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Interactive comment on "Mapping the drivers of formaldehyde (HCHO) variability from 2015–2019 over eastern China: insights from FTIR observation and GEOS-Chem model simulation" *by* Youwen Sun et al.

Anonymous Referee #2

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Review of "Mapping the drivers of formaldehyde (HCHO) variability from 2015-2019 over eastern China: insights from FTIR observation and GEOS-Chem model simulation" (acp-2020-544) by Youwen Sun et al.

This study presents and analyses a 5-year time series of HCHO measurements obtained from ground-based FTIR spectra recorded between 2015 and 2019 at Hefei, eastern China. A statistical model is adjusted in order to reproduce the HCHO abundance and variability based on ground level in situ measurements of CO and Ox (O3 + NO2) taken in the vicinity of the FTIR site. CO and Ox are used as tracers for emitted

and photochemical HCHO, respectively, in order to decipher the contribution of direct emissions and oxidation of gas precursors to the HCHO abundance and variability. Estimates of OH radical production from the photolysis of HCHO at the measurement site are also obtained. Finally, GEOS-Chem model simulations are performed to investigate the contribution of different emission categories and geographical regions in China to the HCHO summertime enhancement captured at the measurement site.

This manuscript is well structured and its topic fits the scope of ACP. However, I have several major concerns that prevent me from recommending this work for publication. Those are summarised here below and detailed in my general comments.

- First, this study presents very little novelty compared to the already abundant literature on HCHO. Indeed, just in the past decade there has been a flurry a major advancements as to the observations of atmospheric HCHO, including extensive studies with various instruments on various platforms and modelling studies (sometimes driven by satellite) providing estimates of the HCHO sources worldwide. To my point of view, since the interpretation of the GEOS-Chem results is quite vague, the only novelty would be the new ground-based FTIR observations of HCHO.

- This is the second issue, because the retrievals are poorly characterized and the dataset is not fully exploited. In light of the recent multi-site studies (e.g., Vigouroux et al., 2018), I do not know if presenting an additional FTIR time series, obtained using a retrieval method from another paper, is enough to justify a publication. - The datasets (from both FTIR and GEOS-Chem) are not fully exploited. Indeed, the study lacks overall discussion of the results and presents few perspectives in relation with the literature. Therefore, this gives to the reader a feeling of "unfinished work".

- I have major concerns as to the use of the datasets. In particular, surface and tropospheric VMRs of HCHO are produced from the FTIR retrievals and then used for analysis. However, these retrievals simply do not provide nor contain the necessary information to produce such datasets. As to GEOS-Chem, the description of the model

runs are incomplete and the authors do not discuss some drawbacks, which makes the interpretation of the results difficult and inconclusive.

- Finally, the conclusions and perspectives that are presented in this study for the whole eastern China are actually derived from analysis made at one site only, which tends to really weaken these conclusions. Therefore, I believe that the current manuscript does not befit the standards of ACP and I fear that the work needed to improve it will be too important for a simple revision step.

General comments

- There is little information on the FTIR retrievals of HCHO used in this work and on the characterization of the retrieved product. For instance, a comprehensive error budget is missing (with the systematic and random error terms). Also, considering that the retrieved information for HCHO is quite small (DOFS close to 1), I am wondering to which extent the retrieved profile is affected by the a priori profile in less favourable observational conditions, e.g., around noontime when the probed atmosphere is thinner, or in winter when HCHO is less abundant. For the analyses made with the HCHO time series, it is important to know if the HCHO measurements are biased during certain periods due to a larger influence from the a priori.

- I am very sceptical on the calculation of a mean tropospheric HCHO VMR that has been averaged between the surface and 10 km altitude. It looks like it has been selected arbitrarily. From the DOFS in Fig. 1, we know that the HCHO retrievals provide only one piece of information (DOFS = 1), which is actually the total column. To my understanding, a DOFS of 1 means that basically you have no information at all on how the HCHO VMR profile is distributed vertically and on how it varies. Therefore, this should not allow the extraction of independent information between the surface and an arbitrary selected levels. The only variable that can be used reliably for further data analysis should be the HCHO total column.

- The manuscript lacks sufficient explanation on the time scales and temporal resolution

of the different datasets investigated. For instance, a regression model for source separation is applied to reproduce the observed HCHO based on in situ CO and Ox measurements. However, it is not said if hourly or daily mean in situ measurements were used, and if the model was adjusted to the individual FTIR observations or daily averages. Considering the high intra-day reactivity of HCHO and Ox, any shifts in time (even a few hours) between the datasets might introduce large biases. Again, the manuscript does not say if hourly or daily mean GEOS-Chem outputs are compared to individual or daily mean FTIR measurements, which – by definition – are performed during daytime only.

- Section 2.4: I am wondering to which extent the results of the source separation are influenced by the fact that tropospheric averaged HCHO VMRs are approximated by surface measurements. In situ measurements are significantly affected by local conditions (e.g., vicinity of a major pollution source), whereas tropospheric averaged HCHO VMRs are already more representative of tropospheric chemistry and are also driven by air masses transported from other regions. In the free troposphere, HCHO production from CH4 should play a key role as well, but it can hardly be accounted for by the in situ tracers. There is no discussion nor evaluation of that point.

