Referee #1

Dear Referee,

Thank you very much for you thoughtful and constructive comments for our work. We have addressed them all in the revised version of the manuscript as described below:

In this work, Nijing Wang and colleagues presented ship-borne measurements of a number of atmospheric carbonyl compounds around the Arabian Peninsula. During the AQABA campaign in 2017, a comprehensive suite of scientific payload provided the simultaneous measurements of an array of trace gases, including the carbonyl compounds measured using a PTR-MS system. The impacts of oceanic emissions and the oil and gas industry in this region are discussed. The manuscript is aligned with the scope of ACP, and the topic is of interest to the community.

A clear focus is given to acetaldehyde in this manuscript. It has been recognized that in situ measurement techniques of acetaldehyde may be subject to substantial inlet artifacts, especially in remote environments. Such artifacts may be related to tropospherically relevant ozone levels and have been reported for the PTR-MS system on research aircraft platforms (e.g., Northway et al., 2004). Compared to airborne measurements, in situ instruments onboard research vessels usually require much longer sampling lines due to logistic reasons; and the ship may had frequently encountered organic-rich air masses (e.g., polluted air, ship exhaust, and marine air with sea spray aerosols and possibly unsaturated organic compounds). The potential inlet artifacts for the ship-borne trace gas measurements have not been thoroughly discussed in many previous ship-borne studies. In this work, the authors presented fairly detailed results and discussion regarding the potential inlet artifacts, and concluded that the inlet artifacts, although cannot be fully ruled out, are unlikely to be significant in this dataset. I do appreciate the efforts the authors have invested in the potential inlet artifacts. Based on the results presented in this manuscript, I cautiously agree with the authors that the inlet artifacts are probably not a huge concern in this work. More thorough inlet tests would provide more information, which should be considered in future cruise deployments.

We thanks the reviewer for noting and agreeing with our assessment of the potential inlet artifact. Through this work we have become more aware of this issue and will characterize the inlet accordingly in future measurement campaigns.

In addition, a global chemical transport model (EMAC) is used to examine the sources and sinks of these compounds in this region. The modeling component of this work certainly provides valuable insights. A more thorough model evaluation should also be carried out for some of the key precursors of these carbonyl compounds, such as ethane, propane, butanes, etc.

A thorough model evaluation of the major alkanes measured during AQABA has been published in *Nature Communications* earlier this year (Bourtsoukidis et al., 2020)., The aforementioned study also included multiple sensitivity analyses by the model to investigate alkane sources. We therefore refer to the published work in this manuscript to avoid duplication. . We have further addressed the model comparison results in terms of carbonyl precursors in section 3.4.3.

However, the empirical analysis presented in Section 3.2.1 is largely based on fundamentally flawed assumptions, especially for formaldehyde, acetaldehyde, and acetone. I will elaborate this later. This is one major drawback of this study. For this reason, it is my opinion that major revision is needed

before publication in ACP. I would recommend the authors remove this section entirely. Or, perhaps some sort of box modeling (e.g., diurnal steady state model constrained to observed hydrocarbons and other measurements available) may be useful.

On reflection, we agree with the reviewer that this section did not contribute significantly to the analysis and we decided to remove this section entirely.

Detailed response:

Page 1, line 14: "In this study we examine carbonyl compounds (CxHyO), …" Clearly the formula CxHyO can be an alcohol, an ether, or an epoxide. I do not think a general formula is useful here.

We agree. CxHyO is removed in the text.

Page 2, line 47: "…important source of free radicals (HOx)" should be hydroxyl radicals here. Also x should be in subscript. Unless the authors have other radicals in mind, in which case it should be more specific.

We revised to "hydroxyl and hydroperoxyl radicals free radicals (HO_x)" in the text.

Page 2, line 48: NOx: please define all abbreviations the first time it appears in the main text. Also x should be in subscript.

We revised to "NO_x (NO and NO_2)" in the text.

Page 2, line 56-57: for the sake of completeness, should cite a few more previous studies here (e.g., Yang et al., 2014; Wang et al., 2019), in which tens of ppt of acetaldehyde were reported in the remote troposphere.

More references were added in the text:

Remote area:

Wisthaler, A.: Organic trace gas measurements by PTR-MS during INDOEX 1999, Journal of Geophysical Research, 107, 10.1029/2001jd000576, 2002.

