Response to reviewers' comments on the paper "Field observational constraints on the controllers in glyoxal (CHOCHO) loss to aerosol"

We sincerely thank both reviewers for their time and valuable comments, which have helped us improve and clarify our paper. In this response, comments from reviewers are in black, our responses in blue, and new text added in **bold blue** with original text in **bold green**, respectively.

Reviewer #1

This paper uses glyoxal measurements from the KORUS-AQ aircraft campaign over S Korea to show evidence for glyoxal uptake by aerosols with reactive uptake coefficient ~0.01 and accounting for ~20% of organic aerosol formation. The authors further find that aromatics are the dominant source of glyoxal, thus representing an important anthropogenic SOA formation pathway. I found the analysis to be carefully done and the results above to be of fundamental importance to support the hypothesis of glyoxal as a significant SOA precursor. The paper is overall very well written. I strongly support publication in ACP. My only significant concern is that the authors are in my opinion over-interpreting their data to reach conclusions that are featured prominently in the abstract but are not based on sound reasoning. I urge the authors to revise or delete these components of the paper, or at a minimum to address my objections in the text.

Specific comments:

1. Abstract, line 28: I suggest "is an important POTENTIAL precursor…"

R.1.1. We appreciate thoughtful word choice and we have updated line 28 to say:

"Glyoxal (CHOCHO), the simplest dicarbonyl in the troposphere, is a potential precursor for secondary organic aerosol (SOA) and brown carbon (BrC) affecting air-quality and climate."

2. Abstract, lines 32-33: I didn't find in the text a statement or reference that the glyoxal yields from aromatics are "relatively well constrained'. In fact, different aromatic oxidation mechanisms disagree in their glyoxal yields and the MCM used here overestimates the yields in chamber data (see Bates et al., ACPD https://doi.org/10.5194/acp-2021-605, Figure 5).

R1.2. To avoid misunderstanding on our intention, we remove the phrase in the abstract as,

"The production of this molecule was mainly from oxidation of aromatics (59%) initiated by hydroxyl radical (OH)."

In addition, we add a sentence in the main text to clarify the possible discrepancies of glyoxal yields from aromatics among mechanisms, models, and chamber studies under extremely high NOx conditions. As reviewer #1 pointed out, glyoxal yield from MCM oxidation mechanisms of aromatic compounds (i.e., toluene) shown in Figure R1 (Figure 5 in Bates et al., 2021) shows higher yield compared to chamber data at very high NO_x conditions (e.g., > 200 ppb). However, in the same figure, MCM shows reasonable agreement with chamber results for toluene and benzene at more KORUS-AQ relevant NO_x ranges (e.g., 0.1-100 ppb). On the other hand, glyoxal yield from isoprene is more variable. For example, Li et al. (2016) showed ~ 10 times higher yield in their updated isoprene mechanisms in AM3 model than MCM v3.3.1 mainly due to the difference in glyoxal yield from isomerization of isoprene peroxy radicals ($ISOPO_2$) and the $ISOPO_2$ + HO_2 reaction. In addition, time and NOx-dependent glyoxal yield from isoprene by modified GEOS-Chem shows a very different distribution pattern compared to MCM v3.3.1, while formaldehyde yield is similar (Fig. 2 in Chan Miller et al., 2017).

