

Interactive comment on “Formation and sink of glyoxal and methylglyoxal in a polluted subtropical environment: observation-based photochemical analysis and impact evaluation” by Zhenhao Ling et al.

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Response to Reviewers

Reviewer #1

This study investigating the contribution of GLY and MGLY to SOA based on observation and box modeling. Overall this study is of interesting based on the methods and results. However, improvements are needed for its publication.

Reply: We appreciate the two anonymous reviewers for their constructive criticisms and valuable comments, which were of great help in improving the quality of the manuscript. We have revised the manuscript accordingly and our detailed responses are shown below. All the revision is highlighted in the revised manuscript.

- 1. The study used VOCs observations to drive the model with MCM mechanism, but there is no list of the VOCs. Are the species enough to drive the mechanism? If major species are missing, the chemistry may be messed up.*

Reply: The reviewer’s comment is highly appreciated. In this study, mixing ratios of 53 C₂-C₁₀ non-methane hydrocarbons (NMHCs), including 27 alkanes, 11 alkenes, ethyne and 14 aromatics were measured continuously from 2-8 January, 2017, which detailed information and statistics has been provided in our previous studies (Chang et al., 2019; Yun et al., 2018). By taking the species incorporating in the MCM mechanism into account (MCM website, <http://mcm.leeds.ac.uk/MCM/roots.htm>, access date: 22 June 2020), observations of total 44 VOC species, including 18 alkanes, 11 alkenes, ethyne and 14 aromatics used as input for the model simulation (Table 1). It should be noted that the selected NMHCs contributed about 98% and 99% to the total mixing ratios and photochemical reactivities of all NMHCs measured

at the Heshan site. Furthermore, the selected VOCs, which were frequently used to drive box model for SOA, photochemical O₃ and photochemical reactivity (Hofzumahaus et al., 2009; Lee-Taylor, et al., 2011), were the major precursors for Gly, Mgly, photochemical O₃ and SOA (Ding et al., 2016, 2017; Hofzumahaus et al., 2009; Li et al., 2014; Lou et al., 2010; Yuan et al., 2013). Therefore, we believe that the selected species were enough to drive the PBM-MCM model and simulate the formation of Gly, Mgly and SOA in this study.

Table 1. The NMHC Compounds which driven the PBM-MCM model

No.	Compound	No.	Compound
1	Ethane	23	1-Butene
2	Propane	24	cis-2-Butene
3	i-Butane	25	1-Pentene
4	n-Butane	26	trans-2-Pentene
5	i-Pentane	27	Isoprene
6	n-Pentane	28	cis-2-Pentene
7	2,2-Dimethylbutane	29	1-Hexene
8	2,3-Dimethylbutane	30	1,3-Butadiene
9	2-Methylpentane	31	Benzene
10	3-Methylpentane	32	Toluene
11	n-Hexane	33	Ethylbenzene
12	2-Methylhexane	34	m/p-Xylene
13	Cyclohexane	35	o-Xylene
14	3-Methylhexane	36	Styrene
15	n-Heptane	37	i-Propylbenzene
16	n-Octane	38	n-Propylbenzene
17	n-Nonane	39	m-Ethyltoluene
18	n-Decane	40	p-Ethyltoluene
19	Ethene	41	1,3,5-Trimethylbenzene
20	Propene	42	o-Ethyltoluene
21	Ethyne	43	1,2,4-Trimethylbenzene
22	trans-2-Butene	44	1,2,3-Trimethylbenzene

To clarify the input of NMHC species, the following text has been added in the revised manuscript:

“By taking the NMHC species incorporating in the MCM mechanism into account (MCM website, <http://mcm.leeds.ac.uk/MCM/roots.htm>, access date: 22 June 2020), observations of total 44 NMHC species, including 18 alkanes, 11 alkenes, ethyne and

14 aromatics were used as input for the model simulation (Table S2). The selected NMHCs contributed about 98% and 99% to the total mixing ratios and photochemical reactivities of all measured NMHCs at the Heshan site. Furthermore, the selected VOCs are the major precursors for Gly, Mgly, photochemical O₃ and SOA (Ding et al., 2016, 2017; Li et al., 2014; Lou et al., 2010; Yuan et al., 2013), and have been frequently used to drive box model for studies on SOA, photochemical O₃ and photochemical reactivity (Hofzumahaus et al., 2009; Lee-Taylor, et al., 2011).”

For details, please refer to Lines 360-370, Page 13 in the revised manuscript. In addition, a table (Table S2) has been provided in the supplementary to list the input NMHCs.