- Overall, the study reads as an unfinished work because the results are briefly presented and there is not real discussion nor comparison with the literature on observation and modelling of HCHO. It is the case, for example, in Section 3.2, where there is no discussion on what drives the temporal variability of HCHO (e.g., in comparison with other Northern Hemisphere mid-latitude sites), or in Section 3.3, where it is not discussed what could explain the relative contribution of the emitted, photochemical and background HCHO. Similarly, there are many missing information on the GEOS-Chem simulations, e.g.: What are the species whose emissions are shut off? What is considered as being background HCHO? What is contributing to the direct HCHO emissions at Hefei? How about the contribution of CH4, a major and ubiquitous precursor of HCHO?

- Section 3.2: The authors are prompt to conclude that GEOS-Chem can be used with confidence for further analysis, while I find the model evaluation to be quite succinct and inconclusive. For example, the manuscript does not indicate if daily means are used for the comparison with FTIR data or hourly model data collocated in time with the individual FTIR measurements. In p8, lines 9-10, it is stated that the daily and seasonal variability can be reproduced by the model. As to the daily variability, it is impossible to see if in Fig. 3 GEOS-Chem is really able to follow the observed day-to-day variability. As to the seasonality, the model overestimates the HCHO abundance in winter, but it is not discussed explicitly.

- Section 3.2: The diurnal modulation of HCHO has already been investigated several times with remote sensing data, e.g., with spaceborne (De Smedt et al., 2015), ground-based FTIR (Vigouroux et al., 2018; Franco et al., 2016) and MAX-DOAS (Peters et al., 2012) observations. In these studies, the typical diurnal modulation of HCHO at mid-latitudes shows a pronounced peak in the early afternoon, when the photochemistry is enhanced. Moreover, several of these studies showed that global models (including GEOS-Chem) are unable to reproduce the observed modulation. Here, investigating the diurnal variations of HCHO, at a specific site, with a coarse-resolution global model such as GEOS-Chem, has very little meaning. Moreover, GEOS-Chem results are not convincing at all since they do not look to be consistent with the Ox in situ measurements that exhibit a peak in the early afternoon (Fig. 4), and also because the entire diurnal modulation is included in the error bars. Hence, no conclusion can be drawn from this exercise.

- De Smedt et al. (2015) Diurnal, seasonal and long-term variations of global formaldehyde columns inferred from combined OMI and GOME-2 observations, Atmos. Chem. Phys., 15, 12519–12545, doi:10.5194/acp-15-12519-2015.

- Franco et al. (2016) Diurnal cycle and multi-decadal trend of formaldehyde in the remote atmosphere near 46°N, Atmos. Chem. Phys., 16, 4171-4189, doi:10.5194/acp-16-4171-2016.

- Peters et al. (2012) Formaldehyde and nitrogen dioxide over the remote western Pacific Ocean: SCIAMACHY and GOME-2 validation using ship-based MAX-DOAS observations, Atmos. Chem. Phys., 12, 11179–11197, doi:10.5194/acp-12-11179-2012.

- Vigouroux et al. (2018) NDACC harmonized formaldehyde time series from 21 FTIR stations covering a wide range of column abundances, Atmos. Meas. Tech., 11, 5049–5073, doi:10.5194/amt-11-5049-2018.

- Section 3.3: I have to question the reliability of the calculated surface HCHO VMRs. If I understood it well, tropospheric HCHO VMRs derived from FTIR measurements – which were already scaled once to obtain these averaged tropospheric VMRs – are used to produce ground level VMRs of HCHO via a second scaling. First, it is not explained how the scaling was performed. This “double” (or even simple) scaling likely generated large uncertainties on the obtained ground level VMRs. Moreover, deciphering surface VMRs of a highly reactive species such as HCHO from ground-based FTIR retrievals with no vertical information (DOFS = 1) and a clear lack of sensitivity in the lowermost layers (Fig. 1) is, to my point of view, not reliable. Hence it casts doubts on the OH production that is deduced from the ground level HCHO VMRs.

- Section 4: Shutting off entire sectors of emissions in global model simulations is relatively “dangerous” and has some feedbacks on the modelled species that are difficult to interpret. For instance, turning off the anthropogenic emissions induces significantly lower atmospheric concentrations in NMVOCs, which mainly react with OH. This results in higher concentrations in OH available for the oxidation of other precursors of HCHO, such as CH4, which eventually enhances the HCHO production from the other sources. In case the NO emissions are also suppressed when shutting off an emission inventory (which is the case I think), what is the impact of the missing NO emissions on the overall HCHO burden since NO plays a key role in both HCHO formation (via the degradation of peroxy radicals) and loss (NO contributes to the recycling of OH)? Therefore, I am wondering to which extent the results from the GEOS-Chem simula-

tions can be impacted by such feedbacks and how they can be interpreted.

