

Reviewer comments are copied below. Our responses are written below each comment in blue font.

REVIEWER 2

This technical note describes the calculation of uptake coefficients for glyoxal and methylglyoxal based on measured values for Henry's Law coefficients as a function of various salt concentrations and on modeled values for OH radical concentrations in cloudwater and aqueous aerosol. These uptake coefficients are sorely needed, and this note is sure to be of value to the field. The results and methods are clearly described and presented.

We thank the reviewer for this overall positive assessment.

The authors at one point compare their calculated uptake coefficients for glyoxal on SNA aerosol at 50% RH to laboratory measurements on ammonium sulfate aerosol at 55% RH by Liggio et al.(1). The new, calculated results are high by a factor of three, which seems like reasonable agreement in this field.

We actually had not done the comparison mentioned by the reviewer, and it is an interesting suggestion. Since the dependence on relative humidity is strong near 50% RH (see Figure 1 top panel), if you use our SNA parameterization for glyoxal from Table 4 and plug in 55% RH, the result is $\gamma_{\text{GLYX}} = 3.6 \times 10^{-3}$, which is only 24% higher than the experimental value of Liggio et al. We now mention this in the Atmospheric Implications section: "The values we calculated under many conditions are lower than those currently used in large-scale models such as GEOS-Chem. However, we note that the parameterization presented in Table 4 at the experimental conditions of Liggio et al. (2005), 55% RH, yields $\gamma_{\text{GLY}} = 3.6 \times 10^{-3}$, which is within 24% of their experimental value (2.9×10^{-3})."

However, values used for certain parameters are not given, and I have two concerns about the scope of the conclusions.

Specific Comments

This study appears to take into account only one kind of irreversible reactivity: oxidation by dissolved OH radicals. Can the authors justify that this reaction is more important than all other irreversible aqueous-phase reactions involving dissolved dicarbonyls, such as organosulfate formation, or the non-radical reactions with ammonium sulfate that the authors have studied in the past? I think it is unlikely that using effective Henry's law coefficients, even ones that include salting in / salting out effects, accounts for all of these processes, and the authors allude to this problem in the final paragraph. Given this problem, could the authors be getting reasonably accurate results for glyoxal uptake for the wrong reasons (due to a second error pushing the results in the opposite direction of the first)? It would be helpful to discuss this limitation and the magnitude of the uncertainties more thoroughly to help readers better interpret the results.

We chose to focus on reactive uptake driven by OH oxidation since this is the dominant irreversible loss process for GLYX and MGLY in aqueous aerosols and cloud droplets. This reaction is the initiation step for most radical-based chemistry of GLYX and MGLY in the aqueous phase, including organic acid formation and organosulfate formation (McNeill et al., 2012; Perri et al., 2010). Other irreversible loss processes such as imidazole formation occur at much longer timescales (Teich et al., 2016; Yu et al., 2011). The consistency of our calculations with the experimental reactive uptake coefficient of Liggio et al.

al., a system in which the multiple aqueous-phase processes mentioned here were active, provides support for our approach. We have expanded our discussion of this matter in the text.

Finally, as we note in the final paragraph of the manuscript, “This representation of aqueous SOA formation by GLYX and MGLY, with the treatment of Henry’s constants described here, does not take into account the contribution of reversible uptake of GLYX, which could be a significant, although transient, source of aerosol mass under some conditions (McNeill et al., 2012; Woo and McNeill, 2015). The use of this parameterization together with simpleGAMMA (Woo and McNeill, 2015) would give representation of both aqSOA formation types by GLYX.”

Second, the authors have chosen to ignore the effects of sulfate / nitrate ratios, sulfate / ammonium ratios, and pH on glyoxal uptake coefficients and focus exclusively on the effects of relative humidity. Based on Figure S1, I acknowledge that RH appears to be more important than these other three factors. However, Figure S1 shows that sulfate / nitrate ratios, sulfate / ammonium ratios, and pH all have non-linear effects on glyoxal uptake that are as large as the effects of increasing the RH from 70 to 99%. In addition, the laboratory experiments of Liggio et al.¹ showed that glyoxal uptake coefficients depend on aerosol acidity. Just because these effects are non-linear does not mean that they can or should be ignored. In the manuscript, the single statement on p. 5 line 18 that discusses Figure S1 (“no clear correlation is apparent”) is at best an oversimplification, and might even be seen as misleading.

We agree with the reviewer that more discussion of the dependence of γ_{GLYX} and γ_{MGLY} on pH, S:N, and S:A is required. In this study, variation in aerosol pH was not controlled independently but rather it developed from the variation in S:N and S:A. Since the correlation with S:N and S:A was not strong, and aerosol pH is not a variable in GEOS-Chem, we chose not to address this dependence in the original manuscript. However, since we see the value of providing this information despite its lack of direct applicability to GEOS-Chem simulations, and we agree with the reviewer that not doing so may lead to misunderstanding, we now discuss this data in more detail and include parameterizations for γ_{GLYX} and γ_{MGLY} as functions of pH.

The dependence of γ_{GLYX} and γ_{MGLY} on S:N and S:A is, as the reviewer noted, weak compared to the dependence on RH and pH. Furthermore, the plots in Figure S1 show scatter and a lack of monotonicity in the dependence that suggest a lack of a mechanistic basis for the variation observed. As a result, we choose not to include these variables in the parameterization. The variation as a function of S:N and S:A is included in the error bars on the RH fits. We now discuss this in more detail in section 3.

Technical comments

While the reported parameter values seem reasonable, I was unable to find the values used for two key parameters: the accommodation coefficient (α) and the aqueous diffusion coefficients. Are the terms that include these parameters not very influential on the overall values calculated for the uptake coefficients?

Thank you for catching this omission, we have now included this information in section 2. The aqueous-phase diffusion coefficient used for glyoxal was $D_{\text{aq}} = 10^{-9} \text{ m}^2/\text{s}$ and the accommodation coefficient used was $\alpha = 0.02$. D_{aq} does not vary much for small species, and this value is typical for small organics (Bird et al., 2006). This value of α is an estimate based assuming that α for GLY and MGLY are similar to that

of formaldehyde uptake to water (Jayne et al., 1992). As the reviewer suggests, the calculation is insensitive to within 10% for a 50% variation in α for values of $\gamma < 10^{-3}$.

Abstract (line 12): I think that the statement “We take into account . . . aqueous-phase chemical kinetics” should be modified given the first concern described above. Only the chemical kinetics of oxidation reactions with OH are taken into account in this study, not the chemical kinetics of other irreversible reactions.

In response to this suggestion, we have modified this sentence to be more specific and accurate, replacing “aqueous-phase chemical kinetics” with “irreversible reaction of the organic species with OH in the aqueous phase”

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