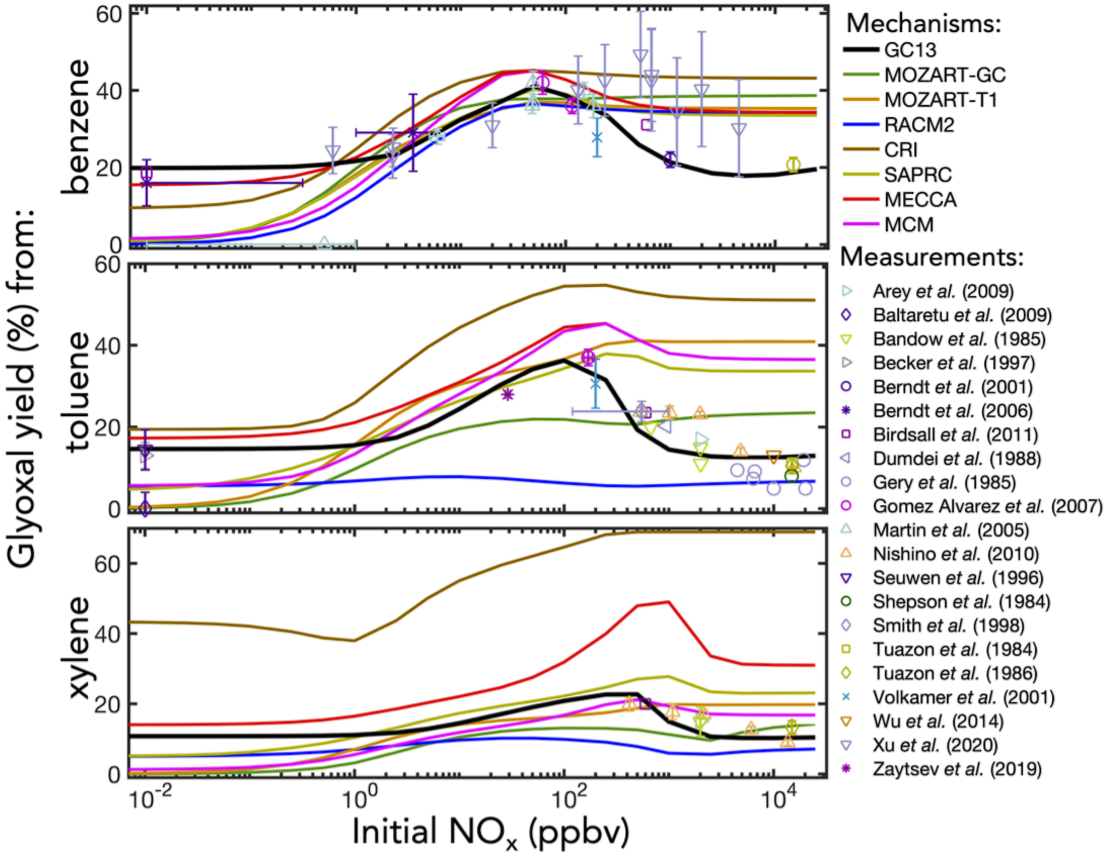

Figure R1. Experimental (points) and simulated (lines) prompt molar yields of glyoxal from BTX oxidation in environmental chambers as a function of the initial NOx mixing ratio (adopted from Bates et al., 2021).

The main text line 265 has been updated as below:

"Even with insufficient understandings on detailed oxidation mechanisms to from CHOCHO, especially under extremely high NO_x condition, CHOCHO yields from aromatic oxidations in MCM v3.3.1 aromatics oxidation mechanisms are reasonably well constrained in the NO_x range relevant to KORUS-AQ campaign (Bates et al., 2021)."

3. Abstract, lines 36-42: in my opinion these findings are based on unsound interpretation of the data. See comments below for specifics.

R.1.3. Please note our replies to the reviewer as described in the individual comments below.

4. Abstract: missing from the Abstract is the glyoxal reactive uptake coefficient inferred from the data, which I think is a robust result and will be an important reason for citing this paper.

R.1.4. We thank the point and have added line 35 to say:

“Reactive uptake coefficient (γ) of ~ 0.008 best represents the loss of CHOCHO by surface uptake during the campaign.”

Also, in order to further highlight the importance of uptake coefficient, we have revised the title in the main manuscript and supplement as below:

“Field observational constraints on the controllers in glyoxal (CHOCHO) reactive uptake to aerosol”

5. Line 127: the authors focus on the boundary layer below 2 km and that makes sense, but they should comment on their free tropospheric glyoxal data reported in the KORUS-AQ database that are surprisingly high with relatively little variability, and imply a background source of glyoxal missing from the models. The same problem was found over the US during SENEX (with the same instrument) by Chan Miller et al. 2017 cited here. I think that the authors need to comment on this. Are their measurements reported in the free troposphere incorrect? It would be very useful (and a relief!) for the community to know this. If they are correct, where does this background glyoxal come from? How would it contribute to boundary layer glyoxal?

