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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Technical note: Updated parameterization of the reactive uptake of glyoxal and methylglyoxal by atmospheric aerosols and cloud droplets" *by* Leah A. Curry et al.

Anonymous Referee #2

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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. 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. 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.

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.

Technical comments

While the reported parameter values seem reasonable, I was unable to find the values used for two key parameters: the accommodation coefficient (alpha) and the aqueous

diffusion coefficients. Are the terms that include these parameters not very influential on the overall values calculated for the uptake coefficients?

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.

Reference cited

1. Liggio, J.; Li, S.-M.; McLaren, R., Reactive uptake of glyoxal by particulate matter. J. Geophys. Res. 2005, 110, D10304. doi:10.1029/2004JD005113

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