Yang, M., Beale, R., Liss, P., Johnson, M., Blomquist, B., and Nightingale, P.: Air–sea fluxes of oxygenated volatile organic compounds across the Atlantic Ocean, Atmospheric Chemistry and Physics, 14, 7499-7517, 10.5194/acp-14-7499-2014, 2014.

Wang, S., Hornbrook, R. S., Hills, A., Emmons, L. K., Tilmes, S., Lamarque, J. F., Jimenez, J. L., Campuzano - Jost, P., Nault, B. A., Crounse, J. D., Wennberg, P. O., Kim, M., Allen, H., Ryerson, T. B., Thompson, C. R., Peischl, J., Moore, F., Nance, D., Hall, B., Elkins, J., Tanner, D., Huey, L. G., Hall, S. R., Ullmann, K., Orlando, J. J., Tyndall, G. S., Flocke, F. M., Ray, E., Hanisco, T. F., Wolfe, G. M., St. Clair, J., Commane, R., Daube, B., Barletta, B., Blake, D. R., Weinzierl, B., Dollner, M., Conley, A., Vitt, F., Wofsy, S. C., Riemer, D. D., and Apel, E. C.: Atmospheric Acetaldehyde: Importance of Air - Sea

Exchange and a Missing Source in the Remote Troposphere, Geophysical Research Letters, 10.1029/2019gl082034, 2019.

Polluted area:

Koss, A. R., de Gouw, J., Warneke, C., Gilman, J. B., Lerner, B. M., Graus, M., Yuan, B., Edwards, P., Brown, S. S., Wild, R., Roberts, J. M., Bates, T. S., and Quinn, P. K.: Photochemical aging of volatile organic compounds associated with oil and natural gas extraction in the Uintah Basin, UT, during a wintertime ozone formation event, Atmospheric Chemistry and Physics, 15, 5727-5741, 10.5194/acp-15-5727-2015, 2015.

Page 2. Line 59: "… in those regions" this can be confusing. Please revise.

We revised to "Generally, secondary photochemical formation from various precursors is the main source for those carbonyl compounds." in the text to avoid the confusion.

Page 4, line 100: VOC container might be misleading here. I'm guessing this is one of the lab containers loaded aboard, but it could be confused with some sort of container for volatile organic compounds.

We revised "VOC container" to "on-board lab container" in the text.

Page 5, line 144: please provide a bit more info about this spectral radiometer, such as manufacturer, model, etc.

The manufacturer and related reference were added in the text as "The actinic flux was measured by a spectral radiometer (Metcon GmbH; Meusel et al., 2016)."

Page 5, line 145: since NMHCs provide vital constraints on the budget analysis in this work, please provide a bit more information here. For example, is this an online or canister-based GC-FID?

More information about NMHCs measurement was added in the text as "Non methane hydrocarbons (NMHC) mixing ratios were measured by a gas chromatograph with flame ionization detector (GC−FID) online with the time resolution of 50 minutes. It measured hydrocarbons (C2 - C8) and aromatics (C6 - C8) with the average LOD < 10 ppt for most of compounds. For a detailed instrumental description see Bourtsoukidis et al. (2019)."

Page 5, line 156: I understand that EMAC is a well established model and many key components are archived elsewhere. Given the importance of oceanic emissions in this work, I am sure the readers would appreciate a bit more details, especially how the oceanic emissions are setup/configured for the compounds discussed in this work, such as acetaldehyde, acetone, etc.

More information on EMAC and in particular the air-sea transfer was added in the text as follows "… a comprehensive chemistry mechanism MOM (Mainz Organic Mechanism) was deployed (Sander et al., 2019). The model considers direct emissions (such as anthropogenic, biogenic, biomass burning etc.), atmospheric transport and mixing, photochemical production of carbonyls (by OH, O_3 and NO_3), as well as physical and chemical removal processes. The exchange of organic compounds between ocean and atmosphere was considered in EMAC via the AIRSEA submodel, described in detail in Pozzer et al. (2006). The transfer velocity is calculated online and the concentration in the water is prescribed by the user. For acetone, a constant water concentration of 15 nmol/L is used, following the suggestion of Fischer et al. (2012), while direct oceanic emission of acetaldehyde was initially not included in the model. The model configuration in the study is the same as the model applied in Bourtsoukidis et al. (2020) in the resolution of T106L31 (i.e. ~ $1.1^o \times 1.1^o$ horizontal resolution and , 31 vertical hybrid pressure levels up to 10 hPa) and the time resolution of 10 minutes. The measurement data of PTR-ToF-MS were averaged to 10-minute resolution to match the model data resolution for further comparison."