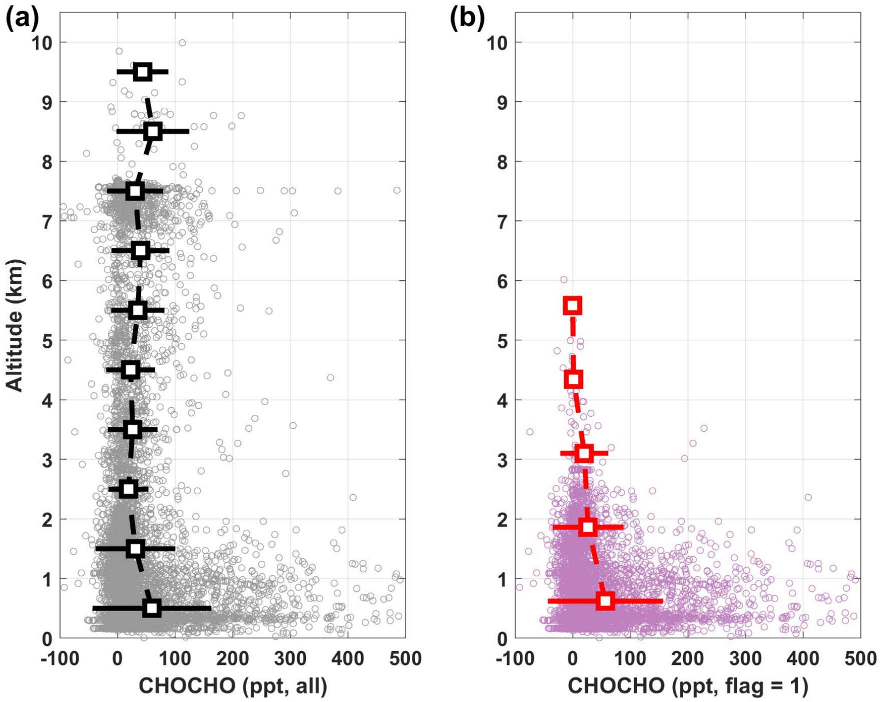

Figure R3. Altitude profiles of (a) CHOCHO with entire KORUS-AQ data including both flag = 0 and 1 (b) CHOCHO with flag = 1 which is finalized data. CHOCHO flag = 0 indicates data that are not suitable for further analysis due to high errors in fitting, possibly not only due to numerical fitting errors with low signal but also the issue related to H_2O fitting and sudden pressure change. The markers and horizontal ranges indicate mean values and ±1 σ variability, respectively.

R.1.5. The presence of glyoxal in the free troposphere is indeed a very interesting topic. Glyoxal has been observed in the free troposphere over the SouthEastern U.S. (Lee et al., 1998; Li et al., 2016; Chan Miller et al., 2017), Los Angeles (Baidar et al., 2013), and the tropical Eastern Pacific Ocean (Volkamer et al., 2015). While the mechanistic understanding of the presence of glyoxal at such high altitudes is still elusive, non-traditional pathways towards glyoxal formation have been shown to proceed from fatty acids in the remote MBL (Coburn et al., 2014; Chiu et al., 2017). The fact that multiple studies with different techniques (HPLC, CEAS, DOAS) observed glyoxal in the free troposphere indicates that the observations shown in Chan Miller et al. (2017) and other earlier studies are not necessarily incorrect.

However, we do not want to over-interpret the data close to the detection limit especially due to the inherent limitations in retrievals; low signals tend to have larger errors in fitting, thus we marked the data with higher fitting errors (> 2 times) than fitted concentration as flag 0. We also would like to point out that the error may also arise from the delay in pressure sensor measurements under the condition of sudden pressure change like spiral ups and downs. We speculate that reviewer #1 has looked at the entire KORUS-AQ data including the data with CHOCHO flag = 0 (Fig.R3a). The data with flag = 0 is not recommended for use or requires discussion with Prof. Kyung-Eun Min (kemin@gist.ac.kr) before use.

On the other hand, when plotting with finalized data only (CHOCHO flag =1), glyoxal concentration goes down to zero above 4 km altitude, although the number of data points is not as abundant as that of under the boundary layer (Fig. R3b). Thus, mentioning the glyoxal abundance in the free troposphere is not adequate in this manuscript, as we can not certainly comment on the background glyoxal concentration during the KORUS-AQ campaign.

