Referee #2

Dear Referee,

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

Overview:

The manuscript presented ship-borne measurements of carbonyl compounds (carbonyls) around the Arabian Peninsula during the AQABA campaign (end of June to end of August 2017) and interpreted the measurement with a chemical transport model (EMAC). Analyses were focused on 3 classified carbonyl groups—aliphatic ($C_nH_{2n}O$), aromatic ($C_nH_{2n-8}O$), and unsaturated ($C_nH_{2n-2}O$)—in terms of their regional distribution, source characterization and partitioning, model-measurement comparison in 8 geographical regions, and finally an in-depth discussion on the missing sources of acetaldehyde (C2 aliphatic carbonyl).

The manuscript is well organized with clear methods, thorough analyses, and coherent arguments. The topic is of great importance given that carbonyls can provide key clues on reactive carbon source and chemistry as an oxygenated and stable group of VOCs, and acetaldehyde has drawn increased attention recently with growing evidence of its large missing sources. This manuscript therefore provided valuable information in advancing the knowledge of atmospheric reactive carbon in the Middle East. I support the publication of the manuscript once the following issues being addressed.

Thank you for noting the key points and significance of the study.

1) **The title is interesting but over-weights the topic towards acetaldehyde and meanwhile lacks a reflection of the large portion of work done on the overview analysis of different carbonyls in this region as presented in the manuscript.**
   On reflection, we agree. The title is now changed to "Measurements of carbonyl compounds around the Arabian Peninsula: overview and model comparison".

2) **L22. "A smaller but still significant discrepancy …", are you trying to say that the model-measurement discrepancy of other carbonyls is smaller comparing to that of acetaldehyde? This sentence is not clear and needs to be reorganized.**
   We revised this sentence in the revised manuscript as "We compared the measurements of acetaldehyde, acetone and methyl ethyl ketone to global chemistry-transport model (EMAC) results. A significant discrepancy was found for acetaldehyde, with the model underestimating the measured acetaldehyde mixing ratio by up to an order of magnitude."

3) **L130. How did you deal with below LOD data?**
   The following statement was added to make this clear: " … and 9 ± 6 ppt for methyl ethyl ketone (MEK) (Table S1). The data below LOD were excluded from the data set instead of giving zero."

4) **L134-140. There are other measurement techniques that can differentiate ketone and aldehyde at a shared mass, better to give an average or a range of in-situ reported ketone:aldehyde ratio (e.g., acetone:propanal ratio) to prove the dominance of ketone.**

   We added this information now in the revised manuscript as "… ketones tend to have longer atmospheric lifetimes and higher photochemical yields than aldehydes as mentioned in the introduction. The ratio of measured propanal to acetone was 0.07 in the western Pacific costal region (Schlundt et al., 2017), 0.06 in an urban Los Angeles (Borbon et al., 2013) and 0.17 - 0.22 in oil &gas production regions (summarized by Koss et al., 2017)…."

5) **Figure2. The description of air mass characteristics is not clear. For example, what's the difference between "source nearby" vs "source nearby, aged", "remote, aged" vs "remote, clean"?**

   We agree that the original description may confuse readers. Now in the revised manuscript we have simplified the labeling of the air mass characteristics by dividing them into source nearby (Suez, Arabian Gulf, Gulf of Oman and Red Sea North) and remote (Mediterranean Sea, Red Sea South, Gulf of Aden and Arabian Sea), which is consistent with what Bourtsoukidis et al. (2019) reported based on the NMHCs variability-lifetime results (b factor).

6) **L246-247. "Interestingly, the mean acetaldehyde mixing ratio (0.62 ± 0.59 ppb) over Suez was twice the level found over the Mediterranean Sea, whilst the acetone level was only slightly higher", why is this interesting? What are you trying to imply?**

   The main message we would like to give is that the air in Suez region was influenced by local formation in addition to the transportation from Mediterranean Sea, since the mixing ratios do not simply scale, we agree that the original text was not clear. We have now rewritten the text as follows: "Another region where abundant aliphatic carbonyls were observed was Suez region. The air in this region was mainly influenced by nearby cities and marine transportation (ship emissions within the Suez Chanel) (Bourtsoukidis et al., 2019;Pfannerstill et al., 2019). Therefore abundant precursors were available in Suez region, producing more carbonyls regionally especially for shorter-lived compounds (formaldehyde and acetaldehyde). Besides the local-scale emissions and photochemical production contribution to the carbonyls over Suez, the longer lived carbonyls (e.g. acetone) 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)….".

