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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Diurnal cycle and multi-decadal trend of formaldehyde in the remote atmosphere near 46 N" *by* B. Franco et al.

Anonymous Referee #1

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This paper presents a multi-decadal time series (01/1988-06/2015) of HCHO total columns retrieved from ground-based high-resolution Fourier transform infrared (FTIR) solar spectra recorded at the high-altitude station of Jungfraujoch (Swiss Alps, 46.5°N, 8.0°E, 3580 m a.s.l.). The HCHO diurnal cycle is first investigated and quantitatively characterized by fitting a parametric model to the observations. A maximum of HCHO is found around noontime and is attributed to the modulation of insolation during daytime and its impact on methane oxidation. Retrieved HCHO columns are then compared to simulations from the 3D-CTM GEOS-Chem and a trend analysis over a 27-year period is performed. All the individual FTIR measurements are scaled beforehand to a fixed local time using the parametric model in order to remove the effects of the intra-day modulation on both comparison and trend results. Sensitivity tests with

GEOS-Chem suggest that the atmospheric CH4 oxidation is the main driver of the seasonal and inter-annual HCHO column variations at Jungfraujoch. The observed trend, with an increase between 1988 and 1995, followed by a decrease between 1996 and 2002, and again an increase from 2003 onwards, is seen to be related to the atmospheric CH4 fluctuations and the short-term OH variability.

This paper is well written and presents very interesting results which fit well with the scope of ACP. I recommend the final publication of the manuscript after addressing the following comments:

Specific comments:

Page 31295, lines 1-4: Ground-based FTIR retrievals with DOFS lower than 0.35 have been discarded from the study. Using a DOFS threshold value lower than 0.5 means that retrievals for which the information content comes mainly from the a priori (DOFS between 0.35 and 0.5 in the present case) and not from measurements are also selected for the study. Figure 2a shows that around local noon the DOFS is very low (<0.5), especially for the summer months. The question is therefore: how the choice of the a priori affects the amplitude of the intra-day variations derived for the different months of the year ? Should we expect larger or lower amplitude when using an a priori profile having higher or lower concentration (and therefore column) values than for the a priori chosen in this study ? Sensitivity tests are needed here in order to investigate this issue.

Page 31305-6, Section 5.1: In addition to the correlation coefficient, the authors should also provide the slope and intercept values corresponding to the scatterplot presented in Figure 6. This Figure shows that the intercept is about 0.3E15 molec/cm2, which is quite higher than 0. Can a possible bias between the LIEGE and BRUCKER data sets be fitted by the bootstrap method ? If yes, the authors should try to include it in their trend analysis and check whether the inferred bias value is indeed not significant.

Page 31311, last paragraph of the conclusions: the parametric model developed in this

study could be potentially used in satellite validation efforts for the conversion of the FTIR HCHO columns to the satellite overpass times. What would be the uncertainties related to this scaling ? I think this should be discussed in the paper.

Page 31331, Figure 5d: The monthly total carbon emissions are given for a large domain over Europe (between 38-86°N and -15-55°E). Does this domain representative of the Jungfraujoch region ? Do emission data exist for a smaller domain closer to Jungfraujoch and to the resolution of the GEOS-Chem model (2°x2.5°) ? If yes, are the evolutions of the anthropogenic, biogenic, and pyrogenic carbon emissions similar to the ones presented in Figure 5d ?

Technical corrections:

Page 31290, lines 21-22: Since MAXDOAS HCHO retrievals are also presented in Vigouroux et al. (2009), I would include this paper in the list here.

Page 31300, lines 3-5: A reference for the Weibull probability distribution should be provided here.

Page 31327, Figure 1: Is there a geophysical reason behind the choice of the different months in the four plots (e.g., Jun/Apr/Dec in plot 1a, Feb/Aug/Oct in plot 1b, etc) or is it only for the sake of clarity ? Maybe plotting separately each month in a different plot, i.e. having Figure 1 composed of 4 x 3 plots (first line: plots for Jan, Feb, Mar; second line: plots for Apr, May, Jun;. . .fourth line: plots for Oct, Nov, Dec), would be preferable since the seasonal variation would then appear more clearly and Figure S1 would not be necessary. I would like also to mention the fact that Figure S1 is not easy to read since the attribution of the different curves to the different months is not easy and straightforward due to the 3D perspective.

References:

Vigouroux, C., F. Hendrick, T. Stavrakou, B. Dils, I. De Smedt, C. Hermans, A. Merlaud, F. Scolas, C. Senten, G. Vanhaelewyn, S. Fally, M. Carleer, J.-M. Metzger, J.-F. Müller,

M. Van Roozendael, and M. De Mazière, Ground-based FTIR and MAX-DOAS observations of formaldehyde at Réunion Island and comparisons with satellite and model data, Atmospheric Chemistry and Physics, 9, 9523-9544, 2009.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 31287, 2015.