Referee 1:

We would like to thank the referee for the detailed review of our manuscript and the highly valuable feedback. The referee's comments (black) our replies (blue) and the resulting modifications to the manuscript (red) are listed below.

The author report measurements of HCHO on a ship cruise. Though this is a measurement report, the discussion is partly not very satisfying and it is not very clear, if there is any deeper meaning in the analysis. It remains unclear, if there is anything to learn from the HCHO yield that is calculated, specifically if the inorganic fraction of the OH reactivity is included. It may make it easier to exclude inorganic species that do not produce HCHO from the beginning.

We modified a few figures and now used the OH reactivity corrected for non-HCHO yielding reactions for the main plots. The NOx dependence of the HCHO production is now discussed in more detail, although we decided to remove the scatter plot of α_{eff} vs NO_x, as a more detailed study would be necessary. In general, the dataset covers modestly polluted air masses with respect to NO_x and thus we think it is likely that we almost never achieved conditions which favor the formation of peroxides through recombination of RO_2 instead of the production of HCHO through RO_2+NO.

One goal of the study was to check whether the calculation of the HCHO yield (α) in different regions correlates with the variation of the measured OH reactivity towards different chemical families (VOCs, OVOCs, aromatics, alkenes) as a proof of concept. The results represent first studies of the HCHO dataset and thus we decided to write a measurement report to highlight the change in chemical composition during AQABA.

Specifically, the dependence on NOx drastically change, because NO and NO2 are OH reactants, but what the authors want to point out is that the fate of RO2 and therefore the product distribution depends on the availability of NO. In addition, the authors want to connect the HCHO production with the chemistry and not just with the presence of OH reactants that do not produce HCHO (e.g. HCHO would be very low in an environment with high NO2 though there might be a lot of efficient HCHO from the chemistry of the organic compounds). The argument that subtracting the inorganic part reduces the data set is weak because the inorganic fraction reducing the chemical meaning of the derived yield. In the discussion the authors mention also the results, if the inorganic fraction is subtracted, but there is no meaningful interpretation.

P3 L2: "reactions" instead of "reaction"

Correction made.

P2/3: It is confusing that Equation 1 is defined for the sum of all reactants, but that later the authors distinguish between different type of OH reactants. This should be consistent specifically regarding the "yield".

In order to clarify this, we have added new indices in Eq. 1 and improved the discussion of the results. The HCHO yield was calculated including all VOCs, we only excluded the contribution of inorganic trace gases and non-HCHO yielding reactions ($R(OH)_{eff}$).

P2 L30 – p3 L4:

In addition to summing up the contributions of individual HCHO production pathways, the production rate of HCHO resulting from reactions involving OH chemistry ($P_{OH}(HCHO)$) can be deduced from the

OH concentration ([OH]), the HCHO yield α_i and the OH reactivity (R(OH)) which represents the summation of all trace gases R_i that react with OH with the rate coefficient k_i (Liu et al. 2017; Wolfe et al., 2019):

$$P_{OH}(HCHO) = \alpha_i \cdot [OH] \cdot R(OH)_i \tag{1}$$

with

$$R(OH)_i = \sum k_i \cdot R_i \tag{2}$$

P4 L14–16:

For further interpretation, the effective HCHO yield α_{eff} was determined for each region by removal of non-HCHO yielding reactions (of NO, NO_2, SO_2, CO, HCHO and O_3 with OH) from the OH reactivity data ($R(OH)_{eff}$, Fig. S1).

$$\alpha_{eff} = \frac{P(HCHO) - P_{add}(HCHO)}{[OH] \cdot R(OH)_{eff}} \tag{6}$$

P3 Equation 4: Could transportation play a role?

Transport of air masses e.g. by advection can have a significant influence on the HCHO mixing ratio, depending on the air mass origin. Since HCHO has many sources and is also released primarily, a strong influence is expected close to primary and secondary sources (e.g. ship plumes and close to cities). Obvious ship plumes were identified with NO_x, CO, SO_2 measurements and excluded from this study, as well as data during the night (sea breeze effect). We address the effects of transport later in the manuscript when interpreting the data and have also added the following at this point in the manuscript.