7) **L247-248. "Besides the local-scale emission and photochemical production contribution to the acetone over Suez, the longer lived acetone could be also transported from the Mediterranean Sea (where acetone was high)". Acetone is quite similar in Mediterranean and Suez. "where acetone was high" seems to indicate that acetone source in Suez is dominated by transport rather than local production. We do not know the contribution of transport vs. local source to acetone in Suez region, so need to provide evidence or reorganize the sentence.**

   Yes. It is correct that we do not know the contribution fraction of transport and local source to acetone. We reorganized the sentences as specified in the last point (6).

8) **L249-250. "Although the mean mixing ratios of aliphatic carbonyls over Suez were lower than those over the Arabian Gulf, larger variations were observed." I'm not seeing the higher variation over Suez than Arabian Gulf.**

   We reorganized the sentence to avoid possible confusion and misunderstanding: "Although the mean mixing ratios of aliphatic carbonyls over Suez were much lower than those over the Arabian Gulf, the variations were still more significant than other regions (not including the Arabian Gulf, see Table 1)."

9) **L254-258. The arguments here are based on back trajectory. How about the proposed deep sea local source of ethane and propane in Bourtsoukidis et al. 2020? This was found not sufficient to explain the model bias of acetaldehyde later in the paper, but it worth being brought up here as one potential explanation of high carbonyls in the Red Sea North and also briefly mention what you found later in the model …**

   We agree that by adding the deep water source information as well as the model comparison result would make the argument more complete. It is now written in the revised manuscript as "…while the sourthern part was more influenced by air from the northern part of the Red Sea mixed with the air masses from desertic areas of central Africa. Therefore, less primary precursors as well as carbonyls were transported to the sourhtern part of the Red Sea compared to the northern part. Moreover, the unexpected sources of hydrocarbons (ethane and propane) from Northern Red Sea deep water reported by Bourtsoukidis et al. (2020) would lead to higher carbonyl levels in the Northen part compared with the Sourthern part due to the additional precursors in the Red Sea North. However, acetaldehdye was still found to be significantly underestimated compared to the model results, even taking the deep-water source into consideration (section 3.3). This indicates that extra sources of acetaldehyde may exist, which will be disscussed in detail in section 3.4."

10) **Table 2. Are you summarizing only surface or tower measurement? Need to clarify.**

   The content of Table 2 was also questioned by reviewer 1. We have now updated the table by including more relevant studies, done in open sea and coastal areas because AQABA measured the marine atmosphere for most of the time. We included the coordinates (latitude and longitude) and measurement sampling height (above sea level) of these studies.

   Too few examples of Oil & Gas region and Forest, add more.

   We now added more studies done related to Oil&Gas regions. However, although lots of studies reported hydrocarbons (NMHCs) from oil&gas region over the world, we could only find OVOCs results of oil&gas related studies done in the US. In terms of the forest category, we excluded together with other categories (urban and rural) because they were less related to AQABA ship campaign atmosphere.

   The revised table is attached in the end of this response.

11) **L291, the title of Section 3.2 "Chemistry of aliphatic carbonyls", does not reflect the source characterization discussed in the two case studies in Section 3.2.2.**

   The section 3.1 and 3.2 were reorganized as we removed one section which will be addressed in the next point. The section titles were changed to

3.1 Aliphatic carbonyls ($C_nH_{2n}O$)
3.1.1 Overview
3.1.2 Case studies of polluted regions: the Arabian Gulf and Suez
3.2 Unsaturated and aromatic carbonyls ($C_nH_{2n-2}O$), ($C_nH_{2n-8}O$)
3.2.1 Overview
3.2.2 Potential precursors and sources of unsaturated carbonyls

