Thank you for your helpful comments our manuscript (EGUSPHERE-2022-854). The comments lead us to re-examine the simulation and observation results and the discussions we had made. Please find our itemized response below and the corrections in the re-submitted files. The original comments are in **black.** The responses to the comments are in **blue**. The changes in the manuscript and the supplementary were highlighted in <mark>**yellow**</mark> and were cited in **purple** in this response.

Comments #2

This paper presented field measurements conducted and a coastal site in Hong Kong from October to November 2020, including OH radical and multiple trace gases. OH, measurement was deployed by a new instrument using Chemical Ionization Mass Spectrometry (CIMS). The maximum OH concentration was 1.5×10^7 cm^-3 on 7 November, which are on the higher end of the measurements obtained in the PRD region. Two groups of air parcels were identified during the campaign, i.e. continental air and coastal air. The OH concentrations were generally higher in continental air (maximum of diurnal average: 5×10^6 cm^-3) compared to that of the coastal air (maximum of diurnal average: 4×10^6 cm^-3), so did for most of the trace gases, e.g. NO, NO2, HONO, VOCs (except O3). A box model based was used to simulate OH concentration, which could reproduce the OH concentration for continental air but overpredicts for coastal air. The overprediction was attributed to missing OH reactivity. As explained by the authors, OH measurement is difficult and additional measurement certainly helps to enrich the data set. Also, the authors made large efforts to review the published results presenting a nice overview of the current OH measurement. The structure may be improved if the authors could balance the introduction and results/discussion. Nevertheless, this reviewer suggests the publication of this paper once the following comments are addressed.

Response: Thanks for your comments and encouragement. The revised manuscript contained a more comprehensive discussion based on the comments of the reviewers which balance the structure of the article.

General comments:

As the instrument was deployed/presented for the first time, the description of the OH instrument is not sufficient. Although the authors referred to the technical details in a preprint (Pu et al. 2020 AMTD), this preprint has not been finally published and readers may have difficulties understanding some of the unaddressed issues. For example, the HO2+NOàOH+NO2 reaction that occurs in the inlet could be a positive bias to the OH measurement, which cannot be subtracted by the scavenge procedure. In the preprint, it's said that the reaction time was reduced to 47ms to minimize interference. However, it will produce a similar amount of OH in this reaction time at 1 ppb of NO and HO2 about 2 orders of magnitude higher than OH.

Response: Thanks for the comments on need to add more details on the CIMS. A similar comment was raised by referee 1. As indicated in our response to referee 1, our preprint in AMT did not move to the full publication stage as one referee of the AMT preprint thought our CIMS did not outperform the CIMS in other groups. Following the suggestions from both referees for the present work, we have moved some of the content in the AMTD preprint to the supplementary to support the measurements in this study. See response to referee 1 for details in added materials.

On the positive bias from the reaction of HO_2+ NO in the inlet, Tanner et al., (1997) calculated the OH produced by the HO_2 recycling reaction under different NO conditions (from <60 ppt to 1-2 ppb) in the inlet by a box model. Their results showed that the positive bias of less than 0.5×10^6 cm^{-3} with a 60 ms conversion time, and the bias does not increase with the increase of NO concentration. In our study, a shorter (47ms) conversion time was used, we thus think positive bias is not large (<10%), but should be quantified in the future. However, such positive bias would increase the model-measurement discrepancy in the coastal air mass.

The discussion for the positive bias in the inlet by NO+HO_2 was added in supplementary as shown below:

"The reaction time affects the positive bias of OH arising from HO_2 + NO in the inlet. To estimate this bias, Tanner et al., (1997) calculated the OH produced by the HO_2 recycling reaction under different NO conditions (from < 60 ppt to 1-2 ppb) in the inlet by a box model. Their results showed that the positive bias of less than 0.5×10^6 cm^{-3} with a 60 ms conversion time, and the bias does not increase with the increase of NO concentration. Thus, the conversion time of 47 ms in our study should further reduce such positive bias."

The box model calculation overestimated the observed OH concentration by 73% for coastal air. The authors attributed the model-measurement discrepancy to missing reactivity. However, more discussion/explanation is needed to explore all possibilities. The justification for missing reactivity is not sufficient. In Line 11 Page 9, it's described that the VOC below the detection limit was set to half of the detection limit, which seems arbitrary. How sensitive are the model results depending on this assumption? The derived missing reactivity only appeared in the coastal air masses but not the relatively more polluted air masses from the continental sector, which is a contradicting feature and needs more explanation.

Response: Thanks for the comments. More discussion and explanation on measurement and simulation uncertainties and HO_2 budgets were added to the revised manuscript (see below). After considering all possibilities, we still incline to attribute the discrepancy to missing reactivity.

"**3.3.1 Uncertainties in OH measurement and simulation**
The OH measurement uncertainties have been calculated as described in Section 2.2

and are shown as the error bars in Figures 5 and 6. The model's overestimation of OH in coastal air masses exceeded the measurement uncertainties (Figures 6 and 7), and thus, the measurement uncertainty is unlikely to be the main reason for the discrepancy.