2. *The SOA simulation was not validated, thus it is not clear if the contribution of GLY and MGLY are in realistic range. For example, the study may underestimate or overestimate SOA from other species. Also, SOA formation pathways are far from accurate, if corrections such as wall loss are considered in this study? It is important to make sure that what recent findings regarding SOA formation have been considered, as the total SOA prediction would be much different.*

Reply: The reviewer’s valuable comments are highly appreciated. We agreed with the reviewer that validation is important for SOA simulation although the aims of this study were to investigate the formation and sink of Gly and Mgly in the PRD region using an observation-based photochemical box model with the incorporation of improved mechanisms of heterogeneous processes of Gly and Mgly, which would be further used to evaluate their contributions to SOA. As there were no direct SOA measurement data such as AMS data, we compared the model simulated SOA concentrations with those calculated by different methods and parameters, to evaluate the performance of SOA simulation.

First, the EC (elemental carbon)-tracer method was used here to estimate the concentration of SOA in the present study, according to the equations 1-2 (Duan et al., 2005):

$$SOC = OC_{tot} - EC \times (OC / EC)_{min} \quad (1)$$

$$SOA = SOC \times Coef_{SOA/SOC} \quad (2)$$

where SOC represents the secondary organic carbon; OC_{tot} represents the measured concentration of total organic carbon; $(OC/EC)_{min}$ is the emission ratio of primary OC (organic carbon) and EC (elemental carbon) from the sources, and can be represented by the minimum OC/EC ratio measured. Based on the $(OC/EC)_{min}$ value which was derived from the hourly observation data during the simulation period (i.e., January 07-08) using the minimum R squared (MRS) method and the hourly measured OC and EC concentrations (details in Chang et al., 2019), as well as the ratio of $Coef_{SOA/SOC}$ (Bae et al., 2006; Turpin and Lim, 2001), the mean concentration of SOA calculated by the EC-tracer method was $2.82 \mu\text{g}/\text{m}^3$, about 1.2 times the model simulated SOA concentration in the present study ($2.47 \mu\text{g}/\text{m}^3$), suggesting that the PBM-MCM model provided a reasonable performance on the simulation of the magnitude of SOA, though uncertainties were frequently observed from the SOA concentrations calculated by the EC-tracer method.

Second, we also conducted the comparison between measurement and simulation on other secondary products, including acetic acid, formic acid and pyruvic acid, which have been recognized as key SOA species, to further evaluate the model performance (Figure 1 as seen below). It was found that the simulated concentrations of acetic acid, formic acid and pyruvic acid were close to those observed at the Heshan site, accounting for ~80%, 70% and 88% of observed values for acetic acid, formic acid and pyruvic acid, respectively. The results confirmed that secondary formation was the dominant source of above species at the Heshan site, and suggested that the PBM-MCM model could provide robust performance on simulating the abundance of above secondary species. The deviations between simulated and observed concentrations may be related to the lack of consideration of primary emissions and/or other production pathways of above species in the model. Nevertheless, the above comparisons confirmed that the PBM-MCM model in this study indeed provided an appropriate description on the formation of SOA and other secondary organic products.

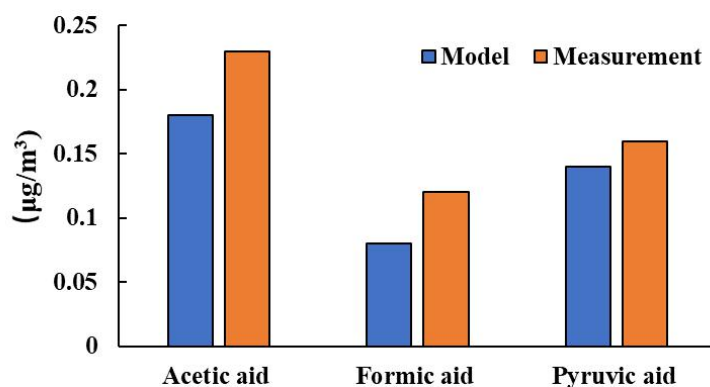


Fig. 1. The concentrations of acetic acid, formic acid and pyruvic acid in filter samples

In the model simulation, SOA formation is generally configured to occur through gas-particle partitioning of low-volatility compounds formed by the gas-phase oxidation of VOCs and other precursors (Aumont, et al., 2012; Lee-Taylor et al., 2011), which have been incorporated in the PBM-MCM model. On the other hand, the recent experimental results suggested that the formation of SOA in chambers may be suppressed due to the losses of SOA onto chamber walls, which leads to underestimation of SOA in air-quality and climate models (Matsunaga and Ziemann 2010; Zhang et al., 2014). We agreed with the reviewer that wall loss of SOA should be considered in the model configuration. Indeed, in this study, the wall loss of SOA has been considered in the model configuration, with the average wall loss rate coefficient of $6 \times 10^{-5} \text{ s}^{-1}$ according to the previous studies on the basis of the calculated organic material using an assumed density of $1 \text{ g}\cdot\text{cm}^{-3}$ (Johnson et al., 2004, 2005). In addition, the wall loss of other gaseous compounds (O_3 , NO_2 and HNO_3) were implemented in the box model with the average parameters of $3 \times 10^{-6} \text{ s}^{-1}$, $1.15 \times 10^{-5} \text{ s}^{-1}$ and $8.2 \times 10^{-5} \text{ s}^{-1}$, respectively. The detailed information for the calculation of above parameters was provided in Bloss et al. (2015).