12) **L303, Eq. (2). Varying formats of this equation have been widely applied but I have one question regarding its application in secondary OVOCs from aged air. Eq. (2) assumes that both precursor hydrocarbon and aliphatic carbonyls experienced the same degree of OH exposure (or Δt). This assumption is reasonable for fresh air mass, but might not be valid for aged air, in which carbonyls kept being produced along the way (depending on its precursor's lifetime) and thus experienced different degrees of OH exposure. So, for long-transported air masses, this could lead to an overestimated OH exposure, an underestimated carbonyl secondary production, and thus an underestimated fractional contribution from oxidation to the measured carbonyl mixing ratio.**

    Would this uncertainty affect your conclusion (P12, L334-336; P22, L615-618) that hydrocarbon oxidation plays a more important role in polluted regions than other less-polluted regions, given the potential low bias in the estimated contribution for less-polluted/more-remote regions?
    Thanks for pointing out the inappropriate application of the Eq. (2) we applied in our study to estimate the carbonyls produced by precursors especially in remote areas where the primary emissions were unknown and the air was aged. The empirical method was also questioned by reviewer 1, because photolysis, primary emissions, dilution effect from the background air and uptake by the land of ocean could influence the small carbonyls to a large extent, factors which were not considered at all in the empirical calculation. We agree that this section does not therefore have much scientific contribution to the whole manuscript. As we compared the measurement with a comprehensive global model in the manuscript, we removed the part regarding the empirical calculations and corrected relevant text in the revised manuscript.
    However, we kept the content of estimating the OH exposure using hydrocarbon ratios for the polluted regions where primary emissions were strong and measured (Arabian Gulf and Suez). And we moved this description to the case studies because we included the OH exposure there to help discuss the air mass age.

13) **L308-312. Table S4. How is the yield being derived from MCM?**
    We assumed the alkylperoxy radicals (RO_2) would mainly react with NO (high NO condition) and then sum 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.

14) L318. Did you use 0-5AM data as in Yuan et al. 2012? Clarify that.
L319. How do your calculated emission ratios compare to literature reported values from similar sources?

Thanks for pointing out this important detail. We now updated the results by using the benzene and toluene data during the night according to actinic flux data and compared the emission ratios to other studies from similar sources. As mentioned in point 12, we moved OH exposure calculation to the case studies. The text is now modified as follows:

For further insight, we focused on a time series of selected trace gases along with the correlations among them 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 these polluted regions to better understand the photochemical aging of the major carbonyls using the following equation:

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

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."

15) L329, "In general, the direct oxidation fraction varied from area to area for C1 to C3 carbonyls (formaldehyde, acetaldehyde and acetone)." Give the range of fractions for each species.

We removed the part regarding the empirical calculations and corrected relevant text in the revised manuscript.

16) L414, Section 3.3 title "air chemistry of unsaturated carbonyls" does not reflect the discussion in this section, which is more focused on potential precursor/source of large unsaturated carbonyls.

We corrected the title to "Potential precursors and sources of unsaturated carbonyls" in the revised manuscript.

17) L470-471. "A strong natural non methane hydrocarbon source from deep water in the Northern Red Sea was implemented in the model (Bourtsoukidis et al., 2020)." This needs to be added in the method section where you describe the model configuration. There you have stated that the

configuration is following Bourtsoukidis et al. 2020, but also worth pointing out that this newly proposed source is applied.

Thanks for the suggestion. We now add this information to the method section 2.5 as “The model configuration in the study is the same as the model applied in Bourtsoukidis et al. (2020), where an extra natural non-methane hydrocarbon source (ethane and propane) was implemented.”

18) **L520. “the air-sea submodel (Pozzer et al., 2006) was augmented to include …” what is the air-sea submodel? Is that a model coupled into the EMAC model? Or it’s a separate model you used to get the ocean acetaldehyde concentration field? Need to clarify. And if it’s coupled to EMAC, need to add in the EMAC model configuration section.**

    This was also mentioned by reviewer 1. The AIRSEA submodel was coupled into the EMAC. We now added this information in the method section 2.5: “The exchange of organic compounds between ocean and atmosphere was considered in EMAC via 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).”