Model uncertainties in our study include the uncertainties in photolysis frequencies correction, uncertainties in the constrained VOCs concentrations when they were below detection limits, and uncertainties from not considering halogen chemistry. On the first possibility, we acknowledge that the correction factor for photolysis frequencies due to cloud presence may be different for different species (Walker et al., 2022), thus, using a single correction factor (based on J_{NO2}) may introduce uncertainty in the model simulations. We think such uncertainty should not be significant because the weather was mostly sunny in the coastal cases. Regarding the uncertainty from the VOCs input, we conducted a sensitivity test to show that the treatment of VOCs that were below the detection limits should have a negligible effect on OH simulation (RUN_{VOC0} and RUN_{VOCDL} in Figure S4). We did not include halogen chemistry in our study as we wanted to compare our results with previous modelling work most of which did not consider halogen chemistry. Our other studies at the same site that did consider the halogen chemistry show a 4% increase in OH concentration from Cl chemistry (Peng et al., 2022) and 2.8% from Br chemistry (Xia et al., 2022), which would even increase the model-measurement discrepancy.

3.3.2 Overestimation of OH sources

Our calculated OH budgets show that the main sources of OH in the coastal air masses were the $HO_2 + NO$ reaction (69%), O_3 photolysis (14%), HONO photolysis (7%), and the reaction between ozone and HO_2 (4%). In the simulation, NO, HONO, and O_3 were constrained by observations. Could HO_2 be overestimated which would cause overprediction of OH?

The main HO_2 sources are the VOCs oxidation by OH and the photolysis of OVOCs. In our study, VOCs and OVOCs were more likely under-measured than over-measured, which would underpredict HO_2 rather than overpredicting it. In addition, not including the halogen chemistry would under-simulate HO_2 at this site (Peng et al., 2022; Xia et al., 2022).

We next examine the possibility of the underestimation of HO_2 sinks as the cause of the overprediction of OH. The major sinks of HO_2 include the reaction of NO to recycle OH, self-reaction to form H_2O_2, and heterogeneous loss by aerosol uptake. The first and second pathways have been considered in the MCM. The heterogeneous uptake … …"

The sensitivity test for the VOCs below the detection limit was added in the supplementary Figure S4 below. In short, those VOCs below detection limits have a negligible effect on the discrepancy. Even if we use the detection limits as model input, the simulation of OH concentration has little changes.
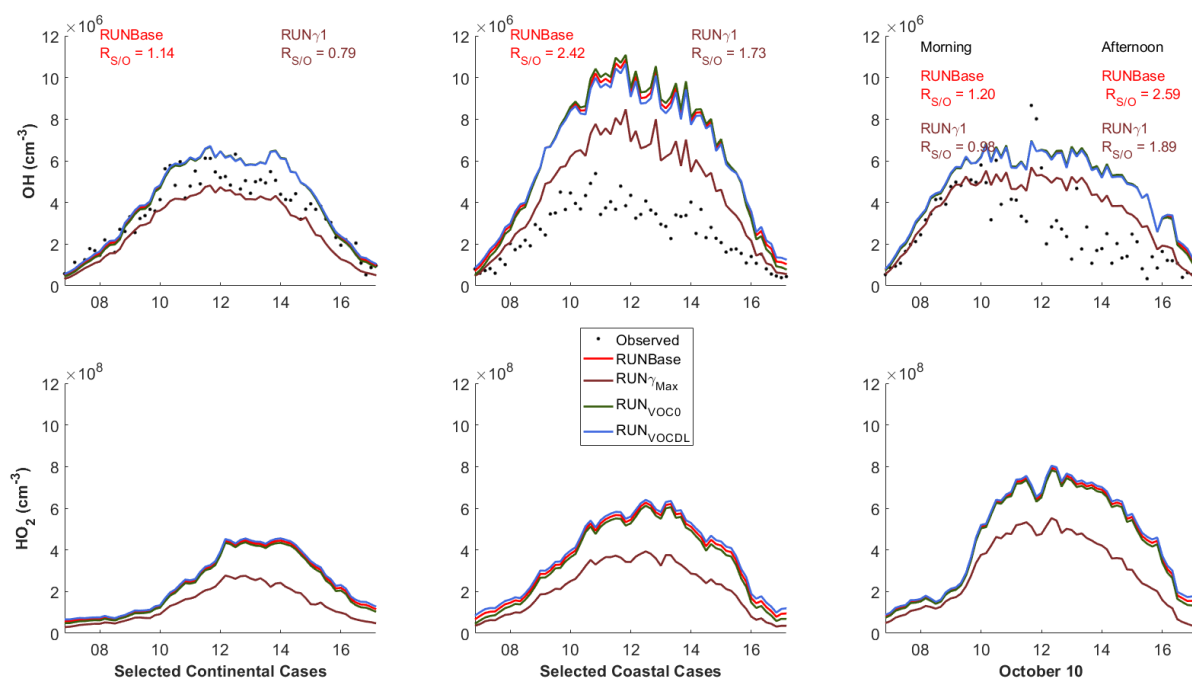
"Figure S4. Sensitivity tests for the simulated OH and HO$_2$ in continental and coastal cases and on 10 October. RUNγ$_{MAX}$ shows the simulated results for the maximum heterogeneous uptake effect of HO$_2$ (γ = 1). The RUN$_{VOC0}$ and RUN$_{VOCDL}$ show the simulated results that constraints "0" and the detection limit value as the concentration of VOCs when their concentration were below detection limits."