6. Line 203: I think you mean equilibrium, not steady state.

R.1.6. Correct and we updated as :

"For simulating volume-controlled processes through aqueous-phase reactions, it was assumed that gas-phase and dissolved-phase CHOCHO (CHOCHO monomer, hydrates, and oligomers) are in equilibrium (more details in section S4 and Fig. S6 in SI)."

7. Lines 246-250: I don't see differences in patterns between glyoxal, formaldehyde, and CO in Figure 1.

R.1.7. The difference in patterns between glyoxal and formaldehyde is most noticeable over the West Coastal Industrial Area (WCIA) as marked in Figure 1a, while formaldehyde and CO show a much similar pattern over the area which makes sense because CO is a good proxy for anthropogenic emission. Unlike formaldehyde, the distribution of glyoxal is more complex as the loss rate depends heavily on ambient aerosol mass loading which also depends on meteorological transportations and photochemical processes (Fig. 6b).

We have added line 244 to say:

"Unlike formaldehyde, distribution of glyoxal is more complex as the loss rate heavily depends on ambient aerosol mass loading which also depends not only on meteorological but also on chemical processes (Fig. 6b)."

And we have revised line 250 to say:

"Meanwhile, aromatic distributions have more similarities with CHOCHO, such as benzene (Fig.1e, in WCIA), toluene (Fig. 1f, in SMA) xylene, and trimethylbenzenes near SMA (Fig. S7)."

8. Line 265: there should be some comment on glyoxal yields from the important VOC precursors. See comment on Abstract.

R.1.8. We have revised lines 285-286 to say:

"Roughly 59 % of CHOCHO in the model originates from aromatics, with the largest contribution from toluene (41 %, Fig. 3a) as supported by previous works with high glyoxal yields (10-40%) from aromatic compounds (i.e., benzene, toluene, p-xylene, and ethylbenzene) under high NO_x conditions (Volkamer et al., 2001, 2007; Bates et al., 2021)."

9. Lines 275-279: this 'test' seems circular?

R.1.9. This test was to plug in the $k_{\mathrm{eff,uptake}}$ that is analytically calculated from the CHOCHO production rate subtracted by CHOCHO loss rate by OH and photolysis, under the assumption of steady-state. The $k_{\mathrm{eff,uptake}}$ was used to estimate γ, $K_{h,eff}$, and glyoxal SOA contribution to the total SOA over Seoul. In the test, we plugged the calculated $k_{\mathrm{eff,uptake}}$ into the box model and ran for 24 hours for each data point, evolving solar zenith angle, and thus evolving loss rate of CHOCHO by photolysis, OH, and evolving production rate from VOCs oxidation. In that sense, this test is circular but still provides useful information. The CHOCHO concentration result of the model run shows good agreement with measured CHOCHO concentration (Fig 2d). This shows that primary glyoxal was insignificant in ambient air. Also, during the 24 hr box model run, if the net glyoxal evolution rate was far from zero, modeled glyoxal and measured glyoxal would have shown larger differences. Thus, this test provides confidence on the initial assumption that glyoxal is in a near steady-state over the course of diurnal evolution.

We have added line 279 to clarify the role of the test as below:

"Also, the agreement between observation and box model indicates that glyoxal is in near steady-state during the 24-hour diurnal evolution."

10. Lines 317-325: I am not convinced at all about the dependence on JNO2. The effect is small and there could be any number of confounding factors.

R.1.10. Light dependence on glyoxal uptake has been shown from previous laboratory study (Volkamer et al., 2009) comparing glyoxal uptake during dark vs irradiated conditions. Unlike laboratory studies, we admit the fact that our setting has a limitation to fully address the significance of irradiation since our measurement happened in the time window of 8:00 AM to 5:00 PM (local time) where sunlight is always present and the intensity changes. Still, the median $k_{\mathrm{eff,uptake}}$ is consistently higher at more irradiated conditions where j_{NO2} is higher than the median j_{NO2} of the campaign, over the wide range of particle surface area (Fig. 4b). When comparing both cases in Regime II, median $k_{\mathrm{eff,uptake}}$ in more irradiated conditions shows ~ 40 % higher value, although interquartile ranges are broad. However, we agree that other factors may have been involved, contributing to the distinction of $k_{\mathrm{eff,uptake}}$ at high/low j_{NO2}.

