

Interactive comment on “Long-term tropospheric formaldehyde concentrations deduced from ground-based fourier transform solar infrared measurements” by N. B. Jones et al.

Anonymous Referee #1

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General Comments

The paper reports a 12 year timeseries of formaldehyde measurements above Lauder (New Zealand) from ground-based FTIR solar absorption measurements. As the H₂CO signal in the infrared is weak, it is rather challenging to derive these data. A strong seasonal variation is observed with a maximum in summer. The data are compared to GOME data - and a good agreement is found, although it is seen that GOME detects a higher seasonal amplitude. The authors find that a simple photochemical box model that produces formaldehyde from the oxidation of CH₄ reproduces the seasonal cycle but not the high column values ($> 4 \times 10^{15}$ molec/cm²) observed regularly:

they pretend that other sources must be present, and indicate isoprene and transport of biomass burning products as candidate additional sources.

The paper includes interesting datasets but is not very convincing in the discussion of the model simulations: this should be improved. Some technical deficiencies are also present - see specific comments hereinafter: these should be corrected before publication.

Specific Comments

Abstract: - The abstract mentions nowhere that the measurements are performed in the infrared. - The abstract indicates that the time series covers the period 1992 to 2004 whereas Section 2 mentions 1992 to 2005 : this should be made consistent.

Introduction:

- Why is so much attention paid to formaldehyde in polar regions, if the paper deals with observations at a mid-latitude station ? Moreover (Rinsland et al., 2003) is not a relevant reference as it does not deal with H₂CO. The isoprene source that is discussed later in Section 3.2 is not mentioned at all in this introduction. - As an advantage of the FTIR measurement technique, it is mentioned that H₂CO and CO can be measured simultaneously. (1) This is not exactly true as they are measured normally in two different optical bandpasses (unless the measurement setup at Lauder is not the typical NDACC one), and (2) this feature is not exploited further in the paper. So why mention it ? More important question: Could this feature not be exploited to identify the origin of the high H₂CO columns that are observed e.g., in 1999, 2000 and 2002 ? - Line 28: I would prefer 'FTIR method' instead of 'gb-FTS' , because that is more specific. - From the introduction, one gets the impression that the paper includes only 3 sections, not 4. - I believe that the introduction should include some more information about alternative ground-based measurement techniques for H₂CO, as compared to FTIR solar absorption measurements.

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Section 2

- Pg. 14546, Line 9: change the sentence to 'The instrumentation and history of the monitoring program at Lauder are described ...' - Pg. 14547, Line 1 mentions interferences from CH₄ and N₂O whereas Table 1 mentions CH₄ and O₃ => ?? - It should be added which spectroscopic data have been used in the retrieval. A discussion of the uncertainties on the spectroscopic parameters for H₂CO would be welcome, especially to justify the assumptions made in the estimation of the systematic uncertainties in Section 3.1. - Pg. 14548 Line 7: I wonder whether the separation of the partial columns into 0-3 km and 3-12 km is a good choice, taking into account that the DOFs for the 0-3 km column is smaller than 1 (see Table 2) and that the averaging kernel does not peak at the correct altitude (see Fig. b). I believe that this partial column cannot be considered semi-independent. It might be better to consider a thicker lower partial column (e.g., from 0 to 6 km) ?

Section 3.1

- Pg. 14548, lines 17-18: The systematic error on the monthly mean should be identical to the systematic error on each individual data point. This is not what is obtained by the approach explained here, taking the total (random + systematic) errors per individual data point. - Pg. 14549, lines 1-3: justification for assumed uncertainties on line strength and air broadening coefficients ? (see remark about spectroscopic databases above) - Equation 1: t should be $(t - 1992.2)$ and normalized to 1 year to have the correct units as given in Table 3? - Pg. 14549, lines 21-22: 'The error bars in Fig. 2...': I don't understand what the significance of this information is at this point in the discussion ? - Pg. 14550, line 5: can you be more quantitative than 'very high values of H₂CO' ? Moreover , the high values that do not follow the seasonal fit seem to occur always around December: does that agree with the biomass burning season in Australia ? - Pg. 14550, line 14: the reference to Notholt, 2000 is not relevant here! The correct references would be Notholt, 1997a and 1997b. Section 3.2: - Par. 5: Are the given mixing ratios from the model surface mixing ratios or mean tropospheric

mixing ratios ? How should one compare these values to the 0-3 km and 3-12 km mixing ratios in Fig. 3 ? - Last line: it is said in the introduction that H₂CO has an atmospheric lifetime of a few hours: can it then be transported from Australia directly or is it the precursors that are transported ?

Section 3.3 - Pg. 14552, line 26: 'with a mean slant column fitting uncertainty...': I think that the word 'slant' is missing. If so, what is the final uncertainty on the vertical column due to the additional uncertainty on the air mass factor ? - How exactly are summer / winter seasons defined ? - Pg. 14554, lines 5 to 7: why taking a monthly mean for the gb FTIR data and a running 21 day mean for the GOME data? Why 21 days ? - The Gratien et al., 2007 compare IR and UV cross sections for H₂CO ? but the IR X sections are not taken in the same wavelength region as the one used here. Therefore the question arises whether the conclusion from Gratien et al can be adopted as such ?

Section 4

Pg 14555, line 25: 'A simple box model reproduces the seasonal cycle': this has not really been demonstrated in the paper.

Acknowledgements: Add the affiliation (BIRA-IASB) behind I. de Smedt .

Table 1:

- Is the first window of Step 1 again included in Step 2? - why are the SNR ratios in step 1 about a factor 10 to 20 smaller than in step 2 ?

Fig. 5: last line: what is meant with: 'The vertical error bars are mean GOME errors derived from the original smoothed GOME data'. What smoothing is referred to here ?

Technical corrections:

- Abstract line 16: 'compare' instead of 'compares' - Table 3: Erroneous footnotes with phi1 and phi2 - Pg. 14554 line 6: 'have' instead of 'has' - Pg. 14554 line 18 indicates a

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red dotted line in Fig. 2 which in reality in the figure is a black solid line. - Pg. 14555,
line 1: four instead of three outliers are marked with green stars.

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