, the CTMs outputs have shown discrepancies embedded in the respective uncertainties of the ground-based measurements, i.e. mainly an underestimation in summer for both models. These results indicate a consistency between the HCHO retrievals from both remote sensing techniques as well as their complementarity for studying the vertical distribution of HCHO above Jungfraujoch."

- Page 10740, line 22-25 – Is the slope expected to be linear at these altitudes? Explain why.

The HCHO profile is not expected to be linear in the lower troposphere, especially during summertime with higher values in the first layers (see Fig. 5). Nonetheless, the slope of a linear regression adjusted to the low-tropospheric profile is a convenient way to gauge and appraise the seasonal variations of the HCHO profile in the first layers.

# **Technical Corrections**

Thank you for your corrections, the manuscript has been amended accordingly. We have responded in italic to your comments when needed.

- Page 10717, line 3 associated with
- Page 10718, line 4 part of the NDACC (Network . . .)
- Page 10718, line 20 Is there a reference for GEOS-Chem v9-01-03 ?
  We have added Bey et al. (2001) as a reference for GEOS-Chem, Page 10718, line 20.
  Also an external link towards the GEOS-Chem v9-01-03 site is given in Sect. 2.4 providing an overview of the model.
- Page 10718, line 20/21 total columns . . . partial columns
- Page 10719, line 5/6 The measurement site . . . 2. A short description . . .
- Page 10719, line 10 reports the results of
- Page 10720, line 15 in Northern Italy . . . of the Alps
- Page 10721, line 2 inverse of
- Page 10723, line 23 consists of two

- Page 10723, line 28 then used as a priori
- Page 10724, line "In brief, it is a dual-channel system composed of two grating spectrometers covering the UV (300–390 nm) and visible (400–580nm here, instead of 400–720nm in Xianghe) wavelength ranges and connected to cooled CCD detectors." Provide some context for Xianghe this is the only mention of it except in the title of the Deng et al. reference.

In the revised manuscript, page 10724, line 16: "(400–580nm here, instead of 400-720nm in Xianghe)" is replaced by "(400–580nm here, instead of 400-720nm for our instrument operating in China; see the three references here above)".

- Page 10726, line 1/2 example of a DOAS fit . . . same date as
- Page 10726, line 10 path through the atmosphere
- Page 10715, line 15 as the forward model . . . (also suggest starting a new paragraph at the end of this sentence)
- Page 10726, line 25 Explicitly state that the SÂn\_e and S\_a matrices are for aerosols, as there is some ambiguity with HCHO.
  - Actually the Sɛ and Sa matrices for <u>both</u> aerosols and HCHO are constructed as in Clémer et al. (2010). We give also details about that just after line 25 (from line 26, page 10726 to line 5, page 10727). For the sake of clarity, the sentence on line 25 has been replaced in the revised manuscript by "Sɛ and Sa matrices for both aerosols and HCHO are constructed as in Clémer et al. (2010; see also Hendrick et al., 2014; Wang et al.; 2014).". The next sentence has been also modified as follows: "...times the maximum partial aerosol optical depth or vertical column density (VCD) of the profiles." instead of "...times the maximum partial vertical column density (VCD) of the profiles.".
- Page 10727, line 26 change "lumping" to combining?
- Page 10728, line 20 closest pixel to
- Page 10729, line 29 consists of HCHO . . . closest pixel to the station
- Page 10730, line 17 Here and throughout the paper, change "1 sigma" to "1sigma". The former looks like the difference between 1 and sigma.
- Page 10731, line 12 reduced to 50%
- Page 10732, line 2 from individual FTIR solar spectra
- Page 10732, line 5 Clarify what is meant by "with an assumed variability close to 50%". It is not clear what this refers to.
  This refers to the assumed covariance (diagonal). We have rephrased it as follows, Page 10732, line 5: "with an assumed variability (i.e. diagonal of covariance) close to 50%".
- Page 10732, line 19 Delete "remained".
- Page 10732, line 21 to the 10% uncertainty assumed here
- Page 10733, line 4 parameters have
- Page 10733, line 13 Make clear that this is the "Retrieval algorithm-related" term in Table 2 use same terminology as in the table.
- Page 10733, line 26 Change "2 sigma" to "2sigma".
- Page 10733, line 22 columns on the
- Page 10734, line 2 The error budget on the retrieved MAX-DOAS HCHO VCDs is
- Page 10734, line 21 For this purpose,
- We have deleted "For this purpose" from this sentence.
- Page 10734, line 25 averaging kernels and a priori profiles from the . . .
- Page 10735, line 5 CTM outputs
- Page 10735, line 9 confronted by MAX-DOAS

- Page 10735, para 2 Change "1 sigma" to "1sigma" (three places). Also page 10738, line 29; page 10739, lines 13 and 29, page 10740, line 20, etc.
- Page 10736, line 9 representing a seasonal amplitude
- Page 10736, line 26 by an amplitude of
- Page 10737, line 8 from Europe to Ny Alesund (correct?) occur especially. . .
- Page 10738, line 29 associated with the
- Page 10739, line 27 IMAGES columns underestimate and overestimate the FTIR
- Page 10740, line 1 associated with these
- Page 10740, lines 7,11,19 and Table 4 caption CTM (or CTMs')
- Page 10740, line 17 for the FTIR and
- Page 10741, line 8/9/10 MAX-DOAS profiles . . . directly above . . . associated with the
- Page 10741, line 14 on average
- Page 10741, line 16 due to lower elevation
- Page 10741, line 19 Delete "instruments".
- Page 10741, line 20 What is meant by "efficient" measurements? Use a better description.

We have reformulated as follows, Page 10741, line 20: "...and provide added-value measurements of HCHO distribution..."

- Page 10742, line 3 of the CTM outputs relative to the FTIR profile extend throughout
- Page 10742, line 11 consist of
- Page 10743, line 3 equal to
- Page 10743, line 5 consistent with
- Page 10743, line 7 equal to
- Page 10744, line 4 CTM
- Page 10744, line 10 precursor concentrations
- Page 10744, line 24 associate with
- Page 10745, line 8 an OEM retrieval process. The MAX-DOAS strategy consists of deriving DSCDs...
- Page 10745, line 20 HCHO columns and profiles simulated by
- Page 10746, line 1 total column
- Page 10746, line 3 Considering that
- Page 10746, line 5 lower-tropospheric
- Page 10746, line 7 CTM
- Page 10746, line 12 remote location
- Page 10746, line 17 combined with CTM outputs . . . an important background test site
- Page 10763, Table 4 caption Mean fractional differences (in % +- 1sigma), calculated
- Page 10763, top row of Table 4 3.6-3.8 km MAX-DOAS column
- Page 10764, Figure 1 caption Typical examples of HCHO spectral fits at ISSJ
- Page 10767, Figure 4 caption Change "1 sigma" to "1sigma". Line 7: associated with
- Page 10768/9, Figure 5/6 captions Change "1 sigma" to "1sigma".
- Page 10770, Figure 7 caption smoothed CTM . . . and the model outputs

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