Page 8, line 210-223: are these carbonyl compounds (i) directly emitted from the oil & gas facilities; (ii) produced from precursors emitted from these facilities; or (iii) associated with other anthropogenic activities in this area?

We discussed these sources (first or secondary formation of carbonyls from anthropogenic activities) in section 3.1.2 (case studies of polluted regions) and also section 3.4.3 (anthropogenic primary sources for underestimated acetaldehyde). In order to make this clear, we now added a sentence in the end of this paragraph as follow: The sources of the major carbonyls in the Arabian Gulf will be discussed in details in section 3.1.2 and 3.4.3.

Page 10, Table 2: I am not entirely sure about the purpose of this table. I understand that the overarching goal here is to put the measurements obtained in this study into the context of previous studies. However, this table itself certainly does not cover the comprehensive characteristics of any of the category. For example, some of the studies cited in the marine category contains clear influence from anthropogenic emissions/ biomass burning; none of the cited studies in this category reflects the pristine marine environments, such as tropics, Southern Ocean, etc. For this reason, this table may create wrong impression to the audience. I understand that this is not a review paper, but a more thorough review of the literature is always appreciated. Therefore, I would suggest that the authors compile a slightly more exhaustive list, and be cautious when quoting/discussing the numbers in the literature. I myself find some of the categories are less relevant for this work (e.g., urban, rural, forest), and the marine category can certainly use more details (e.g., coastal vs open ocean, northern vs southern hemisphere, high latitude oceans, natural vs influenced by anthropogenic/biomass burning emissions). I would also recommend the authors add relevant info (e.g., lat/lon) to this table.

We now updated the table by including 5 more studies done in open sea and coastal areas. We replaced the study of oil and gas influenced regions by other four studies. Studies done in urban, rural and forest regions were removed from the table. We included the coordinates (latitude and longitude) and measurement sampling height for those studies as requested. Accordingly, we updated the text related to the comparison of previous studies in the revised manuscript. The revised table is attached in the end of this response.

Page 9, line 236-237: why do the authors compare the ship-borne measurements obtained in this work to the measurements from a rural site in Cyprus? The numbers may be similar for vastly different reasons. Or do the authors imply a connection with Cyprus? If yes, please clarify & elaborate.

As suggested by the reviewer, the table has been updated and the measurements results from this Cyprus study as well as the related text were removed and rewritten in the revised manuscript as: "… The Mediterranean Sea had somewhat higher levels of aliphatic carbonyls than the clean regions (the Arabian Sea and the Gulf of Aden) but with acetone (above 2ppb) dominating the distribution. Much higher acetone level than acetaldehyde level was also observed for some costal site measurement which was impacted by continental air (White et al., 2008; Schlundt et al., 2017, see Table 2). …"

Page 9, line 247-248: note that acetone may deposit into the ocean, especially in the continental outflow from polluted regions (e.g., Marandino et al., 2005; Schlundt et al., 2017; Yang et al., 2014).

We are aware that under conditions of polluted continental outflow over the ocean uptake of acetone from the air to the sea will occur. Also dilution and mixing with free tropospheric air during transport can modulate acetone mixing ratios. This process is accounted for in the EMAC model used in the subsequent analysis. We now added a sentence in the revised manuscript to make it clear as follow: "… could be also transported from the Mediterranean Sea (where acetone was high). Four-day back trajectories indicate the air reaching Suez region was mostly originated from Europe continent passing over the Mediterranean Sea (Bourtsoukidis et al., 2019). Meanwhile, ocean uptake of acetone from the air due to polluted continental outflow (Marandino et al., 2005) as well as dilution and mixing with free tropospheric air during transport can modulate acetone mixing ratios."

Page 9, line 253: Again, the authors compare the results in Red Sea to that from the other side of the planet (Thompson Farm, a rural site in the US). What is the point of the seemingly random comparison?

With these comparisons we were trying to set the AQABA measurements in a broader context. Since this appears to have been a distraction, we have followed the reviewer's suggestion, and the table has been updated, so that the measurements from the Thompson Farm study as well as the related text were removed in the revised manuscript as follows:"…The mixing ratios of acetaldehyde and acetone over the northern part of the Red Sea were similar to those levels measured in western Pacific coastal regions (South China Sea, Table 2)…."

Page 11, line 269: are these numbers the sum of all measured unsaturated and aromatic carbonyls?