In order to include other possibilities, we have added line 321 as below:

"However, note that the interquartile ranges are broad especially at high j_{NO2} and, thus, we can not rule out possibilities that other unaccounted factors contribute to the distinction of high/low j_{NO2}."

Also, we have revised the interquartile range from lines to shade to better visualize the trend of uptake rate coefficient, as well as the description in Figure 4 as below:

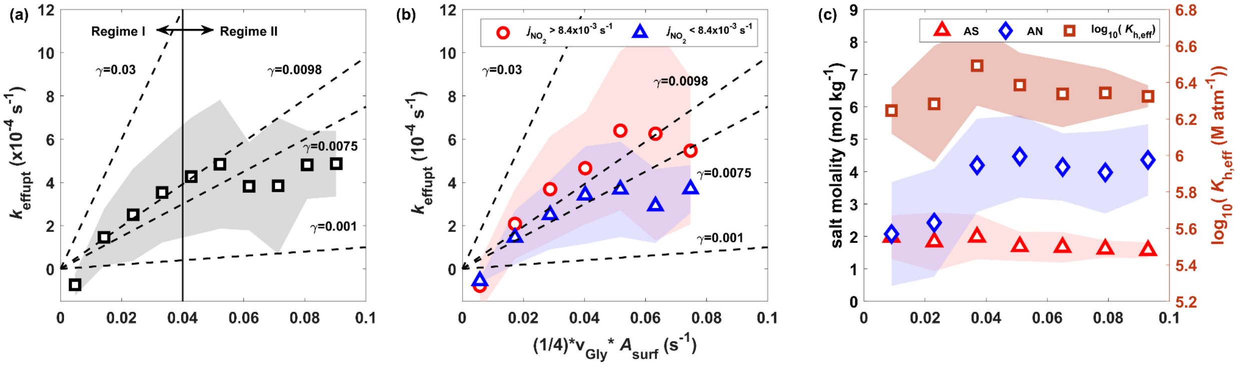

Figure 4. ($1/4*v_{GLY}*A_{surf}$) dependence of (a) $k_{eff,uptake}$, (b) that of high (red) and low (blue) j_{NO2} than 8.6×10^{-3} s^{-1} (median) condition and (c) ammonium sulfate (AS) and ammonium nitrate (AN) molality together with effective Henry's law constant, K_{H,eff_Gly}, parameterized with AS and AN molalities. Black dashed lines in (a) and (b) are few examples of uptake coefficient, γ. Shaded areas represent interquartile ranges.

11. Lines 326-331: I am also not convinced at all about the argument for the flattening of keff as being due to higher aerosol viscosity due to high CAN. That seems really far-fetched. Again, there could be any number of confounding factors.

R.1.11. We agree that there can be many other potential factors that contribute to the flattening of $k_{eff,uptake}$. Although our study does not provide direct evidence of the slowdown of glyoxal uptake rate due to the AN molality, our intention was to provide a potential postulation to explain the flattening based on the insights from previous research (Kampf et al., 2013) which showed that the time scale of equilibrium between the gas-phase CHOCHO and aqueous-phase CHOCHO decrease at high sulfate molality of aerosol seed (called kinetic limitation). It could be attributed to insufficient "free water" for hydration of glyoxal by hydrogen bond at higher molality of inorganics because more water is bound to inorganic ions. And the distinction between "free water" and "bound water" is not well constrained by thermodynamic models. Thus, more laboratory work is needed to elucidate the role of AN on glyoxal uptake kinetics. Please note that we are suggesting a possibility and highlighting the need for more research rather than concluding that AN molality is responsible for the flattening.

In order to clarify, we have revised lines 323-331 to say:

"An increase in salt molality may lead to limited available "free water" and/or more viscous aerosol, retarding mass transfer into the aerosol. Kampf et al. (2013) previously observed slower CHOCHO mass transfer for C_{AS} larger than 12 *m* condition (kinetic limitation) and suggested that, at high salt concentration, fewer water molecules are available for hydration and/or particle viscosity increases leading to slower kinetics. Similarly to this phenomenon, we postulate the possibility that the higher C_{AN} in Regime II may slow down the kinetics, thus, more studies on C_{AN} effect on the time scale of CHOCHO mass transfer into particle are desired."