We agree that the discussion should focus more on the contradicting feature that the missing reactivity appeared in coastal air masses. We believe that this is an indicator of our knowledge gaps in coastal air mass. The K$_{miss}$ discussion in previous manuscript aims to figure out why discrepancy presents in the coastal case only. This discussion remains in the revised updated manuscript. The changes were shown below:

"We next explored the dependence of k_{miss} on different trace gases. Figure 9a shows the correlation between k_{miss} and NO concentration for the nine case days (including 10 October) between 09:00 and 15:00. At NO > 0.5 ppb, k$_{miss}$ is close to zero. At NO < 0.5 ppb, k$_{miss}$ tended to increase with decreasing NO. Similarly, k$_{miss}$ approached zero at high concentrations of NO$_2$ (> 2.5 ppb), TEXs (> 0.25 ppb), and AVOCs (> 5 ppb; Figure 9) and increased with decreasing concentrations of NO$_2$, TEXs, and AVOCs. High k$_{miss}$ also typically occurred at low toluene/benzene ratios and low C$_2$H$_2$/CO ratios (Figure 9), which are indicators of an aged air mass (Xiao et al., 2007; Kuyper et al., 2020).

Therefore, while we cannot completely rule out other possibilities, we argue that the aged coastal air masses could have contained unmeasured species such as oxygenated organic molecules (OOMs; Nie et al., 2022) and ocean-emitted gases (Thames et al., 2020) that contributed to the missing OH reactivity, causing the model to overestimate OH concentrations on the coastal case days. "

How to prove missing OH reactivity for the coastal air masses. On the other hand, discussion on the other possibility of OH model-measurement discrepancy is missing. For example, the OH measurement interference, if existed, could be proportional to the ambient NO level, which could lead to a more significant biased in the continental air. In this case, the model could consistently overpredict OH concentration for both air masses.

Response: Thanks for the comments. As discussed in our response to the general comment, we don't think the positive bias in the inlet due to reaction $NO + HO_2$ would be significant.

As the campaign was conducted at a coastal site, the role of halogen chemistry was not mentioned. It's not clear if the FOAM model contains halogen chemistry. But the original MCM only contains simple Cl reactions with alkanes. It's suggested to include other key halogen chemistry in the box model. Or if the halogen chemistry is not important, more explanation is needed.

Response: We did not include halogen chemistry in our model as we wanted to compare our results with previous modelling work, most of which did not consider halogen chemistry. Our other studies at the same site that did consider the halogen chemistry show a 4% increase in OH concentration from Cl chemistry (Peng et al., 2022) and 2.8% from Br chemistry (Xia et al., 2022), which would even increase the model-measurement discrepancy in the coastal air.

The discussion and citation were added in the updated version as shown below:

"We did not include halogen chemistry in our study as we wanted to compare our results with previous modelling work most of which did not consider halogen chemistry. Our other studies at the same site that did consider the halogen chemistry show a 4% increase in OH concentration from Cl chemistry (Peng et al., 2022) and 2.8% from Br chemistry (Xia et al., 2022), which would even increase the model-measurement discrepancy in the coastal air mass."

Technical comments:

Introduction: It's not clear why isoprene chemistry was discussed here.

Response: The isoprene chemistry partially explains the missing sources of OH in the low NO-high VOCs forest areas in previous observation done by LIF, this we think it is relevant to have a discussion to complete the "map of OH discrepancy".

Line 4-6 Page 10: It's strange to denote the maximum of different compounds while some of these are clearly anticorrelated (e.g. O3 and NO2).

Response: the purpose of the denotation of the maximum of different compounds is to provide the exact maximum value to the reader instead of describing the anticorrelation.

The revised contents are shown as below:

"As a primary source of OH, HONO peaked in the morning at 0.21 ± 0.09 ppb around 7:00 local time (LT), and O_3 peaked in the afternoon at 70 ± 20 ppb at around 16:00 LT."

Line 11 Page 10: (4.0 ± 2.1)

Response: We believed this should be "4.9 ± 2.1" instead of "4.0".

Line 23 Page 13: "fake" suggested to be artificial loss

Response: Thanks for the correction on terminology. We changed "a fake" to "an artificial loss"

Reference:

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