

Interactive comment on “Assessment of dicarbonyl contributions to secondary organic aerosols over China using RAMS-CMAQ” by Jialin Li et al.

Anonymous Referee #1

Received and published: 8 December 2018

SOA is an important but the least understood component of atmospheric aerosols. Generally, the existing models still underestimate the observed organic aerosol concentrations. The SOA formation through aqueous reactions of dicarbonyls is one of potential candidates that can reduce the model-observation gap and thus has attracted many attentions in recent SOA studies. Li et al. 2018 manuscript attempts to implement the aqueous uptake of dicarbonyls in the regional atmospheric chemistry model RAMS-CMAQ and further to estimate the contribution of the uptake of dicarbonyls to the SOA formation in China. The manuscript also tries to quantify the effect of the underestimation of gas-phase glyoxal in the model on the SOA formation. Overall, I think the manuscript would benefit the SOA community after the authors address my

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comments below.

General comments

1. The model accounts for the SOA formation both from the in-cloud oxidation of glyoxal and methylglyoxal and through the aqueous irreversible uptake of glyoxal and methylglyoxal as presented in equation (1). However, the SOA formation pathway through the equation (1) already includes the in-cloud reactions of dicarbonyls. So the model is double-counting the loss of dicarbonyls through the aqueous process and thus the SOA formation from the dicarbonyls.

2. The model does not consider the uptake of dicarbonyls on the aqueous aerosol, but only the uptake on the cloud droplets. Can the authors explain the reason for neglecting the uptake by the aqueous aerosol? Many studies (both lab and model studies) (Lim et al. 2010; Ervens and Volkamer, 2010; Stavrou et al. 2009; Lin et al., 2014) have shown that the SOA formation from the uptake on the aqueous aerosol can be as large as or even larger than that from the cloud processing.

3. The authors only used one station data to evaluate the modeled SOA concentrations. How representative is this station data? Can this single-station data evaluation draw a meaningful and robust conclusion on the model performance?

Specific comments

Line 51-52. The reference list only include the paper previous to 2008. There are many SOA modeling papers published since 2008. The authors should at least include some of them.

Line 53-54. The one-to-two-order-of-magnitude underestimation applies to only the SOA modeling paper before 2007. Recent SOA modeling efforts have closed the gap to some degree, although there are still large uncertainties there (e.g. Tsigaridis et al. 2014). Again, the authors need to clarify this.

Line 70. Again, there are few other recent papers that have made efforts on modeling

aqueous SOA formation. To name a few here, Lin et al. 2012; Lin et al. 2014; Woo and McNeill, 2015.

Line 108-110. How about the lateral boundary conditions?

Line 138. Does the model include the uptake by both aqueous aerosols and cloud droplets? Because according to the the result section, the model seems to include the uptake by the cloud droplets only, excluding the aqueous aerosol uptake?

Line 150. The assumption of the same value of gamma glyoxal and methylglyoxal is not well-grounded. First, Liggio et al. (2005) only gave the value of glyoxal rather than the value of methylglyoxal. Second, the value of glyoxal was derived from the uptake on aqueous aerosols, not on the cloud water. Third, the glyoxal and methylglyoxal uptake by cloud water correlates with their water solubility. So I think scaling of methylglyoxal to that of glyoxal by the ratio of their effective Henry's law constants is a more reasonable assumption.

Line 172-173. Which two episodes?

Section 3.1 Model evaluation. Can the authors compare to surface cloud water content measurements, instead of the vertically integrated water content (CWP)? Because the surface cloud water content is more relevant to the surface SOA concentrations than the CWP.

Line 249-250. The underestimation of aromatics concentrations doesn't necessarily mean that the model underestimates the aromatic emission. The alternate reason could be the chemical consumption rates of aromatics are too fast in the model.

Line 332-334. Why do the authors neglect the aqueous SOA formation in the aqueous aerosols?

Line 338. Distinguish the contribution. . . from what? Please clarify this.

Fig.4. The red line in (a) is different from the one in (c) and (e). So the red line in

different panels represent different case result? Really confusing.

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Interactive comment on *Atmos. Chem. Phys. Discuss.*, <https://doi.org/10.5194/acp-2018-1106>, 2018.

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