P3 L 31 – P4 L2:

The derived production and loss rates of HCHO can be influenced by direct emissions or by advective transport. Obvious direct emissions from ship plumes or other sources were excluded from the dataset. The potential role of transport is addressed in section 4.

P4 Equation 5: It would be easier for the reader, if Eq. was written using the same terms as in the previous Equations.

We changed $P_0(HCHO)$ to $P_{add}(HCHO)$ in Eq. 5. We also added additional information about α, since it represents a lower estimate of the HCHO yield when including non-HCHO yielding reactions of OH. Equation 6 was added as the equivalent for the calculation of α_{eff} via $R(OH)_{eff}$.

P4 L 7–18:

Scatter plots of [OH] × R(OH) versus P(HCHO) yield the lower estimate of the formaldehyde yield α with respect to total OH chemistry (including NO_x, SO_2 and other non-HCHO yielding reactions, Fig. S2), reflecting the transition between rather clean to highly polluted conditions, both with respect to NO_x and VOCs.

$$\alpha = \frac{P(HCHO) - P^{*}_{add}(HCHO)}{[OH] \cdot R(OH)} \tag{5}$$

For further interpretation, the effective HCHO yield α_{eff} was determined for each region by removal of non-HCHO yielding reactions (of NO, NO_2, SO_2, CO, HCHO and O_3 with OH) from the OH reactivity data ($R(OH)_{eff}$, Fig. S1).

$$\alpha_{\mathrm{eff}} = \frac{\mathrm{P(HCHO)} - \mathrm{P_{add}(HCHO)}}{\mathrm{[OH]} \cdot \mathrm{R(OH)_{eff}}} \qquad (6)$$

P7 L11: Are the assumptions about the photo-stationary state plausible, if maximum values were around noontime, when photooxidation is at its maximum and the chemical lifetime is 2.5h? Wouldn't the maximum expected to shift into the afternoon?

The HCHO lifetime is sufficiently short that PSS is, to a good approximation, achieved around local noon. The hypothesis is supported by the fact that values of *d* (HCHO) / dt were quite small on most days (Fig. 2), especially in the clean regions AS and MS. A shift to the afternoon can occur, when P(HCHO) is dominated by VOC oxidation processes which involve further photolysis steps / reactions prior to HCHO release.

Valin et al. 2016 (Fig. 2) also suggests PSS conditions during local noon with a HCHO lifetime of ~ 2.5 h, where L(HCHO) = P(HCHO).

P8 L18: The reason for using of a constant value for the BHL instead of 30 min values is not plausible. Why does an interpolation of values lead to a lower data coverage compared to using a constant value?

The (lack of) variability in the BLH and the associated uncertainty in this parameter do not warrant a more detailed treatment than use of a fixed BLH of 750 ± 113 m (± 1σ). We note additionally, that deposition accounts for only ~ 5–15 % of the HCHO loss, so use of an interpolated BLH would not change the results and/or conclusion of the analysis.

P9 L9–10:

For the BLH we used the mean value (± 1σ) of the ERA-5 results (750 ± 113 m)~~, since the data was unfortunately not available in 5-minute time resolution~~.

P9 L13: A compact linear relationship would only be expected, if chemical conditions do not change and transportation does not play a role. This should be made clear.

We agree and have made this clear in the manuscript where we discuss the correlations between P(HCHO) and [OH]*R(OH) in detail.

P10 L3–6:

A compact linear relationship is expected under PSS conditions if chemical conditions do not change and the air mass is not affected by transport. While data points that do not fulfill PSS, (e.g. due to direct emissions from point sources, or advection of HCHO enriched or depleted air masses) are expected not to follow the regression line, yielding additional scatter.

Figure 4: Error bars would help to see, how much of the scatter is explained by the statistical errors of data points.