The numbers represent the range of the mean mixing ratios of unsaturated carbonyls in each region. To make this clear we added the sentence "…followed by Suez (11 – 68 ppt). The numbers represent the range of the mean mixing ratios of unsaturated carbonyls in each region"

Page 11, line 293-295: these very general descriptions belong to the introduction section.

These lines were removed in the revised manuscript.

Page 11, line 298-299: The first two assumptions do not hold, for the following reasons: (1) Photolysis can be a significant sink for certain carbonyl compounds, such as acetone (Fischer et al., 2012); for formaldehyde it's even the dominant sink (Anderson et al., 2017). (2) Primary emissions contribute substantially to many carbonyl compounds in the atmosphere, such as acetone (e.g., Fischer et al., 2012), acetaldehyde (e.g., Millet et al., 2010). Biomass burning contribution to many of these carbonyl compounds is also substantial, and the authors even discussed the impacts of biomass burning in Section 3.2.2. Therefore, primary emissions certainly cannot and should not be ignored. (3) Dilution (mixing with background air) can lead to rapid decrease of tracer concentrations in freshly emitted plumes. Uptake by the terrestrial vegetation and the ocean may also affect the carbonyl budget on the regional and global scale. The impact of mixing may be cancelled out by scaling the mixing ratios of hydrocarbons and carbonyls to a long-lived compound, such as CO or ethylene, but I cannot think of a simple way to overcome other impacts as I listed above. In addition to the flawed assumptions, the carbonyl yields are not constant and depend on other variables (e.g., NOx levels, oxidation pathways). Table S4 did not clarify how the yields were derived.

Thanks for pointing out the inappropriate application of the empirical method we applied in our study. The intention was to show what the photochemistry alone (without these influences) would generate. We agree that because of the complexity of multiple alternate sources this section does not have much scientific contribution to the whole manuscript. As we compared the measurement with a comprehensive global model in the manuscript (which includes such sources), we removed the part regarding the empirical calculations and corrected relevant text in the revised manuscript. For the yield in Table S4, we assumed the alkylperoxy radicals (RO_2) would mainly react with NO (high NO condition) and then summed up the yield of all the reaction pathways of hydrocarbons that could produce carbonyls. Now as we decided to remove the empirical calculation section, the Table S4 was also removed from the supporting information.

Page 12, line 314-319: This exercise is only valid when the ratio in the source region is well understood, which is clearly not the case here. Figure S3 cannot rule out the possibility that there may be air masses with different initial toluene/benzene ratios mixed together. It certainly looks like there are multiple clusters in Figure S3. This comes back to my previous question regarding the oil and gas industry in this region: do these measurements reflect (i) direct emissions from the oil & gas facilities; or (ii) co-located (anthropogenic) emissions in this area? This type of photochemical clock does not work without a thorough understanding of the source characteristics. To sum up, the OH exposure calculation may be useful, provided that the source contributions are well understood. The empirical analysis based on Equation 1 and 2, however, does not really provide scientifically valuable information, definitely not for acetone, acetaldehyde, and formaldehyde.

We agree that it is not appropriate to use the hydrocarbon ratios to calculate the OH exposure for the remote areas. However, we kept the estimations of OH exposure using hydrocarbon ratios for the polluted regions (Arabian Gulf and Suez) where the major primary emissions were identified based on NMHCs data reported by Bourtsoukidis et al., (2019) and Bourtsoukidis et al. (2020). We now mention this specifically in the section 3.1.2 in the revised manuscript and also compared the toluene to benzene emission ratios to previous studies having similar sources. The revised text is as follows:

…For further insight, we focused on a time series of selected trace-gases and their inter-correlations to better identify the sources of the major aliphatic carbonyls. Meanwhile, we calculated the OH exposure ($[OH]\Delta t$) based on hydrocarbon ratios (Roberts et al., 1984;de Gouw et al., 2005;Yuan et al., 2012) for the polluted regions Arabian Gulf and Suez where primary emissions have been identified (Bourtsoukidis et al., 2019; Bourtsoukidis et al. 2020), to better understand the photochemical aging of the major carbonyls using the following equation:

$$[OH]\Delta t = \frac{1}{k_X - k_Y} \cdot \left(In \frac{[X]}{[Y]}\Big|_{t=0} - In \frac{[X]}{[Y]}\right), \qquad \text{Eq. (1)}$$

