Referee 1

We thank Referee 1 for taking the time to review our manuscript and the valuable feedback. We have corrected our manuscript according to the referee's comments and think it is now significantly improved.

Nussbaumer et al use observations from three campaigns across Europe (CYPHEX in Cyprus, HOPE in southern Germany, and HUMPAA in Finland) to calculate production of HCHO and O3. They find that, across all locations, HCHO production can be closely approximated with only production from methane, acetaldehyde, isoprene, and methanol. They also find that the ozone production regime varies by location. This is an interesting paper that is suitable for publication in ACP once the minor issues below are addressed.

We would like to thank the referee for the positive feedback and the recommendation for publication.

Line 37: You could also cite Fried et al, 2011 who found similar results about methane being the dominant contributor to HCHO production in the remote atmosphere.

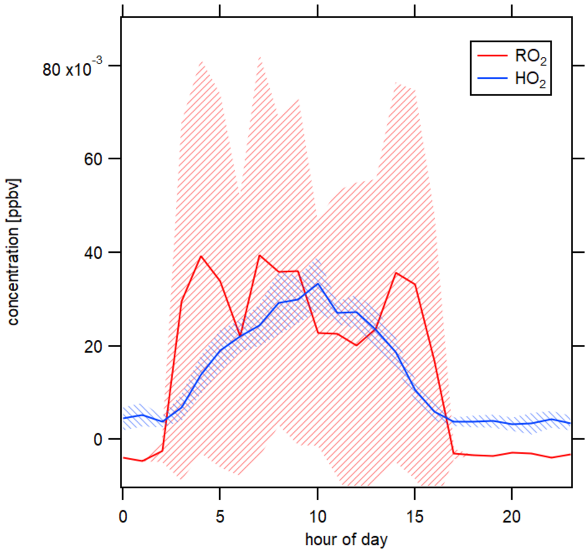
Fried, A., et al. (2011). "Detailed comparisons of airborne formaldehyde measurements with box models during the 2006 INTEX-B and MILAGRO campaigns: potential evidence for significant impacts of unmeasured and multi-generation volatile organic carbon compounds." Atmospheric Chemistry and Physics **11**(22): 11867-11894.

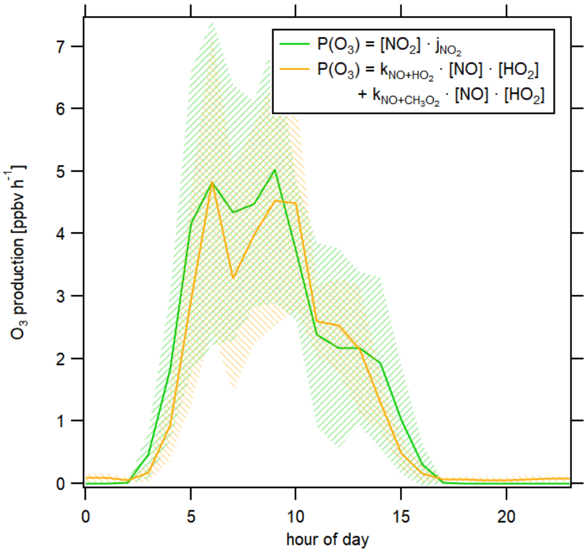
We have added this reference.

Lines 37 ff.: Fried et al. (2011) identified methane to be the main precursor of HCHO in remote regions based on model simulations and measurements during the campaign INTEX-B (Intercontinental Transport Experiment-Phase B) in 2006.

Line 108: I agree that, since the isoprene yield is bounded, this estimate doesn't have a large impact on the overall message of the paper, but for the case of CYPHEX, this range still gives a factor of 2 difference in the potential values of HCHO production from isoprene. I think it's appropriate to either add more discussion to justify your assumption that [HO2] = [RO2] (box modeling studies from similar environments?) or use the data you have to try and estimate the ratio. An alternative method of calculating $P(O_3)$ is from $k_{NO+HO2}[NO][HO_2] + k_{NO+RO2}[NO][RO_2]$. If you equate production from this method with that from the JNO2 method, you could potentially estimate RO2 that way. For HUMPPA, at least, there also appears to be box modeling results by one of the co-authors that could be used to evaluate this assumption.

Thank you for this suggestion. We have calculated [RO2] from equating P(O3) via NO2 photolysis and via the reaction of NO with HO2 and RO2. We used $k_{NO+CH3O2}$ as estimate for k_{NO+RO2}. We have also calculated P(O3) from k_{NO+HO2}[NO][HO2] + k_{NO+RO2}[NO][HO2]. The resulting production term for O3 shows close agreement to the production calculated from the photolytic reaction of NO2 which provides additional justification for our assumption that [HO2] = [RO2].





We have added these Figures to the Supplement and refer to them in the main text. We have also added text referring to the box model study performed by Crowley et al. (2018) showing that [HO2] ≈ [RO2] during HUMPPA.