To maintain clarity of presentation, we included one representative data point with error bars for each plot.

P11 Fig. 4:

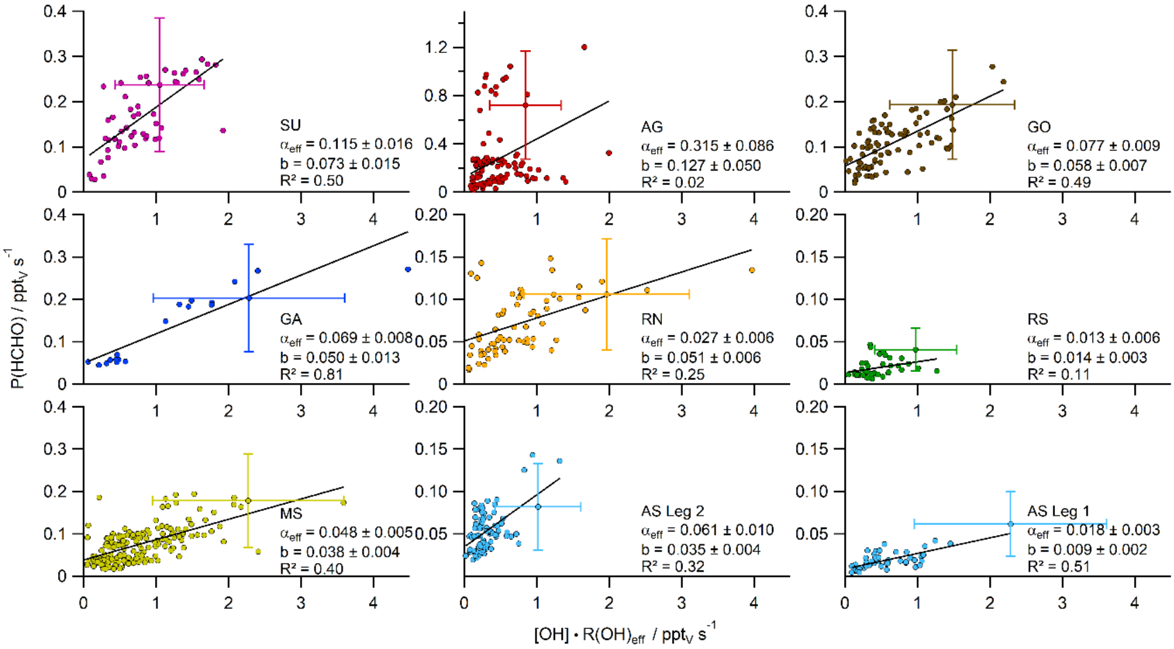

Figure 4: Scatter plots with bivariate fits (York et al., 2004) of the product [OH] × R(OH)$_{eff}$ (± 58 %) versus the HCHO production rate P(HCHO) (± 62 %), subdivided into the different regions probed during the AQABA cruise. The slope of the respective regression represents the HCHO yield α_{eff}, while the intercept can be interpreted as additional HCHO sources not related to OH chemistry (P$_{add}$(HCHO)).

P14 L12-221 and Figure 5: I do not see the meaning of plotting the yield against reactivity. Why should a yield linearly scale with the concentration of a family species? I would rather expect a constant number and a discussion, if the numbers are in any way expected from chemical oxidation mechanisms of these family species. In addition, the slopes in plots in Fig. 5 are essentially determined by one data point, because all other points are scattered in the low range. Please comment.

The HCHO yield of a certain compound class is not expected to change under the same chemical conditions. To clarify this, we modified figure 5 and now use the relative contribution of the separated compound classes (R(OH)$_x$/R(OH)$_{eff}$) on the x-axis vs α_{eff}, color-coded the z-axis with NO$_x$ and removed the bivariate linear fits. We now discuss in more detail how the increase in mixing ratio of a certain compound class would affect the HCHO yield per OH lost. The method used here is not feasible for a detailed study, since we cannot account for chemical aging and inhomogeneity in inflow with our dataset.