19) **P21, Section 3.5.3, “Anthropogenic primary sources”. Comparing to Section 3.5.1 and 3.5.2, this section is lack of analysis-based discussion.**

    Also, if the revised version kept the same length or longer, better to separate into two paragraphs, with the first one discussing the underestimated anthropogenic primary emissions of acetaldehyde precursors (L550-558) and the second talking about underestimated anthropogenic primary emissions of acetaldehyde itself (L558-end).

    We reorganized this section as suggested in the revised manuscript. In terms of the analysis-based discussion, we included one more plot in the supporting information (Figure S9. Time series of ozone mixing ratios and measurement to model ratios of acetaldehyde, propene and ethene over the Arabian Gulf) inspired by the referee’s later comments to further address the potential contribution to carbonyls from nighttime ozonolysis of alkenes.

    L551. Does the carbon number of unmeasured hydrocarbons start from C12? Figure 3 and 6 only considered HCs up to C8.

    Thanks for spotting this mistake. We corrected C12 to C8.

    L552-555. “Bourtsoukidis et al. (2020) compared measured hydrocarbons (ethane, propane, ethene etc.) with the results from model simulations (the same model used in this study) and periodically found significant model underestimation in both regions. This indicates that not all sources were present in the model’s emission inventory”. This seems to argue that the low bias of ethane, propane, ethene, etc. in the model inventory is one reason for the model underestimation of acetaldehyde, which contradicts the argument (P13, L340-349) based on Figure 3. The argument of non-measured and non-modeled HCs before this sentence is fine, but this sentence makes it unclear.

    As the Figure 3 related to empirical calculations of carbonyls was removed from the revised manuscript, there should be no confusion. We modified the sentences to make it clearer to demonstrate the contribution from underestimation of precursors in the revised manuscript: "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 deep water source implemented). The model was able to mostly reproduce the measurement over different regions expect for periodically significant model underestimations in Suez and Arabian Gulf, in which local and small-scale emissions were difficult for 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."

L555-550. 1) need to say that the high ozone in case studies was observed during "nighttime". 2) Does the modeled occasional underestimate of ethene persist in nighttime too?

1) We corrected the sentence to "As mentioned in the previous case studies, high ozone mixing ratios were observed over the Arabian Gulf and Suez especially during the nighttime."
2) Thanks for mentioning the alkene model comparison. We now made a new plot included in the Supporting Information showing the time series of measured to model ratios of alkenes together with ozone. Relevant text was also added in the revised manuscript: "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."

L565-567. "Therefore, the active petroleum industry located in the Arabian Gulf and intensive marine transportation in Suez are likely primary sources of acetaldehyde and other carbonyls which were not well constrained in the model." This is not convincing.

We have rewritten the sentence to raise the point without indicating certainty as follows: A possible explanation for the measurement-model discrepancy is that the active petroleum industry located in the Arabian Gulf and intensive marine transportation in Suez are primary sources of acetaldehyde and other carbonyls which were not well constrained in the model.

20) L587, "Another acetaldehyde formation pathway …" suggest starting a new paragraph to discuss pyruvic acid.

We started a new paragraph to discuss pyruvic acid in the revised manuscript.

L590-592. "Zhou and Mopper (1997) pointed out that the net exchange direction for pyruvic acid is expected to be from the air to the sea due to its high partition coefficient (high solubility). Therefore, only low levels of pyruvic acid would be expected in the remote marine boundary layer". High air-to-sea partition tendency does not guarantee low level of pyruvic acid in the air-side, if gas-liquid partitioning is the only limiting process, its concentration in the air also depends on its degree of saturation in the surface seawater. A review of surface seawater concentration of pyruvic acid and an air-sea equilibrium calculation might be helpful.

L597-598. "As the air-sea exchange of pyruvic acid is limited, low levels of pyruvic acid were expected" Not clear what this means. How did you know that the air-sea exchange of pyruvic acid is limited? And why would that result in low levels of pyruvic acid?