In addition, we also updated some of the relevant expression in the abstract as:

"Dependence on photochemical impact, and changes in the chemical and physical aerosol properties "free water", as well as changes in aerosol viscosity, are discussed as other possible factors influencing the CHOCHO uptake rate."

12. Lines 333-354: that whole subsection is based in my view on flawed interpretation. I don't think that the data provide a distinction between surface- and volume-controlled uptake, because both would show a correlation with surface area. The 'hybrid treatment' is an interesting attempt to explain the glySOA formation mechanistically but it fails miserably. This is important to note but it could be due to any number of reasons.

R.1.12. The uptake of glyoxal is governed by glyoxal solubility (or effective Henry's law coefficient, $K_{h,eff}$) and the time scale of glyoxal equilibrium in the aqueous phase. The surface uptake model is more convenient to use when calculating CHOCHO loss rate, but γ is not often parameterized by the fundamental parameters such as CHOCHO solubility, loss rate in the aqueous phase. Our study shows that ~ 0.01 of γ (when using surface uptake model) and ~ 7×10^{8} M atm^{-1} of $K_{h,eff}$ (when using volume process model) were required to reconcile with a box model which was constrained with measured VOCs and glyoxal.

This study provides a case study where a simple surface uptake model with one fixed γ may fail to describe glyoxal uptake process when aerosol surface area concentration is high (Regime II). We did not mean that a volume process is required at Regime II, as reviewer #1 mentioned as the volume process model also correlates with surface area and thus it does not fully explain the flattening. However, we intentionally introduce it to provide more insights on the parameterization of CHOCHO uptake on the aerosols since it is also clear that the simple surface uptake model fails at Regime II, if one fixed constant γ is assumed. From this exercise, we suggest one possible speculation for the parmeterization regards with AN molality in Regime II (Fig. 4c) as explained in R.1.11.

We also would like to clarify the small role of the volume process in Fig. 5a not necessarily means the failure of the hybrid model since when applying the hybrid method (surface uptake + volume process), we applied $K_{h,eff}$ that is estimated based only on inorganic compounds using the parameters from laboratory studies, and γ was chosen to matches with the box model. For this reason, surface uptake is dominant and the volume process is ~ 2-3 orders of magnitudes slower than surface uptake (Fig. 5a). Thus the large difference between $P_{glySOA,\ hybrid}$ with and without surface uptake indicates that the hybrid method is almost identical to the surface uptake model, rather than the hybrid model fails. More efforts to elucidate the role of organic compounds and AN on $K_{h,eff}$ are needed for better constraints.

For better clarification, we have revised lines 336-338 in the conclusion section to say:

"Calculated P_{glySOA} without surface uptake (volume process only) is 2-3 orders of magnitude slower than that of the steady-state result, suggesting the importance of surface uptake process in Regime I, indicating that volume process by inorganic salts alone is not sufficient to explain the glyoxal uptake rate during the KORUS-AQ, which is consistent with findings in Mexico City (Waxman et al., 2013)"

We have also added line 338:

"By adding surface uptake process, using the median $\gamma_{Gly,uptake}$ in Regime I (0.98×10^{-2}), the hybrid method matches within an order of magnitude with steady-state box model analysis. Thus, the hybrid method in Regime I is almost identical to the surface reactive uptake model."

And we have modified lines 339-341:

"In Regime II, reactive surface uptake process with one fixed $\gamma_{Gly,uptake}$ is unlikely due to the lack of linearity between $k_{eff,uptake}$ and A_{surf} (Fig. 4a-b). In order to apply the surface uptake model, parameterization of $\gamma_{Gly,uptake}$ may be needed in Regime II. When the volume process is applied, the inferred P_{glySOA} from constrained K_{H,eff_Gly} with measured C_{AS} and C_{AN} through Eq. (7) without surface uptake process ($P_{glySOA,\ eff.\ photochem}$, Sect. S4) is 2 orders of magnitude lower than $P_{glySOA,\ eff.\ Steady\ State}$ as shown in colored circles in Fig. 5b."

13. Lines 383-390: these conclusions are in my view not supported by the data, as detailed in my comments above.

R.1.13. Please note our replies to the reviewer as described in the individual comments above.