We also decided to exchange α for α_{eff} in the discussion of the manuscript (also in Table 1), since this adds more value when excluding inorganic and non-HCHO yielding reactions from the beginning. The extensive modifications made to the manuscript in response to these comments are listed below.

P11 L10–13:

For further analysis, we removed the highest contributors to inorganic reactivity (NO, NO_2, SO_2, O_3) and further non-HCHO yielding reactions (CO, HCHO) from R(OH), which resulted in so-called effective OH reactivity ($R(OH)_{eff}$). Even though the dataset was already filtered for stack emissions, NO_x showed the highest contribution of these reactants (Fig. S1, Table S1), especially over the Gulf of Oman and while passing Bab-el-Mandeb (16.08.2017) and the street of Messina (30.08.2017).

P13 L14–15:

Removal of non-HCHO yielding reactions from the OH reactivity ($R(OH)_{eff}$) reduced the overall scatter significantly, which resulted in increased R^2 values for the Suez Canal and the Arabian Sea during leg 2 (Fig. 4, S2).

P14 L12–15:

However, the determined α_{eff} did not change significantly in most regions, which is also the case for the lowest values determined for the Arabian Sea (0.018) and the southern Red Sea (0.013). In these rather clean regions a rather low contribution of inorganic reactivity is expected, which was proven (Fig. S1).

P14 L21–32:

For further analysis, we have recalculated mean OH reactivities ($\pm 1\sigma$ standard deviation) of the individual substance classes ($R(OH)_x$, e.g. alkanes, alkenes, OVOCs, aromatics) based on the findings of Pfannerstill et al. (2019). We assume that the unmeasured VOCs are proportional to the measured compounds of a certain compound class, so that these values represent general trends for the VOC oxidation of the subdivided regions (Table 1). Please note that we removed the reaction of HCHO with OH from the OVOC class, which is usually included when presenting speciated reactivity (Pfannerstill et al., 2019). The ratio of $R(OH)_x/R(OH)_{eff}$ then represents the relative contribution of a certain compound class to the regional so-called effective reactivity, which reflects the measured VOC oxidation plus unattributed reactivity. Alkanes (including CH_4) contributed on average 3 – 10 %, alkenes 2 – 10 %, OVOCs 8 – 33 % and aromatics 1 – 9 % to regional $R(OH)_{eff}$ (Table 1, Fig. 5). All sulfur-containing VOCs together contributed less than 1 % to the total $R(OH)_{eff}$ and were thus neglected. Elevated contribution of alkanes to $R(OH)_{eff}$ was found in SU and AG, while their contribution in the remaining regions was rather constant (~ 5 %) and dominated by the oxidation of methane.

P15 L2–5:

The sum of alkanes, alkenes, OVOCs and aromatic compounds on average contributed 19 % over the Mediterranean Sea, 32 % over the Suez Canal, 31 % over the northern Red Sea, 37 % over the southern Red Sea, 23 % over the Gulf of Aden, 16 % on the first leg over the Arabian Sea, 38 % over the Gulf of Oman, 62 % over the Arabian Gulf and 23 % on the second leg over the Arabian Sea to $R(OH)_{eff}$.

P15 Table 1: now includes α_{eff} instead of α

P15 L13–23:

Methane oxidation via OH and other oxidants (e.g. Chlorine, Cl) is expected to be the main source of HCHO in the remote marine boundary layer, via the production of methylperoxy radicals (CH_3O_2). The fate of these CH_3O_2 radicals determines the main production pathway of HCHO, as they can react to other species e.g. methyl hydroperoxide (CH_3OOH) or methanol (CH_3OH) under low-NO_x conditions (Anderson et al., 2017). The determining step to form HCHO is the reaction of CH_3CO_2 with NO, thus it is expected that HCHO production is suppressed in very clean regimes, since these favor the recombination of CH_3CO_2 and the reaction with HO_2. Emission of rather short-lived alkenes (e.g. ethene, isoprene) can significantly enhance HCHO production via reaction with O_3 or OH and the subsequent formation of OVOCs (e.g. acetone, acetaldehyde). The release of HCHO from these secondary products not only depends on their chemical structure, but also on additional photolysis steps involved. While methane oxidation dominates the HCHO production in the MBL, isoprene is the main precursor over continents for near-surface HCHO. The results of Wolfe et al. (2016) also highlight that the HCHO yield from isoprene oxidation is a nonlinear function of NO_x.