Lines 111 ff.: This assumption is justified when looking at O3 production (P(O3)) terms. P(O3) can either be calculated via the photolytic reaction of NO2 as presented in Section 2.2 or via the reaction of HO2 or RO2 with NO. We equate the two terms, using the rate constant of the reaction of NO and CH3O2 as estimate for the reaction of NO and RO2, and calculate RO2. We show the diurnal profiles of HO2 and calculated RO2 in Figure S1a of the Supplement. Conversely, we calculate P(O3) for both cases, equating RO2 to HO2 which show close agreement and can be seen in Figure S1b of the Supplement. This is also confirmed by findings from Crowley et al. (2018) (presented in Figure 9) based on model simulations of HO2 and RO2 during HUMPPA.

Figure 1: The light grey labels on the blue background are very difficult to read when you print this out. I would recommend changing the font color.

We agree with the referee and have changed the color to a darker gray.

Section 2.3.1: What time resolution are you using for your P(HCHO) and P(O3) calculations? Do you average everything to the photolysis measurement frequency (10 mins?) or something else?

Thank you for pointing this out. We use a 4-minutes time resolution for calculating the production and loss terms which is the integration time of the OH data. All other species used were interpolated to this time stamp. We have added text for clarification and have corrected this in Table S3 of the Supplement.

Lines 212 f.: For the point-by-point calculations, the data were interpolated to the OH time stamp with a 4-minutes time resolution.

Line 202: There’s an extra ß in Hohenpeißenberg.

Thank you, we have corrected that.

Line 211: You need more discussion about how you arrived at the boundary layer heights. At the very least, summarize what was outlined in Fischer et al.

Fischer et al. presents data from five different measurement sites whereas the BLH was measured at four of those sites including CYPHEX and HUMPPA. Unfortunately, there are no BLH measurements for the HOPE campaign available which were estimated based on the observations from the other sites.

Lines 225 f.: These values were derived from BLH measurements at other locations during summertime including the CYPHEX and the HUMPPA measurement site, and a site in central Germany situated at a comparable altitude.

Line 234: Similar to the previous comment, how do you come up with the 20% uncertainty for the boundary layer height, if you don't have observations?

The stated uncertainty of 20% presents the cross section of measurement uncertainty and atmospheric variability based on the BLH in situ measurements performed at the four sites discussed in Fischer et al. Since the BLH for HOPE was derived from these measurements, the uncertainty was also derived in this way. However, an estimated value probably does have a higher uncertainty compared to the value it was derived from. We have therefore increased the uncertainty to 30% for the HOPE BLH values.

Line 226 f.: We assume that this estimate increases the uncertainty from 20% (BLH measurements) to 30%.

Line 272: Here and elsewhere, when discussing diurnal profiles, it's much more intuitive to use local time instead of UTC, especially since you are referencing multiple sites that have different UTC offsets.

The referee is correct that the sites have different UTC offsets. However, we would like to continue using UTC for a consistent presentation and in order to avoid confusion, for example regarding local summer and winter time. We have defined the mean local time difference in Section 2.3 and in the caption of Figure 3 and have added a reference to it when first discussing diurnal profiles. We also show the solar elevation angle in the diurnal profiles.

Lines 288 f.: Please note the mean local time difference stated in Section 2.3 and Figure 3.

Figure 5: Is this in local or UTC? Please label. Also, for panel a, there is a lot of overplotting. Could you redo the figure where production rates from the individual species are stacked on top of each other to prevent the over plotting?

We present all times in UTC and have added a label for clarification. We have added a Figure to the Supplement to show all production terms in individual panels and refer to it in the main text.

Figure 5: Temporal development of (a) HCHO production terms, (b) HCHO loss terms and (c) net HCHO from July 22 to July 31, 2014 during the research campaign

CYPHEX in Cyprus. The NO2 photolysis frequency j(NO2) is shown in (d) as illustration of the diurnal cycle. All times are in UTC.

Lines 295: The individual production terms are shown in Figures S5 of the Supplement.

Figure 6: I assume this is for all data averaged together? Does this include both daytime and nighttime values? Please indicate.

Yes, the pie chart presents an average of all data, including day- and nighttime values. We have clarified this in the text and the Figure caption.

Lines 307 f.: This is additionally illustrated in Figure 6 which presents the daily average share (including day- and nighttime values) of each production term based on a balance to the overall loss rate.

Figure 6: Chemical production terms of HCHO during CYPHEX including daily averages of all data.

Line 302: Where do you get the 20% value from to vary the yields?

While the share of methyl peroxy radicals forming formaldehyde can theoretically be between 0 and 100%, the availability of NO shifts the value towards the upper end. We therefore decided not to include these threshold values in the sensitivity study, but chose a 20% change as an example to demonstrate the sensitivity of the overall result to the α_{CH3O2}. We clarified this in the text.