In addition, the results in the present study can reflect the current knowledge of abundance and evolution of Gly and Mgly, and the contributions of different VOCs to SOA formation in PRD to a certain extent. Since the study of Liggio et al. (2005), there have been many laboratory and model studies that explored and compared the heterogeneous uptake processes of Gly and Mgly on aqueous aerosols. For instance,

the recent studies suggested that salting-in and salting-out effects (*i.e.*, the ammonium-catalyzed reactions) had significant influences on the surface uptake of dicarbonyls (Knote et al., 2014; Kampf et al., 2013; Noziere et al., 2009), and the rate coefficients were found to increase with the increasing ammonium ion activity ($a_{\text{NH}_4^+}$) and pH (Noziere et al., 2009). Those new findings have led to more sophisticated descriptions of Gly and Mgly uptake on aerosols which should be considered accordingly in current model studies (Ervens et al., 2010, 2011). Therefore, in this study, we extended the PBM-MCM (version 3.2) model and included detailed gas-phase chemistry of Gly and Mgly formation, a module describing its partitioning and reactions in the aerosol aqueous-phase, as well as other updated heterogeneous processes, including the irreversible surface uptake process and reversible formation based on recent laboratory studies (Akimoto 2016; Houghton et al., 2017). Based on the above model development, we then compared the different pathways to form SOA from Gly, Mgly and other VOCs, respectively.

In summary, according to the aims of this study mentioned above, our study indeed provided a comprehensive analysis on the abundance and evolution of Gly and Mgly in the PRD region with the incorporation of updated heterogeneous processes and gas-particle partitioning mechanisms, which were further used to investigate contributions of different VOCs to SOA formation. However, we admit that there were still some limitations in the model, e.g., the incorporation of more accurate and precise parameterization of heterogeneous process of Gly and Mgly (such as the surface uptake coefficients), the oxidation of other SOA precursors and complete mechanism of SOA formation, which needs further exploration in the future study (Sumner et al., 2014; Wu et al., 2019; Zhang and Seinfeld, 2013).

The brief discussion on the validation of model simulation has been added in the revised manuscript as follows, while the detailed information of validation has been provided in the supplementary (Section S3).

“.....as there were no direct measured SOA data in this study (Chang et al., 2019), the model performance was evaluated by the comparison between the model

simulated SOA with those calculated using the EC (elemental carbon)-tracer method, and by the comparison between the simulated and observed concentrations of other secondary products, which have been provided in detail in the supplementary (Section 3). For example, the simulated concentration of SOA was about 85% of those calculated by the EC-tracer method based on the observed hourly data (Chang et al., 2019). Furthermore, the simulated concentrations of acetic acid, formic acid and pyruvic acid were close to those observed at the Heshan site, accounting for ~80%, 70% and 88% of observed values for acetic acid, formic acid and pyruvic acid, respectively. The results confirmed that secondary formation was the dominant source of above species at the Heshan site, and suggested that the PBM-MCM model could provide robust performance on simulating the abundance of above secondary species and SOA.”

For details, please refer to Lines 404-417, Pages 14-15 in the revised manuscript and Section 3 in the supplementary.

In addition, to clarify the consideration of the influence of wall loss on SOA formation, the following text has been added in the revised manuscript:

“The above gas-particle partitioning of low volatility compounds formed by the gas-phase oxidation of VOCs and other precursors (Aumont, et al., 2012; Lee-Taylor et al., 2011) was configured in the model to estimate the SOA formation. However, the recent experimental results suggested that the formation of SOA in laboratory chambers may be suppressed due to losses of SOA to chamber walls, which leads to underestimates of SOA in air-quality and climate models (Matsunaga and Ziemann 2010; Zhang et al., 2014). Therefore, to consider the wall loss of SOA, the average wall loss rate coefficient of $6 \times 10^{-5} \text{ s}^{-1}$ was adopted in the model configuration according to previous studies on the basis of calculated organic material using an assumed density of $1 \text{ g} \cdot \text{cm}^{-3}$ (Johnson et al., 2004, 2005). In addition, the wall loss of other gaseous compounds (O_3 , NO_2 and HNO_3) were implemented in the box model with the average parameters of $3 \times 10^{-6} \text{ s}^{-1}$, $1.15 \times 10^{-5} \text{ s}^{-1}$ and $8.2 \times 10^{-5} \text{ s}^{-1}$,

respectively. The detailed information for the calculation of above parameters was provided in Bloss et al. (2015).”