where X and Y refer to two hydrocarbon compounds with different rates of reaction with the OH radical (k). For this study, we chose toluene ($k_{OH+toluene}$: 5.63E-12 cm^3 $molecule^{-1}s^{-1}$) and benzene ($k_{OH+benzene}$: 1.22E-12 cm^3 $molecule^{-1}s^{-1}$) (Atkinson and Arey, 2003), because both compounds were measured by PTR-ToF-MS at high frequency and these values showed a good agreement with values measured by GC-FID (Figure S2). The approach detailed by Yuan et al. (2012) was applied to determine the initial emission ratio $\frac{[X]}{[Y]}\Big|_{t=0}$ in those two regions by only including nighttime data of benzene and toluene. We obtained initial emission ratios (toluene to benzene ratio) of 1.38 for the Arabian Gulf and 2.12 for the Suez region. Koss et al. (2017) summarized the toluene to benzene ratios observed in various locations and showed that urban and vehicle sources tend to have higher toluene to benzene ratio (mean ~ 2.5) than the ratios of oil & gas sources (mean ~ 1.2). Therefore, the toluene to benzene ratios obtained for those two regions agreed well with other studies done with similar emissions sources. The corresponding correlation plots of toluene and benzene for those two regions can be found in Figure S3.

And we removed this description to the case studies because we included the OH exposure to help discuss the air mass age.

Page 14, Figure 4: Units for j_NO2 and OH exposure are missing. Same for Figure 5.

The units were added in the figures of the revised manuscript.

Page 14, line 366: "As mentioned before, photochemical oxidation contributed a large fraction to acetone and the larger aliphatic carbonyls over the Arabian Gulf and Suez areas" I think this statement needs stronger support. The terrestrial biosphere may emit a large amount of acetone. Moreover, the acetone yield from terpene oxidation is quite high, and the acetone production from terpenes may not be a huge source globally, but could well be in certain regions. Similarly, some of these compounds may be from anthropogenic sources or biomass burning.

As the empirical calculation was removed, this conclusion previously derived from the results were removed as well. However, we would like to address the comment given by the reviewer as follows. The high mixing ratios of acetone precursors (propane and several higher alkanes) combined with strong photochemical processing, provide a strong regional secondary source. Regional biogenic sources of acetone are likely to be small given that much of the Arabian Peninsula is desert and little isoprene (a common biogenic emission) and monoterpenes was detected (most of the time below detection limit and the average was around tens of ppt). In the Arabian Gulf no significant increase of acetonitrile was observed so that a biomass burning contribution to acetone can be ruled out. In the region of Suez, acetonitrile levels were somewhat elevated. The lowest of acetone to acetonitrile ratio was only during the biomass burning plum spikes in Suez, ranging from 7-10, which was similar to the ratios of acetone to acetonitrile reported by Holzinger et al. (2005) in aged biomass burning plume over the Eastern Mediterranean. During the rest of the time, the acetone to acetonitrile ratio was above 10 and up to 30. It suggested that the biomass burning source is still likely minor compared to the secondary source.

Holzinger, R., Williams, J., Salisbury, G., Klüpfel, T., de Reus, M., Traub, M., Crutzen, P. J., and Lelieveld, J.: Oxygenated compounds in aged biomass burning plumes over the Eastern Mediterranean: evidence for strong secondary production of methanol and acetone, Atmos. Chem. Phys., 5, 39-46, 10.5194/acp-5-39-2005, 2005.

Page 14, line 374-376: The authors concluded that the carbonyls are co-produced via photochemical oxidation because they show strong correlation with ozone. I am not totally convinced. It is plausible that these carbonyls may be co-emitted with ozone precursors.

We have added support for this statement by referencing the work of Tadic et al., (2020), who studied the production of O3 in the region in detail. In order to retain the reviewer's point we also note that primary emissions may also occur, as follows "Tadic et al. (2020) reported the net ozone production rate over the Arabian Gulf (32 ppb d^{-1}) was the greatest over the Arabian Peninsula. They show that strong ozone forming photochemistry occurred in this region, which would lead to abundant secondary photo-chemically produced products (including carbonyls). However, it should be noted the good correlation between ozone and carbonyls could in part be due to carbonyls co-emitted with ozone precursors (hydrocarbons) as primary emissions."

Tadic, I., Crowley, J. N., Dienhart, D., Eger, P., Harder, H., Hottmann, B., Martinez, M., Parchatka, U., Paris, J.-D., Pozzer, A., Rohloff, R., Schuladen, J., Shenolikar, J., Tauer, S., Lelieveld, J., and Fischer, H.: Net ozone production and its relationship to nitrogen oxides and volatile organic compounds in the marine boundary layer around the Arabian Peninsula, Atmospheric Chemistry and Physics, 20, 6769-6787, 10.5194/acp-20-6769-2020, 2020.