As the two comments above are related, we respond to them together as follows. Regarding the potential contribution of pyruvic acid photolysis to acetaldehyde, we now adopt the reviewers suggestion to make this point clear in the revised manuscript as follows: "…Pyruvic acid has been also observed in seawater (Kieber and Mopper, 1987;Zhou and Mopper, 1997) and was found up to 50 nM in the surface water of eastern pacific Ocean (Steinberg and Bada, 1984), while acetaldehyde was not the major product of aqueous-phase photolysis of pyruvic acid (Griffith et al., 2013). Zhou and Mopper (1997) pointed out that the net exchange direction for pyruvic acid is expected to be from the air to the sea due to high solubility, with Henry's law constant of 3.1×10^3 mol m^{-3} Pa^{-1} (Sander, 2015). Moreover, partitioning to aerosols could be an important sink for pyruvic acid (Reed et al.,2014; Griffith et al., 2013) : an increasing concentration trend of pyruvic acid was observed in marine aerosols over western North Pacific Ocean (Boreddy et al., 2017). Therefore, due to limited terrestrial biogenic sources of pyruvic acid for AQABA campaign, gas-phase level of pyruvic acid was expected to be low. Limited studies reported pyruvic acid level in marine boundary layer, Baboukas et al. (2000) measured 1.1 ± 1.0 ppt of pyruvic acid above the Atlantic Ocean….."

New literature citation:

Steinberg, S. M., and Bada, J. L.: Oxalic, glyoxalic and pyruvic acids in eastern Pacific Ocean waters, Journal of Marine Research, 42, 697-708, 10.1357/002224084788506068, 1984.

Sander, R.: Compilation of Henry's law constants (version 4.0) for water as solvent, Atmospheric Chemistry and Physics, 15, 4399-4981, 10.5194/acp-15-4399-2015, 2015.

Reed Harris, A. E., Ervens, B., Shoemaker, R. K., Kroll, J. A., Rapf, R. J., Griffith, E. C., Monod, A., and Vaida, V.: Photochemical kinetics of pyruvic acid in aqueous solution, J Phys Chem A, 118, 8505-8516, 10.1021/jp502186q, 2014.

L598-599. "Even if we fully assign the m/z 89.024 to pyruvic acid, the contribution to acetaldehyde via photolysis of pyruvic acid is negligible compared other sources" Clarify why it is negligible, using 58 ppt and a 100% yield? Is this contribution detectable by instrument?

We added detailed information to clarify this point in the revised manuscript as follows: "For the AQABA PTR-ToF-MS data set, enhanced signals were observed at m/z 89.024 with the mean mixing ratio of 35-110 ppt over different regions (Table S4), which is much more abundant than reported pyruvic acid levels by Baboukas et al. (2000). This might be due to the uncertainty associated with the theoretical methods of quantification used here or the presence of isomeric compounds on that mass, since pyruvic acid was not calibrated with the standard. Even if we assume the m/z 89.024 to be entirely pyruvic acid, with 60% yield of acetaldehyde via photolysis (IUPAC, 2019), it gave maximum 13 ppt of acetaldehyde over Arabian Gulf, 5-9 ppt over other regions, which were only 0.8% - 6% of the mean mixing ratios (Table S4). Detailed information of the calculation can be found in the Supporting Information. Therefore, we conclude that the contribution from the photolysis of pyruvic acid is not an important source for the unattributed acetaldehyde during the AQABA campaign."

New citation:

IUPAC Task Group on Atmospheric Chemical Kinetic Data Evaluation, (http://iupac.pole-ether.fr).