In order to reflect reviewer comments, we have revised lines 383-386 in the conclusion section as below:

"This slower uptake under high aerosol loading condition correlates with an increase in AN molality, and could reflect limited availability of "free water" or changes in aerosol viscosity or other compounding factors that slow down CHOCHO mass transfer to the aqueous phase. More experimental studies are needed to elucidate the controlling factors of glyoxal mass transfer into aerosols."

Reviewer #2

This study is important since it provides field based constraints on glyoxal SOA formation from KORUS-AQ. The results are very informative. I support publication in ACP after following suggested clarifications/analyses:

1. Line 45: What is meant by the phrase "track parent VOC with formaldehyde"?

R.2.1. Both formaldehyde and glyoxal are oVOCs formed by oxidation of numerous VOCs with often different yields and they have relatively similar lifetimes. Due to such ubiquity, there have been attempts to classify ambient VOCs by measuring the glyoxal to formaldehyde ratio (R_{GF}) as oxidations of different types of VOCs produce results in different R_{GF} (Kaiser et al., 2015). This idea has attracted attention due to the fact that both glyoxal and formaldehyde are satellite observable, which can put constraints on global VOCs speciation by satellite observations.

In order to clarify it, we have revised line 48 as below:

"... volatile organic compounds (VOCs) with different yields. Due to this character, there have been many attempts to use CHOCHO to formaldehyde ratio (R_{GF}) for parent VOC speciation ..."

2. Lines 330 and 385: Why does increase it inorganic content (CAN) increase viscosity? I would expect increase of inorganic aerosol to increase aerosol water and thus reduce viscosity

R.2.2. We think that there was a misunderstanding about the abbreviated terminology. In this work, C_{AN} and C_{AS} were defined as molalities of AN and AS not as the absolute quantities of AN and AS (lines 209). Thus, C_{AN} is an intensive property that was calculated with the consideration of water content by aerosol hygroscopicity.

We have corrected a typo from **salt content** to **salt molality**. See R.1.11.

3. Glyoxal SOA is 20% of OA. What is the rest?

R.2.3. During the KORUS-AQ campaign, glyoxal SOA was estimated to account for ~ 20 % of SOA over the SMA. Although other sources of SOA were not investigated in this study, Nault et al. (2018) have shown a comprehensive analysis of the sources of SOA in SMA during KORUS-AQ. According to Nault et al. (2018), local emissions of short-lived aromatic compounds and S/IVOCs were the main precursors to SOA, accounting for 70 % of the calculated SOA.

We have added line 362 to say:

"According to Nault et al. (2018), local emissions of short-lived aromatic compounds and semi-volatility and intermediate-volatility organic compounds were the main precursors to SOA, accounting for 70 % of the calculated SOA."

4. If glyoxal peaks in anthropogenic regions, does it imply glyoxal SOA is mostly associated with polluted regions compared to natural biogenic regions?

R.2.4. Most of the high glyoxal was observed over the megacities (i.e., SMA) and industrial areas (Fig. 1b). During the KORUS-AQ campaign, ~ 20 % glyoxal was formed from biogenic VOCs (isoprene, alpha, and beta-pinene) while ~ 80 % of glyoxal was formed from anthropogenic VOCs (Fig. 3a). However, the flight paths were intentionally designed for anthropogenic sources. Furthermore, isoprene, a biogenic source of glyoxal, has a shorter lifetime compared to anthropogenic glyoxal sources (i.e., aromatics). When isoprene is sampled by aircraft, potentially a large portion of it may have been oxidized already as it reaches the sampling height. Thus, it is possible that the biogenic portion of the glyoxal source was biased low for such reasons.

To clarify this point, we have revised lines 286-287:

"The biogenic portion of glyoxal production is potentially biased low as the DC-8 flight paths focused more on anthropogenic sources and some portion of isoprene can be oxidized as advected to the sampling altitude due to its short lifetime."

5. What is the role of cloud chemistry in glyoxal SOA? One can expect glyoxal SOA formation in clouds, and then as the cloud evaporates it can be part of interstitial aerosols. It seems a modeling of glyoxal SOA in clouds followed by its evaporation to aerosols needs to be considered.