Specific comments

- Abstract: I think that the abstract should reflect that measurements from only one FTIR site (Hefei) was used for investigating the HCHO variability.

- We can certainly not talk about "trend of HCHO" when it is derived from a 5-year time series only, nor present the current data as representative for real HCHO trends. At best, we can talk about "recent rate of change", with the necessary caveats. Linear regressions adjusted to such a short time series can easily be steered up or down due to exceptionally high or low HCHO levels during a specific season.

- It is repeated several times in the manuscript that the study "should help to improve urban air quality and contributes to the formation of new Chinese clean air policies". It sounds a bit like overselling and a shortcut between scientific works and political decision. I would rather formulate it in a more general way, e.g., "Understanding the sources of VOCs is a necessary step for tackling the problems of poor air quality in eastern China and mitigating the emissions of pollutants."

- p3: I find the first two paragraphs to be quite long while they bring very little useful information to the manuscript, and hence they are not necessary. They can easily be summarised as follows: "The relative contribution of emitted and photochemical sources to atmospheric HCHO has been analysed by using the CO-O3, CO-Ox or CO-CHOCHO tracer pair in various polluted environments (references). In those studies, tropospheric HCHO column measurements were sometimes used as representative of near-surface . . .."

- I disagree with the authors' statement that "the OH radical production rate from HCHO photolysis estimated in this study provides an evaluation of regional photochemical capacity over eastern China". It gauges only part of the OH production rate since OH in the atmosphere is also produced via many other pathways. That statement must be

tempered.

- p4, lines 19-20: Does this average include the days with no measurements? How many days in a year do the FTIR observational operations represent?

- p4, lines 27-8: What type of model simulation is this? What do you mean by "WACCM special run"? In Vigouroux et al. (2018), I understand it as being a long-term standard model run from 1980 to 2020, providing a kind of climatology for various trace gases.

- p4, line 34: Do you mean that you fit the ILS during the retrieval process?

- p4, lines 36-37: What is the percentage of measurements you excluded this way?

- p6, lines 14-17: I do not see the point here. Did you split your dataset into subsets? Below, in the same paragraph, it is quoted that "all measurements were grouped by months". Please specify if your analysis has been performed on a monthly basis.

- Section 2.5: What are the species whose emissions are typically suppressed when an emission inventory is turned off? Does it affect the NMVOCs only, or the NO emissions as well? How about species such as CH4 and CO? Usually, in global model simulations, the CH4 fields are prescribed, and hence cannot be turned off the same way as for the emission inventories.

- p7, lines 2-3: Could you be more specific as to what is included in "biofuel emissions"? "Fossil fuel emissions" look to include already most of the anthropogenic emissions.

- p7, lines 6-13: Is the delimitation of the geographical regions arbitrary or based on specific criteria? Do such regions present different characteristics in terms of population density, presence of polluting industries, agriculture, surface coverage (forests, deserts. . .)?

- p7, line 14: From what I knew about GEOS-Chem, the tagged simulations did not implement the full chemistry. The main fields were prescribed and originated from previous standard (full chemistry) runs. The tagged simulations included instead a

much reduced chemistry of the tagged tracer (basically, the tracer sinks). Is it the case with your tagged simulations?

- Section 3.1: Are the model profiles smoothed by the averaging kernels of individual observations or by a mean of AvKs calculated from multiple observations? Fig. 3 (top panel) displays model data even when there are no FTIR data available. Where do the AvKs used to smooth such profiles come from?

- p8, lines 15-22: I am not entirely convinced by the explanation. If the dilution of the model information inside a grid box was the only issue, we would basically observe an almost "flat" seasonal cycle. Here, GEOS-Chem performs quite well in the summer enhancements while there is a problem mainly in the winter troughs.

- Section 3.2: The interannual variability in the FTIR time series is mentioned several times, but it is actually not discussed. Looking at Fig. 3, it even seems that there is no interannual variability.

- p10, line 25: Where is located the CRDS analyser? At the site where the Ox in situ measurements are performed?

- Section 4.1: It sounds a bit awkward to state that anthropogenic emissions contribute to summertime HCHO enhancement, since usually the anthropogenic emissions are relatively constant throughout the year. Could you explain how they contribute to an enhancement of HCHO in summer? Isn't it just the photochemistry that is simply enhanced?

- p12, lines 39-41: The manuscript does not present any evidence for this. There should be at least references to the rates of change of CH4 and various NMVOCs that are reported in the literature over the same time period.

Typos/errors

- p1, line 39: I think that "FTIR spectroscopy" is more appropriate than "spectrometry"

- p2, line 6: from 2015 to 2019

- p4, line 32: De Mazière

- p6, line 24: YRD is not defined yet

- p11, line 12: accounted

- p11, line 33: Modelled HCHO was decreased by . . .

- p11, line 37: As a short-lived species

- Please check all your references. For example, Vigouroux et al. (2018) is missing.

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