Page 14, line 385: I noticed that the spatial distribution oil fields and refineries is from the year of 2007, while this cruise campaign was conducted in 2017. Is it possible to obtain more recent information on this?

Unfortunately, we do not have access to any newer dataset of the oil and refineries distributions. Previously published work regarding the AQABA (Bourtsoukidis et al., 2019; Pfannerstill et al., 2019) all reported the distribution from 2007 data base.

Page 15, line 405: "The biomass burning emissions were probably transported by on the prevailing northerly wind above Northeast Egypt…" is this supported by backward trajectory analysis or meteorological conditions?

The prevailing wind direction in Suez was shown in Figure S1. We added "…by on the prevailing northerly wind (Figure S1) above Northeast Egypt…" in the revised manuscript.

Page 16, Figure 6: this is interesting. how are these not-assigned ions calibrated/quantified?

Where no calibration gas is available but a mass is detected then the mixing ratios of those ions were calculated based on an established theoretic calculation method (Lindinger et al. 1998) using a fixed proton transfer reaction rate constant (k_{PTR}) of 2.0 × 10^{-9} cm^3 s^{-1}. Fortunately the protonation rate constant for all compounds is very similar, so that this approach yields reasonable results when direct in-field calibration is not possible.

Lindinger, W., Hansel, A., and Jordan, A.: On-line monitoring of volatile organic compounds at pptv levels by means of proton-transfer-reaction mass spectrometry (PTR-MS) medical applications, food control and environmental research, International Journal of Mass Spectrometry and Ion Processes, 173, 191-241, 1998.

Page 17, line 449-452: any GC-FID measurements of isoprene during this period?

Yes. The GC-FID also measured isoprene during AQABA campaign. However, as mentioned already in the text (line 417-418): According to the GC-FID measurement, isoprene was below the detection limit for most of the time during the AQABA cruise with the highest values observed in Suez (10 - 350 ppt).

Page 17, line 460-462: "The model considers direct emissions (such as anthropogenic, biogenic, biomass burning etc.), atmospheric transport and mixing, photochemical production of carbonyls (by OH, O3 and NO3), and physical and chemical removal processes." This belongs to the method section where the model is introduced.

We now moved this part to the method section 2.5.

Page 17, line 465-466: "acetone was overestimated by a factor within 1.5 over the Arabian Sea, Gulf of Aden and Gulf of Oman, and underestimated by a factor within 2.5 over the other regions." Note that previous studies reported downward oceanic fluxes of acetone (ocean being a net sink) in the continental outflow from polluted regions (e.g., Marandino et al., 2005; Schlundt et al., 2017; Yang et al., 2014). How does the model treat the air-sea exchange of acetone? This key information is missing.

The sub-model AIRSEA (Pozzer et al. 2006) implemented in EMAC calculates the exchange of acetone between the ocean and the atmosphere. The transfer velocity is calculated online. In general, ocean is a net sink for acetone, but regionally could be an emitter of acetone. This strongly depends on its concentration, however, away from sources the surface seawater is close to equilibrium with boundary layer air (Williams et al. 2004). For acetone in the model, a constant water concentration of 15 nmol/L is used, following the suggestion of Fischer et al. (2012). We added this information in the method part (section 2.5).

Fischer, E. V., Jacob, D. J., Millet, D. B., Yantosca, R. M., and Mao, J.: The role of the ocean in the global atmospheric budget of acetone, Geophysical Research Letters, 39, n/a-n/a, 10.1029/2011gl050086, 2012.

Pozzer, A., Jöckel, P., Sander, R., Williams, J., Ganzeveld, L., and Lelieveld, J.: Technical Note: The MESSy-submodel AIRSEA calculating the air-sea exchange of chemical species, Atmos. Chem. Phys., 6, 5435-5444, 10.5194/acp-6-5435-2006, 2006.

Williams, J., Holzinger, R., Gros, V., Xu, X., Atlas, E., and Wallace, D. W. R.: Measurements of organic species in air and seawater from the tropical Atlantic, Geophysical Research Letters, 31, 10.1029/2004gl020012, 2004.