P16 L3 – P17 L31:

In clean MBL environments, emissions of alkenes and aromatics are sparse and thus the distribution of HCHO should be primarily controlled via variability of HO_x (= OH + HO_2) and the presence of NO_x (> 0.1 ppbv), while we expect the HCHO budget to be highly complex under more polluted conditions e.g. for the area around Suez Canal and over the Arabian Gulf.

In order to investigate if the change in chemical composition along the different regions correlates with the determined HCHO yield, Fig. 5 shows scatter plots of the HCHO yield α_{eff} versus the measured OH reactivity towards certain compound classes (alkanes, alkenes, OVOCs, aromatics) as the contribution to total OH reactivity without inorganic reactions (e.g. $R(OH)_{alkanes}/R(OH)_{eff}$). We also color-coded the z-axis with the median of measured NO_x as an indicator for air pollution and in order to identify the regions more easily.

The relative contribution of alkanes to $R(OH)_{eff}$ shows low variation at ~ 4 – 5 % except over the area around Suez Canal (~ 7 %) and over the Arabian Gulf (~ 10 %), where methane (Paris et al., 2021) and a variety of NMHCs (Bourtsoukidis et al., 2019) showed significantly elevated mixing ratios. Over the Arabian Gulf, these were likely caused by emission from the oil and gas industry, indicated by the ratio between i-pentane and n-pentane of 0.93 ± 0.03 ppbv $ppbv^{-1}$ while Bourtsoukidis et al. (2019) found a ratio of 1.71 ± 0.06 ppbv $ppbv^{-1}$ over the Suez Canal, which is representative for ship emissions. From a visual inspection, a slight increase of the relative reactivity of alkanes lead to a small, but significant increase of α_{eff} with simultaneously elevated values of NO_x. Alkenes contributed an average between ~ 2 – 10 % to $R(OH)_{eff}$, with the highest values over the Arabian Gulf, the southern Red Sea and the Suez Canal. Interestingly, a high reactivity of alkenes was detected over the southern Red Sea, which was similar to the enhanced values over Suez Canal (Table 1), although with a higher relative contribution. The NO_x measurements indicate rather clean air over the southern Red Sea, where we experienced mostly winds coming from the west (Eritrea, Sudan). As a first result, a higher reactivity of alkenes not necessarily correlated with an increase of α_{eff}, which is expected as the release of HCHO from alkene oxidation depends on photolysis steps and other reactions involved. Also the availability of oxidants was higher over the Suez Canal and the Arabian Gulf, where the highest amounts of O_3 were detected during AQABA (Tadic et al., 2020). This also likely interferes with the here shown results and would lead to an increase of $P_{add}(HCHO)$. In general, emissions of alkenes favor HCHO production, which was also highlighted by the results of Luecken et al. (2018), who found a high sensitivity of HCHO to anthropogenic and biogenic emissions of alkenes with isoprene as the main contributor in summer. We therefore want to emphasize the limits of the here used method, as it only takes into account the `immediate yield` of HCHO. OVOCs showed overall the highest contribution to $R(OH)_{eff}$ with ~ 8 – 33 % during AQABA and high $R(OH)_{OVOCs}$ generally resulted in an increased α_{eff}, with the highest contribution over the Arabian Gulf, the Gulf of Oman and the Suez Canal. The HCHO production through OVOCs strongly depends on the prevalent composition and the amount of anthropogenic emissions. Some C_1-C_3 OVOCs (e.g. methanol, acetaldehyde, acetone, glyoxal) are expected to have a strong influence on the local HCHO distribution depending on their mixing ratio, although this effect is minor in remote marine boundary layer conditions without anthropogenic contribution (Anderson et al., 2017). High amounts of acetaldehyde and acetone over the Arabian Gulf (Wang et al. 2020) occurred with a simultaneous increase of HCHO and highlight this region as unique spot of air pollution, which is reflected by the highest α_{eff} and $R(OH)_{OVOCs}$. $R(OH)_{aromatics}$ showed in general the lowest contribution to $R(OH)_{eff}$ with 1 – 9 % with again highest contribution over the Arabian Gulf, the southern Red Sea and the Suez Canal. Luecken et al. (2018) showed that HCHO concentrations above the United States had about the same sensitivity towards aromatic and alkane emissions. Aromatic compounds do not necessarily release HCHO during their oxidation, but generally they can be useful to identify complex anthropogenic emissions e.g. ship emissions or industry. These results overall highlight the elevated levels and complexity of air pollution detected over the Arabian Gulf and the Suez Canal. Also, an absolute increase in $R(OH)_x$ is mostly accompanied by an increase of α_{eff}, although most of the data is scattered at low α_{eff} and there is no clear correlation towards $R(OH)_{aromatics}$ and $R(OH)_{alkenes}$. Our results are not directly comparable to the findings by Luecken et al. (2018), since in their analysis reactions with O_3 (alkenes) and photolysis of HCHO precursors (OVOCs) were included. These sources of HCHO are not represented by $R(OH)_x$ used in our study and the here used method would be more valuable by 'following' an air mass, to account for air mass aging.