R.2.5. Glyoxal SOA formation by cloud chemistry has been studied from various laboratory studies and it is an important pathway on a global scale. However, we are not able to answer this scientific question in this manuscript not only due to the difference in scope but also due to the insufficient samples to analyze and constrain our model. Although there could be some cases where cloud chemistry is important during the campaign, the intercepts with clouds were rare (according to LARGE cloud marker, including non-cloud intercepts, the potential cloud sampling frequency is less than 3% below 2 km altitude). Thus, the glyoxal SOA process by cloud chemistry is treated as not a major topic in our analysis and results.

We have added line 128 for clarification:

"glySOA formation by cloud chemistry was not included in this work as intercepts with cloud were rare (sampling frequency being less than 3 % below 2 km altitude).

We have revised lines 367-368 to clarify the discussion on CHOCHO wet chemistry:

"Oxalate is a major product from glyoxal reactions in clouds (Tan et al., 2009; Ortiz-Montalvo et al., 2014), but a minor product in wet aerosol (Lim et al., 2013). We find oxalate explains only a minor portion of the glyoxal SOA inferred which is consistent with lower oxalate yields in wet aerosol.

We have revised line 371-372 for clarification:

“From our dataset, in cloud process case, analysis was impossible due to not only low abundance in gas-phase CHOCHO concentration (below detection limit) but also the sparseness of the sample.”

6. If there is a larger source of glyoxal at high altitudes where aerosol surface area is lower, could this be due to direct emissions of glyoxal?

R.2.6. Direct emissions of glyoxal at high altitudes (e.g., airplane exhaust) may be possible, but this type of emission has not been identified to our knowledge. We speculate that the secondary glyoxal formation from the VOCs emitted by aircraft is more likely. Also, VOCs emitted from the surface or fatty acids emitted from the ocean (Chiu et al., 2017) can be advected to high altitudes (especially near the equatorial areas), potentially becoming sources of glyoxal at high altitudes.

Other edits

For clarification, we have revised the following statement at lines 41-42 in the abstract:

"... salting-in effects constrained by inorganic salts only consistent with laboratory findings that show similar high partitioning into water-soluble organics, which urges more …"

Line 76: Changed **"Studies"** to **"works"**

For clarification, we have revised the following statement at lines 383-384 in conclusion:

"We postulate a hypothesis that this slower uptake under high aerosol loading condition is attributed to the increased ..."

For clarification, we have revised the following statement at lines 387-389 in conclusion:

"Finally, our work highlights the limited knowledge in explaining the CHOCHO solubility in real atmospheric circumstances. $K_{\mathrm{H,eff\_Gly}}$ estimated in this study (~ $7\mathrm{x}10^{8}$ M atm^{-1}) was similar to $K_{\mathrm{H,eff\_Gly}}$ of fulvic acid aerosol identified from previous laboratory study (~ $6\mathrm{x}10^{8}$ M atm^{-1}) and was ~ 2 orders of magnitude higher than $K_{\mathrm{H,eff\_Gly}}$ derived from salting-in by AS and AN (~ $3\mathrm{x}10^{6}$ M atm^{-1}). This indicates that inorganic salts were insufficient to account for the solubility of CHOCHO while WSOCs potentially played important roles."

We have revised the following statement at line 797 in Figure 5:

"Figure 5. Comparison of glySOA production rate (P_{glySOA}) between steady-state and hybrid analysis for (a) Regime I with (pink diamonds, $\gamma_{\mathrm{Gly,uptake}}$= 0.0098) and without (green diamonds) surface uptake process, and (b) Regime II with fixed $K_{\mathrm{H,eff\_Gly}}$ (7.0×10^{8} M atm^{-1}, green circles) and calculated $K_{\mathrm{H,eff\_Gly}}$ based on salting-in by AN and AS following Waxman et al. (2015) (colored circles)."

We have corrected the typo in the color scale legend in Fig. 5b and Fig S11: from $K_{\mathrm{H,eff\_Gly}}$ to **$\log_{10}(K_{\mathrm{H,eff\_Gly}})$**.

We have corrected the typo in SI line 34: from **"... to reflect the underestimation the underestimation in LAS volume ..."** to **"... to reflect the underestimation in LAS volume ..."**.

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