Accordingly, we added the detailed information of the acetaldehyde calculation in the supporting information as follow:

Calculation of acetaldehyde yield from pyruvic acid photolysis

In order to verify the contribution from the photolysis of pyruvic acid to acetaldehyde, we calculated the expected acetaldehyde produced through pyruvic acid photolysis over different regions assuming: (1) *m/z* 89.0234 is fully assigned to pyruvic acid; (2) the loss of pyruvic acid is only through photolysis; (3) 60% is the yield of acetaldehyde via pyruvic acid photolysis recommended by IUPAC (2019); (4) the loss of acetaldehyde is only through OH oxidation. The acetaldehyde produced via pyruvic acid photolysis can be calculated using following equation (consecutive reactions):

$$[Acetaldehyde] = [Pyruvic\ acid]\frac{J_{PA}}{k_{OH}[OH]-J_{PA}}[\exp(-J_{PA}\Delta t) - \exp(-k_{OH} \times [OH]\Delta t\,)] \qquad \text{Eq. S1}$$

$[Pyruvic\ acid]$ is the mean of pyruvic acid mixing ratio in each region. J_{PA} represents the mean photolysis rate constant of pyruvic acid during the daytime (dawn to dusk) in each region calculated from the wavelength resolved actinic flux data using quantum yield of 0.2 as suggested by IUPAC (2019). The k_{OH} is the rate constant of acetaldehyde reacting with OH radical (1.5×10^{-11} cm^3 $molecule^{-1}s^{-1}$, Table S3). The $[OH]$ concentrations in each area were the mean values during the daytime obtained from the EMAC model. The maximum acetaldehyde level as well as the corresponding time (Δt) can be derived from Eq. S1 as Δt is the only variable. The results are shown in Table S4.

Table S4. Mean photolysis rate constant of pyruvic acid, OH concentrations, relative time (Δt) needed to reach the maximum acetaldehyde yield from pyruvic acid photolysis, maximum acetaldehyde and its fraction accounting the mean level over regions.

Regions	J_{PA} (s^{-1})	OH (molecules cm^{-3})	m/z 89.0234 pyruvic acid (H^+) (ppt)	Δt (h)	Acetaldehyde maximum (ppt)	Fractions (%)
MS	3.51×10^{-5}	6.52×10^6	39 ± 8	5.6	5.6	1.85
SC	3.44×10^{-5}	7.42×10^6	42 ± 9	5.2	5.3	0.85
RSN	3.52×10^{-5}	7.14×10^6	35 ± 14	5.2	4.7	0.92
RSS	3.00×10^{-5}	8.74×10^6	61 ± 15	4.9	6.2	1.98
GA	3.11×10^{-5}	7.20×10^6	57 ± 12	5.5	6.8	3.60
AS	2.74×10^{-5}	4.35×10^6	59 ± 12	7.8	9.4	5.88
GO	3.31×10^{-5}	7.89×10^6	65 ± 10	5.0	7.6	2.91
AG	3.29×10^{-5}	7.81×10^6	110 ± 53	5.1	12.9	0.75

Minor/technical comments:

L17: typo “3,8”

It is now corrected to “3.8” in the revised manuscript.

L89: section numbering error, should start with 2.1

It is now corrected to “2.1” in the revised manuscript.

L131 and Table S1: 3σ in text but 1σ in Table S1

We corrected Table S1 to 3σ.

L135: add “of” between “number” and “three”

It is now added in the revised manuscript as "… with a carbon number of three…)

L156: "interaction with ocean/land" includes "dry deposition"

We deleted "dry deposition" in the revised manuscript.

Table 1. L193, L207. "Aliphatic CCs, Aromatic CCs, Unsaturated CCS", "CC" hasn't been defined.

We changed "CCs" in the table to "Carbonyls" in the revised manuscript.

Table 1. L193, L207. Extend the name of "S" to "Suez"

We made this change in the revised manuscript.

L348. "section" to "sections"

The related content was deleted. Therefore, this correction was not applicable.

Figures 4, 5, and 7. Change x-axis label from "Dateandtime" to "Dateandtime (UTC)", or clarity in the figure caption.

We made this change in the revised manuscript.

Figures 4 and 5. missing units for several y-axis, "J_NO2", "[OH]t", "Wind_D"

We made this change in the revised manuscript.

Figure 7a left panel. "S" to "Suez"

We made this change in the revised manuscript.

Figure S6. Only 6 regions shown in the figure.

We added the other two regions in the Figure S6 in the revised manuscript.

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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