Page 19, line 504-505: "This indicates that the source of acetaldehyde was probably not from direct biogenic production…" this sentence is confusing. I think "direct oceanic acetaldehyde emissions are probably insufficient to explain…" might be more accurate?

Here we wanted to emphasize that acetaldehyde is more related to a non-biogenic emission mechanism based on its poor correlation with DMS. We now correct the sentence to make it clearer: "This indicates that the direct biogenic acetaldehyde emissions from the ocean are probably insufficient to explain the measured acetaldehyde."

Page 19, line 513-514: "To our knowledge, there is no clear experimental evidence showing the ocean to be a sink for acetaldehyde." This is probably true, but the language is vague (I think "direct" is a better choice than "clear" here), and several important studies are missing here. Schlundt et al. (2017) reported net downward fluxes of acetaldehyde in the polluted marine boundary layer (ocean is a net sink), which is inferred from measurements in the atmosphere and the surface ocean (so not "direct" evidence). Yang et al. (2014) reported oceanic fluxes of acetaldehyde using eddy covariance method (this is direct flux measurements). Indeed the fluxes were mostly upward for acetaldehyde (ocean is a net source), there appears to be a few points indicative of net downward fluxes, although are probably close to the limit detection of that system.

Thanks for pointing out that several important studies were omitted when considering this point. We have added now them in the revised manuscript as follows: "… reporting the upper limit of the net ocean emission of acetaldehyde to be 34 Tg a^{-1}. Yang et al. (2014) quantified the air-sea fluxes of several OVOCs over Atlantic Ocean by eddy covariance measurements, showing ocean is a net source for acetaldehyde. Although Schlundt et al. (2017) reported uptake of acetaldehyde by the ocean from measurement-inferred fluxes in western Pacific coastal regions, to our knowledge, there is no direct experimental evidence showing the ocean to be a sink for acetaldehyde. "

Page 21, line 549-554: can the authors please provide new plots like Figure 7 but for other major acetaldehyde/acetone precursors, such as ethane, propane, butanes, and perhaps ethanol as well? This would certainly make the case stronger, and may provide key insight into the acetaldehyde budget in this region. Even "remote" regions in this work are not really that far from the source regions. Therefore it remains unclear to what degree the underestimation/overestimation of carbonyls is due to their precursors. Emission inventories often have a hard time capturing the anthropogenic emissions, especially from the oil and gas industry.

Exactly these precursor plots are already provided in the paper of Bourtsoukidis et al. (2019). Furthermore, Bourtsoukidis et al. (2020) already published a measurement-model comparison for the key precursors (ethane, propane and butane). After implementing the new deep water source, the model was able to mostly reproduce the measurements of hydrocarbons over most areas except for the large underestimation over Suez and the Arabian Gulf, which was mentioned in the text. Therefore, we think it likely that other precursors which were not included in the model may contribute to the underestimation of acetaldehyde. Ethanol was not measured during the campaign. We do now include model comparison of alkenes (ethene and propene) to further support our argument of nighttime ozonolysis as a potential source in the Arabian Gulf as suggested by the other referee. In order to more clearly demonstrate the anthropogenic contribution to the model bias of acetaldehyde, we revised manuscript as follows:

Over the Arabian Gulf and Suez, the intensive photochemical production of carbonyls is apparent. Bourtsoukidis et al. (2020) compared measured hydrocarbons (ethane, propane, and butane) with the results from model simulations (the same model used in this study with the newly discovered deep water source implemented). The model was able to reproduce the measurement over most regions except for some significant model underestimations in Suez and Arabian Gulf, in which local and small-scale emissions were difficult for the model to capture. Therefore, an underestimation of the precursor hydrocarbons, as well as those large alkanes, alkenes and cyclic hydrocarbons which were not measured (> C8) or included in the model (> C5), could be a reason for the model underestimation of acetaldehyde especially in polluted regions. In addition, as mentioned in the previous case studies, high ozone mixing ratios were observed over the Arabian Gulf especially during the nighttime. Ethene and propene were found to be significantly underestimated during the nighttime high ozone period by a factor over 10 (Figure S9), which indicates that the nighttime ozonolysis of alkenes could be another important source for acetaldehyde, formaldehyde and other carbonyls (Atkinson et al., 1995;Altshuller, 1993) in the Arabian Gulf.

Page 21, line 570: what biomass burning emission inventory is used in this work?

For the biomass burning emissions, we used the global fire assimilation system emissions data from Kaiser et al. (2012). A sentence has been added in the method section 2.4 to address: The global fire assimilation system was used for biomass burning emissions (Kaiser et.al., 2012).”