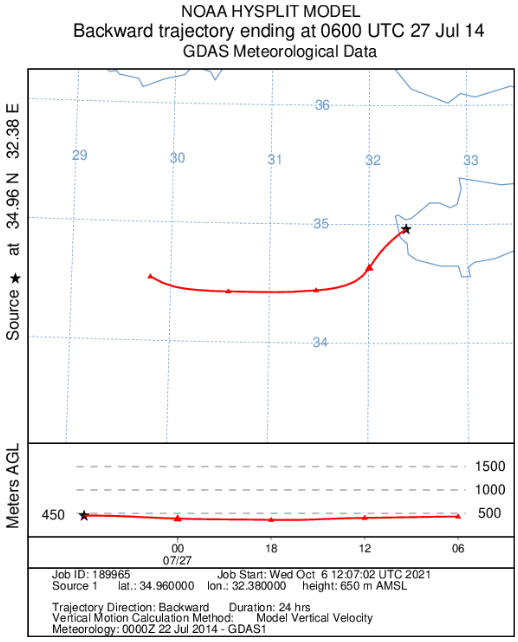
Lines 319 ff.: As an example, a 20% decrease in α_{CH3O2} would give a HCHO production from CH_3O_2 of 38% … .

Line 311: You attribute the nighttime increase in HCHO to local emissions from traffic. Looking at the supplementary figure, it looks like on the night of July 27th, the HCHO concentration nearly doubles. Even with a low boundary layer, that seems too large to be just mobile emissions. Have you looked at ΔHCHO/ΔCO or ΔHCHO/ΔNOx to see if this is in line with what you would expect from traffic emissions? Couldn't this also just be a new air mass moving in or advection from a different region? Do you have meteorological observations you could use to look at this?

The referee is correct that we cannot be certain that this increase is attributed to vehicle emissions and we have changed the text accordingly.

Line 328: The reason for this nighttime HCHO increase is not yet fully understood.

Regarding meteorological aspects, we have run the NOAA Hysplit Model for backward trajectories exemplary for July 27 which shows that air originated from above the Mediterranean Sea which does not reveal any particular HCHO nighttime source. Due to the short atmospheric lifetime of HCHO it is also unlikely that primary HCHO emissions from distant locations are transported to the measurement site.



Line 315: I think you need a little more discussion about the deposition. Instead of there being no deposition, couldn't it just be that the deposition rate is lower than the rate of increase from whatever processes is leading to the increased nighttime HCHO, whether it's advection or direct emission?

We agree with the referee that deposition could be counteracted by a nighttime HCHO source such as advection or secondary formation from terpenes via ozone. The HCHO yields from terpenes are not well known and vary greatly in literature, but are likely very low. The value for the deposition can usually be seen as a lower estimate (previously described by Crowley et al, 2018). We have added some text to the manuscript.

Lines 331 ff.: Please note that the effect of deposition processes could also be counteracted by a nighttime HCHO source, such as terpene oxidation by ozone or advection. The determined value for the deposition can therefore be seen as a lower estimate. As we do not observe net loss of HCHO at night during the CYPHEX campaign, we estimate dry deposition to be negligible.

Line 402: Do you have thoughts as to why the contribution from acetaldehyde photolysis was so much greater at HOPE vs CYPHEX? Is it from larger acetaldehyde concentrations or just a result of higher photolysis related to the higher altitude?

Please note that the contribution from acetaldehyde regarding HCHO production is related to oxidation by OH radicals whereas the contribution from acetaldehyde photolysis is negligibly small. The contribution from acetaldehyde oxidation is greater for HOPE due to a higher acetaldehyde concentration which was 1.21 ± 0.64 ppbv throughout the campaign. In comparison, the average acetaldehyde concentration during CYPHEX was 0.38 ± 0.16 ppbv. This is counteracted by higher temperatures and higher OH radical concentrations during CYPHEX. However, the effect of higher acetaldehyde prevailed.

Line 457: Duncan et al use ratios of tropospheric column $HCHO/NO_2$ to estimate the ozone production regime. While they do show that this is similar for PBL values, here you are just using surface observations. Please give more justification on how the values you use to estimate the ozone production regime could be affected by this difference. Do you have any evidence that the PBL is well-mixed enough to make this assumption? Also, as you cite in your paper, Schroeder et al have pointed out that the values cited in Duncan vary by location. Do your results change at all when taking this into consideration?

We would like to point out that tropospheric columns of HCHO/NO2 from satellite measurements are usually taken into consideration in the absence of ground-based observations. The dominant ozone regime is related to local chemistry taking place and can change with altitude or geographical location. In-situ observations are more reliable when determining the HCHO/NO2 ratio and deducing O3 sensitivity from it, which was also pointed out by Schroeder et al. The considered thresholds were determined via model simulations which assume a well-mixed boundary layer which makes the use of surface observations beneficial. Schroeder et al. present thresholds variations depending on the location particularly in regard to the transition range between a NOx and a VOC limitation, e.g. they find a threshold range from 1.3 to 4.3 in Houston in contrast to the range 1 to 2 presented by Duncan et al. Our results are not affected by higher threshold ranges as the value for the CYPHEX campaign is much higher with an average of 8 and much lower for the HOPE campaign with an average of 0.7. Further, we present two new parameters for determining O3 sensitivity which confirm the findings from in-situ HCHO/NO2 ratios.