For details, please refer to Lines 242-254, Pages 8-9 in the revised manuscript.

3. *The basis for heterogeneous reactions are the comparison of gas phase of GLY and MGLY concentrations, which may cause uncertainties. Without validating the components formed by GLY and MGLY in SOA, it is misleading that GLY and MGLY are actually converted to SOA.*

Reply: Thanks a lot for the reviewer’s comment. We agree with the reviewer that there were possible uncertainties by only comparing the gas phase change of Gly and Mgly concentrations. As the measurement data of SOA and its speciation was not available in our field campaign, we could not directly compare the simulated and observed products formed by Gly and Mgly in SOA. As responded in above comment #2, the model simulated SOA concentration and some SOA components were compared with the measurement or measurement derived values, which show good consistency and suggest acceptable performance of SOA by the PBM-MCM model.

Besides, by comparing with previous studies conducted in different regions in China, particularly those simulation studies of Gly/Mgly in the PRD region (Li et al., 2013, 2014), our study indeed provided a better reproduction of abundance and variations of Gly and Mgly at the Heshan site by adapting the gas-phase chemistry and the updated heterogeneous processes of Gly and Mgly (e.g., irreversible/reversible processes including the reversible partitioning, irreversible volume reactions, and surface uptake). Moreover, we investigated the difference in SOA production with and without the influence of Gly and Mgly in order to evaluate that whether that Gly and Mgly were actually converted to SOA. It was found that the SOA production has been improved apparently from 1.83 to 2.47 $\mu\text{g}\cdot\text{m}^{-3}$ when considering the influence of Gly and Mgly. The total contribution of the Gly and Mgly contributed ~26% to the simulated SOA concentration, of which ~21% was from the heterogeneous processes of Gly and Mgly (SOA_{het}), further demonstrating that the heterogeneous processes have significant influences on the SOA formation of Gly and Mgly.

Furthermore, it should be noted that gas phase Gly and Mgly could be removed by reactions with OH and NO₃ radicals, photolysis and deposition (Sander et al., 2006; Volkamer., et al 2005). However, recent studies found that by comparing the removal of Gly and Mgly through OH, NO₃ reactions, heterogeneous processes were the most important pathway for the destruction of Gly and Mgly (Shi, et al., 2020; De Haan, et al., 2018), and the lifetime of Gly and Mgly was reduced by a factor of two when the heterogeneous uptake processes were considered (Ervens and Volkamer 2010). This is consistent with the results of the present study, which suggested that heterogeneous processes were the most important pathway for the destruction of Gly and Mgly (both with contributions of ~62% during daytime), much higher than the removal of Gly and Mgly through other pathways.

Therefore, by the improvement of SOA production with the consideration of Gly and Mgly and the good performance of the simulation of Gly, Mgly, SOA and other secondary products by the PBM-MCM model, it could be concluded that gaseous Gly and Mgly at Heshan were actually converted to SOA.

To highlight the contributions of Gly and Mgly to the SOA formation, the following text has been added in the manuscript:

“The SOA production from Gly and Mgly was further explored by the model simulation with and without the consideration of Gly and Mgly. It was found that by incorporating the evolution of Gly and Mgly, the SOA production has been improved apparently from 1.83 to 2.47 $\mu\text{g}\cdot\text{m}^{-3}$. The total contribution of the Gly and Mgly contributed ~26% to the simulated SOA concentrations, of which ~21% was from the heterogeneous processes of Gly and Mgly (SOA_{het}), further demonstrating that the heterogeneous processes have significant influences on the SOA formation from Gly and Mgly.”

For details, please refer to Lines 602-609, Pages 22 in the revised manuscript.

- 4. When considering the budget, it is import to considering transport. It is suggested to exclude the effects of transport in/out or note the readers regarding the uncertainties.*

Reply: Thanks a lot for the reviewer's comment. Due to the limitation of the PBM-MCM model and/or other box models (Aumont, et al., 2012; Lam et al., 2013; Lee-Taylor, et al., 2011), the influence of transport on air pollutants could not be estimated, though the simulation was conducted on the basis of observed mixing ratios of air pollutants that could be both influenced by local emissions and those transported from upwind areas (Liu et al., 2019).

To clarify the uncertainty associated with the lack of consideration of transport in the model, the following text has been added:

“Similar to other box models in simulating the degradation of VOCs and formation of SOA (Aumont, et al., 2011; Lee-Taylor, et al., 2011; Zhang and Seinfeld, 2013), the PBM-MCM model was developed by assuming a well-mixed box without consideration of vertical and horizontal transport, and air pollutants were assumed to be homogeneous (Lam et al., 2013; Ling et al., 2014). Thus, the influence of horizontal and vertical transport on air pollutants was not considered in this study.”

For details, please refer to Lines 204-210, Page 7 in the revised manuscript.

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