Kaiser, J. W., Heil, A., Andreae, M. O., Benedetti, A., Chubarova, N., Jones, L., Morcrette, J.-J., Razinger, M., Schultz, M. G., Suttie, M., and van der Werf, G. R. (2012). Biomass burning emissions estimated with a global fire assimilation system based on observed fire radiative power. BG, 9:527-554

Table 2. Mixing ratios (ppb) of OVOCs reported in previous observation in literature

Locations	Lon./Lat.	Height	Time	Technique	Formaldehyde	Acetaldehyde	Acetone	MEK	Literature
Open sea		m							
Tropical Atlantic Ocean	10◦ N-0◦ N 35◦ W-5◦ E	18	Oct.-Nov.	PTR-MS	n.r.	n.r.	0.53	n.r.	(Williams et al., 2004)
Atlantic Ocean	50◦ N-50◦ S 10-60◦ W	18	Oct.-Nov.	PTR-MS	n.r.	0.18 (Northern H) 0.08 (Southern H)	0.6 (North) 0.2 (South)	n.r.	(Yang et al., 2014)
Western North Pacific Ocean	15-20◦ N 137◦ E	6.5 -14	May	PTR-MS	n.r.	n.r.	0.20-0.70	n.r.	(Tanimoto et al., 2014)
Western Indian Ocean	12◦ N-5◦ S 43-55◦ E	15	Feb.-Mar.	PTR-MS	n.r.	n.r.	0.49	n.r.	(Warneke and de Gouw, 2001)
Indian Ocean	19◦ N-13◦ S 67-75◦ E	10	Mar.	PTR-MS	n.r.	0.32-0.42 (continental outflow) 0.18-0.21 (equatorial marine)	1.11-2.08 (continental outflow) 0.51-0.62 (equatorial marine)	n.r.	(Wisthaler, 2002)
Southern Indian Ocean	30◦ S-49◦ S 30-100◦ E	15	Dec.	PTR-MS	n.r.	0.12-0.52	0.42-1.08	n.r.	(Colomb et al., 2009)
Costal									
Caribbean Sea	10-30◦ N 60-80◦ W	10	Oct.	HPLC	0.61	0.57	0.40	0.03	(Zhou and Mopper, 1993)
Cape Verde Atmospheric Observatory	16.86◦ N 24.87◦ W	10	2006-2011	GC-FID	n.r.	0.43 (0.19-0.67)	0.55 (0.23-0.91)	n.r	(Read et al., 2012)
Appledore Island, USA	42.97◦ N 70.62◦ W	5	Jul.-Aug.	PTR-MS	n.r.	0.40	1.5	0.20	(White et al., 2008)
Mace Head, Ireland	53.3◦ N 9.9◦ W	25	Jul.-Sep.	GC-FID	n.r.	0.44 (0.12-2.12)	0.50 (0.16-1.67)	n.r.	(Lewis et al., 2005)
Canadian Archipelago	68-75◦ N 60-100◦ W	Ship cruise	Aug.-Sep.	PTR-MS	n.r.	n.r	0.34	n.r	(Sjostedt et al., 2012)
Barrow Arctic	71.30◦ N 156.77◦ W	6	Mar.-Apr.	TOGA		0.10 ± 0.20	0.90 ± 0.30	0.19 ± 0.05	(Hornbrook et al., 2016)
South China Sea, Sulu Sea	2◦ N-15◦ N 108-124◦ E	10	Nov.	GC-MS	n.r.	0.86	2.1	0.06	(Schlundt et al., 2017)

Oil & Gas								
Horse Pool site, Uintah Basin, USA	Ground site	2012-2013	PTR-MS	3.71	4.27	7.97	2.81	(Koss et al., 2015)
Central United State	<600	Mar.-April	ToF-CIMS	1.13[a]	0.5	1.5	0.2	(Koss et al., 2017)
Eagle Mountain Lake site, Texas, USA	Ground site	June	PTR-MS	n.r.	n.r.	3.2 (1.2-6.7)	0.3 (0.09-0.85)	(Rutter et al., 2015)
Hickory, Pennsylvania, USA	Ground site	June	PTR-MS	n.r.	1.29 (0.28-2.03)	3.22 (1.45-4.99)	0.73 (0.4-0.97)	(Swarthout et al., 2015)

n.r.: not reported in the literature.

a: formaldehyde was measured by laser-induced fluorescence (LIF)

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