P16: In the discussion of the impact of NOx on the HCHO yield, the impact of NO as an OH reactant is missing, if the inorganic part is included. It is hard to see, if there is any meaning in the way the dependence is discussed. This part of the discussion needs significant improvement.

The analysis has been modified with subtraction of the 'inorganic' reactions of OH (including NO, NO_2) from the beginning. The contribution of NO to the total OH reactivity was less than ~ 3 s^{-1} in the used data set, this information is displayed in the SI (Fig. S1).

The discussion of the role of NO for the fate of RO_2 was added to the manuscript and we also added more information about the influence of NO for the recycling of HO_x. The NO_x dependence should be clearer now, although we decided to remove fig. 6 form the manuscript (alpha against NO_x), since a more detailed study would be necessary to identify the role of NO_x for HCHO production.

P17 L32 – P18 L10:

The yield of HCHO can also be interpreted as a function of NO_x levels if there are processes (at low NO) that lead to formation of e.g. peroxides rather than HCHO (Wolfe et al., 2016), this occurs when the recombination of CH_3O_2 radicals is favoured instead of the reaction with NO to form the methoxy radical CH_3O (Stickler et al., 2006). NO also directly influences the abundance of OH, as it recycles HO_2 radicals to OH (Whalley et al., 2010; Wolfe et al., 2016) and thus elevated NO_x generally increases radical turnover. Valin et al. (2016) highlight that in isoprene rich regions, the influence of NO_x on HCHO production is primarily due to its feedback on the production of OH. Wolfe et al. (2016) showed that the NO_x dependence mainly affects the so-called prompt yield of isoprene oxidation as a non-linear function of NO_x. In general, the impact of NO_x on HCHO production can be reduced to two factors: radical recycling and the termination of RO_2 radicals (Wolfe et al., 2016). The fact that very clean background MBL air was rarely sampled during AQABA (lowest median NO_x of 0.16 ppbv over AS Leg 2) indicates that we rarely achieved conditions, in which the production of peroxides through the recombination of CH_3O_2 would dominate the production of HCHO through CH_3O_2 + NO. Tadic et al. (2020) also showed that most regions were in the O_3 production regime (signaling sufficient NO), which indicates that the production of HCHO was rather VOC and not NO_x limited.